QUANTUM AND CLASSICAL OPTICS OF DISPERSIVE AND ABSORPTIVE STRUCTURED MEDIA

by

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Abstract

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This thesis presents a Hamiltonian formulation of the electromagnetic fields in structured (inhomogeneous) media of arbitrary dimensionality, with arbitrary material dispersion and absorption consistent with causality. The method is based on an identification of the photonic component of the polariton modes of the system. Although the medium degrees of freedom are introduced in an oscillator model, only the macroscopic response of the medium appears in the derived eigenvalue equation for the polaritons. For both the discrete transparent-regime spectrum and the continuous absorptive-regime spectrum, standard codes for photonic modes in nonabsorptive systems can easily be leveraged to calculate polariton modes. Two applications of the theory are presented: pulse propagation and spontaneous parametric down-conversion (SPDC).

In the propagation study, the dynamics of the nonfluctuating part of a classical-like pulse are expressed in terms of a Schrödinger equation for a polariton effective field. The complex propagation parameters of that equation can be obtained from the same generalized dispersion surfaces typically used while neglecting absorption, without incurring additional computational complexity. As an example I characterize optical pulse propagation in an Au/MgF$_2$ metallodielectric stack, using the empirical response function, and elucidate the various roles of Bragg scattering, interband absorption and field expulsion. Further, I derive the Beer coefficient in causal structured media.

The SPDC calculation is rigorous, captures the full 3D physics, and properly incorporates linear dispersion. I obtain an expression for the down-converted state, quantify
pair-production properties, and characterize the scaling behavior of the SPDC energy. Dispersion affects the normalization of the polariton modes, and calculations of the down-conversion efficiency that neglect this can be off by 100% or more for common media regardless of geometry if the pump is near the band edge. Furthermore, I derive a 3D three-wave group velocity walkoff factor; due to the interplay of a topological property with a symmetry property, I show that even if down-conversion is into a narrow forward cone, neglect of the transverse walkoff can lead to an overestimate of the SPDC energy by orders of magnitude.
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Chapter 1

Introduction

Structured optical media have been the focus of an intense and sustained research effort over the last two decades, and much progress has been made without fully accounting for realistic material dispersion and absorption. However, recent research has increasingly involved materials for which dispersion and absorption cannot be neglected. Many proposed applications are fundamentally limited by dispersion and absorption, and others leverage these effects explicitly. Thus theoretical tools are needed that incorporate realistic material dispersion and absorption, and at the same time have an intuitive connection to the familiar tools used for nondispersive media. This thesis both presents and applies such a tool.

A “structured medium” is a composite of materials, each with individual optical properties understood in terms of a macroscopic response function, such that the spatial configuration on the scale of an optical wavelength or smaller gives rise to interesting behavior of electromagnetic fields in the system. Fig. 1.1 gives a schematic representation of some typical structured media. From left to right are an optical-frequency plasmonic nanocircuit, a double-negative-index metamaterial, a nanotip, coupled microspheres, coupled rib waveguides (above), a microring SCISSORS waveguide/resonator structure [5] (below), and a 3D photonic crystal. Others include waveguides, multilayer systems, photonic crystal fibers, high-Q torus resonators [6], coupled resonator optical waveguides [7], and plasmonic nanostructures [8][9].

Although the results of this thesis apply to structured media in general, it is illustrative to describe in more detail a common subset. Many systems of interest, including several listed above, have discrete translation symmetry of the macroscopic optical response. They are called photonic crystals, and as a result of this symmetry they support
modes of the Bloch form. These solutions to Maxwell’s equations are functions $B_{uk}(r)$ comprised of a plane wave with wave vector $k$ multiplied by a function $\phi_{uk}(r)$ that shares the discrete translation symmetry: $B_{uk}(r) = \phi_{uk}(r) \exp(i k \cdot r)$. A discrete set of modes labeled by $u$ is found for each wave vector $k$ in the reciprocal space unit cell, one symmetric choice of which is termed the first Brillouin zone (FBZ). The mode frequencies $\omega_{uk}$ comprise the dispersion relation, which is also referred to as the photonic band structure. Besides photonic crystals, for many structured media, the mode fields and photonic band structure provide a useful description of their optical properties.

In Fig. 1.2 is shown a sample photonic band structure and electromagnetic Bloch modes, calculated using a standard code [10]. The system is a 2D photonic crystal made of a periodic triangular array of air columns in a background material with a dielectric function, $\varepsilon_{\text{die}}$, that is 12 times the permittivity of free space, $\varepsilon_0$. The dispersion relation is plotted along high-symmetry lines of the FBZ in $k$-space. Each mode field diagram displays, for a particular mode, a temporal snapshot of the real part of the component of the electric mode field parallel to the air-column axis. An outline of the structure is overlaid on each of these mode field diagrams.

There are numerous methods for obtaining the modes and photonic band structure of structured media. Most often systems are nonmagnetic, and are taken to have a real, piecewise-constant, frequency-independent dielectric function $\varepsilon(r)$. The modes and dispersion relation are then typically obtained from the so-called Maxwell “master equation,” which is a Hermitian eigenvalue equation for the squared frequency $\omega^2_{uk}$ and the Bloch eigenmode $B_{uk}(r)$ for the magnetic field:

$$\nabla \times \left[ \frac{\varepsilon_0}{\varepsilon(r)} \nabla \times B_{uk}(r) \right] = \frac{\omega^2_{uk}}{c^2} B_{uk}(r). \quad (1.1)$$

This equation has been solved using many numerical techniques, and standard, freely available codes will solve it [10][11].
Figure 1.2: Photonic band structure and electromagnetic Bloch modes of a 2D photonic crystal of air columns in a dielectric background with $\varepsilon_{\text{die}}/\varepsilon_0 = 12$.

However, this use of a real, frequency-independent $\varepsilon (r)$ in Eq. (1.1) is approximate. As long as the material responds causally to applied fields, the dielectric function has to satisfy the Kramers-Kronig relations. When there is no singularity in the response function at zero frequency, these are written

\begin{align*}
\text{Re } \varepsilon (r, \omega) &= 1 + \frac{2}{\pi} P \int_0^{\infty} d\Omega \frac{\Omega \text{Im } \varepsilon (r, \Omega)}{\Omega^2 - \omega^2}, \\
\text{Im } \varepsilon (r, \omega) &= -\frac{2\omega}{\pi} P \int_0^{\infty} d\Omega \frac{\text{Re } \varepsilon (r, \Omega) - 1}{\Omega^2 - \omega^2},
\end{align*}

from which it is clear that real and imaginary parts of a causal response function are mutually dependent, and that one serves to specify the other. Now notice that if a system is asserted to have a frequency-independent dielectric function $\varepsilon (r)$, Eqs. (1.2, 1.3) imply that $\varepsilon (r) = 1$, so that the system must be vacuum.

Although neglect of the frequency dependence of $\varepsilon$ is the most common model for the dielectric function in the literature, it lies at one end of a spectrum of models in use, with the full empirically-determined $\varepsilon (r, \omega)$ lying at the other end. Indeed, a constant dielectric function $\varepsilon (r)$ can be a fair approximation if $\varepsilon (r)$ is taken to be real and all material absorption resonances are far above the frequencies of interest. Absorption is often modeled using a frequency-independent complex $\varepsilon (r)$ [12][13], and this can be adequate at a single frequency, but only approximate over a range of frequencies. One step better is a one-resonance model [14], for which we have $\text{Im } \varepsilon (\omega) = \frac{\pi L}{2\omega_0} \delta (\omega - \omega_0) + H (\omega)$, where $H (\omega)$ represents the contribution due to absorption at frequencies far from the
region of interest, and \( F \) is a constant; then the real part of the dielectric function is 
\[
\text{Re} \varepsilon(\omega) \simeq \varepsilon + F (\omega_0^2 - \omega^2)^{-1}
\] at points near (but not at) \( \omega_0 \), where \( \varepsilon \) is the contribution of distant absorption resonances. Next in increasing realism is a model involving a single absorption peak centered at \( \omega_0 \) but broadened in some way. The Drude model [15] for metals is such a model, with \( \omega_0 = 0 \) and with broadening due to a finite microscopic scattering time; so too is the polaritonic model [16], which has positive \( \omega_0 \). Most realistic is use of the full frequency-dependent dielectric function \( \varepsilon(\omega) \) [17][18], which is usually determined empirically [19].

Shown in Fig. 1.3 are response functions of two typical materials used in structured media: GaAs and Au. The GaAs response is based on an \textit{ab-initio} calculation by F. Nastos [20], and the Au response is a fit to empirical optical constants. GaAs appears here to be fairly nondispersive for low frequencies up to about 1 eV, but if the operating frequency is near the 1.42 eV electronic band gap then dispersion is significant. Also depicted is Au; there has been a lot of interest in metallodielectric (plasmonic) nanophotonic media recently [21], and it is a common constituent. Shown in Fig. 1.4 are both the exact empirical dielectric function \( \varepsilon(\omega) \) and the Drude model dielectric function \( \varepsilon_d(\omega) \). It is clear from Fig. 1.4 that metals are inherently absorptive and highly dispersive, so the approximation of frequency-independent \( \varepsilon \) does not hold at any frequency. Other materials in which dispersion and absorption are important in practice include media doped with atoms with excitation energies near photon energies of interest, exciton-polaritonic [22][23] and phonon-polaritonic photonic crystals [16]. However, most analyses presented to date have relied on simple, model dielectric response functions.

Some impetus for the increasing interest in dispersive media comes from the appli-
cations side, and in two distinct ways. First, many applications specifically leverage the absorptive and dispersive properties of host media. Examples include photovoltaic systems, ultrahigh-efficiency incandescent lighting systems [24][25][26], few-photon switches [27][28], low threshold microlasers [29], and infrared emission suppression [25].

For other proposed applications, absorption and dispersion are unavoidable effects that complicate an idealized system. Many of these proposals need to be evaluated and refined in light of realistic models of material absorption and dispersion. Some such proposed applications involve the classical and quantum states of light in high Q defect cavities [30][31][32][33]; others include subwavelength imaging [34] and cloaking devices [35][36]. It has also been shown through numerical [37] and analytical [38] investigations that photonic band gaps can be significantly altered or even closed by very small material absorption.

So for each of the above-mentioned structured media platforms and applications, a physical description is needed that models the material response more realistically. I additionally seek an underlying description in terms of a Hamiltonian formulation. While much work on dispersive media has been done without a Hamiltonian framework, such a framework is either advantageous or ultimately required for many problems, in that symmetries and conserved quantities are more easily identified, in that it is easier to introduce coupling to other systems, and in that quantization can be carried out following a canonical quantization procedure.

While the problem of quantization in nondispersive, nonabsorptive dielectrics has been well-studied since the early work of Jauch and Watson [39], it is significantly more complicated for dispersive and absorptive Kramers-Kronig dielectrics or metals. The reason is that any Hamiltonian, being a state functional, involves dynamical variables at a single given time; however, dispersive response in the medium implies the polarization involves the electromagnetic fields at all previous times, so any naïve attempt to introduce the medium polarization into the Hamiltonian is doomed to failure. The response function simply aggregates too much information about the underlying medium to allow such a Hamiltonian formalism. The solution is the reintroduction of dynamical variables representing the material degrees of freedom. This has been accomplished by various methods [40][41][42][43][44][45][46][47][48][49][50][51], each with its own strengths and limitations. Huttner and Barnett [43][52] introduced the medium as a Hopfield model [53], and were able to construct a Hamiltonian formulation for a uniform infinite dielectric with dispersive and absorptive response. In a Langevin method pioneered by Matloob
and co-workers, and furthered by Gruner and Welsch [54][55][44], the medium is introduced as phenomenological noise currents in the Maxwell equations, and the canonical operators of the theory are noise current operators. However, the electromagnetic fields are obtained only indirectly from the noise current operators via the Green function. A noise current method based on a Hopfield model has provided microscopic justification for this approach [51]. More recently, Tip [45][56][46][47] introduced an Auxiliary Field method, in which two fields $F_2$ and $F_4$ are coupled to the usual electromagnetic fields of the Maxwell equations; the drawback is that the coupling and free-field dynamics of the auxiliary fields are postulated *ad hoc*, so intuition into the resulting physics is more difficult. Both the Langevin and Auxiliary Field methods have been applied to inhomogeneous media [45]. The Huttner-Barnett approach has been applied to quantization of a dielectric half-space [57]. It has been suggested that the scheme would become quite cumbersome to apply to nonuniform media [58]; an early attempt [59] is consistent with that suggestion.

However, as a core result of this thesis in Chapter 2 I present just such a scheme, and demonstrate an approach with an end result that is both elegant and easy-to-use. Instead of relying on a Green function representation of light in the system, I focus directly on the dispersion relation (or photonic band structure) and electromagnetic modes described in Eq. (1.1). Since the majority of studies of structured media are formulated in terms of mode or band structure concepts, the approach here connects more readily with that literature than do others.

The approach is as follows. The medium response is introduced via a Hopfield-type model [53] as an oscillator field, which is well-justified in the linear excitation regime [60]. The full dispersive and absorptive dielectric response of the system, $\chi^{(1)}(\mathbf{r}, t)$, is generated implicitly by the dynamics of the coupled electromagnetic and model medium fields. The dielectric function is easily understood in terms of the model, with dispersion being due to virtual coupling and absorption due to direct coupling. This system is Hamiltonian, and is quantized in a straightforward manner. The polaritons, which are the dressed modes of the system, are obtained through a Fano-type diagonalization procedure and have a natural interpretation. The theory is applicable to inhomogeneous media in general, and I formulate it in terms of a ‘nominal’ nondispersive and nonabsorptive system with index of refraction $n(\mathbf{r})$. While this intermediate step is not necessary—the nominal system can always be taken to be vacuum—it should make this approach particularly useful in the design and analysis of dispersive and absorptive structured media for which the
practitioner has good intuition for the mode structure of the nominal system.

In fact, since the method is based on describing the dynamics in terms of modes, it has practical applicability to any structured medium that is best understood in the nondispersive, nonabsorptive limit in terms of modes.

Having obtained a Hamiltonian formulation of the electromagnetic fields in dispersive and absorptive structured media, in Chapter 3 of the thesis I formulate the propagation of electromagnetic fields in such media. Of particular interest for propagation are nanoplasmonic structures, which can have high nonlinearities and long-range propagation modes [61], but cannot be understood even qualitatively without accounting for absorption and dispersion. Various propagation problems in quantum optics also specifically involve absorptive and dispersive materials. An important class involves few-photon wave packets and generation and propagation of entangled photons [62][63][64][65]. For example, novel propagation effects such as photon-exchange interactions [66] and entangled surface plasmons [67] require absorptive media. It has been argued that a better understanding of the dynamics of few-photon wavepackets in dispersive media [68] is essential to advances in certain practical quantum information systems. A better understanding of the role of material dispersion can also give stronger theoretical input to the optimization of device designs, where improvement in yield has been a priority [69][70].

In the literature, much work on propagation in dispersive and absorptive media has used a phenomenological description of dispersion and absorption usually based on the models described after Eq. (1.3). Sometimes dispersion is included into a Hamiltonian via the Brillouin formula [71][72][73][74], but absorption is nonrigorously treated as being small enough for perturbative approaches to work. However, for many materials and operation regimes of interest, such descriptions are inadequate, and it is necessary to incorporate the full empirically determined response function $\varepsilon (r, \omega)$. While this has been done without a Hamiltonian framework in some work on propagation [17][18], such a framework is ultimately required for many problems.

Besides the inclusion of material absorption and dispersion, a successful treatment of propagation in nanophotonic media must handle a wide array of considerations: high index contrast, fully vectorial fields and divergence conditions, modal dispersion and diffraction, modal absorption effects, nonlinear response, and pulse interactions. Envelope function approaches [2] become arduous beyond second order modal dispersion and diffraction, and have included material dispersion and absorption only at the perturbative level. A photon effective field formulation presented in previous work [3][75] easily
handles modal dispersion and diffraction to higher orders, but not material dispersion or absorption.

I overcome these difficulties, and provide a Hamiltonian approach to propagation in structured media with realistic linear absorption and dispersion. As is done in the formulation of the fields in Ch. 2 of this thesis, the approach in Ch. 3 emphasizes the photonic component of the polariton modes of the system, and is applicable to problems in both classical and quantum optics. Modal and material dispersion, absorption, and diffraction are captured to arbitrary order as coefficients in a Schrödinger equation satisfied by an effective field. In the absorptive regime, the fluctuating part of the field is separated out into a mode expansion over polariton operators. As an example, for a dielectric-metal multilayer stack I give a full description of pulse propagation using the method. I also derive an expression for the Beer coefficient in absorptive and dispersive structured media.

In Ch. 4 I turn to a specific quantum-optical propagation problem: spontaneous parametric down-conversion (SPDC). SPDC is the spontaneous conversion of a photon into two photons in a material with second-order nonlinearity. It is an important method of generating entangled states for quantum optics applications such as quantum communication. However, the generation efficiencies currently achieved in practice are low, around $10^{-9}$ [76]. A promising strategy for improving this is the use of structured media as a generation platform, a strategy which has been very effective for other second-order nonlinear processes such as second harmonic generation (SHG), and has led to $\mathcal{O}(1)$ conversion efficiencies in experiments. Work has begun along these lines for SPDC in a wide variety of structured media [65][77][70][69][78][79][80].

As a stepping stone to a comprehensive theory for structured media, one would like to have a comprehensive theory for SPDC in a uniform medium. Current treatments of SPDC in bulk crystals differ, but usually focus on collinear or degenerate phase matching. Even when the down converted fields are represented in 3D, the group velocity walkoff is 1D [81][82][83], mode polarizations are not fully considered, and either fixed propagation directions of the down-converted fields are ultimately assumed [82], or approximations are tailored to specific directions of propagation [81]. Furthermore, the absolute magnitude of the down-conversion efficiency in a nonlinear crystal has not been calculated. However, in order to design devices with improved efficiency, it is useful to have the absolute magnitude of the down-conversion rate over the full spectrum of down-conversion channels. If devices are to operate at efficiencies beyond the undepleted pump approximation,
designers must consider both how to enhance down-conversion into desired channels and suppress emission into other channels. It is also useful to understand the scaling behavior of the down-conversion rate with respect to various experimental parameters, and to obtain SPDC results for a finite duration pump pulse as well as for CW. To date, for a finite energy pump pulse detailed calculations have been done in a 1D model [84], but not in 3D.

An improved theoretical treatment is challenging because of various features of SPDC. For one, SPDC is inherently broadband. Unlike other nonlinear processes like SHG or sum-frequency generation (SFG), in which an input beam either interacts with itself or with another input beam so that only a single pair (SHG) or triad (SFG) of wave vectors is typically involved, in SPDC a single input beam interacts with the broadband spectrum of vacuum fluctuations. As a result, down-conversion pairs are generated into a wide spectrum of channels, over a broad range of frequencies and directions, and this full spectrum has to be considered.

A rigorous calculation of SPDC is also complicated by general features of quantum optics in dispersive media, which are discussed in Ch. 4 of this work. Of particular importance, for optical excitations in any medium with causal response, the normalization condition for the mode fields depends on the material dispersion [1], as is shown in Ch. 2. Since this normalization condition is involved in the mapping from the energy of a quantum fluctuation to the amplitude of the fluctuation polarization field, the issue will arise in all effects in nonlinear quantum optics in which there is significant material dispersion at frequencies of interest. I emphasize that this normalization effect is even present in the CW limit of a pulse, in which it naively seems that only one frequency is accessed and therefore the variation in index with frequency seems not to be involved.

To address these issues, in Ch. 4 I present a calculation of SPDC with full accounting of the generation in 3D, and careful accounting of polarization effects and especially of the effect of material dispersion. The calculation is done in first order perturbation theory, and the dispersion of the nonlinear response is neglected for simplicity. For the sake of specificity I specialize here to type-I phase matching in a $\beta$-BaB$_2$O$_4$ (BBO) crystal, and note that the same approach can also be applied to type-II phase matching. And since the approach uses a method that leverages arbitrary linear mode structure of the nonlinear system, it can additionally be applied to address structured media.

Although the work in Ch. 4 addresses a purely quantum mechanical problem, possible applications of the work presented in Ch. 2–3 lie both in the classical and quantum
domains. To this end, I have written everything in those chapters so as to apply to both. I formally write commutators instead of Poisson brackets, but keep in mind the correspondence \((\hbar i)^{-1}[[..., ...]] \rightarrow \{..., ...\}\), which yields the associated classical Poisson brackets and classical equations. Similarly, I use \(\dagger\) to denote the adjoint or complex conjugate. Since no commutators appear in any of the final equations, the results are equally applicable to the classical and quantum domains, requiring only that the variables be interpreted appropriately as amplitudes or Heisenberg operators.

The thesis is organized so that Ch. 2 presents the underlying theoretical framework, and Ch. 3–4 present applications. Ch. 2 is structured as follows. In Section 2.1 is presented my canonical Hamiltonian model for dispersive and absorptive dielectrics in terms of electromagnetic fields coupled to model oscillator fields. In Section 2.2 I describe the dielectric response of the model. In Section 2.3 I first diagonalize the Hamiltonian for the dispersive and absorptive system, obtaining a generalized Hermitian eigenvalue problem for the polaritons, which exist at all frequencies in the absorption band. Subsequently, I perform the diagonalization for the nonabsorptive but dispersive frequency regime, in which there is a discrete spectrum of polaritons; these polaritons consist of both an electromagnetic component, which is just the usual dispersive electromagnetic mode, and a medium component, which is slaved to the electromagnetic component. In Section 2.4 I give the equations for systems with continuously labeled modes, choosing photonic crystals as a specific example. Section 2.5 gives a summary and user’s guide, which provides an overview of how to apply the results in this chapter to straightforwardly calculate polariton modes within a Hamiltonian framework for systems of interest; examples are presented for both the transparent and absorptive frequency regimes. For a sense of what is accomplished in Ch. 2, and of the nature of the formalism I produce, the reader may wish to scan Section 2.5 before reading the rest of the chapter. Section 2.6 gives a discussion of the main results of the chapter. Appendices B–C give technical results and protracted calculations which would otherwise impede the flow of the text. Given the number of symbols used in Ch. 2–4, in Appendix A I provide a tabulation of the symbols used in the thesis.

Chapter 3 is structured as follows. In Sec. 3.1 I both summarize the points from Ch. 2 that are needed for Ch. 3, and extend the formulation by introducing a classical source polarization. In Sec. 3.2 I describe a process for formally driving the field to launch a state of physical interest. In Sec. 3.3 I treat propagation in the transparent (but dispersive) regime. In Sec. 3.4 I treat propagation in the absorptive regime. In Sec.
3.5 I show how to use the theory to calculate propagation parameters of a dispersive and absorptive periodic multilayer stack. In Sec. 3.6 I apply the theory to propagation in a periodic metallodielectric stack of current interest, with dispersion and absorption based on the empirical response function. In Sec. 3.7 I derive an expression for the Beer’s coefficient, and apply the results to the metallodielectric stack. In Sec. 3.8 I discuss the main results of the chapter. Finally, Appendices D–H give supporting results which may be of most interest on subsequent readings.

Chapter 4 is structured as follows. In Sec. 4.1, the Hamiltonian and mode fields are introduced. Sec. 4.2 introduces the asymptotic states I use to describe the incident and output states, and also introduces the interaction picture. Sec. 4.3 describes the experimental system of interest. In Sec. 4.4 I derive an expression for the generated bipolariton state. In Sec. 4.5 I calculate properties of this state including the directional distribution and the SPDC energy. In Sec. 4.6 I characterize the scaling behavior of the SPDC energy. Sec. 4.7 gives numerical examples. Finally Sec. 4.8 summarizes the contributions of the chapter, and Appendix I provides a technical detail.

Chapter 2

Hamiltonian formulation of the electromagnetic field in dispersive and absorptive structured media

In this chapter I introduce the formalism that underpins the thesis. It is a Hamiltonian formulation of electromagnetic fields in dispersive and absorptive structured media of arbitrary dimensionality. The Kramers-Kronig relations are satisfied by construction. The method is based on an identification of the photonic component of the polariton modes of the system. Although the medium degrees of freedom are introduced in an oscillator model, only the susceptibility of the medium appears in the derived eigenvalue equation for the polaritons. A discrete polariton spectrum is obtained in the transparent regime below the absorption cutoff frequency, and the normalization condition contains the material dispersion in a simple way. In the absorptive regime, a continuous polariton spectrum is obtained. The expressions for the full electromagnetic field of the system can be written in terms of the modes of a limiting, nondispersive, nonabsorptive system, so the theory is well-suited to studying the effect of dispersion or absorption on photonic dispersion relations and mode structure. Available codes for dispersive photonic modes can easily be leveraged to obtain polariton modes in both the transparent and absorptive regimes.
2.1 Hamiltonian for dispersive and absorptive media

Consider Maxwell’s curl equations for dielectric media. In the absence of free charges and currents, these are

\[ \dot{D} = \nabla \times H, \quad \dot{B} = -\nabla \times E. \quad (2.1) \]

The divergence equations,

\[ \nabla \cdot D = 0, \quad \nabla \cdot B = 0, \quad (2.2) \]

are taken as initial conditions; if the initial fields satisfy (2.2), then evolution according to (2.1) ensures the fields will satisfy (2.2) for all time. However, it is important to note that approximations of (2.1) often do not maintain the conditions (2.2) upon evolution. To ensure the divergencelessness of \( D \) and \( B \) even after making approximations, we take \( D \) and \( B \) to be the primary fields, and consider \( E \) and \( H \) as derived fields. This makes it possible to restrict the space of \( D \) and \( B \) fields to those that satisfy (2.2). In practice this can be simply done, for example, by using a mode expansion in divergenceless fields.

The choice of \( D \) and \( B \) as fundamental fields has its roots in the work of Born and Infeld \[85\]; Bialynicki-Birula and Bialynicka-Birula discuss it in some detail in their text \[86\].

The Maxwell equations require constitutive relations. Neglecting magnetic effects we have

\[ B (r, t) = \mu_0 H (r, t), \quad (2.3) \]
\[ D (r, t) = \varepsilon_0 E (r, t) + P (r, t), \]

where the dielectric response is typically specified by a relation between the polarization and the electric field. However, since we take \( D \) and \( B \) as the primary fields, we write \( P \) in terms of (in general a functional of) \( D \). There is clearly no loss of generality here, since any relation of the first type can be converted into a relation of the second type by the use of the second of (2.3).

Sometimes dispersive and absorptive effects are negligible, and over the frequency range of interest the optical response can be characterized by a nondispersive, nonabsorptive, response of the form

\[ P^i (r, t) = \Gamma^{ij}_0 (r) D^j (r, t), \quad (2.4) \]

which nonetheless allows for material inhomogeneities. Superscript Roman indices indicate Cartesian components, which are to be summed over when repeated. This sort of
description, with $\Gamma^{ij}_0(r) = \Gamma^{ji}_0(r)$ real, follows from considering the response of a nonmagnetic system to the electromagnetic fields in the electric dipole (long-wavelength) limit and at frequencies far below any material resonances. In the special case of isotropic media, the response function $\Gamma^{ij}_0(r)$ is related to the usual (local) index of refraction $n(r)$ by

$$\Gamma^{ij}_0(r) = \left(1 - \frac{1}{n^2(r)}\right) \delta_{ij}, \quad (2.5)$$

where $\delta_{ij}$ is the Kronecker delta. Equation (2.5) can be confirmed by using (2.3) and (2.4) with (2.5) in (2.1), and comparing with the usual equations in this description of the optical response.

To include the effects of absorption and dispersion we need a treatment of the medium response more general than (2.4). The goal here is to model a very general linear response of the form

$$P^i(r, t) = \int ds \Gamma^{ij}(r, t - s) D^j(r, s), \quad (2.6)$$

where the constraint of causality requires that $\Gamma^{ij}(r, \tau) = 0$ for all $\tau < 0$. The response (2.6) is still of course local in space (i.e., we neglect spatial dispersion effects [87]), and we neglect any magnetic aspects of the response by assuming

$$\Gamma^{ij}(r, t) = \Gamma^{ji}(r, t). \quad (2.7)$$

Other than these constraints, we allow for a $\Gamma^{ij}(r, t)$ that is freely specified, either from experimental data or on the basis of a microscopic calculation of the material response. It is the dynamics described by the system of equations (2.1, 2.2, 2.3, 2.6, 2.7) that we wish to generate from a Hamiltonian.

In many treatments, complications arise because the route to canonical formulation begins with a Lagrangian. However, this is not necessary. Even in the canonical formulation of a classical system for ultimate use in quantization, one is only required to provide a set of commutators (or Poisson brackets) and a Hamiltonian such that their use leads in the usual way to the desired dynamical equations, and such that the numerical value of the Hamiltonian in the classical theory is equal to the energy [88]. Earlier we introduced such an approach for nondispersive and nonabsorptive media [3].

However, the kind of very general response (2.6) we consider here requires for its description the inclusion of medium degrees of freedom. We take as our Hamiltonian

$$H = H_{em} + H_{med} + H_{int}, \quad (2.8)$$
where $H_{em}$ is the Hamiltonian of the electromagnetic field in vacuum, $H_{med}$ is the Hamiltonian of the medium, and $H_{int}$ describes the interaction between the vacuum electromagnetic field and the medium. One route to determining $H_{med}$ and $H_{int}$ would be to rely on a microscopic calculation of the actual material medium of interest. My approach here adopts a different philosophy. For a $\Gamma^{ij}(r,t)$ assumed given, I construct effective Hamiltonians $H_{med}$ and $H_{int}$ that lead to the response (2.6) and are convenient for later calculations. Thus I provide formal Hamiltonians $H_{med}$ and $H_{int}$, rather than the actual ones.

2.1.1 Vacuum electromagnetic Hamiltonian

For the vacuum electromagnetic Hamiltonian we take

$$H_{em} = \frac{1}{2\mu_0} \int dr \mathbf{B}(r) \cdot \mathbf{B}(r) + \frac{1}{2\varepsilon_0} \int dr \mathbf{D}(r) \cdot \mathbf{D}(r). \tag{2.9}$$

In conjunction with the equal-time commutators (or alternatively Poisson brackets)

$$[D^i(r), D^j(r')] = [B^i(r), B^j(r')] = 0, \tag{2.10}$$
$$[D^i(r), B^j(r')] = i\hbar \varepsilon^{ilj} \frac{\partial}{\partial r_l}(\delta(r - r')),$$

where $\varepsilon^{ilj}$ is the Levi-Civita symbol ($\varepsilon^{123} = -\varepsilon^{213} = 1$, etc.), Hamilton’s equations

$$i\hbar \dot{\mathbf{D}} = [\mathbf{D}, H], \tag{2.11}$$
$$i\hbar \dot{\mathbf{B}} = [\mathbf{B}, H],$$

with $H = H_{em}$ yield the Maxwell equations (2.1) subject to the constitutive relations (2.3), with $P^i(r,t) = 0$, i.e., the dynamics for the electromagnetic field in vacuum.

2.1.2 Medium and coupling Hamiltonians

We introduce dynamical variables for the medium, and a medium Hamiltonian, in such a way that the addition of coupling to the electromagnetic field results in equations that lead to the desired dispersive and absorptive response. As many workers have realized, harmonic oscillator fields are appropriate for the description of linear excitations of a medium [53][60]. Following the Hopfield model [53], Huttner and Barnett [43] considered optical absorption as resulting from coupling of the electromagnetic field to a discrete oscillator field, with the discrete oscillator field coupled to continuum reservoir fields.
Diagonalization of their medium Hamiltonian alone yields a set of dressed continuum fields that are coupled to the vacuum electromagnetic field. Thus there is a mathematical equivalence between the Hopfield model and the modified model excluding the discrete oscillator as an intermediary between the vacuum electromagnetic and reservoir fields. We are more concerned here with developing a general approach for use in problems in optics, rather than describing any purported model of actual material response by a discrete oscillator field, or the details of the two-step diagonalization that was the focus of earlier workers. Thus we take a simpler model, involving the direct coupling of vacuum electromagnetic modes to a continuum of oscillator fields. It should be emphasized that while the two-step coupling is often adopted, it is certainly not needed to model the response of physical systems. Given the dielectric response function $\Gamma^{ij}(r,t)$ (see e.g. (2.6)), it is possible to immediately produce a model medium of my type that generates it. This is detailed in Section 2.2.

To describe a large class of media, we require medium harmonic oscillators at (possibly) all frequencies. Since we neglect spatial dispersion effects, the harmonic oscillator modes can be identified with individual points in space. In our approach, the coupling between a material excitation at $r$ and the electromagnetic field is encapsulated in a position- and frequency-dependent coupling tensor $\Lambda^{ij}(r,\Omega)$, which is real and satisfies $\Lambda^{ij}(r,\Omega) = \Lambda^{ji}(r,\Omega)$. Furthermore, at each $(r,\Omega)$, the coupling tensor $\Lambda^{ij}(r,\Omega)$ we introduce shares the same set of principal axes as the frequency-domain dielectric response tensor $\Gamma^{ij}(r,\Omega)$. Any material dielectric response can be described by the oscillator model introduced here, with a flat density of oscillator states above a minimum cutoff frequency $\Omega_g$, which may be zero; effects of the density of states in the actual material system can be described through the frequency dependence of the effective coupling constant $\Lambda^{ij}(r,\Omega)$, as is shown in Section 2.2.

We describe the dynamics of our model medium by a coordinate field $X_{\Omega}(r,t)$ and its conjugate momentum field $\Pi_{\Omega}(r,t)$. To avoid introducing superfluous components, two steps are required. For a large class of structured media at frequencies of interest, there is absorption at some positions but not at others; to avoid introducing uncoupled oscillators, we define a (possibly multiply connected) region of space $\mathcal{V}_\Omega$ that identifies the region over which the medium has nonzero absorption at frequency $\Omega$. Then oscillators are only introduced for $(r,\Omega)$ such that $r \in \mathcal{V}_\Omega$.

Furthermore, even in regions with absorption, all three vector components of an oscillator are not necessarily required, because at any given position $r$ the dielectric
response $\Gamma^{ij}(\mathbf{r}, \Omega)$—and therefore the real, symmetric coupling tensor $\Lambda^{ij}(\mathbf{r}, \Omega)$—may have one or more vanishing eigenvalues at a given frequency $\Omega$. For each position $\mathbf{r}$ and frequency $\Omega$ we determine the local principal axes \( \hat{\zeta}_1(\mathbf{r}, \Omega), \hat{\zeta}_2(\mathbf{r}, \Omega), \hat{\zeta}_3(\mathbf{r}, \Omega) \) of the tensor $\Gamma^{ij}(\mathbf{r}, \Omega)$, and then write the coupling tensor $\Lambda^{ij}(\mathbf{r}, \Omega)$ in that position- and frequency-dependent basis. Throughout the chapter, we use $\lambda$ and $\lambda'$ to refer to the components in the basis of the local principal axes, reserving $i$ and $j$ to refer to Cartesian components. Then

$$\Lambda^{\lambda\lambda'}(\mathbf{r}, \Omega) = \Lambda^{\lambda\lambda'}(\mathbf{r}, \Omega) \delta^{\lambda\lambda'} \equiv \Lambda^\lambda(\mathbf{r}, \Omega) \delta^{\lambda\lambda'},$$  \hspace{1cm} (2.12)

where here no summation is taken over repeated indices. The medium fields are then introduced in reference to the local principal axes. For all $\lambda$, if $\Lambda^\lambda(\mathbf{r}, \Omega) \neq 0$, we introduce a local oscillator polarized along $\hat{\zeta}_\lambda(\mathbf{r}, \Omega)$, and write the associated coordinate field component as $X^\lambda(\mathbf{r}, t)$. Of course, the tensors written in the basis of the local principal axes can be rewritten in a global Cartesian basis via an appropriate transformation at each $(\mathbf{r}, \Omega)$, and vice versa. Throughout the chapter, we simply write $X(\mathbf{r}, t)$, and recall that $X(\mathbf{r}, t)$ has 0, 1, 2, or 3 nonvanishing components as required; we revert to the more detailed notation when necessary. Similarly, we write the coupling tensor as $\Lambda(\mathbf{r}, \Omega)$. For notational simplicity, we also typically refrain from indicating the time-dependence of the medium fields, and instead write $X(\mathbf{r})$ and $\Pi(\mathbf{r})$.

The Hamiltonian of the model medium is then

$$H_{med} = \int_{\Omega_\gamma} d\Omega \int_{V_{\Omega}} dr \frac{1}{2} (\Pi(\mathbf{r}) \cdot \Pi(\mathbf{r}) + \Omega^2 X^2(\mathbf{r}) \cdot X(\mathbf{r})),$$ \hspace{1cm} (2.13)

which yields harmonic oscillator field dynamics when we impose the equal-time commutator

$$\left[ X^\lambda(\mathbf{r}), \Pi^{\lambda'}(\mathbf{r}') \right] = i\hbar \delta_{\lambda\lambda'} \delta(\mathbf{r} - \mathbf{r}') \delta(\Omega - \Omega'),$$ \hspace{1cm} (2.14)

with all other commutators (Poisson brackets) vanishing. The association of each medium oscillator with a point in space is apparent in the Hamiltonian (2.13); were there no coupling with the electromagnetic field, the dynamics of oscillators at different field points $\mathbf{r}$ would be completely independent of each other. Making the change of variables

$$\psi(\mathbf{r}) = \sqrt{\frac{1}{2\hbar\Omega}} (\Omega X(\mathbf{r}) + i\Pi(\mathbf{r})),$$ \hspace{1cm} (2.15)

and neglecting the zero point energy in the quantum case, we obtain the form

$$H_{med} = \int_{\Omega_\gamma} d\Omega \int_{V_{\Omega}} dr \hbar \Omega \psi^\dagger(\mathbf{r}) \cdot \psi(\mathbf{r}),$$ \hspace{1cm} (2.16)
Chapter 2. Hamiltonian formulation

and the equal-time commutation relation

\[ \left[ \psi^\lambda_\Omega (r), \psi^{1\lambda'}_{\Omega'} (r') \right] = \delta_{\lambda\lambda'} \delta (r - r') \delta (\Omega - \Omega') , \quad (2.17) \]

with all other equal-time commutators vanishing.

We now turn to the coupling Hamiltonian, which we take to be

\[ H_{\text{int}} = -\frac{1}{\sqrt{\varepsilon_0}} \int_\Omega d\Omega \int_{\Omega} dr D(r) \cdot \Lambda (r, \Omega) \cdot X_\Omega (r) . \quad (2.18) \]

Using this Hamiltonian with \( H_{\text{em}} \) (2.9) and \( H_{\text{med}} \) (2.16) to form the total Hamiltonian \( H \) (2.8), Hamilton’s equations (2.11), evaluated with the commutators (2.10), yield the Maxwell equations (2.1) subject to the constitutive relations (2.3), with

\[ P(r, t) = \sqrt{\varepsilon_0} \int_\Omega d\Omega \Lambda (r, \Omega) \cdot X_\Omega (r, t) . \quad (2.19) \]

Thus we identify the polarization due to the medium fields. Whereas the dispersive constitutive relation involves the displacement fields at all previous times, (2.19) involves the medium fields only at a single time, as is required to build a Hamiltonian formulation.

2.2 Dielectric response of the model

Nonetheless, we can now identify the dielectric response function \( \Gamma^{ij} (r, t) \) generated by this model. To do this we use Hamilton’s equation with the full Hamiltonian (2.8), together with the medium field commutation relations (2.14), to find the dynamical equations for the medium fields:

\[ \dot{X}_\Omega (r, t) = \Pi_\Omega (r, t) , \quad (2.20) \]

\[ \dot{\Pi}_\Omega (r, t) = -\Omega^2 X_\Omega (r, t) + \varepsilon_0^{-1/2} \Lambda (r, \Omega) \cdot D(r, t) . \quad (2.21) \]

Combining these equations yields a second order equation for \( X_\Omega (r, t) \), driven by the displacement field. To allow for Fourier decomposition even on resonance, we allow the free oscillation of the medium fields to be infinitesimally damped in the usual way \[89\], and with the understanding that at the end of the calculation we take \( \eta \to 0_+ \), we modify the second order equation resulting from (2.20, 2.21) to read

\[ (\partial_t^2 + 2\eta \partial_t + (\Omega^2 + \eta^2)) X_\Omega (r, t) = \varepsilon_0^{-1/2} \Lambda (r, \Omega) \cdot D(r, t) . \quad (2.22) \]
Then inverting the differential operator on the left-hand side \([90]\) yields the solution

\[
X_\Omega (r, t) = \frac{1}{\sqrt{\varepsilon_0}} \int ds \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega(t-s)} \frac{\Lambda (r, \Omega) \cdot D (r, s)}{-\omega^2 - 2i\omega\eta + \Omega^2 + \eta^2} + (X_\Omega^0 (r) e^{-i\Omega t} + c.c.),
\]

(2.23)

where the homogeneous solution of (2.20, 2.21) involving the time independent \(X_\Omega^0 (r)\) describes the free (nondriven) oscillation of the medium fields, which we note has 0, 1, 2, or 3 nonvanishing components, as per the discussion concerning \(X_\Omega (r)\) after (2.12). Substitution of (2.23) into the expression (2.19) for the polarization in terms of the medium fields gives

\[
P (r, t) = \int ds \Gamma (r, t-s) \cdot D (r, s) + \int_{\Omega_g}^{\infty} d\Omega \sqrt{\varepsilon_0} \Lambda (r, \Omega) \cdot (X_\Omega^0 (r) e^{-i\Omega t} + c.c.),
\]

(2.24)

where we write the dielectric response of the model as

\[
\Gamma (r, t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma (r, \omega) e^{-i\omega t},
\]

with

\[
\Gamma (r, \omega) = \int_{\Omega_g}^{\infty} d\Omega \frac{\Lambda (r, \Omega) \cdot \Lambda (r, \Omega)}{(\Omega - \omega - i\eta)(\Omega + \omega + i\eta)}.
\]

(2.25)

The second term on the right-hand side of (2.24) is a fluctuation term familiar from reservoir theory \([91]\), and is associated with the fluctuation-dissipation theorem. Note that our model satisfies (2.7), as is expected in the absence of magnetic response effects. We have

\[
\Gamma (r, \omega)^* = \Gamma (r, -\omega),
\]

(2.26)

and it is clear from the form of our expression (2.25) for \(\Gamma (r, \omega)\) that the dielectric response is purely causal, since the poles of \(\Gamma (r, \omega)\) are at \(\omega = \pm \Omega - i\eta\) for all \(\Omega \in [\Omega_g, \infty)\), and are all in the lower half plane. Then using the well-known identity

\[
\lim_{\eta \to 0^+} \frac{1}{x - x_0 \pm i\eta} = \frac{P}{x - x_0} \mp i\pi \delta (x - x_0),
\]

(2.27)

and identifying real and imaginary parts of \(\Gamma (r, \omega)\), we find, for \(\omega > 0\),

\[
\text{Re} \Gamma (r, \omega) = P \int_{\Omega_g}^{\infty} d\Omega \frac{\Lambda (r, \Omega) \cdot \Lambda (r, \Omega)}{(\Omega - \omega)(\Omega + \omega)}
\]

(2.28)

\[
\text{Im} \Gamma^{\lambda \lambda'} (r, \omega) = \pi \frac{\Lambda^{\lambda \lambda'} (r, \omega) \Lambda^{\lambda' \lambda'} (r, \omega)}{2\omega}.
\]

(2.29)

For \(\omega < 0\), \(\Gamma^{\lambda \lambda'} (r, \omega)\) can be determined using (2.26).
A set of Kramers-Kronig relations follow from (2.25), and can be written in the form

\[
\text{Re} \Gamma (r, \omega) = \frac{2}{\pi} P \int_{\Omega_g}^{\infty} d\Omega \frac{\Omega \text{Im} \Gamma (r, \Omega)}{(\omega - \Omega)(\omega + \Omega)}, \tag{2.30}
\]

\[
\text{Im} \Gamma (r, \omega) = \frac{2}{\pi} P \int_{\Omega_g}^{\infty} d\Omega \frac{\Omega \text{Re} \Gamma (r, \Omega)}{(\omega - \Omega)(\omega + \Omega)}. \tag{2.31}
\]

It is now apparent how any specified causal \(\Gamma^{ij} (r, t)\) can be described by our model. From the Fourier transform of \(\Gamma^{ij} (r, t)\) expressed in the basis of its local principal axes, and recalling that the \(\Lambda^{\lambda\lambda'} (r, \omega)\) are chosen to share the same set of principal axes as \(\Gamma^{\lambda\lambda'} (r, \omega)\), we use (2.29) to identify a set of coupling constants \(\Lambda^{\lambda\lambda'} (r, \omega)\). With \(\text{Im} \Gamma (r, \omega)\) correctly described, causality guarantees that the corresponding \(\text{Re} \Gamma (r, \omega)\) follows from (2.30).

Finally, we note that if a medium’s optical response function is given in the more familiar form of a causal \(\varepsilon (r, \omega)\) satisfying the Kramers-Kronig relations (1.2)–(1.3), then the corresponding \(\Gamma (r, \omega)\) is also guaranteed to be causal, as long as the imaginary part of \(\varepsilon (r, \omega)\) is nonvanishing for finite \(\omega > \Omega_g\). This result follows from the property that such a causal \(\varepsilon (r, \omega)\) has no zeros in the upper half-plane [92], and from the expression for \(\Gamma (r, \omega)\) in terms of \(\varepsilon (r, \omega)\), which is given by

\[
\Gamma^{\lambda\lambda'} (r, \omega) = \left(1 - \frac{\varepsilon_0}{\varepsilon^{\lambda\lambda'} (r, \omega)}\right) \delta_{\lambda\lambda'}. \tag{2.32}
\]

### 2.3 Polariton modes

#### 2.3.1 Nominal system

We now identify a useful decomposition of the total Hamiltonian. If dispersive and absorptive effects are small, the simple model (2.4, 2.5) can often provide a good starting point. Whether or not this is the case, we introduce a nominal index profile \(n (r)\) and write the full Hamiltonian (2.8) as

\[
H = H_0 + H_\Delta + H_{\text{med}} + H_{\text{int}}, \tag{2.33}
\]

where we have decomposed the vacuum electromagnetic field Hamiltonian as \(H_{\text{em}} = H_0 + H_\Delta\), with

\[
H_0 \equiv \frac{1}{2\mu_0} \int d\mathbf{r} \mathbf{B} (\mathbf{r}) \cdot \mathbf{B} (\mathbf{r}) + \frac{1}{2\varepsilon_0} \int d\mathbf{r} \frac{\mathbf{D} (\mathbf{r}) \cdot \mathbf{D} (\mathbf{r})}{n^2 (\mathbf{r})}, \tag{2.34}
\]
Chapter 2. Hamiltonian formulation

\[ H_\Delta \equiv \frac{1}{2\varepsilon_0} \int dr \ D^i (r) \Gamma^{ij}_0 (r) \ D^j (r). \] (2.35)

Hamilton’s equations using \( H_0 \) alone as the Hamiltonian, in conjunction with the commutators (2.10), yield the Maxwell equations (2.1) with the constitutive relation

\[ \mathbf{D} (r, t) = \varepsilon_0 n^2 (r) \mathbf{E} (r, t), \] (2.36)

which identifies \( H_0 \) as the Hamiltonian for a nominal, nonabsorptive and nondispersive system with refractive index \( n (\mathbf{r}) \). For the nominal system, \( H_0 \) is numerically equal to the total energy in the electromagnetic fields in the presence of the dielectric [3]. The “counter-term” \( H_\Delta \) in the Hamiltonian \( H \) then guarantees that (2.33) provides the correct sum (2.8).

We emphasize that we do not rely on perturbative results in \((H - H_0)\); in general we exhibit equations whose solutions describe the dynamics that follow from the full \( H \). Yet we see below that having a reference, “nominal system” is useful both in understanding the physics of those exact solutions, and in constructing them. This is in part because there are numerous freely available codes and algorithms for calculating mode fields and dispersion relations of nondispersive, nonabsorptive artificially structured media [93][10][11][94]. Of course, it is always possible to set \( n (\mathbf{r}) = 1 \), in which case the vacuum itself serves as the nominal system, and \( H_\Delta = 0 \).

2.3.2 Nominal system modes

To construct the polariton modes, we first characterize the solutions of the dynamical equations where only \( H_0 \) is used as the Hamiltonian. We begin by identifying stationary solutions,

\[ \mathbf{D} (r, t) = \mathbf{D}_m (r) e^{-i\omega_m t} + c.c., \] (2.37)
\[ \mathbf{B} (r, t) = \mathbf{B}_m (r) e^{-i\omega_m t} + c.c., \]

of the classical equations, where \( c.c. \) stands for complex conjugate. Using the nominal Hamiltonian \( H_0 \) and the form (2.37) in (2.11), we obtain the time-independent Maxwell equations for the nominal system:

\[ -i\omega_m \mu_0 \mathbf{D}_m (r) = \nabla \times \mathbf{B}_m (r), \] (2.38)
\[ i\omega_m \varepsilon_0 \mathbf{B}_m (r) = \nabla \times \left[ \frac{\mathbf{D}_m (r)}{n^2 (r)} \right]. \]
In practice, normal modes are found by combining Eqs. (2.38) into a second-order Hermitian eigenvalue problem for $B_m(r)$:

$$\nabla \times \left[ \frac{\nabla \times B_m(r)}{n^2(r)} \right] = \frac{\omega_m^2}{c^2} B_m(r). \tag{2.39}$$

Solutions of (2.39) for $\omega_m \neq 0$ are sought, subject to the divergence condition

$$\nabla \cdot B_m(r) = 0. \tag{2.40}$$

It is also possible to construct a second-order eigenvalue problem for $D_m(r)$,

$$\nabla \times \nabla \times \frac{D_m(r)}{n^2(r)} = \frac{\omega_m^2}{c^2} D_m(r), \tag{2.41}$$

although the associated operator is not Hermitian. Then solutions of (2.41) for $\omega_m \neq 0$ are subject to the divergence condition $\nabla \cdot D_m(r) = 0$.

Either (2.39) or (2.41) suffices as a master equation for the electromagnetic modes. For example, having obtained $B_m(r)$ satisfying the master equation (2.39), $D_m(r)$ is obtained from the first of (2.38). Then the pair of fields $(D_m(r), B_m(r))$ identifies a stationary solution of the classical Maxwell equations (2.1) and (2.2); the square of the frequency is obtained from (2.39); we take the positive root, associating a real positive frequency $\omega_m$ with each normal mode solution. Taking the complex conjugate of Eqs. (2.38), we note that for every mode $(D_m(r), B_m(r))$ with frequency $\omega_m$ there is another related mode

$$(D_{\overline{m}}(r), B_{\overline{m}}(r)) = (D^*_m(r), -B^*_m(r)). \tag{2.42}$$

with frequency

$$\omega_{\overline{m}} = \omega_m. \tag{2.43}$$

Throughout the chapter, an overbar refers to the physical conjugate in this sense. In problems of interest such pairs of modes are physically easy to identify and pair, so in practice (2.42) and (2.43) can be used to define the mode $\overline{m}$ in terms of the mode $m$, and to thereby obtain half of the modes without resorting directly to the eigenvalue problem (2.39).

Since (2.39) defines a Hermitian eigenvalue problem, solutions associated with different eigenvalues are orthogonal, and we can choose the following normalization conditions for $D_m(r)$ and $B_m(r)$:

$$\int_V \frac{B^*_m(r) \cdot B_{m'}(r)}{\mu_0 n^2(r)} = \frac{\hbar \omega_m}{2} \delta_{mm'}, \tag{2.44}$$

$$\int_V \frac{D^*_m(r) \cdot D_{m'}(r)}{\varepsilon_0 n^2(r)} = \frac{\hbar \omega_m}{2} \delta_{mm'}. \tag{2.44}$$
where we employ box normalization, the spatial integral is over the very large normal-
ization region \( V \) of volume \( V \), and the mode fields \( D_m(r) \) and \( B_m(r) \) are taken to have
periodic boundary conditions. Throughout most of this chapter we use discrete modes
for the electromagnetic field in this fashion. The exception is Section 2.4, where we give
the transition to systems with continuous or mixed labeling.

For the nominal system, arbitrary electromagnetic field solutions can be expanded in
terms of the normal modes as

\[
D(r, t) = \sum_m C_m^{(1)}(t) D_m(r),
\]

\[
B(r, t) = \sum_m C_m^{(2)}(t) B_m(r),
\]

where we guarantee the reality, or Hermiticity in the quantum case, of \( B(r, t) \) and \( D(r, t) \)
by imposing the conditions \( C_m^{(1)} = (C_m^{(1)})^\dagger \) and \( C_m^{(2)} = -(C_m^{(2)})^\dagger \). These conditions can
be satisfied identically by introducing new unrestricted mode amplitudes \( a_m \) such that
\( C_m^{(1)} = a_m + a_m^\dagger \) and \( C_m^{(2)} = a_m - a_m^\dagger \). In conjunction with (2.42), this yields

\[
D(r, t) = \sum_m \left( a_m(t) D_m(r) + a_m^\dagger(t) D_m^*(r) \right),
\]

\[
B(r, t) = \sum_m \left( a_m(t) B_m(r) + a_m^\dagger(t) B_m^*(r) \right),
\]

where the sum is over all modes. From the commutators (Poisson brackets) (2.10) then
follow the equal-time commutators

\[
[a_m, a_{m'}] = 0,
\]

\[
[a_m, a_{m'}^\dagger] = \delta_{mm'}. \tag{2.47}
\]

Using (2.46) in (2.34) we then find that the nominal system Hamiltonian is given by

\[
H_0 = \sum_m \hbar \omega_m a_m^\dagger a_m, \tag{2.48}
\]

with neglect of the zero point energy in the quantum case, and Hamilton’s equation

\[
i\hbar \dot{a}_m = [a_m, H_0] \tag{2.49}
\]

indeed generates the Maxwell curl equations (2.1) with the constitutive relation (2.36);
the Maxwell divergence equations (2.2) are manifestly satisfied due to the form of the
expansion (2.46) in terms of the divergencelessness normal modes.
We can now use the expansion (2.46) of the electromagnetic fields to write the full Hamiltonian in terms of raising and lowering operators of the nominal system modes. First, the counter-term is given by

\[ H_\Delta = \frac{1}{2} \sum_{m,m'} \left( a_{m'} + a_{m'}^\dagger \right) \Gamma^{(0)}_{m'm} \left( a_m + a_m^\dagger \right), \]  

(2.50)

where we define

\[ \Gamma^{(0)}_{m'm} \equiv \frac{1}{\varepsilon_0} \int_V d\mathbf{r} \ D_{m'}^i(\mathbf{r}) \Gamma^{ij}_0(\mathbf{r}) \ D^j_m(\mathbf{r}). \]  

(2.51)

We note that using (2.42) and the permutation symmetry of \( \Gamma^{ij}_0(\mathbf{r}) \) yields the properties

\[ \Gamma^{(0)*}_{m'm} = \Gamma^{(0)}_{m'm} = \Gamma^{(0)}_{mm'}. \]  

(2.52)

The full Hamiltonian then becomes

\[ H = H_{em} + H_{med} + H_{int} \]  

(2.53)

\[ = H_0 + H_\Delta + H_{med} + H_{int} \]  

(2.54)

\[ = \sum_m \hbar \omega_m a_m^\dagger a_m + \frac{1}{2} \sum_{m,m'} \left( a_{m'} + a_{m'}^\dagger \right) \Gamma^{(0)}_{m'm} \left( a_m + a_m^\dagger \right) \]

\[ + \int_{\Omega_g} d\Omega \int_{V_\Omega} d\mathbf{r} \hbar \Omega \left( \psi_{\Omega}^\dagger(\mathbf{r}) \cdot \psi_{\Omega}(\mathbf{r}) \right) \]

\[ + \sum_{m'} \int_{\Omega_g} d\Omega \int_{V_\Omega} d\mathbf{r} \left( a_{m'} + a_{m'}^\dagger \right) \Lambda_{m'}(\mathbf{r},\Omega) \cdot \left( \psi_{\Omega}(\mathbf{r}) + \psi_{\Omega}^\dagger(\mathbf{r}) \right), \]  

(2.55)

where again we have neglected the zero-point energy in the quantum case, and where we have defined a mode coupling constant

\[ \Lambda^\lambda_m(\mathbf{r},\Omega) \equiv -\sqrt{\frac{\hbar}{2\varepsilon_0\Omega}} D^{\lambda*}_m(\mathbf{r}) \Lambda^{\lambda\lambda}(\mathbf{r},\Omega). \]  

(2.56)

Recall that the overbar always refers to physical conjugation of a mode as described after (2.43), and that sums over modes in (2.55) are over all modes.

### 2.3.3 The polariton problem

It is the Hamiltonian (2.55) we seek to diagonalize, and find the full, or polariton, modes of the system. Generally, some of the nominal electromagnetic modes lie below the cutoff frequency \( \Omega_g \) for absorption, and for our calculation with periodic boundary conditions a discrete spectrum of modes can be found there. Above the cutoff frequency for absorption
the polariton spectrum forms a continuum. At these frequencies, the problem of finding the polariton modes amounts to a Fano diagonalization with many discrete modes coupled to many continuous modes — a “many-many” Fano problem — with counterrotating wave terms, and an extra coupling between the bare discrete modes. The single-single, single-many, and many-single rotating-wave approximation (RWA) cases were solved by Fano [95]; the many-many RWA case was subsequently solved by Mies [96] in a way specific to Fano’s configuration interaction problem; that solution does not apply here. The single-single non-RWA problem was solved by Huttner and Barnett in their seminal work\footnote{We take this opportunity to correct a typographical error in Huttner and Barnett’s [43] Hamiltonian. In their Equation (2.25b), the expression $\hat{b}_\omega (\lambda, \pm k, t) + \hat{b}_\omega (\lambda, \mp k, t)$ should read $\hat{b}_\omega (\lambda, k, t) + \hat{b}_\omega (\lambda, -k, t)$. In their Equation (A1), the expression $\hat{b}_\omega (-k) + \hat{b}_\omega (k)$ should read $\hat{b}_\omega (k) + \hat{b}_\omega (-k)$. We thank Stephen Barnett [97] for distilling this problem.}; here we must address an augmented many-many non-RWA case, which has not been previously solved. In addition, virtual coupling to the medium modes shifts the nominal system modes in the discrete spectrum below the cutoff frequency $\Omega_g$.

We begin with an important classification of the polariton modes. Below we identify two types of modes, which we call longitudinal polarization (LP) polaritons and transverse polarization (TP) polaritons. For the first type, $H_{\text{int}}$ does not contribute to the dynamics, and the modes consist solely of contributions from the medium oscillators. We formally separate the Hamiltonian of these uncoupled modes from the Hamiltonian of the TP modes as

$$ H = H_{LP} + H_{TP}, $$

(2.57)

where $H_{LP}$ and $H_{TP}$ are respectively the parts of the Hamiltonian corresponding to longitudinal polarization and transverse polarization modes; then we seek polariton operators that diagonalize the Hamiltonian in the form

$$ H_{LP} = \sum_m \int_{\Omega_g}^{\infty} d\Omega \, \hbar \Omega \, s_{\Omega m}^{\dagger} s_{\Omega m}, $$

(2.58)

$$ H_{TP} = \sum_m \hbar \tilde{\omega}_m c_m^{\dagger} c_m + \sum_m \int_{\Omega_g}^{\infty} d\Omega \, \hbar \Omega \, c_{\Omega m}^{\dagger} c_{\Omega m}, $$

(2.59)

where the $c_{\Omega m}$ are the TP operators for the continuous polariton spectrum, and the $c_m$ are the TP operators of the discrete polariton spectrum; we will find, for all $m$, $\tilde{\omega}_m < \Omega_g$. The $s_{\Omega m}$ are the LP operators, and by definition they satisfy

$$ [s_{\Omega m}, H_{\text{int}}] = 0. $$

(2.60)
The various modes are plotted schematically in Fig. 2.1. The polariton operators are required to satisfy harmonicity conditions

\[
[s_{\Omega m}, H] = \hbar \Omega s_{\Omega m}, \quad (2.61)
\]
\[
[c_m, H] = \hbar \tilde{\omega}_m c_m, \quad (2.62)
\]
\[
[c_{\Omega m}, H] = \hbar \Omega c_{\Omega m}, \quad (2.63)
\]

and the (equal-time) canonical commutation relations that apply are

\[
\left[ s_{\Omega m}, s_{\Omega' m'}^\dagger \right] = \delta_{mm'} \delta (\Omega - \Omega'), \quad (2.64)
\]
\[
\left[ c_m, c_m^\dagger \right] = \delta_{mm'}, \quad (2.65)
\]
\[
\left[ c_{\Omega m}, c_{\Omega' m'}^\dagger \right] = \delta_{mm'} \delta (\Omega - \Omega'), \quad (2.66)
\]

with all other equal-time commutators between the various \( c \) and \( s \) operators vanishing. The number of discrete modes is not known at the outset, although for sufficiently small absorption and dispersion it is expected to approach the number of nominal modes below the cutoff frequency in typical systems.
The picture that then arises is that there are three types of polaritons. In the absorption regime \((\Omega \geq \Omega_g)\) we have LP and TP polaritons, which form continua, and in the purely dispersive regime \((\Omega < \Omega_g)\) we have discrete TP polaritons.

We mention now, and prove in Section 2.3.8, that the displacement and magnetic induction fields can be written entirely in terms of the TP polariton operators and their corresponding mode fields as

\[
D(r, t) = \sum_m \left[ c_m \tilde{D}_m(r) + c^\dagger_m \tilde{D}^*_m(r) \right] + \sum_m \int_{\Omega_g}^{\infty} d\Omega \left[ c_{\Omega m} \tilde{D}_{\Omega m}(r) + c^\dagger_{\Omega m} \tilde{D}^*_{\Omega m}(r) \right],
\]

\[
B(r, t) = \sum_m \left[ c_m \tilde{B}_m(r) + c^\dagger_m \tilde{B}^*_m(r) \right] + \sum_m \int_{\Omega_g}^{\infty} d\Omega \left[ c_{\Omega m} \tilde{B}_{\Omega m}(r) + c^\dagger_{\Omega m} \tilde{B}^*_{\Omega m}(r) \right],
\]

where the \(\tilde{D}_m(r)\) and the \(\tilde{B}_m(r)\) are the mode fields for discrete TP mode \(m\), and the \(\tilde{D}_{\Omega m}(r)\) and the \(\tilde{B}_{\Omega m}(r)\) are the mode fields for the continuum TP mode \(\Omega m\).

We now turn to the task of identifying three polariton types.

### 2.3.4 Longitudinal polarization (LP) polariton modes

We first identify the longitudinal polarization polariton modes. These are modes in the absorption regime composed entirely of medium fields which, although not necessarily longitudinal themselves, produce a longitudinal polarization. Thus they do not couple through \(H_{int}\) with the displacement field, which is purely transverse. It is possible to formulate the medium fields in \(k\)-space so as to avoid inclusion of longitudinal polarizations from the outset. However, this approach leads to more complicated expressions elsewhere, especially if \(V_\Omega\) is not all of space. The real-space formulation of the medium fields we have used here, in contrast, respects the spatial locality of the dielectric response of the model.

Writing the lowering operators of the LP polaritons as \(s_{\Omega m}\) (2.58), we expand them in terms of the medium fields as

\[
s_{\Omega m} = \int_{V_\Omega} dr \rho^{\Omega m}(r) \cdot \psi_{\Omega}(r),
\]

where the condition (2.60) requires that the expansion coefficients \(\rho^{\Omega m}(r)\) obey

\[
\int_{V_\Omega} dr D^*_{\Omega m'}(r) \cdot \Lambda(r, \Omega) \cdot \rho^{\Omega m}(r) = 0
\]
for all $m'$; we normalize them according to

$$\int_{\mathcal{V}_\Omega} d\mathbf{r} \, \mathbf{\rho}^{\Omega m'}(\mathbf{r}) \cdot \mathbf{\rho}^{\Omega m}(\mathbf{r}) = \delta_{mm'}.$$  \hfill (2.71)

In conjunction with the Hamiltonian (2.55) and the medium field commutators (2.17), it follows from (2.69), (2.70) and (2.71) respectively that the LP lowering operators $s^{\Omega m}$ have harmonic time dependence (2.61), and satisfy canonical commutation relations (2.64).

To identify the $\mathbf{\rho}^{\Omega m}(\mathbf{r})$, we first note that we can extend the integrals in (2.69, 2.70, 2.71) to all space, by extending the functions $\mathbf{\rho}^{\Omega m}(\mathbf{r})$ to vanish for $\mathbf{r}$ outside $\mathcal{V}_\Omega$. Now the $\mathbf{D}^{*}_{m'}(\mathbf{r})$ span the space of transverse vector fields, so since the integral over all space of the dot product of a longitudinal field with any transverse field vanishes, and since no transverse field has a vanishing overlap in this sense with every transverse field, $\Lambda(\mathbf{r}, \Omega) \cdot \mathbf{\rho}^{\Omega m}(\mathbf{r})$ must be a longitudinal vector field. Recalling the periodic boundary conditions employed, and neglecting any uniform fields over the volume, we must have

$$\Lambda(\mathbf{r}, \Omega) \cdot \mathbf{\rho}^{\Omega m}(\mathbf{r}) = \nabla \phi^{\Omega m}(\mathbf{r})$$  \hfill (2.72)

for some scalar field $\phi^{\Omega m}(\mathbf{r})$. Since the left hand side of (2.72) must be finite, we require that $\phi^{\Omega m}(\mathbf{r})$ be continuous and once-differentiable. Taking components along the principal axis direction $\lambda$, it then follows from (2.72) and from the requirement that $\mathbf{\rho}^{\Omega m}(\mathbf{r})$ vanish outside $\mathcal{V}_\Omega$ that (no summation)

$$\rho^{\Omega m}_{\lambda}(\mathbf{r}) = f^{\Omega}_{\lambda}(\mathbf{r}) \partial_\lambda \phi^{\Omega m}(\mathbf{r}),$$  \hfill (2.73)

with

$$f^{\Omega}_{\lambda}(\mathbf{r}) = \frac{1}{\Lambda^\lambda(\mathbf{r}, \Omega)} \quad \text{if } \Lambda^\lambda(\mathbf{r}, \Omega) \neq 0.$$  \hfill (2.74)

We need not specify $f^{\Omega}_{\lambda}(\mathbf{r})$ for $\mathbf{r}$ such that $\Lambda^\lambda(\mathbf{r}, \Omega) = 0$, since

$$\partial_\lambda \phi^{\Omega m}(\mathbf{r}) = 0 \text{ if } \Lambda^\lambda(\mathbf{r}, \Omega) = 0,$$  \hfill (2.75)

which follows immediately from (2.72). Eqs. (2.73, 2.74, 2.75) describe the full set of LP polariton solutions.

It follows from (2.75) that $\phi^{\Omega m}$ is constant within any connected region outside of $\mathcal{V}_\Omega$. Furthermore, given a local unit vector $\hat{n}$ normal to the boundary of $\mathcal{V}_\Omega$, which we denote $\mathcal{S}_\Omega$, we have the boundary condition $\hat{n} \times \nabla \phi^{\Omega m}(\mathbf{r}) = 0$ on $\mathcal{S}_\Omega$. Note that $\mathcal{V}_\Omega$ may be
Figure 2.2: Schematic illustration of the scalar potential $\phi^{\Omega m}$ for the longitudinal polarization modes. The hatched regions represent $\mathcal{V}_\Omega$, the region in which absorption is present at frequency $\Omega$. The scalar potential is constant within connected non-absorbing regions.

multiply-connected, and that we make no assumptions about the number of connected components. This is illustrated schematically in Fig. 2.2.

The procedure for constructing the full set of LP polariton modes is as follows. Find a set of continuous scalar fields $\tilde{\phi}^{\Omega m}(r)$ that span the space of continuous once-differentiable functions on the normalization volume up to addition of an overall constant, subject to the condition (2.75). Obtain the corresponding set of $\tilde{\rho}^{\Omega m}(r)$ from (2.73). Then the $\rho^{\Omega m}(r)$ can be constructed from the $\tilde{\rho}^{\Omega m}(r)$ by Gram-Schmidt orthogonalization of the $\tilde{\rho}^{\Omega m}(r)$ with respect to the normalization condition (2.71).

There is a formal mathematical analogy here to electrostatics with perfect conductors and free charges. The field $\phi^{\Omega m}(r)$ corresponds to the electromagnetic scalar potential, and the quantity $-\Lambda(r, \Omega) \cdot \rho^{\Omega m}(r)$ corresponds to the electric field. The local charge density is proportional to $-\nabla^2 \phi^{\Omega m}(r)$. And the region of space outside of $\mathcal{V}_\Omega$, where there is no coupling, plays the role of a perfect conductor. The complete set of fields $\tilde{\phi}^{\Omega m}(r)$ then corresponds to those potentials that can be generated by arbitrary charge distributions in the region $\mathcal{V}_\Omega$.

While a route to constructing the LP polariton modes has been identified here for completeness, the explicit construction of these modes is unnecessary for most applications. This is because the displacement and magnetic induction fields only involve the transverse polarization polariton modes, as mentioned before (2.67), and we now turn to
the task of finding them.

### 2.3.5 Transverse polarization (TP) polariton modes

We first consider the transverse polarization polaritons in the absorptive regime. To find them, we begin with a general expansion of the $c_{\Omega m}$ in terms of nominal system modes and medium oscillator modes that are coupled together in the Hamiltonian (2.55). We write

$$c_{\Omega m} = \sum_n \left( \begin{array}{c} \alpha_{\Omega m}^n a_n + \beta_{\Omega m}^{\dagger n} a_{\Pi}^{\dagger} \end{array} \right) + \int_{\Omega_g} d\Omega' \int_{\mathcal{V}_{\Omega'}} dr \left( \alpha_{\Omega' m}^{\dagger} \cdot \psi_{\Omega'} (r) + \beta_{\Omega' m}^{\dagger} (r) \cdot \psi_{\Omega'}^{\dagger} (r) \right),$$

(2.76)

where we recall that the sums include all nominal system modes in the sum, regardless of whether they lie in the frequency range of the discrete or continuous polariton band. We refer to the terms containing $\alpha$’s and $\beta$’s as respectively as corotating and counterrotating terms.

The harmonicity condition (2.63), in conjunction with (2.76) and (2.55), yields an equation that we do not reproduce here, which provides the conditions on the expansion coefficients $\alpha_{\Omega m}^n$, $\alpha_{\Omega' m}^{\dagger n}$, $\beta_{\Omega m}^{\dagger n}$, and $\beta_{\Omega' m}^{\dagger n}$. We refer to it as the “harmonicity equation.” Taking commutators of the harmonicity equation with $a_{\Pi m}$, $a_{\Pi m}^{\dagger}$, $\psi_{\Omega''} (r)$ and $\psi_{\Omega''}^{\dagger} (r)$, and relabeling the primes, we obtain the following system of equations:

$$(\hbar \Omega - \hbar \omega_{\Omega'}) \alpha_{\Omega m}^n - \sum_n \Gamma_{n n'}^{(0)} \left( \alpha_{\Omega m}^n - \beta_{\Omega m}^{\dagger n} \right) - \int_{\Omega_g} d\Omega' \int_{\mathcal{V}_{\Omega'}} dr \Lambda^*_{n'} (r, \Omega') \cdot \left( \alpha_{\Omega' m}^{\dagger n} (r) - \beta_{\Omega' m}^{\dagger n} (r) \right) = 0,$$

(2.77)

$$(\hbar \Omega + \hbar \omega_{\Omega'}) \beta_{\Omega m}^n - \sum_n \Gamma_{n n'}^{(0)} \left( \alpha_{\Omega m}^n - \beta_{\Omega m}^{\dagger n} \right) - \int_{\Omega_g} d\Omega' \int_{\mathcal{V}_{\Omega'}} dr \Lambda^*_{n'} (r, \Omega') \cdot \left( \alpha_{\Omega' m}^{\dagger n} (r) - \beta_{\Omega' m}^{\dagger n} (r) \right) = 0,$$

(2.78)

$$(\hbar \Omega - \hbar \Omega') \alpha_{\Omega' m} (r) - \sum_n \Lambda_n (r, \Omega') \left( \alpha_{\Omega m}^n - \beta_{\Omega m}^{\dagger n} \right) = 0,$$

(2.79)

$$(\hbar \Omega + \hbar \Omega') \beta_{\Omega' m} (r) - \sum_n \Lambda_n (r, \Omega') \left( \alpha_{\Omega m}^n - \beta_{\Omega m}^{\dagger n} \right) = 0,$$

(2.80)

where we recall that $\Lambda_n (r, \Omega)$ is defined in (2.56). We note that the harmonicity equation can be reconstructed from (2.77, 2.78, 2.79, 2.80) by multiplying each of these equations by an appropriate operator and then composing the results, so (2.77, 2.78, 2.79, 2.80) are necessary and sufficient for the harmonicity equation to be satisfied, and thus for
harmonicity condition (2.63), to hold. For every solution to Eqs. (2.77, 2.78, 2.79, 2.80) labeled by \( m \), i.e. \( (\alpha_{m}^{n'}, \beta_{m}^{n'}, \alpha_{m}^{\Omega_{m}} (r), \beta_{m}^{\Omega_{m}} (r)) \), there is another solution, which we label \( \overline{m} \), given by

\[
(\alpha_{m}^{\overline{n'}} = \alpha_{m}^{\overline{n'}*}, \beta_{m}^{\overline{n'}} = \beta_{m}^{\overline{n'}*}, \alpha_{m}^{\Omega_{m}} (r) = \alpha_{m}^{\Omega_{m}} (r), \beta_{m}^{\Omega_{m}} (r) = \beta_{m}^{\Omega_{m}} (r) ),
\]

where \( \alpha_{m}^{\overline{n'}*} \equiv (\alpha_{m}^{n'})^{*} \) etc. To confirm (2.81), let \( m \to \overline{m} \) and \( n' \to \overline{n'} \) in Eqs. (2.77, 2.78, 2.79, 2.80), take the complex conjugate of these equations, use properties (2.43), (2.52), and reorganize the sums. This yields the original set of equations, i.e. the set (2.77, 2.78, 2.79, 2.80), but with the replacements

\[
\{ \alpha_{m}^{n'} \to \alpha_{m}^{\overline{n'}*}, \beta_{m}^{n'} \to \beta_{m}^{\overline{n'}*}, \alpha_{m}^{\Omega_{m}} (r) \to \alpha_{m}^{\Omega_{m}} (r), \beta_{m}^{\Omega_{m}} (r) \to \beta_{m}^{\Omega_{m}} (r) \},
\]

and (2.81) results.

We now solve the system (2.77, 2.78, 2.79, 2.80). Comparing (2.77) and (2.78) immediately gives

\[
\beta_{m}^{\Omega_{m}} = \frac{\hbar \Omega - \hbar \omega_{n'}}{\hbar \Omega + \hbar \omega_{n'}} \alpha_{m}^{\Omega_{m}},
\]

so the contributions of the corotating nominal system operators determine the contributions of the associated counterrotating operators. Making use of (2.83), Equation (2.79) is formally solved in terms of the \( \alpha_{m}^{\Omega_{m}} \) as

\[
\alpha_{m}^{\Omega_{m}} (r) = \left( P \frac{1}{\hbar \Omega - \hbar \Omega'} + Z_{\Omega m} \delta (\hbar \Omega - \hbar \Omega') \right) \sum_{n} \Lambda_{n} (r, \Omega') \gamma_{n}^{\Omega_{m}},
\]

where we have suppressed the homogeneous solution, \( P \) denotes the Cauchy principal value, and we have defined \( \gamma_{n}^{\Omega_{m}} \) according to

\[
\gamma_{n}^{\Omega_{m}} \equiv \alpha_{n}^{\Omega_{m}} - \beta_{n}^{\Omega_{m}} = \frac{2 \hbar \omega_{n}}{\hbar \Omega + \hbar \omega_{n}} \alpha_{n}^{\Omega_{m}}.
\]

The factor \( Z_{\Omega m} \delta (\hbar \Omega - \hbar \Omega') \) in (2.84) picks up the contribution from the singularity, which must be determined self-consistently, as is usual in Fano-type problems [98]. We call \( Z_{\Omega m} \) the “resonant oscillator amplitude,” because it is proportional to the contribution to the polariton from medium fields with natural frequency equal to that of the polariton. Suppressing the homogeneous solution in (2.84) prevents the inclusion of any LP polariton mode components in \( c_{\Omega m} \). This is verified in (B.10) of Appendix B, where explicit calculation shows that \( \left[ c_{\Omega m}, s_{\Omega_{m'}}^{\dagger} \right] \) vanishes for all \( (\Omega, \Omega') \) without requiring any further condition on the \( c_{\Omega m} \) beyond dropping the homogeneous solution, as done here.
Hence using (2.84) for our solution of (2.79, 2.80) restricts our otherwise general form (2.76) to yield only TP polariton modes.

Using (2.85), from (2.80) we have

$$\beta_{\Omega'}^{\Omega m}(r) = \frac{1}{\hbar \Omega + \hbar \Omega'} \sum_n \Lambda_n(r, \Omega') \gamma_n^{\Omega m},$$

and it is useful to combine the $$\alpha_{\Omega'}^{\Omega m}(r)$$ and $$\beta_{\Omega'}^{\Omega m}(r)$$ to give the combined effect including the counterrotating wave as

$$\gamma_{\Omega'}^{\Omega m}(r) \equiv \alpha_{\Omega'}^{\Omega m}(r) - \beta_{\Omega'}^{\Omega m}(r)$$

$$= \left( P \frac{2h \Omega'}{(\hbar \Omega)^2 - (h \Omega')^2} + Z_{\Omega m} \delta (h \Omega - h \Omega') \right) \sum_n \Lambda_n(r, \Omega') \gamma_n^{\Omega m}. \tag{2.88}$$

Combining (2.87) and (2.77) yields, in terms of the parameters of the underlying oscillator model,

$$\gamma_n^{\Omega m} = \frac{Z_{\Omega m}}{\hbar} \sum_n \int dV \Lambda^*_n(r, \Omega) \cdot \Lambda_n(r, \Omega) \gamma_n^{\Omega m}. \tag{2.89}$$

Equation (2.89) is a generalized Hermitian eigenvalue problem with the eigenvalue-eigenvector pair $$(Z_{\Omega m}, \gamma_n^{\Omega m})$$. To our knowledge, such an equation has never appeared in a previous treatment of a multichannel Fano-diagonalization problem [96][99]. Since the resonant oscillator amplitude $$Z_{\Omega m}$$ is the eigenvalue in a generalized Hermitian eigenvalue problem, it is guaranteed to be real.

Making use of the expressions (2.28, 2.29) for the medium response, (2.89) becomes

$$\sum_n \left[ \frac{(\hbar \Omega)^2 - (\hbar \omega_n)^2}{2 \hbar \omega_n} \delta_{n'n'} - (\Delta \text{Re } \Gamma)^O_{nn'} - P \int_{\Omega_0}^\infty d\Omega' \left( \frac{2h \Omega' \int_{V_{\Omega'}} dr \Lambda^*_{n'}(r, \Omega') \cdot \Lambda_n(r, \Omega')}{(\hbar \Omega)^2 - (h \Omega')^2} \right) \right] \gamma_n^{\Omega m} = \frac{Z_{\Omega m}}{\hbar} \sum_n (\text{Im } \Gamma)^O_{nn'} \gamma_n^{\Omega m}, \tag{2.90}$$

where

$$\Delta \text{Re } \Gamma^O_{nn'} \equiv \frac{1}{\varepsilon_0} \int_V dr \mathbf{D}^*_n(r) \cdot [\text{Re } \Gamma(r, \Omega) - \Gamma_0(r)] \cdot \mathbf{D}_{n'}(r), \tag{2.91}$$

$$\text{Im } \Gamma^O_{nn'} \equiv \frac{1}{\varepsilon_0} \int_V dr \mathbf{D}^*_n(r) \cdot \text{Im } \Gamma(r, \Omega) \cdot \mathbf{D}_{n'}(r). \tag{2.92}$$

Physical quantities appear in (2.90) in a very natural way. Here the information about the model coupling constant $$\Lambda(r, \Omega)$$ has completely given way to the aggregate real and imaginary parts of the dielectric response; for the real part of the response, the quantity
that appears is in fact just the dispersive part of the response beyond the nominal value. The eigenvalue equation for the underlying polaritons is now expressed in terms of the physically important quantities, and the dispersive and absorptive contributions to the dielectric response appear in separate roles here, rather than as real and imaginary parts of a single quantity, reflecting their physical difference. Furthermore, the real response appears as its difference from the nominal response, so there is a simple connection to the nominal system. From a practical standpoint, (2.90) can be applied straightforwardly; one takes matrix elements consisting of overlap integrals between nominal system modes and the medium response; the generalized Hermitian eigenvalue problem can be solved using standard routines to obtain $Z_{\Omega m}$ and $\gamma_{\Omega m}$. We note that Eqs. (2.90, 2.91, 2.92) hold for any choice of nominal index profile $n(r)$; no perturbative analysis has been used. We can also isolate the effect of inclusion of the counterrotating terms in the expansion (2.76), since performing the above derivation in the rotating wave approximation, i.e. with all $\beta_{\Omega m} = 0$, yields the eigenvalue equation (2.90), but with the replacement $\gamma_{\Omega m} \rightarrow \alpha_{\Omega m}$ and with the term multiplying the $\delta_{n,n}$ replaced by $(\hbar \Omega - \hbar \omega_n)$. Furthermore, although it is not immediately apparent from (2.90), the eigenvalues $Z_{\Omega m}$ depend only on $\Gamma(r, \omega)$, and are independent of the choice of nominal index profile $n(r)$. This is verified in Section 2.3.7.

Finally, imposing the equal-time commutation relation (2.66) yields the normalization condition for the $\gamma_{\Omega m}$. We defer the details to Appendix B, where the normalization condition is determined to be

$$
\sum_{n,n'} \gamma_{\Omega m'} \langle \text{Im } \Gamma \rangle_{nn'} \gamma_{\Omega m} = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m})^2} \delta_{nn'}.
$$

(2.93)

2.3.6 Polaritons in the transparent regime

Below the absorption cutoff frequency $\Omega_g$, the polariton spectrum is discrete, and the TP polariton operators there are the $c_m$ introduced in (2.59). We begin with a general expansion of the $c_m$ in terms of nominal system modes and medium modes that are coupled together in the Hamiltonian (2.55):

$$
c_m = \sum_n \left( \alpha_n^m a_n + \beta_n^m a_n^\dagger \right) + \int_{\Omega_g} d\Omega' \int_{V_{\Omega'}} d\mathbf{r} \left( \alpha_{\Omega'}^m (\mathbf{r}) \cdot \psi_{\Omega'} (\mathbf{r}) + \beta_{\Omega'}^m (\mathbf{r}) \cdot \psi_{\Omega'}^\dagger (\mathbf{r}) \right).
$$

(2.94)
Then, following the approach used in the absorptive regime, the harmonicity condition (2.62) leads to the following system of equations:

\[
(\hbar \bar{\omega}_m - \hbar \omega_{n'}) \alpha^m_n - \sum_n \Gamma^{(0)}_{nn'} (\alpha^m_n - \beta^m_n) - \int_{\Omega_n} d\Omega' \int_{V_{\Omega'}} d\mathbf{r} \mathbf{\Lambda}^*_{n'} (\mathbf{r}, \Omega') \cdot (\mathbf{\alpha}^m_{n'} (\mathbf{r}) - \mathbf{\beta}^m_{n'} (\mathbf{r})) = 0,
\]

\[
(\hbar \bar{\omega}_m + \hbar \omega_{n'}) \beta^m_n - \sum_n \Gamma^{(0)}_{nn'} (\alpha^m_n - \beta^m_n) - \int_{\Omega_n} d\Omega' \int_{V_{\Omega'}} d\mathbf{r} \mathbf{\Lambda}^*_{n'} (\mathbf{r}, \Omega') \cdot (\mathbf{\alpha}^m_{n'} (\mathbf{r}) - \mathbf{\beta}^m_{n'} (\mathbf{r})) = 0,
\]

\[
(h \bar{\omega}_m - h \Omega') \mathbf{\alpha}^{m*}_{m'} (\mathbf{r}) - \sum_n \mathbf{\Lambda}_n (\mathbf{r}, \Omega') (\alpha^m_n - \beta^m_n) = 0,
\]

\[
(h \bar{\omega}_m + h \Omega') \mathbf{\beta}^{m*}_{m'} (\mathbf{r}) - \sum_n \mathbf{\Lambda}_n (\mathbf{r}, \Omega') (\alpha^m_n - \beta^m_n) = 0.
\]

Physical conjugate solutions are given by

\[
(\alpha^m_{m'} = \alpha^{m*}_{m}, \beta^m_{m'} = \beta^{m*}_{m}, \mathbf{\alpha}^{m}_{m'} (\mathbf{r}) = \mathbf{\alpha}^{m*}_{m'} (\mathbf{r}), \mathbf{\beta}^m_{m'} (\mathbf{r}) = \mathbf{\beta}^{m*}_{m'} (\mathbf{r})).
\]

As with (2.77) and (2.78), comparing Eqs. (2.95) and (2.96) immediately gives

\[
\beta^m_{m'} = \frac{\hbar \bar{\omega}_m - \hbar \omega_{n'}}{\hbar \bar{\omega}_m + \hbar \omega_{n'}} \alpha^m_{n'}.
\]

Making use of (2.100), Eqs. (2.97) and (2.98) have the following solutions in terms of the \( \alpha^m_n \):

\[
\begin{cases}
\mathbf{\alpha}^m_{m'} (\mathbf{r}) \\
\beta^m_{m'} (\mathbf{r})
\end{cases}
= \frac{1}{h \bar{\omega}_m + h \Omega'} \sum_n \mathbf{\Lambda}_n (\mathbf{r}, \Omega') \gamma^m_n.
\]

Here we define the coefficients \( \gamma^m_n \) as

\[
\gamma^m_n \equiv \alpha^m_n - \beta^m_n = \frac{2 \hbar \omega_n}{\hbar \bar{\omega}_m + \hbar \omega_n} \alpha^m_n.
\]

The system (2.95, 2.96, 2.97, 2.98) yields a nullspace equation, and as before it is possible to combine factors involving the model coupling tensor into a term involving only the dielectric response:

\[
\sum_n G_{n'n'} (\bar{\omega}_m) \gamma^m_n = 0,
\]

where

\[
G_{n'n'} (\bar{\omega}_m) \equiv \frac{(\hbar \bar{\omega}_m)^2 - (\hbar \omega_{n'})^2}{2 \hbar \omega_{n'}} \delta_{n'n} + (\Delta \Gamma)_{nn'}^m,
\]

with

\[
(\Delta \Gamma)_{nn'}^m \equiv \frac{1}{\varepsilon_0} \int_V d\mathbf{r} \mathbf{D}_n^* (\mathbf{r}) \cdot [\Gamma (\mathbf{r}, \bar{\omega}_m) - \Gamma_0 (\mathbf{r})] \cdot \mathbf{D}_{n'} (\mathbf{r}).
\]
We note that \((\Delta \Gamma)_{nn'}^{m} = (\Delta \Gamma)_{n'n}^{m*}\) due to the permutation symmetry of \(\Gamma^{ij}\) (2.7). Equation (2.103) plays the role for discrete modes that (2.90) plays for the modes at absorbing frequencies. And the homogeneous system of equations (2.103) plays the role here for the transparent regime that the generalized Hermitian eigenvalue problem (2.90) plays for the absorptive regime. Solutions to (2.103) exist for the frequencies \(\tilde{\omega}_m, \tilde{\omega}_m \in (0, \Omega_g)\), such that \(\det G(\tilde{\omega}_m) = 0\). The \(\gamma_m^n\) are then obtained by solving the system (2.103), which is linear once \(\tilde{\omega}_m\) has been found. Note that it is not clear \textit{a priori} how many solutions there are, and this number is generally not the same as the number of eigenvalues below the absorption cutoff frequency in the nominal system.

Next, we impose canonical commutation relations on the \(c_m\). As in the absorptive regime this calculation is lengthy, so we defer details to Appendix B, where it is found that the equal-time commutation relation (2.65) holds if the dispersive electromagnetic modes are normalized as specified there. It is also shown there that for equal times,

\[
\left[ c_m', s_{\Omega m}^\dagger \right] = 0, \quad (2.106)
\]

as required.

### 2.3.7 Master equations

We define

\[
\tilde{D}_m(r) \equiv \sum_n \gamma_n^{m*} D_n(r), \quad (2.107)
\]

\[
\tilde{B}_m(r) \equiv \sum_n \gamma_n^{m*} B_n(r), \quad (2.108)
\]

which are shown below to be the respective displacement and magnetic induction mode fields of polariton \(m\) in the transparent regime.

Equation (2.103) can be reworked into a master equation in terms of \(\tilde{D}_m(r)\) or \(\tilde{B}_m(r)\). We multiply (2.103) by \(\omega_n^2\), and make use of (2.44) and the nominal system master equation (2.41) for the displacement field. Employing standard vector identities, this then yields master equations for the \(D\) and \(B\) fields

\[
\frac{\tilde{\omega}_m^2}{c^2} \tilde{D}_m(r) - \nabla \times \nabla \times (1 - \Gamma(r, \tilde{\omega}_m)) \cdot \tilde{D}_m(r) = 0, \quad (2.109)
\]

\[
\frac{\tilde{\omega}_m^2}{c^2} \tilde{B}_m(r) - \nabla \times (1 - \Gamma(r, \tilde{\omega}_m)) \cdot \nabla \times \tilde{B}_m(r) = 0, \quad (2.110)
\]
It is also possible to obtain equations analogous to (2.38). Substituting the expressions (2.67, 2.68) into the first of (2.1), making use of the harmonicity of the $c$ operators to take the time derivative explicitly, and taking the commutator of the resultant equation with $c_m^\dagger$ yields an equation analogous to the first of (2.38); the second can be obtained by from the first in conjunction with (2.110). Thus,

$$-i\tilde{\omega}_m \mu_0 \tilde{D}_m(r) = \nabla \times \tilde{B}_m(r),$$  

(2.111)

$$i\tilde{\omega}_m \varepsilon_0 \tilde{B}_m(r) = \nabla \times \left[ (1 - \Gamma(r, \tilde{\omega}_m)) \cdot \tilde{D}_m(r) \right],$$

and analogous to (2.42), we see that for every mode $m$, there is another mode $\tilde{m}$ with mode fields and frequency given by

$$\left( \tilde{D}_m(r), \tilde{B}_m(r) \right) = \left( \tilde{D}_m^*(r), -\tilde{B}_m^*(r) \right), \quad (2.112)$$

$$\tilde{\omega}_m = \tilde{\omega}_m.$$

Eqs. (2.111) are just the time independent Maxwell curl equations for dispersive media, which can be obtained in na"ıve ways, without a Hamiltonian or reference to a medium. Thus the curl equations (2.111) and the master equations (2.109) and (2.110) are completely model-independent, retaining no trace of our microscopic model whatsoever, but depending only on the response tensor $\Gamma$. Therefore the dispersive modes can be obtained by any method at all, for example using an FDTD method [93], a local eigensolver [11], the cutting surface method [94], or an iterative scheme. Given the dispersive electromagnetic modes and dispersion relation, the associated medium component of the polariton modes is given immediately by (2.101). Thus polariton solutions can be constructed from dispersive modes with little extra effort.

What is not obvious from such na"ıve ways of finding the mode fields are their normalization conditions. In terms of the mode fields, the field normalization conditions, derived in Appendix B, become

$$\frac{1}{\varepsilon_0} \int_V d\mathbf{r} \; \tilde{D}_m^*(r) \cdot \left[ 1 - \Gamma(r, \tilde{\omega}_m) + \frac{\tilde{\omega}_m}{2} \Gamma'(r, \tilde{\omega}_m) \right] \cdot \tilde{D}_m(r) = \frac{\hbar \tilde{\omega}_m}{2},$$  

(2.113)

$$\frac{1}{\mu_0} \int_V d\mathbf{r} \; \tilde{B}_m^*(r) \cdot \tilde{B}_m(r) + \left( \frac{c}{\tilde{\omega}_m} \right)^2 \frac{1}{\mu_0} \int_V d\mathbf{r} \; \left( \nabla \times \tilde{B}_m(r) \right) \cdot \tilde{D}_m^*(r) \cdot \left( \tilde{\omega}_m \Gamma'(r, \tilde{\omega}_m) \cdot (\nabla \times \tilde{B}_m(r)) \right) = \frac{\hbar \tilde{\omega}_m}{2},$$  

(2.114)

---

\(^2\)An alternative form for (2.114) is given in (B.19).
where we have defined the material dispersion
\[ \Gamma' (\mathbf{r}, \tilde{\omega}_m) \equiv \frac{\partial}{\partial \omega} \Gamma (\mathbf{r}, \omega) \bigg|_{\tilde{\omega}_m}. \] (2.115)

Note that (2.114) follows from (2.113) using (2.111). By comparison with (2.44) for the nondispersive case, we see that in the dispersive case we have obtained an extra term in each normalization condition which is proportional to \( \Gamma' \), and which is due to the medium contribution. We note here that although only a single frequency derivative appears in (2.113) and (2.114), the results are exact—they do not rely on perturbation theory. Furthermore, these normalization conditions are independent of the choice of nominal system and are straightforward for the practitioner to use. Having obtained the dispersive modes and dispersion relation using any preferred method, the modes can be normalized using only the known macroscopic response function \( \Gamma (\mathbf{r}, \tilde{\omega}_m) \).

Having obtained the \( \gamma_{n \mathbf{r}}^m \) or \( \tilde{\mathbf{D}}_m (\mathbf{r}) \), the the coefficients for the medium component of the polariton mode field is given by (2.101), which using the definition of \( \tilde{\mathbf{D}}_m (\mathbf{r}) \) can be written in the form
\[ \left\{ \begin{array}{c} \alpha_{\mathbf{r}' \mathbf{r}}^m (\mathbf{r}) \\ \beta_{\mathbf{r}' \mathbf{r}}^m (\mathbf{r}) \end{array} \right\} = -\sqrt{\frac{\hbar}{2 \varepsilon_0 n^2 (\mathbf{r}, \omega) \Omega' (\mathbf{r})}} \left( \frac{1}{\hbar \omega_m + \hbar \Omega'} \right) \tilde{\mathbf{D}}^*_m (\mathbf{r}) \cdot \Lambda (\mathbf{r}, \Omega'). \] (2.116)

At this point we can make an interesting connection with much earlier work. In 1921, Brillouin [71] derived an expression for the cycle-averaged energy density of electromagnetic fields in dispersive dielectrics, which has been used to quantize the electromagnetic field in dispersive but completely nonabsorptive systems [100][101]. Brillouin’s derivation made use of slow-variation approximations, including the the requirement of near monochromaticity and the rotating wave approximation; in our notation and system of units, and for nonmagnetic media, his expression applied to the cycle-averaged energy of a single excited transparent-regime transverse polariton mode is
\[ H_{BR} = \int_V d\mathbf{r} \frac{1}{\varepsilon_0} \left( \frac{\partial (\omega n^2 (\mathbf{r}, \omega))}{\partial \omega} \right) \frac{1}{n^4 (\mathbf{r}, \omega)} \tilde{\mathbf{D}}^*_m (\mathbf{r}) \cdot \tilde{\mathbf{D}}_m (\mathbf{r}) + \frac{1}{\mu_0} \tilde{\mathbf{B}}^*_m (\mathbf{r}) \cdot \tilde{\mathbf{B}}_m (\mathbf{r}), \] (2.117)
where \( n^2 (\mathbf{r}, \omega) \equiv \varepsilon (\mathbf{r}, \omega) / \varepsilon_0 \). Performing the integral and making use of the normalization conditions (2.113) and (2.114), we find agreement with our result for the energy of a single polariton as obtained from (2.59), although here we have not resorted to any approximations in the derivation, and our expressions are not cycle-averaged. Further interpretation of the normalization condition Eq. (2.113) in terms of energy propagation is given in Ch. 3 after Eq. (3.18). Finally, we note here that for a medium with a countable
number of poles in $\varepsilon(\omega)$, Candelas [102] presented an expression for the vacuum state energy that parallels Eq. (2.117) but is applicable in the absorptive regime; the caveat is that $\varepsilon(\omega)$ employed must be subject to Feynman boundary conditions.

Returning now to the absorptive regime, we define

$$\tilde{D}_{\Omega m}(\textbf{r}) \equiv \sum_n \gamma_{n m}^\Omega \textbf{D}_n(\textbf{r}),$$

(2.118)

$$\tilde{B}_{\Omega m}(\textbf{r}) \equiv \sum_n \gamma_{n m}^\Omega \textbf{B}_n(\textbf{r}),$$

(2.119)

which we demonstrate below to be the respective $\textbf{D}$ and $\textbf{B}$ field modes of the polariton $\Omega_m$, although we note their units differ from those of the displacement and magnetic induction fields because of the continuous frequency index $\Omega$.

We find that (2.90) can also be reworked into a master equation in terms of either $\tilde{D}_{\Omega m}(\textbf{r})$ or $\tilde{B}_{\Omega m}(\textbf{r})$. Following the procedure used to obtain (2.109) and (2.110) from (2.103), Equation (2.90) can be written as a master equation for $\textbf{D}$ or $\textbf{B}$:

$$\frac{\Omega^2}{c^2} \tilde{D}_{\Omega m}(\textbf{r}) - \nabla \times \nabla \times \left( (1 - \text{Re} \Gamma (\textbf{r}, \Omega)) \cdot \tilde{D}_{\Omega m}(\textbf{r}) \right) = \frac{Z_{\Omega m}}{\pi} \nabla \times \left( \text{Im} \Gamma (\textbf{r}, \Omega) \cdot \nabla \times \tilde{D}_{\Omega m}(\textbf{r}) \right),$$

(2.120)

$$\frac{\Omega^2}{c^2} \tilde{B}_{\Omega m}(\textbf{r}) - \nabla \times \nabla \times \left( (1 - \text{Re} \Gamma (\textbf{r}, \Omega)) \cdot \nabla \times \tilde{B}_{\Omega m}(\textbf{r}) \right) = \frac{Z_{\Omega m}}{\pi} \nabla \times \left( \text{Im} \Gamma (\textbf{r}, \Omega) \cdot \nabla \times \tilde{B}_{\Omega m}(\textbf{r}) \right).$$

(2.121)

Eqs. (2.120, 2.121) have a number of salient features, which for brevity we consider only for the $\textbf{B}$ master equation (2.121) in what follows. First, since the nominal system mode amplitudes and index profile $n(\textbf{r})$ do not appear explicitly in (2.121), we see that the eigenvalue $Z_{\Omega m}$ is independent of the choice of nominal system. Furthermore, while we have introduced an underlying oscillator model to describe the optical response of the medium, in the eigenvalue equation for the polaritons all the details of the oscillator model have disappeared, except that it leads to the physically significant $\Gamma^{ij}(\textbf{r}, \Omega)$.

Second, we note that (2.121) can be written in the form

$$\frac{\Omega^2}{c^2} \tilde{B}_{\Omega m}(\textbf{r}) - \nabla \times (1 - \Gamma (\textbf{r}, \Omega; Z_{\Omega m})) \cdot \nabla \times \tilde{B}_{\Omega m}(\textbf{r}) = 0,$$

(2.122)

with

$$\Gamma (\textbf{r}, \Omega; Z) \equiv \text{Re} \Gamma (\textbf{r}, \Omega) - \frac{Z}{\pi} \text{Im} \Gamma (\textbf{r}, \Omega),$$

(2.123)

where since $Z$ is real, $\Gamma (\textbf{r}, \Omega; Z)$ is also real. Equation (2.122) is just a master equation that would result in the artificial model of a dispersive, nonabsorptive dielectric response, with an effective dispersive dielectric tensor given by $\Gamma (\textbf{r}, \Omega; Z)$ for a given fixed $Z$. 


This means that we can find the modes of the physical absorptive system by solving for the modes and discrete band structure of the family of artificial but nonabsorptive systems characterized by \( \Gamma (r, \Omega; Z) \) for all \( Z \in (-\infty, \infty) \), and thereby scanning the \((Z, \Omega)-plane\) (positive \( \Omega \)) for solutions. This is the inverse process of solving the eigenvalue equation (2.90) for eigenvalues \( Z \) at each \( \Omega \). Yet it yields the same image in the \((Z, \Omega)-plane\). For \( Z = 0 \), (2.122) becomes just the master equation that arises if absorption is neglected. Thus the mode fields and dispersion relation obtained by making the replacement \( \Gamma (r, \Omega) \rightarrow \text{Re} \Gamma (r, \Omega) \) are solutions to the full absorptive problem with \( Z = 0 \).

As in the transparent regime, we can identify effective Maxwell curl equations analogous to (2.38), even in the absorptive regime. The derivation is parallel to that which leads to (2.111). The equations are

\[
-i \Omega \mu_0 \tilde{D}_{\Omega m}(r) = \nabla \times \tilde{B}_{\Omega m}(r),
\]

\[
i \Omega \varepsilon_0 \tilde{B}_{\Omega m}(r) = \nabla \times \left[ (1 - \Gamma (r, \Omega; Z_{\Omega m})) \cdot \tilde{D}_{\Omega m}(r) \right],
\]

and for every mode \( \Omega m \), there is a physical conjugate mode \( \Omega \overline{m} \) with mode fields and eigenvalue \( Z_{\Omega \overline{m}} \) given by

\[
\left( \tilde{D}_{\Omega \overline{m}}(r), \tilde{B}_{\Omega \overline{m}}(r) \right) = \left( \tilde{D}_{\Omega m}^*(r), -\tilde{B}_{\Omega m}^*(r) \right), \quad Z_{\Omega \overline{m}} = Z_{\Omega m}.
\]

In terms of the mode fields \( \tilde{D}_{\Omega m}(r) \) and \( \tilde{B}_{\Omega m}(r) \), defined in (2.118, 2.119), the normalization condition equivalent to (2.93) is

\[
\frac{1}{\varepsilon_0} \int_V dr \tilde{D}_{\Omega m}^*(r) \cdot \text{Im} \Gamma (r, \Omega) \cdot \tilde{D}_{\Omega m'}(r) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m})^2} \delta_{mm'},
\]

\[
c^2 \frac{1}{\mu_0} \int_V \left( \nabla \times \tilde{B}_{\Omega m}^*(r) \right) \cdot \text{Im} \Gamma (r, \Omega) \cdot \left( \nabla \times \tilde{B}_{\Omega m'}(r) \right) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m})^2} \delta_{mm'},
\]

as derived in Appendix B. Note that (2.127) follows from (2.126) using (2.124), and furthermore that like the eigenvalue equation (2.120), the normalization conditions are independent of the choice of nominal system.

The coefficients that identify the contribution from the medium fields to the TP polariton mode amplitudes \( c_{\Omega m} \) can be written in a simple form using the \( \tilde{D}_{\Omega m}(r) \). Eqs. (2.84) and (2.86) reduce to

\[
\alpha_{\Omega m}^\Omega (r) = -\sqrt{\frac{\hbar}{2 \varepsilon_0 \Omega'}} \left( P \frac{1}{\hbar \Omega - \hbar \Omega'} + Z_{\Omega m} \delta (\hbar \Omega - \hbar \Omega') \right) \tilde{D}_{\Omega m}^*(r) \cdot \Lambda (r, \Omega'),
\]

where

\[
\Lambda (r, \Omega') = \frac{\mu_0}{2 \varepsilon_0} \left( P \frac{1}{\hbar \Omega - \hbar \Omega'} + Z_{\Omega m} \delta (\hbar \Omega - \hbar \Omega') \right) \frac{\Gamma (r, \Omega; Z_{\Omega m})}{\mu_0} \tilde{D}_{\Omega m}(r) \cdot \Lambda (r, \Omega'),
\]

and

\[
\frac{1}{\varepsilon_0} \int_V \frac{\Gamma (r, \Omega; Z_{\Omega m})}{\mu_0} \tilde{D}_{\Omega m}(r) \cdot \Lambda (r, \Omega') = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m})^2} \delta_{mm'},
\]

as derived in Appendix B.
\[ \beta_{\Omega m}^\text{Im} (r) = -\sqrt{\frac{\hbar}{2\varepsilon_0 \Omega'}} \left( \frac{1}{\hbar \Omega + \hbar \Omega'} \right) \tilde{D}_m^\ast (r) \cdot \Lambda (r, \Omega'), \tag{2.129} \]

so that given the electromagnetic component mode field \( \tilde{D}_m^\ast (r) \) of a polariton, the medium component field follows immediately.

### 2.3.8 Inverse transformation

Having obtained the canonical transformation to the polariton basis, we now determine the inverse transformation. The procedure is to expand the medium and nominal system operators in polariton operators, then calculate the various commutators of the nominal system and medium operators with the polariton operators two ways: once using the expansion (2.69, 2.76, 2.94) of the polariton operators in terms of nominal system and medium operators, and once using the expansion of the nominal system and medium operators in terms of the polaritons. Comparing these two results, and then using the relations (2.81, 2.99), yields the inverse transformation

\[ a_m = \sum_n \int_{\Omega_y}^\infty d\Omega \left\{ (\alpha_n^\Omega m)^* c_{\Omega m} - (\beta_n^\Omega m)^* c_{\Omega m}^\dagger \right\} + \sum_n \left\{ (\alpha_n^m)^* c_n - (\beta_n^m)^* c_n^\dagger \right\}, \tag{2.130} \]

\[ \psi_{\Omega} (r) = \sum_m \int_{\Omega_y}^\infty d\Omega' \left\{ (\alpha_{\Omega}^m (r))^* c_{\Omega m} - (\beta_{\Omega}^m (r))^* c_{\Omega m}^\dagger \right\} \]

\[ + \sum_m \left\{ (\alpha_{\Omega}^m (r))^* c_m - (\beta_{\Omega}^m (r))^* c_m^\dagger \right\} + \sum_m (\rho_{\Omega m} (r))^* s_{\Omega m}. \tag{2.131} \]

Making use of the inverse transformation (2.130) and the definitions (2.118) and (2.107) of \( \tilde{D}_{\Omega m} (r) \) and \( \tilde{D}_m (r) \) in the expression (2.46) for the displacement field \( D (r, t) \) in terms of the nominal system operators and modes \( a_m \) and \( D_m (r) \), we obtain the expression (2.67) for the displacement field in terms of the polariton operators and polariton displacement field modes. Similarly, we obtain the corresponding expression (2.68) for the field \( B (r, t) \) in terms of polariton operators and polariton \( B \) field modes. Clearly, \( \tilde{D}_{\Omega m} (r) \) and \( \tilde{B}_{\Omega m} (r) \) are the displacement and magnetic induction field modes for the polariton \( c_{\Omega m} \), and \( \tilde{D}_m (r) \) and \( \tilde{B}_m (r) \) are mode fields for the \( c_m \). We note that the dispersive (tilde) electromagnetic mode fields provide a nonorthogonal basis for the physical electromagnetic fields.
2.3.9 Other fields

Here we briefly describe the electric and polarization fields in terms of the polariton modes. Substituting (2.15) and (2.131) into the expression (2.19) for the polarization field, then making use of (2.28, 2.29, 2.72, 2.112, 2.116, 2.123, 2.125, 2.128, 2.129) yields an expression for the polarization, separated into transverse and longitudinal pieces as

\[
P(r, t) = P_T(r, t) + P_L(r, t),
\]

where

\[
P_T(r, t) = \sum_m c_m \tilde{P}_m(r) + \sum_m \int_{\Omega_g} d\Omega c_{\Omega m} \tilde{P}_{\Omega m}(r) + c.c.,
\]

\[
P_L(r, t) = \sum_m \int_{\Omega_g} d\Omega s_{\Omega m} \tilde{P}^{(L)}_{\Omega m}(r) + c.c.,
\]

with the mode polarization fields given by

\[
\tilde{P}_m(r) = \Gamma(r, \tilde{\omega}_m) \cdot \tilde{D}_m(r),
\]

\[
\tilde{P}_{\Omega m}(r) = \Gamma(r, \Omega; Z_{\Omega m}) \cdot \tilde{D}_{\Omega m}(r),
\]

\[
\tilde{P}^{(L)}_{\Omega m}(r) = \sqrt{\frac{\hbar \varepsilon_0}{2\Omega}} \left( \nabla \phi^{\Omega m}(r) \right)^*.
\]

It then follows that

\[
E(r, t) = \frac{1}{\varepsilon_0} \left( D(r, t) - P(r, t) \right) \equiv E_T(r, t) + E_L(r, t),
\]

where

\[
E_T(r, t) = \sum_m c_m \tilde{E}_m(r) + \sum_m \int_{\Omega_g} d\Omega c_{\Omega m} \tilde{E}_{\Omega m}(r) + c.c.,
\]

\[
E_L(r, t) = \sum_m \int_{\Omega_g} d\Omega s_{\Omega m} \tilde{E}^{(LP)}_{\Omega m}(r) + c.c.,
\]

with

\[
\tilde{E}_m(r) = \frac{1}{\varepsilon_0} \left( (1 - \Gamma(r, \tilde{\omega}_m)) \cdot \tilde{D}_m(r) \right),
\]

\[
\tilde{E}_{\Omega m}(r) = \frac{1}{\varepsilon_0} \left( (1 - \Gamma(r, \Omega; Z_{\Omega m})) \cdot \tilde{D}_{\Omega m}(r) \right),
\]

\[
\tilde{E}^{(L)}_{\Omega m}(r) = -\sqrt{\frac{\hbar}{2\Omega \varepsilon_0}} \left( \nabla \phi^{\Omega m}(r) \right)^*.
\]

so that \( \tilde{E}_{\Omega m}(r) \) is of the form expected from (2.111, 2.124).
Whereas the Hamiltonian (2.8) with commutators (2.10,2.14), yields dynamics of the Maxwell equations augmented by a fluctuation polarization, here we find that the polariton mode fields satisfy the time-independent Maxwell equations with an effective dielectric response \( \Gamma (\mathbf{r}, \Omega; Z_{\Omega m}) \), but without any additional fluctuation term.

Of course, \( \mathbf{H} \) field expressions are obtained from corresponding \( \mathbf{B} \) field expressions through division by \( \mu_0 \).

### 2.4 Systems with continuously labeled modes

For many systems of interest, the modes are labelled continuously or both continuously and discretely in combination. Examples include waveguides, Bragg gratings, photonic crystals, multilayer thin films, coupled microring structures, and periodic quantum well structures. Often the modes of interest are labeled by a discrete-continuous pair \((u, k)\), where \( k \) is the crystal wave vector and \( u \) is a “band index.” Then in the equations of Section 2.3, the discrete mode index \( m \) is replaced by \( m \rightarrow (u, k) \). As an example, we consider photonic crystals of arbitrary dimensionality; the equations involving other systems with discrete-continuous labeling can be derived analogously. Since a related discussion for nondispersive, nonabsorptive systems was presented earlier [3], we here focus only on the full dispersive system (tilde) modes, which are solutions to the master equations described in Section 2.3.

Systems with continuously labeled modes arise from systems subject to periodic boundary conditions for which the normalization region \( V \) is extended infinitely in one or more orthogonal directions. The cross-section of the original finite \( V \) is always invariant in any direction perpendicular to the direction(s) of extension. Then the extended \( V \) can be written as \( V = D_\infty \otimes F \), where \( F \) is a finite “cross-section” in dimensions in which \( V \) is of finite extent, and \( D_\infty \) is the subspace of the dimensions of infinite extent, which has dimensionality \( d \in \{1, 2, 3\} \).

For these continuous systems, the derivation of Section 2.3 holds, but subject to the extension of \( V \), and replacement of summations according to

\[
\sum_m \rightarrow \sum_u C_\infty \int d^d k,
\]

where the integral is over the region of reciprocal space corresponding to the real space region \( D_\infty \). Making these replacements requires simultaneous modification of the other
quantities introduced in Section 2.3, which involve a factor $C_\infty$ which depends on $D_\infty$ according to

$$
\begin{array}{c|c}
\text{direction} & \text{factor} \\
\hline
i \text{ axis} & \frac{L_i}{2\pi} \\
ij \text{ plane} & \frac{A_{ij}}{(2\pi)^2} \\
ijl \text{ space} & \frac{V}{(2\pi)^3}
\end{array}
$$

(2.145)

where $L_i$ is the extent of the original finite $V$ in the $i$-direction, $A_{ij}$ is the area of the original finite $V$ in the $ij$-plane, and recall that $V$ is the volume of the original finite normalization region $V$. In the interest of brevity, we quote only end results whenever possible, and in what follows we work out the continuous-$k$ equations in the context of a photonic crystal.

### 2.4.1 Example: photonic crystal

Photonic crystals [103] are a particularly interesting examples of inhomogeneous media, defined by the property

$$
\Gamma^{ij} (r + \mathbf{R}, \Omega) = \Gamma^{ij} (r, \Omega)
$$

(2.146)

where $\mathbf{R}$ is any lattice vector. Interesting systems exist for 1D, 2D, and 3D periodic lattices. We thus take the coupling constant and nominal index to have the periodicity of the lattice:

$$
\Lambda^{ij} (r + \mathbf{R}, \Omega) = \Lambda^{ij} (r, \Omega),
$$

$$
\Gamma^{ij}_0 (r + \mathbf{R}) = \Gamma^{ij}_0 (r).
$$

(2.147)

Numerous codes for the calculation of nondispersive, nonabsorptive band structures are freely available [10][11], so it is easy to obtain the nominal bands and nominal system modes.

As discussed above, we write $m \to (u, k)$, where $k$ is the wave vector which is restricted to the first Brillouin zone, and $u$ is a discrete band index. Under the transition from discrete to continuous $k$, the governing equations (2.111, 2.124) and thus also the master equations (2.109, 2.110) are unchanged. They retain their physical conjugate properties (2.112, 2.125), and the physical conjugate to a nominal mode $(u, k)$ is another mode at $-k$. Using this property to label the bands at $-k$, we can write

$$
(u, k) = (u, -k).
$$

(2.148)
So the modes are first determined in one half of \( \mathbf{k} \)-space (e.g. in 3D, the first Brillouin zone for \( k_z \geq 0 \)), with the modes in the other half of \( \mathbf{k} \)-space given by

\[
\left( \tilde{\mathbf{D}}_{u(-k)}(\mathbf{r}), \tilde{\mathbf{B}}_{u(-k)}(\mathbf{r}) \right) = \left( \mathbf{D}^*_{uk}(\mathbf{r}), -\mathbf{B}^*_{uk}(\mathbf{r}) \right),
\]

(2.149)

with \( \tilde{\omega}_{u(-k)} = \tilde{\omega}_{uk} \). The analogous relation holds between the \( \mathbf{D}_{\Omega uk}(\mathbf{r}) \) and \( \mathbf{B}_{\Omega uk}(\mathbf{r}) \).

The electromagnetic fields are then expanded in terms of polariton operators and mode fields as

\[
\mathbf{D}(\mathbf{r}, t) = \sum_u \int d^d k \left( c_{uk} \mathbf{D}_{uk}(\mathbf{r}) + c_{uk}^\dagger \mathbf{D}^*_{uk}(\mathbf{r}) \right)
+ \sum_u \int d^d k \int_{\Omega_\gamma} d\Omega \left[ c_{\Omega uk} \mathbf{D}_{\Omega uk}(\mathbf{r}) + c_{\Omega uk}^\dagger \mathbf{D}^*_{\Omega uk}(\mathbf{r}) \right],
\]

(2.150)

\[
\mathbf{B}(\mathbf{r}, t) = \sum_u \int d^d k \left( c_{uk} \mathbf{B}_{uk}(\mathbf{r}) + c_{uk}^\dagger \mathbf{B}^*_{uk}(\mathbf{r}) \right)
+ \sum_u \int d^d k \int_{\Omega_\gamma} d\Omega \left[ c_{\Omega uk} \mathbf{B}_{\Omega uk}(\mathbf{r}) + c_{\Omega uk}^\dagger \mathbf{B}^*_{\Omega uk}(\mathbf{r}) \right],
\]

(2.151)

where the \( \mathbf{k} \)-integrals are over the \((d\)-dimensional\) first Brillouin zone, and where we have used (2.149). The \( (\text{equal-time}) \) canonical commutation relations are then

\[
\left[ c_{uk}, c_{uk'}^{\dagger} \right] = \delta_{uu'} \delta^d(\mathbf{k} - \mathbf{k}'), \quad (2.152)
\]

\[
\left[ c_{\Omega uk}, c_{\Omega' uk'}^{\dagger} \right] = \delta_{uu'} \delta(\Omega - \Omega') \delta^d(\mathbf{k} - \mathbf{k}'), \quad (2.153)
\]

\[
\left[ s_{\Omega uk}, s_{\Omega' uk'}^{\dagger} \right] = \delta_{uu'} \delta(\Omega - \Omega') \delta^d(\mathbf{k} - \mathbf{k}'), \quad (2.154)
\]

where \( \delta^d(\mathbf{k} - \mathbf{k}') \) is a delta function with dimensionality \( d \), and where all other equal-time commutators between the \( c \) and \( s \) operators vanish. The mode fields are normalized according to

\[
\frac{1}{\varepsilon_0} \int d\mathbf{r} \tilde{\mathbf{D}}_{uk'}^* (\mathbf{r}) \cdot \left[ 1 - \Gamma(\mathbf{r}, \tilde{\omega}_{uk}) + \frac{\tilde{\omega}_{uk}}{2} \Gamma'(\mathbf{r}, \tilde{\omega}_{uk}) \right] \cdot \tilde{\mathbf{D}}_{uk}(\mathbf{r}) = \frac{\hbar \tilde{\omega}_{uk}}{2} \delta^d(\mathbf{k} - \mathbf{k}'), \quad (2.155)
\]

\[
\frac{1}{\varepsilon_0} \int d\mathbf{r} \tilde{\mathbf{D}}_{\Omega uk'}^* (\mathbf{r}) \cdot \text{Im} \Gamma(\mathbf{r}, \Omega) \cdot \tilde{\mathbf{D}}_{\Omega uk}(\mathbf{r}) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega uk})^2} \delta_{uu'} \delta^d(\mathbf{k} - \mathbf{k}'). \quad (2.156)
\]

The normalization of the \( \mathbf{B}_{uk} \) and \( \mathbf{B}_{\Omega uk} \) fields follows from (2.111) and (2.124).
Since the full dielectric function $\Gamma^{ij}(r, \Omega)$ is periodic, Bloch’s theorem applied to the master equations (2.109, 2.110) guarantees the existence of mode fields in Bloch form. Choosing convenient scaling factors, we then write the $\tilde{D}_{\omega k}(r)$, $\tilde{B}_{\omega k}(r)$, $\tilde{D}_{\Omega \omega k}(r)$, and $\tilde{B}_{\Omega \omega k}(r)$ in Bloch form as

$$\tilde{D}_{\omega k}(r) \equiv \sqrt{\frac{C_{\infty}}{V}} \tilde{d}_{\omega k}(r) e^{ik \cdot r},$$

$$\tilde{B}_{\omega k}(r) \equiv \sqrt{\frac{C_{\infty}}{V}} \tilde{b}_{\omega k}(r) e^{ik \cdot r},$$

$$\tilde{D}_{\Omega \omega k}(r) \equiv \sqrt{\frac{C_{\infty}}{V}} \tilde{d}_{\Omega \omega k}(r) e^{ik \cdot r},$$

$$\tilde{B}_{\Omega \omega k}(r) \equiv \sqrt{\frac{C_{\infty}}{V}} \tilde{b}_{\Omega \omega k}(r) e^{ik \cdot r},$$

where lower case characters represent the periodic Bloch fields, which have the periodicity of the lattice; so for any lattice vector $\mathbf{R}$,

$$\tilde{d}_{\omega k}(r) = \tilde{d}_{\omega k}(r + \mathbf{R}),$$

$$\tilde{b}_{\omega k}(r) = \tilde{b}_{\omega k}(r + \mathbf{R}),$$

and the same holds for the $\tilde{d}_{\Omega \omega k}$ and $\tilde{b}_{\Omega \omega k}$. Normalization of these periodic Bloch fields involves a unit cell $V_{\text{cell}}$, which can be chosen to be any region which tiles the full normalization region $V$ via translations by lattice vectors.

In the transparent regime, the normalization condition for the $\tilde{d}_{\omega k}(r)$ is

$$\frac{1}{\varepsilon_0} \int_{V_{\text{cell}}} \frac{dr}{V_{\text{cell}}} \tilde{d}_{\omega k}^*(r) \cdot \left(1 - \Gamma(r, \tilde{\omega}_{\omega k}) + \frac{\tilde{\omega}_{\omega k} \Gamma'(r, \tilde{\omega}_{\omega k})}{2}\right) \tilde{d}_{\omega k}(r) = \frac{\hbar \tilde{\omega}_{\omega k}}{2},$$

where $\Gamma'$ is defined in (2.115), and where the spatial integral is over the unit cell, which has volume $V_{\text{cell}}$. In the absorptive regime, the normalization condition is

$$\frac{1}{\varepsilon_0} \int_{V_{\text{cell}}} \frac{dr}{V_{\text{cell}}} \tilde{d}_{\Omega \omega k}^*(r) \cdot \text{Im} \Gamma(r, \Omega) \cdot \tilde{d}_{\Omega \omega' k}(r) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega \omega k})^2} \delta_{\omega \omega'}. $$

We note that under a transition from discrete to continuous wave vectors, the variables which appear in Sections 2.1–2.3 are slightly modified as follows. Below, we use $\hat{Q}$ to denote a generic quantity with discretely labeled wave vector and $Q$ to denote the corresponding quantity with continuously labeled wave vector. Then the transition to from discrete to continuous labeling is given by

$$\hat{Q} \rightarrow \left( \frac{1}{C_{\infty}} \right)^{N/2} Q,$$
for some $N$; $N = 1$ gives the transition for $Q$ which is any of the variables $a_{uk}$, $D_{uk}(r)$, $B_{uk}(r)$, $A_{uk}(r, \Omega)$, $c_{uk}$, $s_{\Omega k}$, $\rho^{\Omega k}(r)$, $\phi^{\Omega k}(r)$, $\alpha^{\Omega k}_{\Omega k}(r)$, $\beta^{\Omega k}_{\Omega k}(r)$, $\gamma^{\Omega k}_{\Omega k}(r)$, $\alpha^{uk}_{\Omega k}(r)$, $\beta^{uk}_{\Omega k}(r)$ and $\gamma^{uk}_{\Omega k}(r)$; and $N = 2$ gives the transition for $c^{\Omega k}_{\Omega k}$, $\beta^{\Omega k}_{\Omega k}$, $\gamma^{\Omega k}_{\Omega k}$, $\alpha^{uk}_{\Omega k}$, $\beta^{uk}_{\Omega k}$, and $\gamma^{uk}_{\Omega k}$.

2.5 Calculating polaritons: summary and user’s guide

We emphasize here that the effort in constructing the formalism has yielded a theory that is easy to use in the end. Calculating the polariton modes is in fact straightforward, and here we review the process of calculating polariton modes for artificially structured inhomogeneous media. Most importantly, in both the transparent and absorbing regimes, the photonic component of the polariton modes can be obtained from the dielectric response function, after which the corresponding medium field component modes are obtained trivially; together a photonic component and its associated medium field comprise the components of a polariton. For the sake of simplicity, we take the dielectric response to be isotropic in this section: $\varepsilon_{ij}(r, \omega) = \varepsilon(r, \omega) \delta_{ij}$. We employ box normalization, and seek first the photonic component of the polariton modes of the system. We label modes by $m$; for example, for a photonic crystal we might have $m = (u, k)$, where $u$ is a band index and $k$ denotes the wave vector, which until Section 2.3.3 we take to be a discrete label.

We suppose the user is given a causal $\varepsilon(r, \omega)$ that describes the system of interest. In this thesis we work in terms of a response function $\Gamma(r, \omega)$ defined in (2.6), which we take to be isotropic in this section, and which is constructed from $\varepsilon(r, \omega)$ using (q.v. Eq. (2.32))

$$\Gamma(r, \omega) = \left(1 - \frac{\varepsilon_0}{\varepsilon(r, \omega)}\right) \delta_{ij}. \quad (2.162)$$

The coupling tensor of the underlying model of the medium is chosen to satisfy Eq. (2.29), in which it is defined in terms of the dielectric response function; thus

$$\Lambda(r, \omega) \equiv \sqrt{\frac{2\omega}{\pi} \Im \Gamma(r, \omega)}. \quad (2.163)$$

The electromagnetic fields are given in terms of the polariton basis by

$$\mathbf{D}(r, t) = \sum_m \left[ c_m \mathbf{\bar{D}}_m(r) + c^\dagger_m \mathbf{\bar{D}}^*_m(r) \right] + \sum_m \int d\Omega \left[ c_{\Omega m} \mathbf{\bar{D}}_{\Omega m}(r) + c^\dagger_{\Omega m} \mathbf{\bar{D}}^*_{\Omega m}(r) \right], \quad (2.164)$$
\[ \mathbf{B}(r,t) = \sum_m \left[ c_m \tilde{B}_m(r) + c_m^\dagger \tilde{B}^*_m(r) \right] + \sum_m \int d\Omega \left[ c_{\Omega m} \tilde{B}_{\Omega m}(r) + c_{\Omega m}^\dagger \tilde{B}^*_{\Omega m}(r) \right], \quad (2.165) \]

where the \( \tilde{D}_m(r) \) and \( \tilde{D}_{\Omega m}(r) \) are the respective mode fields of the polaritons in the transparent and absorbing frequency regimes, and the frequency integral is taken over the absorbing regime.

### 2.5.1 Transparent (dispersive) regime

First we consider calculating the mode fields and eigenfrequencies in the nonabsorbing regime, i.e., for frequencies at which \( \text{Im} \varepsilon(r,\omega) \) vanishes. The dispersion relation \( \tilde{\omega}_m \) and electromagnetic field modes \( \tilde{B}_m(r) \) of the system described by \( \varepsilon(r,\omega) \) are calculated according to the usual Maxwell master equation (2.110) using any preferred method, such as an FDTD method [93], a local eigensolver [11], or an iterative scheme. For iterative schemes, the modes of a nominal, nondispersive and nonabsorptive medium characterized by an index profile \( n_0(r) \) can be used to provide initial guesses for the modes. The \( \tilde{D}_m(r) \) are then obtained from the \( \tilde{B}_m(r) \) using (2.111). The normalization condition on the displacement fields (2.113) takes the form

\[ \int_V d\mathbf{r} \tilde{D}^*_m(\mathbf{r}) \left[ \frac{1}{\varepsilon(r,\tilde{\omega}_m)} - \frac{\tilde{\omega}_m}{2} \frac{\partial}{\partial \omega} \left( \frac{1}{\varepsilon(r,\omega)} \right) \right] \cdot \tilde{D}_m(\mathbf{r}) = \frac{\hbar \tilde{\omega}_m}{2}. \quad (2.166) \]

This differs from the nondispersive case by the appearance of the second term in the square parentheses.

If so desired, the associated medium fields can be explicitly constructed with little effort from (2.116) using (2.163).

### 2.5.2 Absorbing regime

At frequencies at which the medium is absorbing, the polariton spectrum is continuous, with solutions at all frequencies – even for a single given wave vector, for example. At frequencies close to those of the bands of the limiting nonabsorptive but dispersive structure, the polariton modes typically have more electromagnetic character, while away from the dispersive bands, the modes have more medium field character to them. Since the spectrum is continuous, the frequency \( \omega \) is no longer an eigenvalue. Instead the eigenvalue is the resonant oscillator amplitude \( Z \), which is proportional to the contribution to the polariton from medium fields with natural frequency equal to that of the polariton.
Here are two methods for calculating polaritons:

Method (i): This is a method that can easily leverage codes for the photonic modes of dispersive systems. It is shown in Section 2.3 that in the absorptive regime, the polariton mode displacement fields are solutions to the dispersive, nonabsorptive Maxwell master equation, but with an effective $Z$-dependent dielectric tensor $\varepsilon (r, \omega; Z)$. Thus

$$\nabla \times \frac{\varepsilon_0}{\varepsilon (r, \omega; Z)} \cdot \nabla \times \tilde{B}_{\omega m} (r) = \frac{\omega^2}{c^2} \tilde{B}_{\omega m} (r),$$

with $\varepsilon_0/\varepsilon (r, \omega; Z) \equiv 1 - \Gamma (r, \omega; Z)$, with

$$\Gamma (r, \omega; Z) \equiv \text{Re} \Gamma (r, \omega) - Z \pi \text{Im} \Gamma (r, \omega).$$

The modes are found by scanning through values of the parameter $Z$ (which are real, $-\infty < Z < \infty$), and solving the dispersive Maxwell master equation (2.167) for eigenvalues $\omega$ and eigenmodes which give the displacement fields of the modes. Often, the modes with small $|Z|$ are the ones of most interest; they span the band width around the limiting dispersive but nonabsorptive band, and that is the band width which represents the electromagnetic fields dressed by the medium, rather than the other way around.

The displacement field modes are obtained from the $\tilde{B}_{\omega m} (r)$ using (2.124), and are then normalized using (2.126):

$$\frac{1}{\varepsilon_0} \int_V d\mathbf{r} \tilde{D}_{\omega m}^* (r) \cdot \text{Im} \Gamma (r, \omega) \cdot \tilde{D}_{\omega m'} (r) = \frac{\pi \hbar}{\pi^2 + Z^2} \delta_{m m'},$$

where $Z$ is the eigenvalue $Z_{\omega m}$.

Method (ii): This method may be computationally faster in many cases, and mostly follows the text after Equation (2.90), so for the sake of brevity we do not rewrite the equations here. A nominal, nondispersive, nonabsorptive system is selected, and its displacement field modes are taken to be a basis. The generalized Hermitian eigenvalue problem (2.90) is constructed by performing the overlap integrals (2.91) and (2.92). When the system of interest is composed of only a few types of underlying materials, the overlap integrals need not be redone at each frequency; rather, the spatial overlaps of nominal modes with each type of material can be done separately, and the dielectric response tensor components can be left as frequency-dependent parameters. The eigenproblem is then solved at each of a set of frequencies spanning the range of interest, and the eigenvectors are normalized in a way that is eigenvalue-dependent using (2.93). Then the normalized eigenvectors give the components of the displacement field modes in terms
of the nominal basis as defined in (2.118). Again, once the electromagnetic component of the polariton modes has been obtained, the medium component follows from (2.128) and (2.129) if so desired. This method has the advantage of avoiding the techniques (e.g. iteration) required to solve for the frequencies and modes of the effective dispersive system described in method (i).

2.5.3 Example calculations

Here we give two examples which demonstrate the simplicity of the approach and its applicability to real systems of interest.

2D Photonic crystal polariton bands: transparent regime

We consider a photonic crystal consisting of a 2D triangular lattice of cylindrical air holes of radius \( r/a = 0.417 \) in a dispersive dielectric background, for a frequency range in which there is no absorption. The structure is shown schematically in Fig. 2.3. Here the modes are appropriately labeled by \( m = (u, k) \) where \( u \) is the band index and \( k \) labels the (2D) wave vector which lies in the crystal plane. In the frequency range \( \nu a/c \in [0, 0.66] \) for which the bands are given, the medium exhibits normal dispersion and its index of refraction spans \( n \in [3.1, 3.59] \); the corresponding \( \Gamma \) is shown in the inset to Fig. 2.4. The nominal response in the dielectric is indicated by the pink dashed line in the inset to Fig. 2.4, and corresponds to index \( n = 3.325 \). We reiterate here that, despite the use of a nominal crystal, our results are nonperturbative. Calculation of the polariton modes begins with a calculation of the dispersive photonic component modes \( \tilde{D}_{uk}(r) \) and \( \tilde{B}_{uk}(r) \). Here is how it is done: given \( \Gamma (r, \omega) \), the dielectric constant
Figure 2.4: In-plane polariton band structure for TM modes of 2D photonic crystal with lattice constant $a$ (black lines with symbols) and the photonic bands of a corresponding nominal, nondispersive crystal (pink lines). The crystal is composed of a triangular lattice of air cylinders of radius $r/a = 0.417$ in a dispersive background medium with dielectric response $\Gamma (\omega)$ shown inset (black curve), along with the nominal nondispersive response (pink), which corresponds to index $n = 3.325$. 
\[ \varepsilon \left( \mathbf{r}, \omega \right) \] is calculated using (2.162). The photonic bands are then found using the MIT Photonic Bands Package (MPB) [10], a freely available frequency domain eigensolver for the photonic band structure and modes of nondispersive, nonabsorptive systems. An outer iterative loop over the MPB code is used to solve for the dispersive modes. If the code is thought of at each \( k \) as a mapping from a dielectric function to a set of mode frequencies, i.e. \( MPB : \varepsilon \left( \mathbf{r}, \omega \right) \to \{ \tilde{\omega}_u \} \), then the iterative loop seeks fixed points at which for some \( u \), \( \tilde{\omega}_u = \omega \). These give the dispersive mode frequencies, and their associated eigenvectors give the dispersive photonic modes, which are just the photonic component of the polariton modes. The nominal crystal frequencies are used as a starting point for the iteration at the first \( k \) point, and the frequencies obtained at a given \( k \) point are used as a starting point for the subsequent adjacent \( k \) point. The modes are normalized according to (2.166). The TM (\( E \) parallel to rod axis) dispersive band structure is plotted in black in Fig. 2.4, along with the nominal crystal band structure in pink.

We demonstrate that the medium components of the polariton modes have reasonable behavior by calculating them from the photonic components of the mode fields and eigenvalues according to (2.116). To make a definite calculation, we assume the dielectric material of the photonic crystal is a semiconductor with an electronic band gap of \( \omega_g a / 2 \pi c = 0.85 \). Now consider any dispersive mode with wave vector \( \mathbf{k} \) and unitless frequency \( \tilde{\omega}_u k a / 2 \pi c = 0.8 \) (dashed line in Fig. 2.5), which is just below the electronic band gap. In Fig. 2.5 we plot the magnitudes of the corotating and counterrotating medium coefficients \( |\alpha_{\omega k}(\mathbf{r})| \) and \( |\beta_{\omega k}(\mathbf{r})| \) as a function of unitless natural oscillator frequency \( \omega a / 2 \pi c \), in units of \( \left( a / 2 \pi c \right) \sqrt{\text{Im} \Gamma (\mathbf{r}, \omega) / (\hbar \varepsilon_0 \pi) \tilde{D}_{\omega k}(\mathbf{r})} \). The patterned area below \( \omega_g \) in Fig. 2.5 indicates the transparent regime, in which there are no medium oscillators. Other things being equal, the amplitudes of the medium coefficients are larger wherever the displacement mode field is concentrated and wherever the dielectric material is more absorptive at the medium oscillator’s natural frequency. The amplitudes also fall off with increasing frequency away from the polariton frequency.

**GaAs slab waveguide near the electronic band gap**

We next consider a single-polariton state in a dielectric slab waveguide. We take the structure to be composed of a 262 nm thick GaAs layer, and to be of infinite extent in the \( xy \) plane and bounded by perfect conductors, as depicted in Fig. 2.6. We take as
Figure 2.5: For the dispersive photonic crystal with band structure given in Fig. 2.5, medium field amplitudes in a transparent-regime polariton with unitless frequency \( \omega a/2\pi c = 0.8 \). The medium contribution is given as \( |\alpha^m_\omega| \) (left scale) and \( |\beta^m_\omega| \) (right scale) in units of \( \left| \frac{a}{2\pi c} \right| \sqrt{\text{Im} \Gamma (r, \omega) / (\hbar \varepsilon_0 \pi)} \tilde{D}^*_m (r) \). The patterned area indicates the absence of medium oscillators, i.e. the transparent frequency regime, and the dotted line indicates the frequency of the polariton mode of interest. The electronic gap is at \( \omega_g a/2\pi c = 0.85 \).

Figure 2.6: Schematic diagram of a GaAs waveguide structure.
input the dielectric constant $\varepsilon(\omega)$ of GaAs. This could come from experimental data, semi-empirical formulae, or \textit{ab initio} calculations. Here we take data for $\varepsilon(\omega)$ based on \textit{ab initio} calculations [20]. The real and imaginary parts of $\varepsilon(\omega)$, which obey the Kramers Kronig relations, are given in Fig. 2.7. The corresponding $\Gamma(\omega)$ is obtained from $\varepsilon(\omega)$ using (2.162), and is plotted in Fig. 2.8. The absorption cutoff frequency $\omega_g$ is taken to be $1.42\text{eV}/\hbar$, which corresponds to the electronic band gap of GaAs from Fig. 2.7.

We consider here only the polaritons with an in-plane (continuous) wave vector $\mathbf{k} = (k_x, k_y)$, and perpendicular but in-plane displacement field polarization. The normalization region $V$ comprises the slab region of height $L_z$ in the $z$-direction, and the (infinite) $xy$-plane. The unit cell $V_{\text{cell}}$ is then taken to be of height $L_z$, and its extent in the $xy$-plane can be freely chosen, as described after (2.159). Since the electromagnetic fields vanish on the bounding conductors, the periodic boundary condition on $V$ is guaranteed to hold. Equation (2.167) is solved readily. We can neglect solutions with radiation incident from outside the waveguide region, since they do not interact with the fields inside the GaAs slab. The electromagnetic component modes inside the waveguide are characterized by $m = (u, \mathbf{k})$, where $u \in \{1, 2, 3, \ldots\}$ gives the $z$-direction “quantum number” and $\mathbf{k}$ is the in-plane wave vector. Then the eigenvalues $Z_{\omega u \mathbf{k}}$ are used to normalize the mode fields using (2.160).

Assuming for the moment no excitation in the transparent regime, an arbitrary one-polariton state in the Hilbert space is given by

$$|\Phi\rangle = \sum_{u} \int_{\omega_g}^{\infty} d\omega \int d^2k \ A_{\omega u \mathbf{k}} c_{\mathbf{k}}^\dagger (0) |0\rangle ,$$

(2.170)
where $|0\rangle$ is the vacuum state, and

$$
\sum_u \int_{\mathcal{Q}} \int d^2k |A_{\omega uk}|^2 = 1. \quad (2.171)
$$

We consider a single-band “windowed” state from band $u$ centered at the point $(\omega, \mathbf{k})$, which we write as $|\omega, u, \mathbf{k}\rangle$. That is, we take $A_{\omega uk}$ to have support on $u = \pi$, and on a small window around $(\omega, \mathbf{k})$, with area $Q$ in 2D $\mathbf{k}$-space and full width $W$ in frequency space. We consider $\sqrt{Q}$ and $W$ to be much smaller than any other inverse length or inverse time scale in the problem, except for the inverse of the evolution time coordinate. Inside the window, $A_{\omega uk}$ is taken to have uniform value $(WQ)^{-1/2}$, as is required to satisfy (2.171).

The degree of electromagnetic character of this polariton state is given by the quantity $V(t)$, which we define as the ratio of electromagnetic energy to total energy, per unit of window energy $\hbar W$, neglecting zero point energy. Defining $H_{\text{nonvac}} (H_{\text{em,nonvac}}(t))$ to be the total (electromagnetic) Hamiltonian neglecting zero-point energy, q.v. (2.8, 2.9), we have

$$
V(t) \equiv \frac{\langle \Phi | H_{\text{em,nonvac}}(t) | \Phi \rangle}{\langle \Phi | H_{\text{nonvac}} | \Phi \rangle} \frac{1}{\hbar W}.
$$

(2.172)

For a window state $|\omega, u, \mathbf{k}\rangle$ we write this ratio as $V_{\omega uk}(t)$.

We note here that the time evolution does not just exhibit loss in electromagnetic energy. $V_{\omega uk}(t)$ vanishes in the limit $t \to -\infty$. Before $t = 0$ the evolution is such that energy flows into the electromagnetic fields. Then for $t > 0$ the dynamics show a decay in $V_{\omega uk}(t)$ as expected. We note also that while $V_{\omega uk}(t)$ describes the electromagnetic character of the modes, it does not describe the state obtained by driving the electromagnetic fields with a source, which is addressed in Ch. 3.

In Fig. 2.9 we plot $\max_{\pi} V_{\omega uk}(0)$ for $\pi \in \{1, 2, 3\}$, on the $(\omega, \mathbf{k})$ plane $(\omega > \omega_g)$. Superimposed are the dotted curves giving $\omega_{uk}$ for $u \in \{1, 2, 3\}$ for the formal dispersive but nonabsorptive system obtained if the absorption is switched off, i.e. $\Gamma (\mathbf{r}, \omega) \to \text{Re} \Gamma (\mathbf{r}, \omega)$. These curves also indicate points $(\omega, k)$ at which $Z_{\omega uk}$ vanishes for the appropriate band $u$.

We note some other features of Fig. 2.9. First, polaritons close in frequency to dispersive-limit bands tend to be highly electromagnetic in character, and appear to define broadened bands. A cross-section at $k = 25 \mu m^{-1}$ is given in Fig. 2.10. Note also that as a broadened band approaches the electronic band gap frequency, it tapers out.
Figure 2.9: The lowest three effective broadened polariton bands for a GaAs waveguide with 1D confinement. Brightness indicates the degree of electromagnetic character of the polariton modes in the absorption regime ($\omega \geq \omega_g$). Dashed lines give the transparent regime bands below $\omega_g$; in the absorption regime above $\omega_g$ they give the three lowest bands of the transparent system obtained if absorption is switched off: $\Gamma(\omega) \rightarrow \text{Re} \Gamma(\omega)$. 
and merges with the discrete transparent-regime polariton band, which is indicated by the dotted line at frequencies below the electronic band gap frequency $\omega_g$.

We demonstrate that the medium components of the mode fields have reasonable behavior by calculating them from the electromagnetic mode fields and eigenvalues according to (2.128, 2.129). Consider the second polariton band $m = (u, k) = (2, 25 \mu m^{-1})$, for which $\hbar \omega_{\text{disp}} = 1.824 \text{eV}$ is the energy of the corresponding discrete band in the dispersive but nonabsorptive limit, i.e., $\Gamma (r, \omega) \to \text{Re} \Gamma (r, \omega)$. The medium field excitation is shown in Fig. 2.11, where $c_{\text{max}} \left| \alpha^m_{\omega'} (r) \right|$ is plotted as a function of $\hbar \omega'$ for $m = (u, k) = (2, 25 \mu m^{-1})$, for polaritons of energies $\hbar \omega = \{\hbar \omega_{\text{disp}} - 0.1 \text{eV}, \hbar \omega_{\text{disp}}, \hbar \omega_{\text{disp}} + 0.1 \text{eV}\}$.

## 2.6 Discussion

In this chapter I have presented a Hamiltonian formulation of dispersive and absorptive structured (inhomogeneous) media explicitly obeying the Kramers-Kronig relations. The main results for the practitioner are summarized in Section 2.5. My method focuses on the photonic component of the polariton modes of the system. This has an advantage over noise-current methods in that existing codes and computational methods for photonic band structures can easily be leveraged to calculate the polariton modes while maintaining a Hamiltonian formulation. The medium components of the polariton
Figure 2.11: For the GaAs waveguide, $c \max_r |\alpha_{\omega}^m(r)|$ versus $\hbar \omega'$ for $m = (u, k) = (2, 25 \mu m^{-1})$, for $\hbar \omega = \{\hbar \omega_{\text{disp}} - 0.1 \text{eV}, \hbar \omega_{\text{disp}}, \hbar \omega_{\text{disp}} + 0.1 \text{eV}\}$, where $\hbar \omega_{\text{disp}} = 1.824 \text{eV}$ is the energy of the corresponding discrete band in the dispersive but non-absorptive limit, i.e., $\Gamma(r, \omega) \to \Re \Gamma(r, \omega)$.

modes are easily obtained once the photonic components have been determined. This is demonstrated in Section 2.5, in which I construct polaritons for an example 2D dispersive photonic crystal and for a dispersive and absorptive waveguide. Given an arbitrary space- and frequency-dependent dielectric response function, the model and canonical description of the modes of the coupled medium and electromagnetic field can immediately be found.

In the formulation the medium is represented using an intuitive model. There is a clear connection between the coupling constants of the model medium and the dielectric response, and only the susceptibility of the medium appears in the final equations for the polariton modes. The numerical value of the Hamiltonian of the system is equal to its energy, so this formulation gives both quantum and classical results; furthermore, having a Hamiltonian allows conserved quantities to be identified and understood for dispersive and absorptive frequency regimes. Throughout, I have explicitly treated transparent and absorptive frequency regimes, which have qualitatively different polariton characteristics.

In the transparent regime below the (optionally vanishing) absorption cutoff frequency, I obtain a discrete polariton spectrum, and the polariton dispersion relation is exactly the dispersion relation of the electromagnetic modes of the dispersive system. Each polariton mode consists of its dispersive electromagnetic component augmented by
the associated medium excitation component, and the electromagnetic components are obtained from the usual dispersive Maxwell master equation.

In the absorptive regime, I obtain a continuous spectrum of polaritons; however, it is possible to identify effective broadened polariton bands. Furthermore, the electromagnetic component of each polariton mode is found to obey the familiar nonabsorptive Maxwell master equation, but with an effective dielectric function parameterized by the eigenvalue. It is inevitable that a continuous spectrum will arise in the absorptive regime; absorption causes a temporal decay of electromagnetic fields, which implies a linewidth in the (real) polariton spectrum. This has implications for the treatment of wave packet propagation, which is done in Ch. 3.
Chapter 3

Propagation in dispersive and absorptive structured media

In this chapter I present a Hamiltonian formulation of pulse propagation in structured media with arbitrary absorptive and dispersive response subject to the Kramers-Kronig relations. In the transparent regime the dynamics are formulated in terms of a Schrödinger equation satisfied by a polariton effective field, with emphasis on the photonic component of the polariton modes of the system. The absorptive regime fields separate into a fluctuating part described in terms of vacuum polariton operators and modes, and a non-fluctuating part with Schrödinger dynamics. States that represent an arbitrary classical-like beam incident on a quiescent sample are shown to be a subset of the polariton coherent states, and I express them naturally using an initial value problem for the nonfluctuating part of the polariton effective field. I show that the full dynamics of the absorptive and dispersive system can be obtained from the same generalized dispersion surfaces typically used while neglecting absorption; only a deformation of the cutting surfaces is needed. As an example of the method presented in this chapter, I calculate complex propagation parameters (loss, group velocity, dispersion/diffraction) as a function of carrier wave vector for an optical pulse traversing a 1D Au/MgF$_2$ metallodielectric stack, using the full dispersive and absorptive $\varepsilon (r, \Omega)$, and elucidate the various roles of Bragg scattering, (electronic) interband absorption and field expulsion on propagation in that sample. Further, I both correct a result in the literature for the Beer coefficient in structured media, and provide a result for fully causal absorptive structured media.
Chapter 3. Propagation

3.1 Hamiltonian dynamics in dispersive and absorptive media

This chapter uses as its foundation the Hamiltonian formalism of Ch. 2. Here I both summarize the points from Ch. 2 that are essential for this chapter, and extend the formulation by introducing a classical source polarization that is used in Sec. 3.2 to generate states of interest.

Recall we take $D$ and $B$ as the fundamental electromagnetic fields. We assume the medium of interest can be characterized by a causal linear response $\Gamma (\mathbf{r}, t)$ of the form of Eq. (2.6), where we neglect magnetic effects so that $\Gamma^{ij} (\mathbf{r}, t) = \Gamma^{ji} (\mathbf{r}, t)$. To consider frequency-space quantities we take the usual Fourier transform convention

$$f (t) = \int_{-\infty}^{\infty} \frac{d\Omega}{2\pi} f (\Omega) e^{-i\Omega t}. \quad (3.1)$$

The response function $\Gamma (\mathbf{r}, \Omega)$ is simply related to the familiar dielectric function $\varepsilon (\mathbf{r}, \Omega)$. For an isotropic medium to which we restrict ourselves in this chapter, it is given by

$$\Gamma^{ij} (\mathbf{r}, \Omega) = \left[ 1 - \varepsilon_0 \varepsilon^{-1} (\mathbf{r}, \Omega) \right]\delta_{ij}, \quad (3.2)$$

although the results that follow trivially generalize to describe non-isotropic media.

For many media of interest there is a cutoff frequency $\Omega_g$ for absorption, below which $\text{Im} \varepsilon (\mathbf{r}, \Omega) = 0$ for all $\mathbf{r}$. We explicitly allow such a cutoff here, but we can take $\Omega_g = 0$ if desired. The medium is modeled by harmonic oscillator fields $X_\Omega (\mathbf{r})$ of all frequencies $\Omega$ at all positions $\mathbf{r}$, and by an appropriate coupling tensor function $\Lambda (\mathbf{r}, \Omega)$ that couples these medium fields to the electromagnetic fields; for the isotropic media we consider here, $\Lambda (\mathbf{r}, \Omega)$ is diagonal. Since we wish to describe a response (2.6) that is local in space, it suffices to take the coupling between the medium oscillators and the electromagnetic fields to be local in space.

The sole input to the model is a specified or calculated response tensor $\Gamma (\mathbf{r}, \Omega)$. Alternatively $\varepsilon (\mathbf{r}, \Omega)$ can be specified (see Eq. (3.2)). For the isotropic media considered here we then model the physics by setting the coupling according to the prescription (q.v. Eq. (2.29))

$$\Lambda^{ii} (\mathbf{r}, \Omega) \equiv \sqrt{\frac{2\Omega}{\pi}} \text{Im} \Gamma^{ii} (\mathbf{r}, \Omega), \quad (3.3)$$

where there is no summation over $i$. A nice feature of the approach is that, unlike $\varepsilon (\mathbf{r}, \Omega)$, the function $\Gamma (\mathbf{r}, \Omega)$ does not diverge at $\Omega = 0$ in metals, and therefore $\Lambda (\mathbf{r}, \Omega)$ does not diverge either.
The Hamiltonian of the system [1] is

\[ H = H_{\text{sys}} + H_{\text{drive}} = (H_{\text{em}} + H_{\text{med}} + H_{\text{int}}) + H_{\text{drive}}, \]  

(3.4)

where \( H_{\text{em}}, H_{\text{med}}, \) and \( H_{\text{int}} \) are respectively the bare electromagnetic, bare medium, and medium-electromagnetic interaction Hamiltonians. For the purpose of generating states of interest we have added an additional term \( H_{\text{drive}} \) representing coupling of the electromagnetic fields with an external (classical) source polarization \( P_s(r,t) \):

\[ H_{\text{drive}} = -\frac{1}{\varepsilon_0} \int d\mathbf{r} \mathbf{D}(\mathbf{r}) \cdot \mathbf{P}_s(\mathbf{r},t). \]  

(3.5)

Using a standard set of commutation relations between the various fields, Hamilton’s equation \( i\hbar \partial_t f = [f,H] \) yields the macroscopic Maxwell equations with no free charges or currents, and with constitutive relation (2.6), but with an additional polarization

\[ \mathbf{P}_{\text{extra}}(r,t) = \int_\Omega g d\Omega \sqrt{\varepsilon_0 \Lambda(\mathbf{r},\Omega)} \cdot (X_\Omega^0(\mathbf{r}) e^{-i\Omega t} + \text{c.c.}) + \mathbf{P}_s(\mathbf{r},t). \]  

(3.6)

The first term is the fluctuation polarization associated with dissipation, and the term in parentheses is the medium oscillator coordinate field in the absence of any coupling to the electromagnetic fields.

A Fano-like diagonalization of \( H_{\text{sys}} \) yields the polariton modes of the system. These separate into longitudinal polarization (LP) and transverse polarization (TP) varieties, so (q.v. Eq. (2.57))

\[ H_{\text{sys}} = H_{\text{TP}} + H_{\text{LP}}. \]  

(3.7)

The LP polaritons are composed entirely of medium fields, and do not enter into the physics discussed here. The TP polaritons are composed of a superposition of medium and electromagnetic fields. When we have periodic boundary conditions, the TP polaritons have a discrete spectrum below the absorption cutoff frequency \( \Omega_g \) and a continuous spectrum above \( \Omega_g \). That is, even at a single given wave vector in a uniform medium, for example, there exist polaritons at a continuum of frequencies including the entire absorptive frequency regime. Thus (q.v. Eq. (2.59))

\[ H_{\text{TP}} = \sum_m \hbar \tilde{\omega}_m c_m^\dagger c_m + \sum_m \int_{\Omega_g}^\infty d\Omega \hbar \Omega c_{\Omega m}^\dagger c_{\Omega m}, \]  

(3.8)

where the \( c_m \) and \( c_{\Omega m} \) are the polariton lowering operators for the discrete and continuous spectra respectively, and we neglect the zero point energy. Whereas the \( m \) labels the
modes for the transparent regime, the absorptive regime modes require an additional frequency label Ω for their specification. The operators have harmonic time dependence with frequencies $\tilde{\omega}_m$ and Ω respectively, and they satisfy the canonical commutation relations of Eqs. (2.65, 2.66).

The electromagnetic fields can be expanded in terms of the TP polariton operators and the electromagnetic components of their corresponding mode fields as (q.v. Eqs. (2.67, 2.68))

$$D(r, t) = \sum_m \left[ c_m \tilde{D}_m(r) + c^\dagger_m \tilde{D}^*_m(r) \right] + \sum_m \int_{\Omega_y} d\Omega \left[ c_{\Omega m} \tilde{D}_{\Omega m}(r) + c^\dagger_{\Omega m} \tilde{D}^*_{\Omega m}(r) \right],$$

$$B(r, t) = \sum_m \left[ c_m \tilde{B}_m(r) + c^\dagger_m \tilde{B}^*_m(r) \right] + \sum_m \int_{\Omega_y} d\Omega \left[ c_{\Omega m} \tilde{B}_{\Omega m}(r) + c^\dagger_{\Omega m} \tilde{B}^*_{\Omega m}(r) \right],$$

where we have left the time dependence of the $c$ operators implicit.

In the transparent regime, the mode field $\tilde{B}_m(r)$ and mode frequency $\tilde{\omega}_m$ are determined from the usual (dispersive) Maxwell master equation, (q.v. Eq. (2.110))

$$\nabla \times \left[ \varepsilon_0 \varepsilon^{-1}(r, \tilde{\omega}_m) \nabla \times \tilde{B}_m(r) \right] = -i\tilde{\omega}_m \mu_0 \tilde{D}_m(r).$$

Eq. (3.11) must be solved self-consistently for the eigenvalue $\tilde{\omega}_m$ and the eigenfunction $\tilde{B}_m(r)$. The mode field $\tilde{D}_m(r)$ is subsequently calculated from

$$-i\tilde{\omega}_m \mu_0 \tilde{D}_m(r) = \nabla \times \tilde{B}_m(r).$$

We note that for each mode $m$ there is a physically conjugate mode $\overline{m}$ with frequency $\tilde{\omega}_{\overline{m}} = \tilde{\omega}_m$, and with mode fields

$$\left( \tilde{D}_{\overline{m}}(r), \tilde{B}_{\overline{m}}(r) \right) = \left( \tilde{D}^*_m(r), -\tilde{B}^*_m(r) \right).$$

Employing box normalization with $V$ being the normalization region of volume $V$, the mode fields must be normalized so that

$$\frac{1}{\varepsilon_0} \int_V dr \tilde{D}^*_m(r) \cdot T(r, \tilde{\omega}_m) \cdot \tilde{D}_m(r) = \frac{\hbar \tilde{\omega}_m}{2},$$

with

$$T^{ij}(r, \omega) \equiv \left( \frac{1}{\varepsilon_0^{-1} \varepsilon(r, \omega)} - \omega \frac{\partial}{\partial \omega} \frac{1}{2 \varepsilon_0^{-1} \varepsilon(r, \omega)} \right) \delta_{ij}. $$
We note that the expression for $T(r, \omega)$ in Eq. (3.15) does not rely on perturbation theory, but is exact; it can be written as

$$T_{ij}(r, \omega) = \varepsilon_0 \varepsilon^{-1}(r, \omega) \frac{v_p(r, \omega)}{v_g(r, \omega)} \delta_{ij},$$

(3.16)

where we introduce the local group and phase velocities $v_g(r, \omega)$ and $v_p(r, \omega)$. These are the respective group and phase velocities of electromagnetic fields of frequency $\omega$, propagating through a uniform infinite sample of whichever material is at point $r$ in the actual system, so

$$v_p(r, \omega) = c / \sqrt{\varepsilon_0 \varepsilon^{-1}(r, \omega)},$$

(3.17)

and

$$v_g(r, \omega) = c / \sqrt{\varepsilon_0 \varepsilon^{-1}(r, \omega) + \omega \frac{d}{d\omega} \sqrt{\varepsilon_0 \varepsilon^{-1}(r, \omega)}}.$$  

(3.18)

The normalization condition Eq. (3.14) has a simple interpretation. For specificity, consider a single polariton state in a uniform medium. Then use of Eq. (3.14) to calculate the Poynting vector $\mathbf{E}(r, t) \times \mathbf{H}(r, t)$ yields the expression $(\hbar \tilde{\omega}_m / V) v_g(\tilde{\omega}_m)$ for the expected intensity. This can be understood as the intensity associated with a single polariton of energy $\hbar \tilde{\omega}_m$ contained in the normalization volume $V$, propagating at the group velocity $v_g(\tilde{\omega}_m)$.

We next describe polariton modes in the absorptive regime. In that regime, since even for a given index $m = (u, k)$ there exist polaritons at all frequencies $\Omega > \Omega_g$, the modes are determined in a different manner than they are in the transparent regime. A useful quantity is the real variable $Z$, which is proportional to the contribution to the polariton from the resonant medium fields [1], i.e., those medium field oscillators of natural frequency $\Omega$; recall from the discussion after Eq. (2.85) that we call $Z$ the “resonant oscillator amplitude.” It is sometimes convenient to consider $\Omega$ to be the independent variable and $Z$ the dependent variable, but often the opposite is more natural; both approaches are used in this thesis, with the obvious notations $\Omega_m(Z)$ and $Z_m$. For a given $Z$, the mode fields and mode frequency $\Omega$ are given by (q.v. Eq. (2.122))

$$\nabla \times \varepsilon_0 \varepsilon^{-1}(r, \Omega; Z) \nabla \times \tilde{\mathbf{B}}_\Omega (r) = \frac{\Omega^2}{c^2} \tilde{\mathbf{B}}_\Omega (r).$$

(3.19)

Here $\varepsilon^{-1}(r, \Omega; Z)$ is a real function defined by $[1 - \varepsilon_0 \varepsilon^{-1}(r, \Omega; Z)] \delta_{ij} \equiv \Gamma(\Omega; \Omega; Z)$, where we recall the definition of the effective response function (q.v. Eq. (2.123))

$$\Gamma(r, \Omega; Z) \equiv \text{Re} \Gamma(r, \Omega) - \frac{Z}{\pi} \text{Im} \Gamma(r, \Omega).$$

(3.20)
The effective dielectric function $\varepsilon(r, \Omega; Z)$ is not a physically intuitive quantity, except in that it plays the role in the equation (3.19) for $\{\Omega, \vec{B}_{\Omega m}(r)\}$ that $\varepsilon(r, \tilde{\omega}_m)$ plays in the equation (3.11) for $\{\tilde{\omega}_m, \vec{B}_m(r)\}$. For the special choice $Z = 0$, Eq. (3.19) is exactly equivalent to the transparent regime Maxwell master equation (3.11) that would be obtained were the imaginary part of $\Gamma(r, \Omega)$ disregarded but the real part kept. We define a “central frequency” $\Omega_m$ of the continuum corresponding to mode index $m$ via

$$\Omega_m \equiv \Omega_m(0).$$

(3.21)

Note that although $\Omega$ depends on $m$, this dependence is sometimes left implicit for notational convenience. Typically the modes at frequencies near $\Omega_m$, i.e., those with small $|Z|$, are the ones of most interest; they are the modes that, when combined into pulses, contribute most to the electromagnetic component of the state.

Having obtained the $\vec{B}_{\Omega m}(r)$ from Eq. (3.19), we obtain the $\vec{D}_{\Omega m}$ from

$$-i\Omega\mu_0 \vec{D}_{\Omega m}(r) = \nabla \times \vec{B}_{\Omega m}(r).$$

(3.22)

Again there exist physical conjugate modes:

$$(\vec{D}_{\Omega m}(r), \vec{B}_{\Omega m}(r)) = (\vec{D}^*_{\Omega m}(r), -\vec{B}^*_{\Omega m}(r)),$$

(3.23)

with

$$Z_{\Omega m} = Z_{\Omega m}.\quad (3.24)$$

The mode fields here are normalized so that (q.v. Eq. (2.126))

$$\frac{1}{\varepsilon_0} \int_V d\mathbf{r} \vec{D}^*_{\Omega m}(r) \cdot \text{Im} \Gamma(r, \Omega) \cdot \vec{D}_{\Omega m'}(r) = \hbar \frac{\pi}{Z_{\Omega m}^2 + \pi^2} \delta_{mm'}.\quad (3.25)$$

When material absorption is small enough for propagation to be of interest, the factor on the right-hand side (RHS) of Eq. (3.25) varies rapidly in $\Omega$ over a width comparable to a pulse decay rate, as is seen in Sec. 3.4. For that reason it is useful to define a new mode field that is more slowly varying in frequency and that is well-suited to propagation problems, as will become apparent. To do so, we introduce a function $L_{\Omega m}$ such that its amplitude is set according to

$$|L_{\Omega m}| = \sqrt{\frac{\pi}{Z_{\Omega m}^2 + \pi^2}}.\quad (3.26)$$

For the description of pulse propagation, there is a single good choice for the phase of $L_{\Omega m}$ up to an overall constant. We introduce the choice here and show it to be unique in
Appendix D. Since the RHS of Eq. (3.26) is real and finite on the real axis, the poles of any analytic continuation into the complex plane occur in pairs \( \{ \Omega_m, \Omega_m^* \} \), i.e., with one pole in the lower half-plane (LHP) and the other in the upper half-plane (UHP). Assume they are all single poles\(^1\). Then we specify \( L_{\Omega_m} \) so that it satisfies (3.26) and so that all of its poles are in the LHP, i.e.,

\[
L_{\Omega_m} \equiv C_m \prod_i \frac{1}{\Omega - \Omega_m^{(i)}},
\]

where \( C_m \) is a real normalization constant chosen so that Eq. (3.26) holds. By convention we take \( C_m \) to be positive. Note that \( L_{\Omega_m} \neq \sqrt{\pi}/(Z_{\Omega_m} + i\pi) \) here\(^2\). We then define a new set of mode fields via

\[
D_{\Omega_m}(r) \equiv \hbar^{-1/2} L_{-1}^{-1} \tilde{D}_{\Omega_m}(r).
\]

We choose the dependence of the phase of \( \tilde{D}_{\Omega_m}(r) \) on \( \Omega \) so that \( D_{\Omega_m}(r) \) is continuous and smooth in \( \Omega \) and slowly varying over the bandwidth of any wave packet we wish to describe. It follows from the normalization condition (3.28) that

\[
\frac{1}{\varepsilon_0} \int_V d^3r \, D_{\Omega_m}(r) \cdot \text{Im} \, \Gamma(r, \Omega) \cdot D_{\Omega_m'}(r) = \delta_{mm'}.
\]

Often systems of interest are periodic structured media or uniform media. In such systems the polariton modes are labeled by \( m = (u, k) \), and it is commonly convenient to take \( V \) to be \( \mathbb{R}^3 \). Then the wave vector \( k \) becomes a continuous label. Sometimes we will use the general \( m \) notation; other times we will specifically refer to a photonic crystal using \((u, k)\) notation.

This transition from discrete labels to continuous wave vectors is discussed in Section 2.4, in which equations are given for continuous-\( k \) labeling in 1D, 2D and 3D. In the interest of keeping uncluttered notation, in this chapter the system of interest is taken to be a 3D periodic system with a continuous wave vector. For this 3D continuous-\( k \) system, the sum over \( m \) is replaced according to

\[
\frac{1}{V} \sum_m \rightarrow \sum_u \frac{1}{(2\pi)^3} \int_D d^3k,
\]

\(^1\)From the form of Eq. 3.26, that \( Z_{\Omega_m} \) is real implies that \( L_{\Omega_m} \) never diverges on the real axis, so it can always be approximated arbitrarily closely over the domain of interest by a function with only single poles.

\(^2\)However, the equality does hold if \( Z_{\Omega_m} \) is considered only at lowest order in \( \Omega \).
where $D$ is a reciprocal space unit cell such as the first Brillouin zone, and recall from after Eq. (3.13) that $V$ is the volume of the normalization region $V$. The normalization condition (3.14) becomes
\[ \frac{1}{\varepsilon_0} \int_V d\mathbf{r} \left( D_{\mathbf{k}'k}^* (\mathbf{r}) \cdot \mathbf{T}(\mathbf{r}, \tilde{\omega}_{\mathbf{u}k}) \cdot \mathbf{D}_{\mathbf{u}k} (\mathbf{r}) = \frac{\hbar \tilde{\omega}_{\mathbf{u}k}}{2} \delta (\mathbf{k} - \mathbf{k}') ; \right) \tag{3.31} \]
the normalization conditions (3.25, 3.29) also hold, with it remembered that $V$ is $\mathbb{R}^3$ and with the replacement $\delta_{\mathbf{mm}'} \rightarrow \delta_{\mathbf{uu}'} \delta (\mathbf{k} - \mathbf{k}')$.

For periodic media Bloch functions are of interest. We denote the periodic part of a Bloch function by lower case letters, and for the respective transparent and absorptive regimes we have
\[ \mathbf{\tilde{D}}_{\mathbf{k}k} (\mathbf{r}) \equiv (2\pi)^{-3/2} \mathbf{d}_{\mathbf{k}k} (\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}} , \tag{3.32} \]
\[ \mathbf{\tilde{D}}_{\mathbf{\Omega k}} (\mathbf{r}) \equiv (2\pi)^{-3/2} \mathbf{d}_{\mathbf{\Omega k}} (\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}} , \tag{3.33} \]
and the replacements $\mathbf{D} \rightarrow \mathbf{B}$ and $\mathbf{d} \rightarrow \mathbf{b}$ in Eqs. (3.32, 3.33) give the analogous definitions of the periodic parts of the magnetic field Bloch functions. The functions $f \in \{ \mathbf{d}_{\mathbf{k}k}, \mathbf{b}_{\mathbf{k}k}, \mathbf{d}_{\mathbf{\Omega k}}, \mathbf{b}_{\mathbf{\Omega k}} \}$ satisfy $f(\mathbf{r}) = f(\mathbf{r} + \mathbf{R})$ for any lattice vector $\mathbf{R}$. For the mode fields $\mathbf{D}_{\mathbf{\Omega m}} (\mathbf{r})$ we define the corresponding periodic part of the Bloch functions $\mathbf{d}_{\mathbf{\Omega m}} (\mathbf{r})$ in the same way with the same prefactor $(2\pi)^{-3/2}$. Then the normalization of the fields $\mathbf{d}_{\mathbf{k}k} (\mathbf{r})$ for continuous $\mathbf{k}$ is given by
\[ \frac{1}{\varepsilon_0} \int_{\text{cell}} d\mathbf{r} \left( \mathbf{d}_{\mathbf{k}k}^* (\mathbf{r}) \cdot \mathbf{T}(\mathbf{r}, \tilde{\omega}_{\mathbf{u}k}) \cdot \mathbf{d}_{\mathbf{k}k} (\mathbf{r}) = \frac{\hbar \tilde{\omega}_{\mathbf{u}k}}{2} ; \right) \tag{3.34} \]
the normalization of the $\mathbf{d}_{\mathbf{\Omega k}} (\mathbf{r})$ is given by
\[ \frac{1}{\varepsilon_0} \int_{\text{cell}} d\mathbf{r} \left( \mathbf{d}_{\mathbf{\Omega k}}^* (\mathbf{r}) \cdot \mathbf{\Im} \Gamma (\mathbf{r}, \mathbf{\Omega}) \cdot \mathbf{d}_{\mathbf{\Omega k}} (\mathbf{r}) = \frac{\hbar \pi}{2Z_{\mathbf{\Omega k}}} + \frac{\pi^2}{4} \delta_{\mathbf{uu}'} ; \right) \tag{3.35} \]
and the normalization of the fields $\mathbf{d}_{\mathbf{\Omega k}} (\mathbf{r})$ is given by
\[ \frac{1}{\varepsilon_0} \int_{\text{cell}} d\mathbf{r} \left( \mathbf{d}_{\mathbf{\Omega k}}^* (\mathbf{r}) \cdot \mathbf{\Im} \Gamma (\mathbf{r}, \mathbf{\Omega}) \cdot \mathbf{d}_{\mathbf{\Omega k}} (\mathbf{r}) = \delta_{uu'} . \right) \tag{3.36} \]
In Eqs. (3.34, 3.35, 3.36) the domain of integration is taken to be a unit cell of volume $V_{\text{cell}}$; the unit cell can be chosen to be any region which tiles the full normalization region $V$ via translations by lattice vectors.
3.2 Strategies for generating the field

An arbitrary absorptive-regime one-polariton state belonging to the Hilbert space is given by

$$|\Phi\rangle = b^\dagger (A, t) |0\rangle ,$$

(3.37)

where $|0\rangle$ is the vacuum state, with

$$b^\dagger (A, t) \equiv \sum_u \int_{\Omega_g} d\Omega \int_{D} d\mathbf{k} A_{\Omega u k} c_{\Omega u k}^\dagger (t) ,$$

(3.38)

for some $A_{\Omega u k}$ normalized according to

$$\sum_u \int_{\Omega_g} d\Omega \int_{D} d\mathbf{k} |A_{\Omega u k}|^2 = 1.$$  

(3.39)

Thus it is possible to generate states of the system by specifying some $A_{\Omega u k}$, and this approach has been used in the literature [43]. By taking the expectation value of the number operator describing modes of the electromagnetic field in the state $|\Phi\rangle$, it can be seen that it is possible to choose $A_{\Omega u k}$ such that the expected photon number can evolve erratically, rising and falling many times. So an arbitrary $A_{\Omega u k}$ (or $A_{\Omega m}$ more generally) does not in general describe absorption of a propagating electromagnetic wave packet, and it is not obvious which subset of the $A_{\Omega u k}$ specify such states.

Similarly, it is possible to define a coherent state creation operator in the usual way via

$$B^\dagger (\lambda, A, t) \equiv \exp \left( \lambda b^\dagger (A, t) - \lambda^* b(A, t) \right) ,$$

(3.40)

for some $A_{\Omega u k}$, but again the expected photon number can rise and fall in a way that does not correspond to absorption.

However, there are subsets of the one-polariton states and coherent states with the variation of the phase of $A_{\Omega u k}$ with $\Omega$ such that the state evolves in a physically desirable way: The electromagnetic component begins in its vacuum state, and over some finite “generation period” the expected photon number increases as the energy flows from the medium fields into the electromagnetic fields as the phases of the polariton creation operators acquire the appropriate values. Subsequently as the phases of the polariton operators further evolve, the electromagnetic energy decays away as the energy flows back into the medium.

But this picture of the electromagnetic fields being generated by contrived interference of the fluctuations is unappealing from a physical standpoint. We prefer to define that
physical subset of states for which the relative phase relation for $A_{\Omega k}$ is sensible by generating its members via a robust physical process, employing the classical driving polarization field $P_s$. This will ensure states of physical interest are created, and indeed these states comprise a subset of the coherent states. Once generated, the electromagnetic component of such a state subsequently decays as the energy flows into the medium fields.

We envision formally turning on the field via the following process. Before some time $-\tau_s < 0$, there are only fluctuations; depending on the system of interest, this could be the vacuum state or thermal equilibrium at some finite temperature. The driving field $P_s(r, t)$ is then applied over a time interval with support $t \in [-\tau_s, 0)$. Ultimately, we will specify which states can be generated in this way, so that in practice the detailed profile of $P_s(r, t)$ will not be needed.

While this process is useful for the absorptive regime, we also use it in the transparent regime, in which the subtleties discussed in this section do not arise. In either regime, we take $c_0^\alpha(t)$ to be the polariton operators in the absence of $P_s(r, t)$, and separate the full polariton operators into two pieces by writing

$$c_\alpha(t) = \tau_\alpha(t) + c_0^\alpha(t),$$

(3.41)

where $\alpha = m$ and $\alpha = (\Omega, m)$ in the transparent and absorptive regimes respectively, and where $\tau_\alpha(t)$ is a c-number. Then

$$\left[ c_0^m(t), (c_0^{m'}(t))^\dagger \right] = \delta_{mm'}$$

(3.42)

$$\left[ c_0^{\Omega m}(t), (c_0^{\Omega m'}(t))^\dagger \right] = \delta_{mm'} \delta(\Omega - \Omega').$$

The dynamics of $c_\alpha(t)$ are given by Hamilton’s equation, using the full Hamiltonian $H$,

$$\frac{d}{dt} c_\alpha(t) = -i\omega_\alpha c_\alpha(t) - \frac{1}{i\hbar \varepsilon_0} \int dr \bar{D}_\alpha^*(r) \cdot P_s(r, t),$$

(3.43)

where $\omega_\alpha = \{\Omega \text{ or } \bar{\omega}_m\}$. Here we have used the expansion (3.9) and the commutators (2.65, 2.66). The fluctuation dynamics are given by Hamilton’s equations using $H_{TP}$,

$$\frac{d}{dt} c_0^\alpha(t) = -i\omega_\alpha c_0^\alpha(t).$$

(3.44)

The dynamics of $\tau_\alpha(t)$ then follow from the difference of (3.43) and (3.44), and the definition (3.41):

$$\frac{d}{dt} \tau_\alpha(t) = -i\omega_\alpha \tau_\alpha(t) - \frac{1}{i\hbar \varepsilon_0} \int dr \bar{D}_\alpha^*(r) \cdot P_s(r, t).$$

(3.45)
Now $\tau_\alpha(-\tau_s) = 0$, so integrating (3.45) we have

$$
\tau_\alpha(t) = -\frac{1}{i\hbar\varepsilon_0} \int dr \bar{D}_\alpha^*(r) \cdot \int_{-\tau_s}^t dt' e^{i\omega_\alpha t'} P_s(r, t'),
$$
(3.46)

and since $P_s$ has support on $[-\tau_s, 0)$, we have for $t \geq 0$,

$$
\tau_\alpha(t) = -\frac{1}{i\hbar\varepsilon_0} \int dr \bar{D}_\alpha^*(r) \cdot P_s(r, \omega_\alpha).
$$
(3.47)

Note that using Eq. (3.41) and performing an expectation value of the polariton operators yields

$$
\langle c_\alpha(t) \rangle = \tau_\alpha(t),
$$
(3.48)

since the expectation value of any fluctuation operator vanishes.

### 3.3 Propagation in the transparent regime

Consider propagation in a frequency regime below the cutoff frequency for absorption. It is shown in Appendix D that by suitably choosing $P_s(r, t)$, it is possible to generate any $\tau_{\alpha k}(0)$ with at most negligible excitation in the absorptive-regime part of the Hilbert space. It follows that we need not consider in detail the profile of $P_s(r, t)$, but can begin the analysis at $t = 0$—i.e. after $H_{drive}$ has subsided—by specifying some “initial condition” $\tau_{\alpha k}(0)$ for the polariton amplitudes generated by $H_{drive}$. Then the relevant part of the Hamiltonian (3.8) is

$$
H_{TP} = \sum_u \int_D dk \hbar \tilde{\omega}_{\alpha k} c^\dagger_{\alpha k} c_{\alpha k}.
$$
(3.49)

To describe propagation it is common to introduce slowly-varying amplitudes, or “envelope functions.” While this has been done in the past for structured media with at most perturbative material dispersion and absorption [2], the approach has two serious disadvantages: Slowly varying amplitudes do not have simple commutators, and therefore the dynamics are not simply obtained using Hamilton’s equations; and slowly varying amplitudes from different bands have coupled dynamics, even in the linear regime. Instead of envelope functions, for nondispersive, nonabsorptive media an effective field [3] has been introduced that has simple commutators, and effective fields from different bands obey decoupled dynamical equations; so the effective field dynamics are easily obtained to all orders. The effective field is equivalent to an envelope function to lowest order, and it
Figure 3.1: Schematic diagram of a band structure and effective field. Lower half of the diagram: a transparent-regime dispersion relation and the effective field content. Upper half of the diagram: the effective field content for the absorptive regime.

is possible to reconstruct the electromagnetic fields in a similar way as is done from an envelope function. Here we take an approach that retains all of these advantages of the effective field, but is capable of treating arbitrarily dispersive causal media. To do so, we introduce a polariton effective field,

\[ g_u (r, t) = (2\pi)^{-3/2} \int_D d\kappa c_{\kappa u} (t) e^{i(\kappa - \bar{\kappa}) \cdot r}, \]

where \( \bar{\kappa} \) is the central wave vector around which \( g_u \) is built, but we leave the \( \kappa \)-dependence implicit. Recall from after Eq. (3.30) that \( D \) is the relevant reciprocal-space domain. For a uniform medium \( D \) is all of \( k \)-space; for a periodic system \( D \) is the first Brillouin zone. The lower half of Fig. 3.1 shows a schematic diagram of a transparent-regime dispersion relation and the content of a transparent regime effective field \( g_u (r, t) \). We note that the \( c_{\kappa u} (t) \) can be recovered from \( g_u (r, t) \) by inverting the Fourier transform in Eq. (3.50).

It follows from the canonical commutation relations for the \( c_{\kappa u} \) in Eq. (2.66) that the polariton effective fields obey the following equal time commutation relations:

\[
\begin{align*}
\{ g_u (r, t), g_{\kappa' u'} (r', t) \} &= \delta_{uu'} \tilde{\Pi} (r - r') , \\
\{ g_u (r, t), g_{\kappa u'} (r', t) \} &= 0 , \\
\{ g_u (r, t), g_{\kappa' u} (r', t) \} &= 0 ,
\end{align*}
\]
where
\[ \tilde{\Pi} (r) = (2\pi)^{-3} \int_D d\mathbf{k} e^{i(\mathbf{k} - \mathbf{K}) \cdot r}, \]  
(3.52)
and we note that
\[ \int d\mathbf{r}' \tilde{\Pi} (\mathbf{r} - \mathbf{r}') = 1. \]  
(3.53)

If \( D \) is all of reciprocal space \( \tilde{\Pi} (\mathbf{r}) \) is the Dirac delta function \( \delta (\mathbf{r}) \). If instead \( D \) is a first Brillouin zone, then the domain of \( \tilde{\Pi} (\mathbf{r}) \) is localized to roughly the size of the unit cell in real space; but when the pulse being described is slowly varying in space compared with \( \tilde{\Pi} (\mathbf{r}) \), then \( \tilde{\Pi} (\mathbf{r}) \) can be treated as a delta function. Here for simplicity of notation we allow at most one wave packet per band. The analysis of multiple wave packets from a single band has been treated [75] for nondispersive, nonabsorptive media, and it can be applied here straightforwardly when needed.

The form (3.50) is tailored to representing pulses with carrier wave vector \( \mathbf{k} \), and for pulses which extend over many lattice periods. Writing \( \mathbf{k} = \mathbf{K} + \mathbf{\kappa} \) we note that for states of interest, it is reasonable to assume that the expectations of \( c_{\mathbf{uK}+\mathbf{\kappa}}^\dagger c_{\mathbf{uK}} \) and its multiples are small except for small \( |\mathbf{\kappa}| \). And for small \( |\mathbf{\kappa}| \), a Taylor expansion provides a good description of the polariton dispersion relation of each discrete band:
\[ \tilde{\omega}_{\mathbf{uK}+\mathbf{\kappa}} = \tilde{\omega}_{\mathbf{uK}} + \kappa^i \tilde{\omega}_{\mathbf{uK}}^{(1)i} + \frac{1}{2} \kappa^i \kappa^j \tilde{\omega}_{\mathbf{uK}}^{(2)ij} + ..., \]  
(3.54)
where \( \tilde{\omega}_{\mathbf{uK}}^{(2)ij} \), etc., give derivatives of the dispersion relation with respect to wave vector at \( \mathbf{k} \), and where the number of raised indices gives the order of the derivatives:
\[ \tilde{\omega}_{\mathbf{uK}}^{(1)i} \equiv \left[ \frac{\partial \tilde{\omega}_{\mathbf{uK}}}{\partial k^i} \right]_{\mathbf{K}}, \]  
(3.55)
\[ \tilde{\omega}_{\mathbf{uK}}^{(ij)} \equiv \left[ \frac{\partial^2 \tilde{\omega}_{\mathbf{uK}}}{\partial k^i \partial k^j} \right]_{\mathbf{K}}. \]  
(3.56)
Here we assume that the polariton bands are nondegenerate in the neighborhood of \( \mathbf{K} \) that supports the bulk of the pulse. The relevant part of the Hamiltonian can then be written in terms of the \( g_{\mathbf{uK}} \) up to second order in the Taylor expansion for \( \tilde{\omega}_{\mathbf{uK}} \) as
\[ H_{TP} = \sum_n \int d\mathbf{r} \hbar \tilde{\omega}_{\mathbf{uK}} \left( g_{\mathbf{uK}}^\dagger \mathbf{g}_u (\mathbf{r}) + i \hbar \tilde{\omega}_{\mathbf{uK}}^{(1)i} \left( \frac{\partial g_{\mathbf{uK}}^\dagger}{\partial r^i} \mathbf{g}_u (\mathbf{r}) - \text{c.c.} \right) \right) + \frac{1}{4} \hbar \tilde{\omega}_{\mathbf{uK}}^{(2)ij} \left( \frac{\partial}{\partial r^i} g_{\mathbf{uK}}^\dagger \frac{\partial}{\partial r^j} \mathbf{g}_u (\mathbf{r}) + \text{c.c.} \right). \]  
(3.57)
Using Hamilton’s equation and integrating by parts yields a Schrödinger equation for the polariton effective fields,
\[
\frac{i}{\partial t} g_u (r, t) - \tilde{\omega}_{\mathbf{k}} g_u (r, t) + i \tilde{\omega}_{\mathbf{k}}^{(1)} \frac{\partial}{\partial r^i} g_u (r, t) + \frac{1}{2} \tilde{\omega}_{\mathbf{k}}^{(2)} \frac{\partial^2}{\partial r^i \partial r^j} g_u (r, t) = 0. \tag{3.58}
\]
Propagation of a slowly varying pulse is thus reduced to a single Schrödinger equation, and both modal and material effects are aggregated in the coefficients obtained from the Taylor expansion (3.54) of the polariton dispersion relation. The coefficients \( \tilde{\omega}_{\mathbf{k}} \) and \( \tilde{\omega}_{\mathbf{k}}^{(1)} \) give the carrier frequency and group velocity of the wave packet, and if desired the terms involving these coefficients can be eliminated from Eq. (3.58) using a simple transformation [2]. The coefficient \( \tilde{\omega}_{\mathbf{k}}^{(2)} \) gives both the group velocity dispersion and the diffraction. Higher order dispersion and diffraction terms are easily obtained from successive terms in the Taylor expansion, if desired.

The non-fluctuating part of Eq. (3.58) is an initial value problem for \( \overline{g}_u (r, t) \), which we define in terms of \( \overline{c}_u \) in the same way that \( g_u (r, t) \) is defined in terms of the \( c_u \) in Eq. (3.50). Since any initial \( \overline{c}_u (0) \) can be generated, it follows that we can have any desired initial condition for the non-fluctuating part \( \overline{g}_u (r, 0) \) of (3.58), as long as its \( k \)-space Fourier spectrum is supported on \( D \). We make a mild approximation by allowing a negligible tail of the Fourier spectrum of \( \overline{g}_u (r, 0) \) to extend outside of \( D \).

It is possible to write the displacement field in terms of the effective field \( g_u (r, t) \). To do this we expand \( d_{u \mathbf{k}} (r) \) in a Taylor series in \( \mathbf{k} \) about \( \overline{\mathbf{k}} \), then integrate by parts in (3.9). The result is
\[
D (r, t) = e^{i \mathbf{k} \cdot \mathbf{r}} \sum_u \left( d_{u \mathbf{k}} (r) g_u (r, t) - i [\partial_{\mathbf{k}} d_{u \mathbf{k}} (r)]_{\mathbf{k}=\overline{\mathbf{k}}} \cdot \frac{\partial}{\partial r} g_u (r, t) + \ldots \right) + c.c.. \tag{3.59}
\]
We wish to ensure that slow spatial variation in \( g_u (r, t) \) implies slow spatial variation in \( D (r, t) \), and this requires that \( d_{u \mathbf{k}} (r) \) be a continuous function of \( \mathbf{k} \) over the range of wave vectors involved in the wave packet. This issue has been discussed in more detail for nondispersive systems [3], and the same discussion applies here.

### 3.4 Propagation in the absorptive regime

Propagation in the absorptive regime differs significantly from the transparent regime in several ways. First, polaritons exist at all (real) frequencies and wave vectors in the
absorption regime. Differences also arise because fluctuations are intimately linked with the dissipation. Furthermore, since any realistic field decays into the future, it must also formally be generated by some source in the past.

As in the transparent regime, here in our analysis of the absorptive-regime dynamics we need not consider the specific profile of $P_s(r,t)$. Instead we begin our analysis after $H_{\text{drive}}$ has subsided by specifying an initial condition for the nonfluctuating part of the state at $t = 0$ that is consistent with generation via $P_s(r,t)$. Specifically, it is seen in what follows that any absorptive regime pulse profile can be generated using some $P_s(r,t)$. However, the situation here is somewhat more nuanced than that of the transparent regime; it is described in the subsections below, and is analyzed in detail in Appendix D. For simplicity, in this section we take $\Omega_g = 0$, so we can neglect the transparent regime and focus on the absorptive one. However, since the transparent regime operators are left undisturbed by $P_s(r,t)$ except for perhaps some negligible effect, the results of this section generalize in an immediate and obvious way to the case of $\Omega_g \neq 0$.

It is common in the literature on absorptive media to calculate a dispersion relation $k(\omega)$ with $\omega$ a real variable, and with $k$ that is in general complex. From this one can construct dynamical equations for the electromagnetic fields which treat the dynamical variables as functions of space rather than functions of time, i.e. space is taken as the evolution parameter. But for pulse propagation we would like to be able to specify the fields at an initial time and be able to describe forward evolution, and the space-as-evolution-parameter approach is not naturally an initial value problem.

An alternative is to calculate a dispersion relation $\omega(k)$ with $k$ real, so that $\omega$ is in general complex; but this has the drawback that realistic dispersive and absorptive response functions are generally only specified at real frequencies. The complex $k$, real $\omega$ dispersion relation can be quite different from the complex $\omega$, real $k$ dispersion relation, and in some parts of $(k,\omega)$-space the relationship between the two is nontrivial [16] even at lowest order. The cost involved here in modeling the system by a continuum of oscillator fields is that the response function $\Gamma(r,\omega)$ is only equal to the actual physical...
response function for real $\omega$ (and for frequencies in the upper half-plane)\(^3\), so care is needed. But use of the response function only at real frequencies is of course sufficient, since for materials of interest the actual physical response function is never tabulated for complex $\omega$ anyway [19]; in principle a calculation at complex $\omega$ could be done within an \textit{ab-initio} study of the material response function, but \textit{ab-initio} calculations of material response functions are an involved affair [104].

Instead of using a complex dispersion relation for the electromagnetic fields, we rely on the real $\omega$, real $k$ dispersion relation of the polariton fields that formally arises from Fano-type diagonalization of the full Hamiltonian of the system. The relevant part of the polariton dispersion relation can be calculated using the methods of Sec. 3.1, and we find that the complex $\omega$, real $k$ dispersion relation of the electromagnetic fields arises naturally in the dynamical equation we obtain for the polariton effective field. Indeed decay, and with it all orders of absorption, dispersion, and diffraction—material and modal—are captured in the various $k$ and $\Omega$ derivatives of the real function $Z_{\Omega k}$, the resonant oscillator amplitude.

### 3.4.1 Composite polariton operators

Since polaritons exist at all frequencies for each given label $m$ in the absorptive regime, there is no polariton dispersion relation in the usual sense of having a discrete set of $\Omega$ for any given label $m$. However, it is possible to recover “composite” polariton operators $c_m(t)$ that are labeled only by usual mode labels $m$, that additionally have simple commutation relations, and that play a role in the absorptive regime similar to the role played by the $c_m(t)$ in the transparent regime. We define these composite polariton operators according to

$$c_m(t) \equiv \int d\Omega \phi_{\Omega m} c_{\Omega m}(t),$$

where

$$\phi_{\Omega m} \equiv L_{\Omega m}/\sqrt{\nu_m},$$

---

\(^3\)This can be seen from the expression for the model response function in terms of the actual response function based on the detailed underlying physics. Writing these model and actual response functions respectively as $\Gamma_M(r, \omega)$ and $\Gamma_P(r, \omega)$,

$$\Gamma_M(r, \omega) = \frac{2}{\pi} \int_{\Omega_g} d\Omega \frac{\Omega \text{Im} \Gamma_P(r, \Omega)}{(\Omega - \omega - i0)(\Omega + \omega + i0)},$$

which implies a branch cut along the real axis when $\Omega_g = 0$. Details of the model response properties are elaborated in Sec. 2.2.
\[
\nu_m \equiv \int_{\Omega_n} d\Omega |L_{\Omega m}|^2. \tag{3.62}
\]

We will see that the operator \(c_m (t)\) is an absorptive regime analogue of the transparent regime operator \(c_m (t)\), although \(c_m (t)\) is actually a frequency integral over polariton operators, with the \(c_{\Omega m} (t)\) weighted by the factor \(\phi_{\Omega m}\). We note that in an expansion of \(Z_{\Omega m}\) in terms of \(\Omega\) kept to linear order, \(|\phi_{\Omega m}|^2\) is a normalized Lorentzian in \(\Omega\), and the real and imaginary parts of the complex frequency pole of \(\phi_{\Omega m}\) are respectively the band center frequency \(\Omega_m\) defined in Eq. (3.21), and the decay rate due to absorption. This is elaborated upon in Appendix E.

It follows from (3.60, 3.61, 2.66) that
\[
\left[ c_m (t), c_{m'}^\dagger (t) \right] = \delta_{mm'}. \tag{3.63}
\]

We define \(c_m\) and \(\tilde{c}_m\) operators in terms \(\tilde{c}_{\Omega m}\) and \(\phi_{\Omega m}\) analogously to (3.60). Then
\[
c_m (t) = \tilde{c}_m (t) + \tilde{c}_m (t), \tag{3.64}
\]
where \(\tilde{c}_m (t)\) is a c-number and we have
\[
\left[ \tilde{c}_m (t), (\tilde{c}_{m'} (t))^\dagger \right] = \delta_{mm'}. \tag{3.65}
\]

Recalling from the discussion after Eq. (3.28) that \(D_{\Omega m} (r)\) is continuous and smooth in \(\Omega\), and slowly varying in \(\Omega\) over the frequency band width of the state of interest, we can take a Taylor expansion of the displacement field modes about the central frequency \(\Omega_m\) defined in Eq. (3.21):
\[
D_{\Omega m} (r) = D_{\Omega m} (r) + (\Omega - \Omega_m) \left[ \partial_\Omega D_{\Omega m} (r) \right]_{\Omega = \Omega_m} + \frac{1}{2} (\Omega - \Omega_m)^2 \left[ \partial^2_\Omega D_{\Omega m} (r) \right]_{\Omega = \Omega_m} + \ldots \tag{3.66}
\]

After \(P_s\) subsides, i.e. for \(t > 0\), the displacement field operator can be written simply in terms of the \(c_{uk} (t)\). We find it convenient to separate the non-fluctuating and fluctuating parts of \(D (r, t)\) according to \(D (r, t) = \overline{D} (r, t) + D^0 (r, t)\). Using Eqs. (3.9, 3.28) we obtain
\[
\overline{D} (r, t) = \hbar^{1/2} \sum_u \int_{D} dke^{-i\Omega_k t} \sqrt{\nu_{uk}} \left[ \tilde{c}_{uk} (t) D_{\Omega_m, uk} (r) + [\partial_\Omega D_{\Omega m} (r)]_{\Omega = \Omega_m} + \ldots \right] + c.c., \tag{3.67}
\]
where \(\tilde{c}_{uk} (t) \equiv \exp \left( i\Omega_m t \right) \tilde{c}_{uk} (t)\) and \(m = (u, k)\). The fluctuating part of the displacement field can be written as
\[
D^0 (r, t) = \sum_m \int_{\Omega_n} d\Omega \left[ c_{\Omega m}^0 (t) \tilde{D}_{\Omega m} (r) + (c_{\Omega m}^0 (t))^\dagger \tilde{D}_{\Omega m}^* (r) \right]. \tag{3.68}
\]
Comparing Eq. (3.67) to Eq. (3.9) we see that when absorption is weak, at lowest order, up to factors, $\bar{c}_m(t)$ and $D_{\Omega m}(r)$ play roles for the absorptive regime analogous to those played by $\bar{c}_m(t)$ and $\tilde{D}_m(r)$ for the transparent regime. That is, the non-fluctuating part of the composite polariton operator of the absorptive regime is analogous to the non-fluctuating part of the transparent-regime polariton operator.

### 3.4.2 Effective fields

We are interested in propagation of a field containing some narrow band of frequencies and wave vectors composed of neighboring modes in $\Omega$ and $\mathbf{k}$. The pulse is taken to be centered at wave vector $\mathbf{K}$, and only modes with wave vector $\mathbf{k}$ near $\mathbf{K}$ contribute to the pulse. Furthermore, there will typically only be a significant contribution from modes in the frequency bandwidth around $\Omega \omega_k$, i.e., such that $|Z_{\Omega \omega_k}|$ is small, since these are the modes that are primarily electromagnetic in character, rather than primarily composed of medium modes [1].

Analogous to Eq. (3.50) for the transparent regime, here we posit a field of the form

$$g_u(r, t) \equiv (2\pi)^{-3/2} \int_D d\mathbf{k} c_{\omega_k}(t) e^{i(\mathbf{k} - \mathbf{K}) \cdot \mathbf{r}},$$

(3.69)

where the dependence of $g_u$ on $\mathbf{K}$ is kept implicit. The upper half of Fig. 3.1 shows schematically the content of the effective field $g_u(r, t)$ in the absorptive regime.

From the definitions of $g_u(r, t)$ and $\phi_{\Omega m}$ and (2.66) it follows that

$$\left[ g_u(r, t) , g^\dagger_{u'}(r', t) \right] = \delta_{uu'} \tilde{\Pi}(r - r'),$$

(3.70)

where we recall the definition of $\tilde{\Pi}(r)$ in Eq. (3.52). Of general use are the commutators

$$\left[ g_{u'}(r, t) , c^\dagger_{\omega_k}(t) \right] = (2\pi)^{-3/2} \delta_{uu'} \phi_{\Omega \omega_k} e^{i(\mathbf{k} - \mathbf{K}) \cdot \mathbf{r}},$$

(3.71)

$$\left[ g_{u'}(r, t) , c_{\omega_k}(t) \right] = (2\pi)^{-3/2} \delta_{uu'} e^{i(\mathbf{k} - \mathbf{K}) \cdot \mathbf{r}}.$$  

(3.72)

Again, we find it convenient to separate the fluctuating and nonfluctuating pieces of the displacement field. For the nonfluctuating part, it is convenient to work in terms of the field

$$\bar{g}_u(r, t) \equiv (2\pi)^{-3/2} e^{i\Pi_{\omega_k} t} \int_D d\mathbf{k} (\nu_{\omega_k})^{1/2} \bar{c}_{\omega_k}(t) e^{i(\mathbf{k} - \mathbf{K}) \cdot \mathbf{r}}.$$  

(3.73)

The definition (3.73) differs in two ways from the field obtained by using $\bar{c}_{\omega_k}(t)$ instead of $c_{\omega_k}(t)$ in Eq. (3.69). First, the new field contains a factor $\exp (i\Pi_{\omega_k} t)$, so it is slowly
varying in time. Second, the integrand differs by a factor of $\sqrt{\nu_k}$, so $\bar{g}_u (\mathbf{r}, t)$ depends on $L_{\Omega u k}$ rather than $\phi_{\Omega u k}$, and is in fact independent of $\nu_k$ (see Eq. (3.60)). We note here that it is shown in Appendix D that any initial effective field $\bar{g}_u (\mathbf{r}, 0)$ can be generated using a suitable driving polarization $\mathbf{P}_s (\mathbf{r})$ that subsides before time $t = 0$.

For positive times, the field $\mathbf{D} (\mathbf{r}, t)$ can be written simply in terms of the $\bar{g}_u (\mathbf{r}, t)$ field. We have

$$
\mathbf{D} (\mathbf{r}, t) = \hbar^{1/2} \sum_u e^{i (k \cdot \mathbf{r} - \Omega u k t)} \left[ \mathbf{d}_{\Omega u k} (\mathbf{r}) \bar{g}_u (\mathbf{r}, t) + [\partial_k \mathbf{d}_{\Omega u k} (\mathbf{r})] \frac{1}{i} \frac{\partial}{\partial t} \bar{g}_u (\mathbf{r}, t) \right. 
$$

$$
\left. + [\partial_\Omega \mathbf{d}_{\Omega u k} (\mathbf{r})] \frac{1}{i} \frac{\partial}{\partial \Omega} \bar{g}_u (\mathbf{r}, t) + \ldots \right] + \text{c.c.}
$$

(3.74)

The fluctuating part of the displacement field, $\mathbf{D}^0 (\mathbf{r}, t)$, is given by the expression (3.68). Using Eqs. (3.74) and (3.68) we can express the field $\mathbf{D} (\mathbf{r}, t) \equiv \mathbf{D} (\mathbf{r}, t) + \mathbf{D}^0 (\mathbf{r}, t)$ in a convenient way. The non-fluctuating (expectation value) part of the $\mathbf{D}$ field is expressed simply in terms of the effective field $\bar{g}_u (\mathbf{r}, t)$, which will be a slowly-varying quantity in propagation problems of interest, and for which we obtain a dynamical equation in the following subsections. And the fluctuating part of the field is expressed in terms of the vacuum polariton operators and mode fields.

### 3.4.3 Differential equation for polariton effective field

Our goal is to obtain a dynamical equation for $\bar{g}_u (\mathbf{r}, t)$. We begin with an intermediate step, which is to derive a differential equation for $\bar{g}_u (\mathbf{r}, t)$, involving spatial and temporal derivatives up to a finite but arbitrary truncation order $n$.

We will see that it is useful to consider the function

$$
\gamma_{uk} (\Omega) \equiv \frac{1}{L_{\Omega u k}},
$$

(3.75)

which we establish is continuous and differentiable at $\Omega$ and $k$ of interest for propagation as follows. First it is seen from the forms of the effective master equation (3.19) and effective response function (3.20) that $Z_{\Omega u k}$ changes continuously in both $k$ and $\Omega$, as long as $\Gamma (\mathbf{r}, \Omega)$ is sufficiently well-behaved at frequencies of interest, and as long as modes are labeled to ensure continuous mode fields across points of degenerate $\Omega$. Next it is shown in Appendix E that the frequency dependence of $Z_{\Omega u k}$ is approximated to arbitrary order as a polynomial in frequency over the relevant domain, and for such a $Z_{\Omega u k}$ it follows from the form in Eq. (3.27) of $L_{\Omega u k}$ in terms of the roots of that polynomial that $L_{\Omega u k}$
is continuous and differentiable in both $\Omega$ and $k$. Finally, it follows from the reality of $Z_{\Omega k}$ and the form of Eq. (3.26) that $L_{\Omega k} \neq 0$ for any $\Omega$ or $k$, and in conjunction with the fact that $L_{\Omega k}$ is continuous and differentiable this implies that $\gamma_{\Omega k}(\Omega)$ is continuous and differentiable.

Performing a Taylor expansion of $\gamma_{\Omega k}(\Omega)$ in $k$ about $\bar{k}$ yields

$$\gamma_{\Omega k}(\Omega) = \gamma_{\Omega k}^{(0)}(\Omega) + (k - \bar{k}) \cdot \gamma_{\Omega k}^{(1)}(\Omega) + (k - \bar{k}) (k - \bar{k}) : \gamma_{\Omega k}^{(2)}(\Omega) + \ldots,$$

(3.76)

where

$$\gamma_{\Omega k}^{(n)}(\Omega) = \frac{1}{n!} \left[ (\nabla_k)^n \gamma_{\Omega k}(\Omega) \right]_{k=\bar{k}}.$$

(3.77)

Next we consider the differential operator $Y_u^{(n)}$, defined according to

$$Y_u^{(n)} g_u(r, t) \equiv \gamma_{\Omega k}^{(n)}(\Omega) \bar{g}_u(r, t) + \left[ \partial_\Omega \gamma_{\Omega k}^{(n)}(\Omega) \right]_{\Omega=\bar{\Omega}} (i\partial_t) \bar{g}_u(r, t)$$

$$+ \frac{1}{2} \left[ (\partial_\Omega)^2 \gamma_{\Omega k}^{(n)}(\Omega) \right]_{\Omega=\bar{\Omega}} (i\partial_t)^2 \bar{g}_u(r, t) + \ldots,$$

(3.78)

The expansion is taken up to some suitable finite order and, using the argument of Appendix F, the remainder can be shown to be negligible. Using the result (F.14) from Appendix F, (3.78) can be written as the convolution integral

$$Y_u^{(n)} g_u(r, t) \equiv \int_{-\infty}^{\infty} ds y_u^{(n)}(t - s) \bar{g}_u(r, s),$$

(3.79)

where $y_u^{(n)}(t)$ is any function such that its Fourier transform matches $\gamma_{\Omega k}^{(n)}(\Omega)$ over the entire frequency bandwidth over which $\bar{g}_u(r, t)$ is non-negligible.

The effective field $\bar{g}_u(r, t)$ obeys the partial differential equation

$$\sum_{n \geq 0} (-i \nabla_r)^n Y_u^{(n)} \bar{g}_u(r, t) = \bar{f}(r, t),$$

(3.80)

where

$$\bar{f}(r, t) \equiv (2\pi)^{-3/2} e^{i\bar{\Omega} \cdot \bar{r}} \int d\kappa \int d\Omega_{\Omega_\Omega k} (t) e^{i\kappa \cdot r}.$$

(3.81)

This can be verified by substitution of (3.73) for $\bar{g}_u(r, t)$ into the LHS of (3.80). The field $\bar{f}(r, t)$ is associated with the generation of the “initial” field $\bar{g}_u(r, 0)$. In Appendix D we show that for all $t \geq 0$, we have $\bar{f}(r, t) = 0$, after making very mild assumptions about the pulse and material response.
3.4.4 Multiple scales and dynamical equation

In order to evolve $\mathbf{g}_u(r, t)$ in time we require a dynamical equation to replace (3.80) that is first order in time. We here derive one for a narrowband pulse via the asymptotic method of multiple scales [2]. The idea is to consider the pulse to evolve on many scales in both space and time, and to relate these scales to a small parameter $\eta$. Then we keep terms in (3.80) up to a given order in $\eta$, so in an asymptotic sense we capture the dynamics of the exact solution at that order. We explicitly separate the different space and time scales by writing the effective field as

$$g_u(r, t) = G(x, \eta x, \eta^2 x, \ldots, y, \eta y, \eta^2 y, \ldots, z, \eta z, \eta^2 z, \ldots; t, \eta t, \eta^2 t, \ldots)$$

(3.82)

$$\equiv G(r_0, r_1, r_2, \ldots; t_0, t_1, t_2, \ldots),$$

where $G$ is considered to vary equally significantly as each of its arguments varies over a distance $d$ or period $\tau_{cw}$ associated with the wavelength and carrier wave period respectively:

$$d \equiv \frac{2\pi}{k} \quad \text{and} \quad \tau_{cw} \equiv \frac{2\pi}{\Omega_u k},$$

where $k \equiv |\mathbf{k}|$. The ranges and periods $d^{(p)} = d/\eta^p$ and $\tau^{(p)} = \tau_{cw}/\eta^p$ for $p \geq 0$ define the multiple scales of the problem. The field $g_u(r, t)$ is defined (3.73) such that the most rapid time and space variations of the operator $D(r, t)$ are included in the carrier frequency factor and spatial mode. So we take $g_u(r, t)$ to be slowly varying in space and time, i.e., $G$ is taken to be independent of $r_0$ and $t_0$.

Derivatives are given by

$$\frac{\partial g_u(r, t)}{\partial t} = \eta \frac{\partial G}{\partial t_1} + \eta^2 \frac{\partial G}{\partial t_2} + \ldots + \eta^n \frac{\partial G}{\partial t_n} + O(\eta^{n+1}),$$

(3.83)

$$\frac{\partial g_u(r, t)}{\partial r} = \eta \frac{\partial G}{\partial r_1} + \eta^2 \frac{\partial G}{\partial r_2} + \ldots + \eta^n \frac{\partial G}{\partial r_n} + O(\eta^{n+1}),$$

(3.84)

where $\partial/\partial \mathbf{r}$ denotes the gradient. For convenience we define coefficients $\widetilde{C}_m^{(n,l)} (m = (u, \mathbf{K}))$ according to

$$\widetilde{C}_m^{(n,l)} = l^{1-n-1} \frac{\Omega_k^{-1} k^n \left[ (\partial_\Omega)^l \gamma_{uK}^{(n)} (\Omega) \right]}{\left[ \partial_\Omega \gamma_{uK}^{(0)} (\Omega) \right]_{\Omega}},$$

(3.85)

with $\Omega = \Omega_u k$. Note that by construction we have $\widetilde{C}_m^{(0,1)} = 1$. Then (3.80) can be written for $t \geq 0$ as

$$\sum_{n,l \geq 0} \widetilde{C}_m^{(n,l)} \left( k^{-1} \nabla_r \right)^n \left( \Omega_k^{-1} \partial_t \right)^l g_u (r, t) = 0.$$
Here we note that \( \tilde{C}_m^{(n,l)} \) is a unitless \( n \)-component tensor, and we take the convention that wherever \( \tilde{C}_m^{(n,l)} (\nabla_r)^n \) appears, an inner product over the \( n \) factors of \( \nabla_r \) is implied.

In past work we have given a detailed discussion of scaling in the context of pulse propagation [2]. Here, the three most important time scales in the problem are i) the carrier wave period \( \tau_{cw} \), as defined after Eq. (3.82); ii) a decay time \( \tau_{de} \), which we take to be the time for the electromagnetic energy in the pulse to halve; iii) a characteristic propagation time \( \tau_{pr} \), which is taken to be the time for the pulse to travel a distance comparable to its spatial length. We are interested in pulses for which the carrier wave period is the shortest of these three time scales, and in which the decay time is longer than the propagation time, i.e. \( \tau_{cw} < \tau_{pr} < \tau_{de} \). The carrier frequency has already been factored out. Propagation such that \( \tau_{pr} < \tau_{de} \) has the following scaling relationships of the coefficients in Eq. (3.86), which are taken to be scaling assumptions:

\[
\tilde{C}_m^{(0,0)} = \mathcal{O} (\eta),
\]

and for all other \( n \) and \( l \) we take

\[
\tilde{C}_m^{(n,l)} = \mathcal{O} (1).
\]

The choice that \( \tilde{C}_m^{(0,0)} \) is smaller by a factor in \( \eta \) than the other coefficients reflects that \( \tilde{C}_m^{(0,0)} \) will determine the decay time.

Now consider Eq. (3.86) at various orders. The lowest order is \( \mathcal{O} (\eta^1) \). At that order we have

\[
\tilde{C}_m^{(0,0)} G + \frac{1}{\Omega} \frac{\partial G}{\partial t_1} + \tilde{C}_m^{(1,0)} \frac{1}{k} \frac{\partial G}{\partial r_1} = 0.
\]

At order \( \mathcal{O} (\eta^2) \) we have

\[
0 = \tilde{C}_m^{(0,1)} \left( \frac{1}{\Omega} \frac{\partial G}{\partial t_1} \right) + \tilde{C}_m^{(0,2)} \left( \frac{1}{\Omega} \frac{\partial}{\partial t_1} \right)^2 G + \tilde{C}_m^{(1,1)} \frac{1}{k \Omega} \frac{\partial}{\partial r_1} \frac{\partial G}{\partial t_1} + \tilde{C}_m^{(2,0)} \left( \frac{1}{k} \frac{\partial}{\partial r_1} \right)^2 G + \tilde{C}_m^{(1,0)} \frac{1}{k} \frac{\partial G}{\partial r_2}.
\]

Similarly we construct the order \( \mathcal{O} (\eta^3) \) equation, but do not write it here. Note that (3.89) gives \( \partial G/\partial t_1 \) in terms of spatial derivatives at lowest order. Substituting (3.89) into (3.90) we obtain an equation for \( \partial G/\partial t_2 \) in terms of spatial derivatives.

Finally, we construct an equation for \( \partial \mathcal{g}/\partial t \) by substituting (3.89) and (3.90) into the RHS of (3.83). Then identifying space and time derivatives according to (3.84) and (3.83) we obtain the Schrödinger equation. The final dynamical equations at first and second order in \( \eta \) are both independently of interest.
Including all terms up to order $O(\eta^2)$ we have, for $t \geq 0$,
\[
i \frac{\partial g_u(r,t)}{\partial t} - w_{u\mathbf{k}}^{(0)} g_u(r,t) + i w_{u\mathbf{k}}^{(1)} \frac{\partial g_u(r,t)}{\partial r} + \frac{1}{2} w_{u\mathbf{k}}^{(2)} \frac{\partial^2 g_u(r,t)}{\partial r^2} = 0, \quad (3.91)
\]
where the propagation parameters are
\[
w_{u\mathbf{k}}^{(0)} = - \left[ C_m^{(0,0)} + C_m^{(0,2)} \left( C_m^{(0,0)} \right)^2 \right], \quad (3.92)
\]
\[
w_{u\mathbf{k}}^{(1)} = - \left[ C_m^{(1,0)} - C_m^{(1,1)} C_m^{(0,0)} + 2 C_m^{(0,0)} C_m^{(1,0)} C_m^{(0,2)} \right], \quad (3.93)
\]
\[
w_{u\mathbf{k}}^{(2)} = -2 \left[ C_m^{(2,0)} - C_m^{(1,0)} C_m^{(1,1)} + C_m^{(2,0)} \left( C_m^{(1,0)} \right)^2 \right], \quad (3.94)
\]
where
\[
C_m^{(n,l)} \equiv \frac{1}{n!l!} \frac{\left[ (\partial_\Omega)^l \left[ (\nabla_k)^n \gamma_{u\mathbf{k}}(\Omega) \right] \right]}{\left[ \partial_r \gamma_{u\mathbf{k}}(\Omega) \right]}, \quad (3.95)
\]
Eq. (3.91) is a Schrödinger equation. Here the dynamical equation for the nonfluctuating part of the effective field has the same form as the dynamical equation (3.58) obtained for the effective field in the transparent regime, but with coefficients that are complex in contrast with the real coefficients in Eq. (3.58). The imaginary part of the coefficient $w_{u\mathbf{k}}^{(0)}$ is a decay constant associated with absorption, and a negative value implies fields decay into the future. The real part of $w_{u\mathbf{k}}^{(0)}$ represents a carrier-wave frequency shift. Here $\text{Re} w_{u\mathbf{k}}^{(0)}$ is analogous to $\tilde{\omega}_{u\mathbf{k}}^{(0)}$ in Eq. (3.58) for the transparent regime, but represents a carrier shift rather than a carrier frequency because, unlike the transparent regime effective field, the absorptive regime $g_u(r,t)$ is defined after factoring out the carrier frequency to make it slow-varying. Contributing to $w_{u\mathbf{k}}^{(0)}$, the first term in Eq. (3.92) accounts for absorption at the lowest order, and the second term is the second-order correction. The coefficient $w_{u\mathbf{k}}^{(1)}$ is the complex group velocity vector, and we recall from the discussion after Eq. (3.86) that in Eq. (3.91) a dot product is implied between the factor $w_{u\mathbf{k}}^{(1)}$ and the gradient of $g_u(r,t)$. The real part of $w_{u\mathbf{k}}^{(1)}$ is the pulse group velocity, analogous to $\tilde{\omega}_{u\mathbf{k}}^{(1)}$ in Eq. (3.58) for the transparent regime effective field. The imaginary part of $w_{u\mathbf{k}}^{(1)}$ captures an absorption effect that depends on pulse width. The first term in Eq. (3.93) is the lowest order contribution to the group velocity; the second and third terms are 2nd order corrections. The coefficient $w_{u\mathbf{k}}^{(2)}$ is a second rank tensor, and again it is complex. Its imaginary part is found to be small in the example calculation of Section 3.6. Like its transparent regime analogue $\tilde{\omega}_{u\mathbf{k}}^{(2)}$ in Eq. (3.58), the real part of $w_{u\mathbf{k}}^{(2)}$ captures both pulse dispersion and diffraction.
Also like Eq. (3.58) for the transparent regime, Eq. (3.91) is an initial value problem. It is shown in Appendix D that there exists an infinite set of $P_s(r,t)$ to generate any initial condition $\overline{g}_u(r,0)$ consistent with the multiple-scales expansion (3.82) and with the requirement of no variation on the shortest time and space scales $t_0$ and $r_0$ described after Eq. (3.82). It is also shown there that there is an infinite set of $P_s(r,t)$ to generate any initial condition for the non-fluctuating part of the transparent regime effective field $\overline{g}_u(r,0)$ in Eq. (3.58). However, unlike in the transparent regime, here in the absorptive regime the initial condition $\overline{g}_u(r,0)$ actually underdetermines the state $\overline{c}_{\Omega u}(0)$; this is discussed in more detail in Appendix D. However, as long as the solution can be taken to be of the form (3.82) and the scaling assumptions (3.87, 3.88) are met, the evolution of $\overline{g}_u(r,t)$ is given approximately by the initial value problem (3.91), and additional information about which specific $P_s(r,t)$ is used to formally generate $\overline{g}_u(r,0)$ makes no difference to the dynamics in this multiple-scales approximation. Thus it suffices to consider Eq. (3.91) as an initial value problem for $t \geq 0$ only, and $\overline{g}_u(r,0)$ can be taken as an initial condition to be freely chosen as long as it is slowly varying on the spatial scale of a unit cell for a periodic system.

We note also that the multiple-scales approach can be used to write the displacement field in terms of the $g$ field at a single time. Substituting (3.91) into (3.74) so as to convert the time derivative into a spatial derivative, we obtain for $t \geq 0$,

$$D(r,t) = \frac{\hbar}{2} \sum_u e^{i(\mathbf{k} \cdot \mathbf{r} - \Omega u t)} \left[ (d_{\Omega u k}(r) + w_u^{(0)} \frac{\partial}{\partial t} d_{\Omega u k}(r)) \overline{g}_u(r,t) \right] \overline{g}_u(r,t) + \left( \frac{\partial k d_{\Omega u k}(r)}{\partial k} + w_u^{(1)} \left( \frac{\partial}{\partial t} d_{\Omega u k}(r) \right) \right) \frac{1}{i} \frac{\partial}{\partial t} \overline{g}_u(r,t) + \ldots + c.c.$$  \hspace{1cm} (3.96)

As a further aside we note here that if $\overline{g}_u(r,t)$ is uniform in space, Eq. (3.91) reduces to a dynamical equation for $\overline{c}_{\Omega u}(t)$.

### 3.4.5 Propagation parameters in terms of the polariton dispersion relation

A key advantage of the dynamical equation (3.91) is that its coefficients are easily calculated despite the multitude of physical effects included in its derivation. Comparing Eqs. (3.92, 3.93, 3.94) with Eqs. (3.95, 3.77, 3.75, 3.61, 3.27, 3.26, 3.19), it is clear that the propagation parameters $w_u^{(n)}$ are functions of the resonant oscillator amplitude $Z_{\Omega u k}$ and its $\Omega$- and $k$- derivatives around the pulse carrier wave at $(\mathbf{k},\Omega_{\Omega u k})$. So all aspects
of pulse propagation can be extracted from photonic band structure calculations for the nonabsorptive system with effective response $\Gamma (r, \Omega; Z)$ defined in (3.20), and only $Z_{\Omega_uk}$ values in the neighborhood of $Z = 0$ need be considered. This is a key advantage of this method: Values of $Z_{\Omega_uk}$ are easily calculated using standard codes for calculating photonic band structures of structured media [10][11][94].

In fact, for the cutting surface method of Toader and John [94], the $\Omega_{uk} (Z)$ dispersion relation for various $Z$ sits within the generalized dispersion surfaces, so it can be extracted without any additional computations involving the eigenvalue equation beyond those needed to obtain the dispersion relation of a dispersive, nonabsorptive system.

This would work as follows. The cutting surface method is designed for composite structured media consisting of one nondispersive material (e.g. air) and one dispersive material. It is a two-step process: first is the computationally intensive step of computing the generalized dispersion surfaces (GDS); second is the computationally non-intensive step of intersecting the GDS with a cutting surface corresponding to $\Gamma (\Omega)$ for the dispersive material to obtain the band structure $\Omega_{uk}$. In my application, the computationally intensive step is exactly the same. In the second step we intersect the GDS with the effective response $\Gamma (\Omega, Z)$ for the dispersive material to obtain $\Omega_{uk} (Z)$. In other words, the full dispersive and absorptive polariton dispersion relation can be obtained with only negligible extra computing time compared to obtaining the electromagnetic dispersion relation while neglecting absorption.

The analytic expressions for the $w^{(j)}_{uk}$ for $j = 0, 1, 2$ are composed solely from $\gamma_{uk} (\Omega)$, as can be seen from Eqs. (3.92, 3.93, 3.94) in conjunction with Eq. (3.95). To calculate these parameters we expand $Z_{\Omega_uk}$ to quadratic order in $\Omega$ around $\Omega_{uk}$. The details of this calculation are given in Appendix E. Here we define the quantities

$$Z'_{uk} \equiv \left[ \frac{\partial Z_{\Omega_uk}}{\partial \Omega} \right]_{\Omega_{uk}},$$

$$Z''_{uk} \equiv \left[ \frac{\partial^2 Z_{\Omega_uk}}{\partial \Omega^2} \right]_{\Omega_{uk}}.$$  (3.97)

From the reciprocal of Eq. (E.16), the $\gamma_{uk} (\Omega)$ are given by

$$\gamma_{uk} (\Omega) = \frac{|Z''_{uk}|}{2\sqrt{\pi}} \left( \Omega - \Omega^{(1)}_{uk} \right) \left( \Omega - \Omega^{(2)}_{uk} \right),$$  (3.99)

where $\Omega^{(1)}_{uk}$ is the one of $\{ \Omega^+_{uk}, (\Omega^+_{uk})^* \}$ that is in the LHP, and where $\Omega^{(2)}_{uk}$ is the one of
\{ \Omega_{\pm}^u, (\Omega_{\pm}^u)^* \} in the LHP, where \( \Omega_{\pm}^u \) is given by

\[
\Omega_{\pm}^u = \Omega_{u}^k + \frac{Z_{u}^u Z_{u}^u}{Z_{u}^u} \left[ -1 \pm \sqrt{1 - 2 \pi i \frac{Z_{u}^u}{(Z_{u}^u)^2}} \right].
\]

(3.100)

We note that in many systems of interest, including the lowest band of the example structure that is treated in Secs. 3.6 and 3.7, we have that \( |Z_{u}^u| \ll |Z_{u}^u|^2 \); intuition for this relation is given after Eq. (3.103). A binomial approximation can then be used in Eq. (3.100), and we have

\[
\Omega_{u}^{(1)} = \Omega_{u}^k - \frac{i \pi}{|Z_{u}^u|},
\]

(3.101)

\[
\Omega_{u}^{(2)} = \left( 1 - \frac{2Z_{u}^u}{\Omega_{u}^k Z_{u}^u} \right) \Omega_{u}^k - \frac{i \pi}{|Z_{u}^u|}.
\]

(3.102)

It is no more difficult to calculate \( \Omega_{u}^{(1)} \) and \( \Omega_{u}^{(2)} \) using the full expression (3.100) than it is using the binomial approximation, so in numerical calculations this binomial approximation need not ever be used.

### 3.4.6 First order dynamical equation

Often the first order propagation dynamics are all that is needed, for example in systems in which loss and group velocity are all that need to be considered, so the term with the second order derivatives in Eq. (3.91) can be neglected. It is common in the literature to calculate propagation at this level [18]. Then the Schrödinger equation becomes for \( t \geq 0 \),

\[
\frac{i}{\hbar} \frac{\partial g_u(r,t)}{\partial t} - w_{u}^{(0)} g_u(r,t) + iw_{u}^{(1)} \frac{\partial g_u(r,t)}{\partial r} = 0,
\]

(3.103)

To obtain the simplest expressions for the coefficients in Eq. (3.103) we additionally assume that \( |Z_{u}^u| / |Z_{u}^u|^2 \) is negligible\(^4\). This assumption is reasonable for obtaining the first-order dynamics of systems in which dispersion and absorption are not too large, including the first-order dynamics treated in the example calculation of Secs. 4.6 and 4.7. Under this assumption we take a linear expansion of \( Z_1 u_k \) in \( \Omega \) around \( \Omega \), and it is shown in detail in Appendix E that the expressions for the \( w_{u}^{(j)} \) for \( j = 1, 2 \) are then simply given by

\[
w_{u}^{(0)} \equiv i \text{Im} \Omega_{u}^{(1)}.
\]

(3.104)

\(^4\)It can be seen by writing the resultant Eq. (3.104) for \( w_{u}^{(0)} \) in terms of \( Z_{u}^u \) and substituting it into the relation \( |Z_{u}^u| \ll |Z_{u}^u|^2 \) that this assumption is equivalent to the assumption that at a fixed wave vector \( \overline{k} \), \( Z_{u}^u \) does not vary significantly over a frequency range of width \( |w_{u}^{(0)}| \), the temporal decay rate.
Chapter 3. Propagation

\[ w_{uK}^{(1)} \equiv \left[ \frac{\partial \Omega_{uK}^{(1)}}{\partial k} \right]_k, \tag{3.105} \]

where

\[ \Omega_{uK}^{(1)} = \Omega_{uK} - i\pi \left| \Omega'_{uK} \right|, \tag{3.106} \]

and where \( \Omega'_{uK} \equiv \left[ \frac{\partial}{\partial Z} \Omega_{uK} (Z) \right]_{Z=0} \). Recall from the definition of \( \Omega_{uK} (Z) \) before Eq. (3.19) and the definition of \( Z'_{uK} \) in Eq. (3.97) that \( \left[ \frac{\partial}{\partial Z} \Omega_{uK} (Z) \right]_{Z=0} = (Z'_{uK})^{-1} \), so Eq. (3.106) gives the same result for \( \Omega_{uK}^{(1)} \) here as is obtained in Eq. (3.101) for the second order dynamics under the binomial approximation.

The first order propagation parameters of Eqs. (3.104, 3.105) are functions of only \( \Omega_{uK} \) and \( \Omega'_{uK} \), and determining these either involves calculation of \( \Omega_{uK} (Z) \) for small \( Z \) using Eqs. (3.19, 3.20), or involves perturbation theory as discussed in Appendix G.

### 3.5 Propagation parameters of a periodic multilayer stack

In this section we show how to calculate the propagation parameters \( w_{uK}^{(0)}, w_{uK}^{(1)}, \) and \( w_{uK}^{(2)} \) of Eqs. (3.92, 3.93, 3.94) for a periodic multilayer stack. We consider the system to have interfaces normal to the \( z \)-axis, and consider propagation normal to the interfaces. Accordingly we write for the wave vector \( k = k \hat{e}_z \), where \( \hat{e}_z \) is the unit vector associated with the \( z \)-axis. In the absence of absorption, the periodic multilayer is known to have a simple analytic expression for the band structure [105]. In absorptive media, this would normally require a numerical solution because it involves either a complex wave vector or frequency. But here since we have a real frequency and a real wave vector, a simple analytic solution is obtained. Although the result applies for any \( \varepsilon \), anticipating the use of a negative \( \varepsilon \) for material 2 in Section 3.6, for propagation normal to the interfaces we write it in the form

\[ k (\omega, Z) a = \arccos \left[ \cos (k_1 d_1) \cosh (\kappa_2 d_2) - \frac{1}{2} \left( \frac{k_1}{\kappa_2} - \frac{\kappa_2}{k_1} \right) \sin (k_1 d_1) \sinh (\kappa_2 d_2) \right], \tag{3.107} \]

\[ k_1 (\omega, Z) = \frac{\omega}{c} \sqrt{\varepsilon_1 (\omega, Z) / \varepsilon_0}, \tag{3.108} \]

\[ \kappa_2 (\omega, Z) = \frac{\omega}{c} \sqrt{-\varepsilon_2 (\omega, Z) / \varepsilon_0}, \tag{3.109} \]
where each layer of medium \( j \) is of thickness \( d_j \), where the unit cell is of size \( a \equiv d_1 + d_2 \), where \( k (\omega, Z) \) is the Bloch wave vector, \( \pm k_1 (\omega, Z) \) is the local wave vector inside medium 1, \( \pm i k_2 (\omega, Z) \) is the local wave vector inside medium 2, and recall the effective dielectric function \( \varepsilon_j (\omega, Z) \) is defined after Eq. (3.19).

In addition to the dispersion relation of Eq. (3.107) the \( Z = 0 \) mode fields are also of interest. To obtain these, we start with the plane wave (possibly with imaginary wave vector) solutions to Eq. (3.19) in regions of bulk material 1 and of bulk material 2, then solve a linear system which expresses the boundary conditions at the interfaces and the Bloch condition \( E_{\Omega u_k} (z + a) = e^{i k (\Omega, Z = 0) a} E_{\Omega u_k} (z) \) (note \( z \) is a spatial coordinate and \( Z \) is the resonant oscillator amplitude described before Eq. (3.19) for Bloch wave vector \( k (\omega, Z) \) obtained from Eq. (3.107)).

Here Eq. (3.107) can be differentiated analytically with respect to \( \omega \) and \( Z \) in order to calculate propagation parameters. For structures in which an analytic form for the dispersion relation is not available, high-order finite differences can be used [107] instead to calculate propagation parameters for the Schrödinger equation (3.91) according to the prescription of Eqs. (3.95, 3.99, 3.100).

It is also possible to use perturbation theory to calculate the frequency derivatives \( Z'_{uk} \) and \( Z''_{uk} \) defined in Eqs. (3.97, 3.98), and to use these values to obtain the propagation parameters. In Appendix G \( Z'_{uk} \) is explicitly derived in terms of the band center mode fields and band center polariton dispersion relation, and an approach to obtain \( Z''_{uk} \) in terms of these same quantities is outlined. Using the perturbation result, we obtain

\[
\omega^{(0)}_{u_k} = - \frac{i \pi \Omega}{2} \frac{\int_{cell} d^3 r \left( \frac{\partial}{\partial r} d^3 r_{\Omega u_k}(r) \cdot \left[ \text{Im} \Gamma (r, \Omega) \right] \cdot \frac{\partial d^3 r_{\Omega u_k}(r)}{\partial r} \right) \int_{cell} d^3 r \left( \frac{\partial}{\partial r} d^3 r_{\Omega u_k}(r) \cdot \left[ \text{Re} T (r, \Omega) \right] \cdot \frac{\partial d^3 r_{\Omega u_k}(r)}{\partial r} \right)}{\int_{cell} d^3 r \left( \frac{\partial}{\partial r} d^3 r_{\Omega u_k}(r) \cdot \left[ \text{Im} \Gamma (r, \Omega) \right] \cdot \frac{\partial d^3 r_{\Omega u_k}(r)}{\partial r} \right) \int_{cell} d^3 r \left( \frac{\partial}{\partial r} d^3 r_{\Omega u_k}(r) \cdot \left[ \text{Re} T (r, \Omega) \right] \cdot \frac{\partial d^3 r_{\Omega u_k}(r)}{\partial r} \right)},
\]

which is applicable to periodic systems of arbitrary dimensionality, as long as the integration domains are taken to be the appropriate unit cell in 3D, 2D or 1D. Compared with Eq. (3.110) for \( \omega^{(0)}_{u_k} \), the expressions for \( \omega^{(1)}_{u_k} \) and \( \omega^{(2)}_{u_k} \) are more complicated, and we will typically find it easier to calculate these via direct differentiation of the polariton dispersion relation \( k (\omega, Z) \) given in Eq. (3.107).

Transverse diffraction of a pulse with finite spot size could also be calculated using the methods outlined in this section. Although we discuss propagation normal to the

---

5When materials with negative dielectric constants are involved, this method involves ill-conditioned matrices, so it must be used carefully and is not suitable for all such systems and frequencies of interest; scattering matrix methods have been used in the literature to circumvent the ill-conditioning problem [106].
interfaces here, note also that off-axis propagation can be treated easily as well in the same way, making use of the analytic expression for the dispersion relation but including transverse components of the wave vector.

### 3.6 Example calculation: periodic metallo-dielectric stack with realistic response

Here we apply the results of Section 3.5 to a system of current interest [108][109]: a periodic 1D metallo-dielectric multilayer structure. Such structures have been analyzed in the literature several times. For example, band structure calculations have been done for a 1D metallo-dielectric stack using a Drude model for the metal [15], and transmission calculations have been done using empirical optical constants [110]. In the last few years, the 1D metallo-dielectric stack has been proposed as a subwavelength imaging lens [12] and as an artificial nonlinear material [13][109]. However, these works involved absorption for CW fields at a single frequency only [12][13], or did transmission calculations using a Drude model [109]. Clearly it would be useful to be able to understand pulse propagation, and to do it based on the full empirical response function of the metal.

The specific system we consider here is shown in the inset of Fig. 3.2: It is an Au/MgF$_2$ stack of unit cell thickness $a = 193$ nm. We neglect fabrication issues and assume an MgF$_2$ layer thickness $d_1 = 177$ nm and an Au layer thickness $d_2 = 16$ nm; the interfaces are taken to be normal to the $z$-axis. To characterize this system we make use of the polariton dispersion relation given in Eq. (3.107). Recognizing that the effective dielectric function $\varepsilon_2(\omega, Z)$ of the metal is real and negative over the $\omega$ and $Z$ regime of interest for propagation we have that $k(\omega, Z)$, $k_1(\omega, Z)$, and $\kappa_2(\omega, Z)$ are all real, and that $\kappa_2(\omega, Z)$ is a real spatial decay/growth constant of the field inside the metal. Although the differentiation of $k(\omega, Z)$ to obtain the propagation parameters can be done analytically for this structure using Eq. (3.107), we instead use high-order finite differences [107] to demonstrate the feasibility of this more broadly applicable method.

Shown in Fig. 3.2 is the dielectric function of Au. We use a fit to empirical data that satisfies the Kramers-Kronig relations [4]. Also shown in Fig. 2 for comparison is the Drude model dielectric function for Au (labeled $\varepsilon_d$), which is given by

$$\varepsilon_d(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma_d)},$$  \hspace{1cm} (3.111)
with plasma frequency $\omega_p = 9.03 \text{ eV}/\hbar$ and relaxation rate $\gamma_d = 0.053 \text{ eV}/\hbar$ [4]. MgF$_2$ is much less dispersive and we take it to have refractive index $n = 1.38$, so that $k_1(\omega, Z) = \frac{\omega}{c} \sqrt{\varepsilon_1}$.

In Fig. 3.3 is shown $\bar{\omega}_{zk}$, which is the $Z = 0$ band structure (dispersion relation) defined in Eq. (3.21). This is shown for the lowest two bands for both the empirical response function (blue) and the Drude response (dashed; violet), from $ka/2\pi = 0$ (the $\Gamma$-point) to $ka/2\pi = 0.5$ (the $X$-point). Also shown inset are the mode fields $|E_{\text{Emp}}(r)|^2$ corresponding to the empirical response function.

Only in the lowest band of the dispersion relation is the structure sufficiently transparent to be of interest for propagation. This is consistent with results of transmission calculations for an Ag/MgF$_2$ structure [110] with comparable parameters. So from here on we restrict our attention to the lowest frequency band.

Observe that there is a low-frequency cutoff for propagation. The band spans a range of frequencies above the cutoff, and throughout this range the mode frequencies are sufficiently high that interband transitions in the metal are in play. Since the Drude model does not include interband transitions, it might therefore be expected to be an
Figure 3.3: Band center band structure $\overline{\omega}_{ak}$, for both the empirical response function (blue) and the Drude response (dashed; violet). Overlaid are mode fields displayed as $|E_{\pi ak}(r)|^2$. (System shown in the inset of Fig. 3.2)

inadequate model for use at any frequency of interest in this structure.

Plotted in Fig. 3.4 is the mode field $|E_{\Omega ak}(r)|^2$ for three k-points in the first band. Shown are $ka/2\pi = \{0.09, 0.39, 0.47\}$. To allow direct comparison of the spatial concentrations of these mode fields all on a single plot, we normalize them in Fig. 3.4 so that $|E_{\Omega ak}(r)|^2$ has unit integral over a 1D unit cell. Note that the mode field changes slowly in $k$ over the interval $ka/2\pi \in [0.09, 0.39]$, but then as $ka/2\pi$ approaches the band edge over the interval $ka/2\pi \in [0.39, 0.47]$, the mode fields are expelled from the metal.

Next we consider the propagation parameters defined in Eq. (3.104, 3.105), to compare the empirical response (upper curve) and the Drude response (lower curve), and show in Fig. 3.5 that the Drude model is inadequate to determine even the first order dynamics. In Fig. 3.5(a) is plotted the decay rate $-\text{Im} w_{uk}^{(0)}$ in units of $2\pi c/a$ for the empirical response (upper curve) and the Drude response (lower curve). In Fig. 3.5(b) is the group velocity $\text{Re} \left(w_{ak}^{(1)}\right)$ in units of $c$ for the empirical response (upper curve) and the Drude response (lower curve).

We next turn to the propagation parameters $w_{uk}^{(n)}$ of Eqs. (3.92, 3.93, 3.94) for the full Schrödinger equation (3.91) at second order. Although we could use the binomial approximation of Eqs. (3.101, 3.102) here we do not; the results for $\Omega_{uk}^{(1)}$ and $\Omega_{uk}^{(2)}$ are practically identical both ways. Shown in Fig. 3.6 is $Z(\Pi_{ak} + \Delta \Omega)_{uk}$ vs. $\Delta \Omega$ for several k-points.
Figure 3.4: Normalized mode field $|E_{\text{THz}}(r)|^2$ for three k-points in the first band. Shown are $ka/2\pi = \{0.09, 0.39, 0.47\}$.

Figure 3.5: (a) Decay rate $-\text{Im} w^{(0)}$ in units of $2\pi c/a$ for the empirical response (upper curve) and the Drude response (lower curve); (b) Group velocity $\text{Re} \left( w^{(1)} \right)$ in units of $c$ for the empirical response (upper curve) and the Drude response (lower curve).

Figure 3.6: $Z_{\text{(THz+D)}\text{Thz}}$ vs. $\Delta \Omega$ for several k-points in the lowest band. From closest to furthest from the orange dot superimposed on the figure are plots for $ka/2\pi = 0.05, 0.38, 0.43, 0.46$. 
in the lowest band. From closest to furthest from the orange dot mark superimposed in the figure are plots for $ka/2\pi = 0.05, 0.38, 0.43, 0.46$. All curves vanish at $\Delta \Omega = 0$, which reflects the definition of $\Omega$. It is clear that at low $k$, there is little curvature in $Z_{(\Omega_{uk}+\Delta\Omega)}$ with respect to $\Delta \Omega$. Closer to the photonic band edge at the $X$-point, for $ka/2\pi > 0.38$ the curvature increases, along with the slope.

In Fig. 3.7 is $w_{1k}^{(0)}$. Note that the left and right scales differ, and that both the real and imaginary parts are negative. The quantity $\text{Re} w_{1k}^{(0)}$ (left scale) is a small carrier frequency shift term that is of little interest here. The quantity $-\text{Im} w_{1k}^{(0)}$ (right scale) is the carrier wave decay rate, and is very similar to the result obtained at first order in $\Omega$ for $Z_{\Omega_{uk}}$ shown in Fig. 3.5(a). The decay rate reduces slowly in $k$ from the $\Gamma$-point up to about half way to the $X$-point. Then as the absorptivity of the metal increases in frequency due to interband transitions, the decay rate increases as well. As $k$ gets close to the $X$-point, however, the mode field is largely expelled from the metal and becomes spatially concentrated in the dielectric region, as is shown in Fig. 3.4. As a result the decay rate decreases significantly toward the $X$-point.

In Fig. 3.8 we plot $w_{1k}^{(1)}$. The real part of $w_{1k}^{(1)}$ (left scale) is the group velocity of the pulse. It is low near the $\Gamma$-point and $X$-point band edges due to Bragg scattering, and higher away from them as would be expected. The imaginary part of $w_{1k}^{(1)}$ (right scale) is a decay term that depends on the pulse width, and can be significant for sufficiently short pulses near the $X$-point.

Finally in Fig. 3.9 is shown the real part of the $zz$-component of the tensor $w_{1k}^{(2)}$. This component governs dispersion dynamics in the Schrödinger equation (3.91).
imaginary part is small and is not shown. This dispersion coefficient is monotonic here, and is positive near the Γ-point and negative near the X-point.

As mentioned in Sec. 3.5, these calculations could be straightforwardly extended to include off-axis propagation or transverse diffraction.

3.7 Beer’s law for dispersive structured media

3.7.1 Formula

We next turn to the Beer coefficient, which characterizes the exponential decay in intensity of a beam propagating through a dispersive but not too strongly absorptive structured medium. This coefficient is of current interest in the literature [111]. Here we present a result which incorporates the full effect of material dispersion consistent with the Kramers-Kronig relations, and which is applicable to 3D periodic media as well to structures of lower dimensionality. The result is derived from Eq. (3.103). We consider a single-band, smooth classical beam of finite energy that rises but then falls such that \( \mathcal{G}_u(\mathbf{r}, t) \) decays exponentially in the positive \( s \)-direction. In this exponential tail the \( \mathcal{G}_u(\mathbf{r}, t) \) field is time-independent for some finite period of time, and we are interested in that part of the pulse while it is time-independent. The pulse in the region of interest is then of the form

\[
\mathcal{G}_u(s, t) \propto e^{-\alpha s/2},
\]

where \( \alpha \) is the Beer coefficient, which is to be determined. The factor of 2 is introduced in the exponential so that \( \alpha \) will be the spatial decay coefficient of the pulse intensity,
which is proportional to $|\overline{g}_u(s,t)|^2$ when $\overline{g}_u(s,t)$ varies sufficiently slowly in space, which is the situation considered here. Alternatively, we could have considered a long single-carrier-mode pulse of, say, Gaussian shape, and associated a spatial decay coefficient with the carrier wave. That approach yields the same coefficient $\alpha$ that is obtained in what follows.

We substitute Eq. (3.112) into Eq. (3.103), and neglect the imaginary part of $w^{(1)}_{u\mathbf{k}}$. Solving the first order ordinary differential equation yields

$$\alpha = \frac{2 |\text{Im} w^{(0)}_{u\mathbf{k}}|}{(\text{Re} w^{(1)}_{u\mathbf{k}}) \cdot \hat{e}_s}, \quad (3.113)$$

that is, that $\alpha$ is proportional to the ratio of carrier mode temporal decay rate to the group velocity in the direction of interest. Making use of Eqs. (3.104, 3.105, 3.106), using $\Omega'_{u\mathbf{k}} = (Z'_{u\mathbf{k}})^{-1}$ it is possible to write $\alpha$ in terms of the band center dispersion relation and resonant oscillator amplitude as $\alpha = 2\pi/([Z'_{u\mathbf{k}} [\nabla_k \Omega_{u\mathbf{k}}]_k])$. Evaluating this using the perturbation theory expression (G.18) for $Z'_{u\mathbf{k}}$ obtained in Appendix G yields

$$\alpha = \frac{\Omega}{[\nabla_k \Omega_{u\mathbf{k}}]_k} \left| \frac{\int_{\text{cell}} d\mathbf{r} d^2 \mathbf{d}_{\Omega_{u\mathbf{k}}}^e(\mathbf{r}) \cdot [\text{Im} \Gamma(\mathbf{r}, \Omega)] \cdot \mathbf{d}_{\Omega_{u\mathbf{k}}}^e(\mathbf{r})}{\int_{\text{cell}} d\mathbf{r} d^2 \mathbf{d}_{\Omega_{u\mathbf{k}}}^e(\mathbf{r}) \cdot [\text{Re} T(\mathbf{r}, \Omega)] \cdot \mathbf{d}_{\Omega_{u\mathbf{k}}}^e(\mathbf{r})} \right|, \quad (3.114)$$

where $\hat{e}_s$ is the unit vector in the $s$-direction. Besides 3D periodic media, Eq. (3.114) is valid for media with 2D or 1D periodicity, provided the domains of integration are replaced by the appropriate unit cell in 2D or 1D. Note that although the factor $T(\mathbf{r}, \Omega)$ is defined in Eq. (3.15) in the context of fields in the transparent regime, here it appears in Eq. (3.114) characterizing fields in the absorptive regime. For an absorptive medium, define the effective local phase and group velocities $\tilde{v}_p(\mathbf{r}, \Omega)$ and $\tilde{v}_g(\mathbf{r}, \Omega)$ as the respective local velocities of the modified system that would be obtained from Eqs. (3.17, 3.18) were $\text{Im} \Gamma$ neglected, i.e., the local velocities of the nonabsorptive system with response $\text{Re} \Gamma(\mathbf{r}, \Omega)$. Then we have

$$\text{Re} T^{ij}(\mathbf{r}, \Omega) = \varepsilon_0 \varepsilon^{-1}(\mathbf{r}, \Omega) \frac{\tilde{v}_p(\mathbf{r}, \Omega)}{\tilde{v}_g(\mathbf{r}, \Omega)} \delta_{ij}, \quad (3.115)$$

where we define $\varepsilon_0 \varepsilon^{-1}(\mathbf{r}, \Omega)$ to be any diagonal component of the tensor$^6$ $1 - \text{Re} \Gamma(\mathbf{r}, \Omega)$. Using Eqs. (3.114, 3.115) we can understand the Beer coefficient $\alpha$ in terms of the Bloch

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$^6$Recall from before Eq. (3.2) that we are considering materials with isotropic response here, so that all diagonal components of the tensor $1 - \text{Re} \Gamma(\mathbf{r}, \Omega)$ are the same.
modes of the system with response $\text{Re} \Gamma(r, \bar{\Omega})$. The constant $\alpha$ is proportional to the amount the mode fields overlap with the material absorption in the form of $\text{Im} \Gamma(r, \bar{\Omega})$, and is inversely proportional to the mode field sampling of the inverse effective local group velocity \( \tilde{v}_g^{-1}(r, \bar{\Omega}) \) considered in units of the inverse effective local phase velocity \( \tilde{v}_p^{-1}(r, \bar{\Omega}) \). It is also inversely proportional to the mode group velocity \( \nabla_k \Omega_{ak} \) parallel to the direction of interest.

My result for the Beer coefficient agrees with the standard absorptive uniform medium result $\alpha = 2 \text{Im} [n(\bar{\Omega})] \bar{\Omega}/c$, where $n$ is the index of refraction. This work can also be compared with derivations in the literature that describe other simple limiting cases. Recently, an expression for $\alpha$ was derived for absorptive structured media in the absence of material dispersion [111]. The no-dispersion limit of my Eq. (3.114) is inconsistent with that result. However, besides being nominally applicable only for dispersionless absorptive media, that result does not reduce to the standard absorptive uniform medium expression, so it cannot be correct.

Eq. (3.114) can also be compared to a less simple limiting case that arises in past work [2], in which a dynamical equation is given for a pulse propagating in a photonic crystal made of nondispersive material with weak absorption. In brief, if the material dispersion is completely neglected in Eq. (3.114), then that result agrees exactly with a coefficient determined earlier [2], with the exception being that the earlier result lacks the absolute value bars that appear in Eq. (3.114). So as long as $\text{Re} \varepsilon > 0$ everywhere—which is a restriction under which the earlier result was obtained—the results agree. Note that in absorptive media for which $\text{Re} \varepsilon < 0$, the expression for $\alpha$ in Eq. (3.114) has the correct sign while the earlier result would not: With $\text{Re} \varepsilon < 0$ the denominator inside the absolute value bars of Eq. (3.114) can be negative while the rest of the expression for $\alpha$ is positive for propagation in the positive direction, so the absolute value bars that appear in Eq. (3.114) are necessary.

### 3.7.2 Example: Beer coefficient of Au/MgF\(_2\) stack

We next apply the results of this section to the multilayer Au/MgF\(_2\) stack considered in Section 3.6. In Fig. 3.10 is plotted the Beer coefficient $\alpha$ defined in Eq. (3.112) and given in Eqs. (3.114, 3.113). As a check we calculated $\alpha$ using each of these two equations and obtained identical results. As seen in Eq. (3.113), $\alpha$ is proportional to the carrier wave temporal decay rate and inversely proportional to the group velocity, so the curve shown
Figure 3.10: Beer coefficient $\alpha$ in units of $a^{-1}$, as a function of wave vector $ka/2\pi$. Inset shows the same function zoomed in near $ka/2\pi = 0.5$. (System shown in the inset of Fig. 3.2)

in Fig. 3.10 arises from the interplay of the $k$-dependence features of $\text{Im} w_{1k}^{(0)}$ and $\text{Re} w_{1k}^{(1)}$ described in the Section 3.6. The dominant contributions to $\alpha$ are from its proportionality to mode field sampling of the absorptive (metal) material and its inverse proportionality to the group velocity. As $k$ increases from the $\Gamma$-point to the $X$-point, the $E$ field is expelled monotonically from the metal, and on its own this has the effect of reducing $\alpha$. However, the group velocity slows to zero at both photonic band edges, and this has an upward effect on the size of $\alpha$. It is interesting that as $k$ approaches the $X$-point that the downward effect of field expulsion on $\alpha$ is largely counterbalanced by the upward effect of group velocity reduction, so $\alpha$ is flat even quite close to the $X$-point. But while the mode field is never fully expelled from the metal, the group velocity actually does reach zero at the band edge, so very close to the $X$-point the group velocity effect wins out and $\alpha$ diverges. This is shown in the inset to Fig. 3.10. Of course, very near the band edges the scaling assumptions made after Eq. (3.86) do not hold so this behavior is outside the scope of this thesis.

### 3.8 Discussion

In summary, in this chapter I have presented a Hamiltonian-based formulation of electromagnetic propagation in structured media with arbitrary dispersive and absorptive linear response consistent with the Kramers-Kronig relations.
Like the underlying formalism of Ch. 2, the approach here emphasizes the photonic component of the polariton modes of the system. In both the transparent and absorptive frequency regimes, I have formulated the dynamics in terms of a Schrödinger equation satisfied by an effective field. The coefficients in that dynamical equation capture the various physical effects in the problem: modal and material dispersion, absorption, and diffraction.

In the transparent regime, I have shown that the field operator $D(r,t)$ can be written simply in terms of the effective field and the photonic component of the polariton mode field at the carrier wavelength.

In the absorptive regime, the fluctuating and non-fluctuating pieces of the field separate. The fluctuating part is written as a mode expansion over all frequencies and wave vectors involving the vacuum polariton operators and photonic components of the polariton mode fields. The dynamics of the non-fluctuating part are prescribed by a Schrödinger equation for an effective field, and the field operator $D(r,t)$ can be recovered from the effective field and its spatial derivatives combined with the polariton mode fields and their derivatives with respect to frequency and wave vector.

I have also shown that only a subset of the states in the Hilbert space are of physical interest and characterize propagating and decaying electromagnetic fields such as would be present, for example, in an experiment in which a laser pulse is incident on a quiescent absorptive and dispersive material sample. These states are the dispersive/absorptive analog of photon coherent states, and I formally treat their generation via a classical source polarization, but then hide these details from the user by allowing specification of the initial effective field $\psi_u(r, t=0)$, so the user can naturally consider pulses with this formalism without having to deal with the details of state generation.

As a numerical example, I calculated propagation parameters (loss, group velocity, dispersion/diffraction etc.) for a pulse in a 1D metallodielectric periodic structure, using the full dispersive and absorptive $\varepsilon(r, \omega)$ of the metal. Since both the frequency and wave vector are real, the polaritonic band structure for the metallodielectric can be obtained analytically in explicit closed form, despite the presence of absorption. The propagation parameters are obtained from derivatives of the band structure, including derivatives involving the resonant oscillator amplitude factor $Z$. My results give a view of the interplay of Bragg scattering, electronic interband absorption, and field expulsion in shaping pulse propagation. In calculating the propagation parameters I also showed that the Drude model is inadequate for even determining the first order dynamics of a pulse in that
particular sample. In addition to the propagation parameters calculated here, transverse
diffraction of a 3D pulse and off-axis propagation could be treated straightforwardly.

For other more complicated dispersive and absorptive systems, the propagation pa-
rameters can be calculated straightforwardly at all points in the band structure using
numerical techniques. For example, by using modified cutting surfaces in a method
such as Toader and John’s cutting surface method [94], the loss parameter (and others)
can be extracted from the same generalized dispersion surfaces used for calculations of
dispersive media that neglect absorption, essentially without incurring any additional
computational cost.

As a final example of the theory I derived the Beer coefficient for dispersive and
absorptive structured media.
Chapter 4

Spontaneous parametric

down-conversion in dispersive media

In this chapter I present a fully 3D calculation of spontaneous parametric down-conversion (SPDC) of a pump pulse, based on the Hamiltonian formulation of Ch. 2 that incorporates arbitrary material dispersion consistent with causal response. I rigorously derive an expression for the down-converted state and characterize the scaling behavior of the SPDC energy. Dispersion affects the normalization of the polariton modes of the system, and calculations of the down-conversion efficiency that neglect this can be off by 100% or more for common media regardless of geometry if the pump is near the band edge. Furthermore, I derive a 3D three-wave group velocity walkoff factor; due to the interplay of a topological property with a symmetry property, I show that even when the down-converted polaritons are generated into a narrow forward cone, neglect of the transverse walkoff can lead to an overestimate of the SPDC energy by orders of magnitude. Indeed in a $\beta$-BaB$_2$O$_4$ (BBO) crystal this is seen to occur for every possible pump frequency below the BBO band gap.

4.1 Hamiltonian and modes

In this chapter we consider non-magnetic media that are non-absorptive at frequencies up to and including the frequency components of the pump pulse. The linear response is taken to be spatially uniform but dispersive, subject to consistency with the Kramers-Kronig relations. We include a second-order nonlinear response, and neglect the dispersion in the nonlinear response.
4.1.1 Linear Hamiltonian

The linear theory is the Hamiltonian formulation of the electromagnetic field in dispersive and absorptive structured media presented in Ch. 2. The linear response is again of the form of Eq. (2.6). Unlike Ch. 3, in this chapter we deal with anisotropic media; we note that the response function $\Gamma (\mathbf{r}, \omega)$ for such media is simply written in terms of the familiar dielectric function. For $i$ and $j$ that refer to principal axis components of the linear dielectric tensor $\varepsilon$, we have $\Gamma^{ij} = 0$ for $i \neq j$, and

$$\Gamma^{ii} (\mathbf{r}, \omega) = 1 - \frac{\varepsilon_0}{\varepsilon_{ii} (\mathbf{r}, \omega)}, \quad (4.1)$$

(no summation), where $\varepsilon_{ii} (\mathbf{r}, \omega) = \varepsilon_0 n_i^2 (\mathbf{r}, \omega)$ relates the $ii$ principal-axis component of the dielectric tensor to its associated index of refraction. Here we take the usual Fourier transform convention from Eq. (3.1). The physics is modeled in the usual way by setting the coupling tensor according to the prescription of Eq. (3.3) (q.v. Eq. (2.29)).

Since the process considered in this chapter takes place entirely at frequencies below the cutoff frequency for absorption $\omega_g$ (e.g. the electronic band gap), we leave out the polaritons in the absorptive regime, dealing only with polariton modes of eigenfrequency less than $\omega_g$. The relevant Hamiltonian is then

$$H_L = \sum_u \int d\mathbf{k} \hbar \tilde{\omega}_{uk} c_{uk}^\dagger c_{uk}, \quad (4.2)$$

where $c_{uk}$ is the polariton lowering operator, which is labeled by $(u, \mathbf{k})$. Here $\mathbf{k}$ is a three-dimensional wave vector and $u$ is a label that more generally would indicate a band in a photonic crystal or mode type in a more general structured medium; in this chapter where we consider only uniform media, it simply labels the polarization of the mode. The polariton energy is $\hbar \tilde{\omega}_{uk}$; and in Eq. (4.2) we have neglected the zero-point energy. The polariton operators satisfy canonical commutation relations (q.v. Eq. (2.152))

$$[c_{uk}, c_{uk'}^\dagger] = \delta_{uu'} \delta (\mathbf{k} - \mathbf{k'}), \quad (4.3)$$

with all other commutators between the $c$ operators vanishing.

4.1.2 Nonlinear Hamiltonian

For the sake of simplicity we neglect the dispersion in the second-order nonlinearity and augment the linear Hamiltonian by a nondispersive, second-order nonlinear term. This
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has been done for systems in the absence of linear dispersion [3], and here in the presence of linear dispersion it is done in the same way, so we refer the reader to the literature for a more detailed discussion. The nonlinear Hamiltonian is

\[ H_{NL} = -\frac{1}{3\varepsilon_0} \int d\mathbf{r} \Gamma_{ijl}^2(\mathbf{r}) D^i(\mathbf{r}, t) D^j(\mathbf{r}, t) D^l(\mathbf{r}, t), \] (4.4)

where \( \Gamma_{ijl}^2(\mathbf{r}) \) is the second-order nonlinear response associated with the nonlinear polarization

\[ P_{2}^i(\mathbf{r}, t) = \Gamma_{ijl}^2(\mathbf{r}) D^j(\mathbf{r}, t) D^l(\mathbf{r}, t). \] (4.5)

With both dispersion and magneto-optical effects neglected, \( \Gamma_{ijl}^2(\mathbf{r}) \), like the more familiar \( \chi_{ijl}^2(\mathbf{r}) \), is symmetric under permutation of its tensor indices [112]. Were linear dispersion neglected, the response \( \Gamma_{ijl}^2(\mathbf{r}) \) could be written straightforwardly in terms of \( \chi_{ijl}^2(\mathbf{r}) \) and the linear response. This is done by comparing the power series expansion for \( P_{2}^i(\mathbf{r}, t) \) in terms of \( \chi \) and \( E \) to that in terms of \( \Gamma \) and \( D \), writing them both in terms of powers of \( D \), and equating the various coefficients. Although we do have linear dispersion in this chapter, we follow the same process; here and only here for obtaining the relation between \( \Gamma_{ijl}^2(\mathbf{r}) \) and \( \chi_{ijl}^2(\mathbf{r}) \), the linear response is taken to be frequency-independent. This is just a manifestation of the neglect of dispersion in the nonlinear response. When \( (i, j, l) \) label the principal axes of the linear response \( \Gamma(\mathbf{r}) \), in the MKS system of units the result is

\[ \Gamma_{ijl}^2(\mathbf{r}) = \frac{1}{\varepsilon_0 n_i^2(\mathbf{r}) n_j^2(\mathbf{r}) n_l^2(\mathbf{r})} \chi_{ijl}^2(\mathbf{r}), \] (4.6)

where there is no summation over repeated indices on the right-hand side of Eq. (4.6). In evaluating this equation, we will take the linear indices of refraction at typical pump and down-conversion frequencies. Indeed, comparing a 1D version of the work done in this chapter to a 1D formulation of quantum optics that incorporates dispersive nonlinearity [113], we establish that at least for 1D models the dependence of the two results on the linear refractive indices is the same in the approximation that the linear indices vary little over the range of frequencies in which the bulk of down-conversion occurs. In BBO for example, these linear indices individually vary by no more than a few percent over the range of frequencies in which the bulk of down-conversion occurs.

4.1.3 Modes of linear uniaxial media

In this chapter we focus on linear uniaxial media, which are a common platform for SPDC. The optical properties of these media are anisotropic, and their specification involves an
 optic axis $\hat{q}$. Displacement fields polarized perpendicular to $\hat{q}$ experience the ordinary refractive index $n_o(\omega)$, and those polarized parallel to $\hat{q}$ experience the extraordinary index $n_e(\omega)$. Of particular interest for SPDC are negative uniaxial media, which are those for which $n_e(\omega) < n_o(\omega)$.

The electromagnetic fields can be expanded in terms of the polariton operators and the electromagnetic components of their corresponding mode fields as (q.v. Eq. (2.150))

\[
D(r, t) = \sum_u \int dk \left[ c_{uk} e^{-i\omega_{uk} t} \tilde{D}_{uk}(r) + c_{uk}^{\dagger} e^{i\omega_{uk} t} \tilde{D}_{uk}^*(r) \right], \tag{4.7}
\]

\[
B(r, t) = \sum_u \int dk \left[ c_{uk} e^{-i\omega_{uk} t} \tilde{B}_{uk}(r) + c_{uk}^{\dagger} e^{i\omega_{uk} t} \tilde{B}_{uk}^*(r) \right], \tag{4.8}
\]

where the integral is over all of $k$-space and $u$ labels the different modes corresponding to any given $k$. In Eqs. (4.7, 4.8) we note that $D(r, t)$ and $B(r, t)$ are Heisenberg field operators, in contrast with the $c_{uk}$ which are Schrödinger operators. Indeed, unlike Chs. 2 and 3 in which only Heisenberg operators are used and time dependencies of those operators are often left implicit, here in Ch. 4 we make use of Heisenberg, Schrödinger, and other operators. Accordingly, time dependencies of operators in this chapter are always made explicit.

In the expansion of Eq. (4.8), the polariton mode magnetic field $\tilde{B}_{uk}(r)$ and mode frequency $\tilde{\omega}_{uk}$ are determined from the usual (dispersive) Maxwell master equation (q.v. Eq. (2.110))

\[
\nabla \times \left[ \varepsilon_0 \varepsilon^{-1}(r, \tilde{\omega}_{uk}) \cdot \nabla \times \tilde{B}_{uk}(r) \right] = \frac{\tilde{\omega}_{uk}^2}{c^2} \tilde{B}_{uk}(r), \tag{4.9}
\]

where recall $\varepsilon^{-1}$ is a second rank tensor equal to $(1 - \Gamma)/\varepsilon_0$. The field $\tilde{D}_{uk}(r)$ is subsequently calculated from Eq. (3.12). The mode fields $\tilde{D}_{uk}(r)$ are normalized according to (q.v. Eq. (2.155))

\[
\int d\mathbf{r} \tilde{\mathbf{D}}_{uk}^*(\mathbf{r}) \left[ \varepsilon^{-1}(\mathbf{r}, \tilde{\omega}_{uk}) - \frac{\tilde{\omega}_{uk}}{2} \left[ \frac{\partial}{\partial \omega} \varepsilon^{-1}(\mathbf{r}, \omega) \right] \right] \cdot \tilde{\mathbf{D}}_{uk}(\mathbf{r}) = \frac{\hbar \tilde{\omega}_{uk}}{2} \delta(\mathbf{k} - \mathbf{k}'), \tag{4.10}
\]

where the integral is over all space. We recall that expression (4.10) does not only hold for a weakly dispersive medium, but is exact.

The solution to Eqs. (4.9, 3.12) for linear uniaxial media is well-known in the literature [114]. For any given wave vector $\mathbf{k}$, there are two possible modes $u = \{o, e\}$ termed the ordinary ($o$) and extraordinary ($e$) wave. These two modes have $D$ fields that are mutually orthogonal, and that are also each orthogonal to $\mathbf{k}$. Taking the well-known
classical solution for the mode fields, and applying the quantum normalization condition that arises from our Hamiltonian formulation of the fields, we have

\[ \tilde{D}_{uk}(r) = (2\pi)^{-3/2} \hat{e}_{uk} \sqrt{\frac{\hbar \omega_{uk} c_0}{2}} \zeta_{uk} e^{i k r}, \]

(4.11)

where \( \hat{e}_{uk} \) is the mode polarization and \( \zeta_{uk} \) is the normalization factor that arises from enforcing Eq. (4.10), and is given by

\[ \zeta_{uk} = \left[ \frac{1}{n_u^2(\bar{\omega}_{uk})} + \frac{n'_u(\bar{\omega}_{uk})}{n_u^2(\bar{\omega}_{uk})} \right]^{-1/2}, \]

(4.12)

where

\[ n'_u(\omega) \equiv \frac{dn_u(\omega)}{d\omega}. \]

(4.13)

There exist both \( o \) and \( e \) waves for all directions \( \hat{k} \), and the dispersion relation \( \bar{\omega}_{uk} \) is found by solving

\[ |k_u| = \frac{n_u(\omega, \theta) \omega}{c}. \]

(4.14)

Here \( n_u(\omega, \theta) \) is given by

\[ n_o(\omega; \theta) = n_o(\omega), \]

(4.15)

\[ n_e(\omega; \theta) = (n_o^{-2}(\omega) \cos^2 \theta + n_e^{-2}(\omega) \sin^2 \theta)^{-1/2}, \]

(4.16)

where \( \theta \) is the angle between \( \hat{k} \) and the optic axis \( \hat{q} \), and where \( n_o(\omega) \) and \( n_e(\omega) \) are the ordinary and extraordinary indices of refraction of the nonlinear medium described before Eq. (4.7).

The mode polarizations are

\[ \hat{e}_{ok} = \frac{k \times \hat{q}}{|k \times \hat{q}|}, \]

(4.17)

\[ \hat{e}_{ek} = \frac{k \times (k \times \hat{q})}{|k \times (k \times \hat{q})|}. \]

(4.18)

From Eqs. (4.17) and (4.18) we observe that the ordinary wave (\( o \)-wave) polarization is normal to the \( kq \)-plane, and the extraordinary wave (\( e \)-wave) polarization is in the \( kq \)-plane, perpendicular to \( k \).

### 4.2 Asymptotic states and operator dynamics

Since the process of SPDC is due to a nonlinear interaction spatially confined to a nonlinear crystal, it is appropriate to represent incoming and outgoing fields in terms of the
asymptotic states familiar from scattering theory. We do this following the strategy of Yang et al. [80]. The interaction picture propagator $U (t', t)$ is defined according to

$$U (t', t) = e^{iH_L t'/\hbar} U_S (t', t) e^{-iH_L t/\hbar}, \quad (4.19)$$

where $U_S (t', t)$ is the Schrödinger picture propagator $e^{-i(H(t'-t)/\hbar)}$, where $H \equiv H_L + H_{NL}$, and where we recall that $H$, $H_L$ and $H_{NL}$ are independent of time in the Schrödinger picture. It follows from differentiating Eq. (4.19) that

$$-i\hbar \frac{\partial}{\partial t} U (t', t) = U (t', t) H_{NL} (t), \quad (4.20)$$

where we define the interaction picture Hamiltonian, $H_{NL} (t)$, according to

$$H_{NL} (t) \equiv e^{iH_L t/\hbar} H_{NL} e^{-iH_L t/\hbar}. \quad (4.21)$$

Other interaction picture operators are defined analogously.

Next we take $t_0 < 0$ and $t_1 > 0$ and define asymptotic-in and-out states according to

$$|\psi_{in}\rangle = e^{iH_L t_0/\hbar} |\psi (t_0)\rangle, \quad (4.22)$$

$$|\psi_{out}\rangle = e^{iH_L t_1/\hbar} |\psi (t_1)\rangle. \quad (4.23)$$

Here is the physical significance of these states: If the initial ket $|\psi (t_0)\rangle$ were to evolve to $t = 0$ according to $H_L$, then $|\psi_{in}\rangle$ would be the result; and $|\psi_{out}\rangle$ is the ket that upon evolution from $t = 0$ to $t = t_1$ according to $H_L$ would yield the final ket $|\psi (t_1)\rangle$. Processing the equation for the actual evolution

$$|\psi (t_1)\rangle = U_S (t_1, t_0) |\psi (t_0)\rangle, \quad (4.24)$$

together with Eqs. (4.19, 4.22, 4.23) we have that

$$|\psi_{out}\rangle = U (t_1, t_0) |\psi_{in}\rangle. \quad (4.25)$$

It is clear from Eqs. (4.25, 4.20, 4.22, 4.23) that by working with the interaction Hamiltonian $H_{NL} (t)$ we confine the nonlinear dynamics to the mapping from $|\psi_{in}\rangle$ to $|\psi_{out}\rangle$; evolution from $|\psi (t_0)\rangle$ to $|\psi_{in}\rangle$ is trivially calculated, and once $|\psi_{out}\rangle$ is determined from $|\psi_{in}\rangle$, further evolution from $|\psi_{out}\rangle$ to $|\psi (t_1)\rangle$ is again trivial.

We next treat the dynamics of a class of operators that is encountered in what follows. Letting $|0\rangle$ be the vacuum state, and taking $\mathcal{O}$ to be an arbitrary Schrödinger picture operator, we consider an asymptotic-in state of the form

$$|\psi_{in}\rangle = e^{\mathcal{O}} |0\rangle, \quad (4.26)$$
and consider its evolution according to Eq. (4.25).

It follows from the form of Eq. (4.2) that $H_L |0⟩ = |0⟩$. As an approximation, we similarly set $H_{NL} |0⟩ = |0⟩$. It is seen in Sec. 4.4 that this amounts to neglecting terms associated with processes other than SPDC, and that it in fact trivially holds under an approximation made and discussed there. It follows from $H_{NL} |0⟩ = H_L |0⟩ = |0⟩$ that $U^\dagger (t_1, t_0) |0⟩ = |0⟩$. Using this in Eq. (4.26), substituting the result into Eq. (4.25), and making use of the unitarity of $U$ to bring the propagators up into the exponential, we obtain

$$|ψ_{out}⟩ = e^{\mathcal{O}(t_0)} |0⟩,$$  \hspace{1cm} (4.27)

where

$$\mathcal{O} (t) \equiv U (t_1, t) \mathcal{O} U^\dagger (t_1, t).$$  \hspace{1cm} (4.28)

The operator $\mathcal{O} (t)$ satisfies the “initial” (more properly, the “final”) condition

$$\mathcal{O} (t_1) = \mathcal{O}.$$  \hspace{1cm} (4.29)

To obtain the dynamical equation for $\mathcal{O} (t)$, we differentiate Eq. (4.28), apply Eq. (4.20), and make use of the Hermiticity of $H_{NL} (t)$ and unitarity of $U$, yielding

$$i\hbar \frac{d}{dt} \mathcal{O} (t) = \left[ \mathcal{O} (t), \hat{H}_{NL} (t) \right],$$  \hspace{1cm} (4.30)

where

$$\hat{H}_{NL} (t) \equiv U (t_1, t) H_{NL} (t) U^\dagger (t_1, t).$$  \hspace{1cm} (4.31)

Our goal is then to integrate Heisenberg’s equation (4.30) from $t = t_1$ back to $t = t_0$, subject to the final condition (4.29). This gives $\mathcal{O} (t)$ in terms of the Schrödinger operators, from which $|ψ_{out}⟩$ is obtained by using Eq. (4.27). We carry out this procedure in Sec. 4.4; first it will be necessary to specify the experimental system in detail.

### 4.3 Experimental geometry

The system we consider is depicted in Fig. 4.1, and consists of a pump pulse incident on a negative uniaxial nonlinear crystal. There are two relevant coordinate systems: the lab frame is $\{x, y, z\}$ and the crystal frame is $\{x', y', z'\}$. The nonlinear crystal has optic axis $\hat{q} = \hat{e}_z$ and has faces cleaved normal to $\hat{e}_z$. The crystal is of length $L$, positioned symmetrically about the origin. It has refractive index $n_o (\omega)$ associated with the $x'x''$-
and $y'y'$-principal-axis components of the dielectric tensor $\epsilon$, and index $n_e(\omega)$ associated with the $z'z'$-component. To avoid trivial issues relating to linear surface reflections, we take the nonlinear crystal to be embedded in a material that shares its linear properties; reflection coefficients can easily be included at the end of the calculation.

We consider only type-I phase matching here [115], which is the process in which an $e$-polarized pump polariton generates two $o$-polarized down-converted polaritons. The approach could be applied to type-II phase matching as well.

In Ch. 3 we discussed the formal generation of coherent states. Here we take the pump pulse to be an $e$-polarized Gaussian polariton coherent state, within a very good approximation to be specified below. It is taken to have carrier frequency $\omega_P$ corresponding to carrier wave vector $\mathbf{k}_P = k_P \hat{e}_z$, to have spatial band width $\kappa_z$ in the propagation direction $z$, and to have spatial band width $\kappa_{\perp}$ in the perpendicular directions $x$ and $y$.

We take the following relationship between the length $L$ of the crystal and the main length scales associated with the pump pulse:

$$k_P^{-1} \ll \kappa_z^{-1} \ll L \ll \kappa_{\perp}^{-1}. \quad (4.32)$$

That is, the pump carrier wavelength is much shorter than the pulse length, which in turn is much shorter than the crystal length, as shown in Fig. 4.2. We take the pulse to have large spatial extent in the transverse direction; in early work it was noted that the down-conversion efficiency is independent of the degree of pump beam focusing [116], so at no cost and to keep things simple, $\kappa_{\perp}^{-1}$ is taken to be large in (4.32).
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Figure 4.2: Relationship between key length scales.

An additional length scale is the magnitude of ratio of the group velocity dispersion to the group velocity, all taken in the $z$-direction, for the pump at its carrier wave vector. We write this as $R_D \equiv \left| v_{P}^{-1} \frac{\partial^2 \omega_{ek}/\partial k_z^2}{k_p} \right|$, where we define $v_p$ to be the $z$-component of the group velocity of the pump at its carrier wave vector, $v_p \equiv \left[ \frac{\partial \omega_{ek}/\partial k_z}{k_p} \right]$. Then in addition to (4.32), as a further scaling relation we take

$$R_D \ll \kappa_z^{-2} L^{-1}.$$  

(4.33)

Intuitively, the purpose of (4.33) is to ensure the longitudinal spreading of the pump pulse is not significant over the time taken to traverse the nonlinear crystal; this intuition is made explicit in Sec. 4.4.

The detailed specification of the pump pulse is as follows. We write the pump pulse asymptotic in-state according to

$$|\psi_{in}\rangle = e^{A_{P}^\dagger - A_{P}} |0\rangle,$$  

(4.34)

where

$$A_{P}^\dagger \equiv \alpha_{P} \int d{k} \phi_{ek} e_{ek}^\dagger.$$  

(4.35)

Here $\phi_{ek}$ is normalized according to

$$\int d{k} |\phi_{ek}|^2 = 1,$$  

(4.36)

and we note that $\langle \psi_{in} | \psi_{in} \rangle = 1$. Specifically, $\phi_{ek}$ is taken to be Gaussian:

$$\phi_{ek} \equiv \left( \frac{2}{\pi} \right)^{3/4} \kappa_z^{-1/2} \kappa_{\perp}^{-1} e^{-\left( k_z^2 + k_{\perp}^2 \right)/\kappa_z} e^{-\left( k_z - k_P \right)^2/\kappa_{\perp}^2}.$$  

(4.37)
Using the coherent state property

$$c_{ek} |\psi_{in}\rangle = \alpha_P \phi_{ek} |\psi_{in}\rangle,$$

(4.38)

and making the approximation that for components of the pump we have \(\hbar \tilde{\omega}_{ek} \approx \hbar \omega_P\), we find the expectation value of the linear energy operator \(H_L\) defined in Eq. (4.2) is

$$E_P \equiv \langle \psi_{in} | H_L | \psi_{in}\rangle \approx |\alpha_P|^2 \hbar \omega_P,$$

(4.39)

and we note that clearly \(|\alpha_P|^2\) is the expected number of pump polaritons. To the extent that both \(\tilde{\omega}_{ek}\) is approximately given by \(\omega_P + v_P (k_z - k_P)\) and \(\tilde{D}_{ek}(r)\) is approximately given by \(\int \tilde{D}_{ekP}(r) e^{i(k-k_P) \cdot r}\), we have

$$\langle \psi_{in} | D(r, t) | \psi_{in}\rangle \approx \alpha_P (2\pi)^{3/4} k_z^{1/2} k_{\perp} e^{-i\omega_P t} \tilde{D}_{ekP} e^{i(k-k_P) \cdot r}.$$

(4.40)

Thus the pump pulse expected displacement and electric fields approximately have a Gaussian profile.

Since \(\kappa_z/k_P \ll 1\) and \(\kappa_{\perp}/k_P \ll 1\), it follows that wave vectors of components of the pump are indeed near-parallel to \(\hat{e}_z\). In the calculations that follow we accordingly make the very good approximation that for the polarization of pump components it suffices to take the polarization of the pump mode at the carrier wave vector \(k_P\), which we write as \(\hat{e}_P\); that is,

$$\hat{e}_{ek} \approx \hat{e}_{ekP} \equiv \hat{e}_P = \cos \theta \hat{e}_x + \sin \theta \hat{e}_y.$$

(4.41)

Finally, we note that while the pump carrier wave vector \(k_P\) is parallel to the \(z\)-axis, the group velocity of the pump pulse has a transverse component \(v_{P\perp}\) in any uniaxial crystal due to the birefringence, and \(v_{P\perp}\) is small in any crystal of interest because the birefringence is small. For example, \(|v_{P\perp}/v_P| = 0.07\) in the numerical example considered in Sec. 4.7. The \(z\)-component of the pump group velocity, \(v_P\), is what is important in the physics considered here. It is given by

$$v_P = \frac{c}{n_e (\omega_P; \theta)} \frac{1}{1 + s_{\text{disp}}}.$$

(4.42)

$$s_{\text{disp}} \equiv n_e^2 (\omega_P; \theta) \left[ \cos^2 (\theta) n_o^{-3} (\omega_P) \omega P n_o' (\omega_P) + \sin^2 (\theta) n_e^{-3} (\omega_P) \omega P n_e' (\omega_P) \right],$$

(4.43)

where we recall from Eq. (4.13) that \(n_{o,e}'(\omega) \equiv dn_{o,e}(\omega)/d\omega\). This expression for \(v_P\) is obtained by differentiation of the dispersion relation (4.14) with respect to \(\omega\) at \(\omega_P\), using that \([\partial \tilde{\omega}_{ek}/\partial k_z]_{k_P} = 1/ [\partial k_z/\partial \omega]_{\omega_P}\).
4.4 State of the generated bipolaritons

4.4.1 Schrödinger operator mapping from asymptotic-in to -out state

In this section we turn to the task of integrating Heisenberg’s equation (4.30) to obtain an expression for the asymptotic-out state \(|\psi_{\text{out}}\rangle\) in terms of \(|\psi_{\text{in}}\rangle\) and Schrödinger operators.

The state \(|\psi_{\text{out}}\rangle\) is given according to Eq. (4.27); for the pump defined in Eq. (4.34) we have \(\mathcal{O} = A_{\text{p}}^{\dagger} - A_{\text{p}}\), so

\[
|\psi_{\text{out}}\rangle = e^{\overline{A}_{\text{p}}(t_0) - \overline{A}_{\text{p}}(0)} |0\rangle,
\]

where

\[
\overline{A}_{\text{p}}(t_0) = \alpha_{\text{p}} \int d\mathbf{k} \phi_{\text{ph}} \overline{c}_{\text{ck}}(t_0) .
\]

Here \(\overline{c}_{\text{ck}}(t_0)\) is a polariton operator with the time dependence of Eq. (4.28); it is given a different typeface to distinguish it from the polariton operator of Ch. 3 defined in Eq. (3.41). In Eq. (4.45), the evolution of \(\overline{A}_{\text{p}}(t)\) is due to the nonlinear Hamiltonian. We are interested in type-I spontaneous parametric down-conversion here, so in the nonlinear Hamiltonian \(H_{\text{NL}}\) of Eq. (4.4), after substituting in the mode expansion (4.7) we keep only terms associated with type-I SPDC, namely terms with two \(o\)-wave creation operators and one \(e\)-wave annihilation operator \((c_{o}^{\dagger}c_{o}^{\dagger}c_{e})\), as well as their Hermitian conjugates. Amongst other processes, this neglects type-II phase matched SPDC \((c_{o}^{\dagger}c_{o}^{\dagger}c_{e}c_{e})\). Note that under this approximation, we have \(H_{\text{NL}}|0\rangle = |0\rangle\) since each term in \(H_{\text{NL}}\) contains a lowering operator on the right. Thus the approximation made for \(H_{\text{NL}}\) after Eq. (4.26) is contained entirely in the one made here.

Using Eqs. (4.31, 4.21) \(\hat{H}_{\text{NL}}(t)\) is obtained from \(H_{\text{NL}}\); subsequently use of the mode commutator \([c_{uk}, H_{L}] = \hbar \omega_{uk} c_{uk}\) yields

\[
\hat{H}_{\text{NL}}(t) = -\int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k} S_{ooe}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}; t) \overline{c}_{\text{ok}_1}^{\dagger}(t) \overline{c}_{\text{ok}_2}^{\dagger}(t) \overline{c}_{\text{ck}}(t) + \text{h.c.},
\]

where we define

\[
S_{u_1u_2u}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}; t) \equiv \frac{1}{\varepsilon_0} \int d\mathbf{r} \frac{\hat{D}_{u_1k_1}^{ij}(\mathbf{r})}{\tilde{D}_{u_1k_1}^{ij}(\mathbf{r})} \left( \tilde{D}_{u_2k_2}^{ij}(\mathbf{r}) \right)^* \hat{D}_{uk}^{ij}(\mathbf{r}) e^{-i(\tilde{\omega}_{uk} - \tilde{\omega}_{u_1k_1} - \tilde{\omega}_{u_2k_2}) t}.
\]

From the Heisenberg equations (4.30) we have

\[
i\hbar \frac{d}{dt} \overline{c}_{\text{ck}}(t) = \int d\mathbf{k}_1 d\mathbf{k}_2 S_{ooe}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}; t) \overline{c}_{\text{ok}_1}^{\dagger}(t) \overline{c}_{\text{ok}_2}^{\dagger}(t) ,
\]
and corresponding equations for $\bar{c}^{\dagger}_{ok_1}(t)$ and $\bar{c}^{\dagger}_{ok_2}(t)$. The zeroth order solution of these equations is given by

$$\left(\bar{c}^{\dagger}_{uk}(t)\right)^{(0)} = \bar{c}^{\dagger}_{uk}(t_1) = c^{\dagger}_{uk}, \quad (4.49)$$

where recall the right hand side is the Schrödinger operator. Then to first order the solution for $c^{\dagger}_{ek}(t)$ is given by integration of Eq. (4.48) from $t_1$ to $t_0$, making use of the initial condition (4.29). We obtain

$$\bar{c}^{\dagger}_{ek}(t_0) = c^{\dagger}_{ek} - \frac{1}{i\hbar} \int d\vec{k}_1 d\vec{k}_2 \int_{t_1}^{t_0} dt' S_{ooe}(\vec{k}_1, \vec{k}_2, \vec{k}; t') c^{\dagger}_{ok_1} c^{\dagger}_{ok_2}, \quad (4.50)$$

Using this in Eq. (4.45) and taking $t_0 \to -\infty$ and $t_1 \to \infty$, we have

$$A^{\dagger}_P(-\infty) = A^{\dagger}_P + A^{\dagger}_{oo}, \quad (4.51)$$

where $A_P$ is given by Eq. (4.35) and we define

$$A^{\dagger}_{oo} = \frac{\alpha_P \xi}{\sqrt{2}} \int d\vec{k}_1 d\vec{k}_2 \phi_{oo}(\vec{k}_1, \vec{k}_2) c^{\dagger}_{ok_1} c^{\dagger}_{ok_2}, \quad (4.52)$$

where

$$\phi_{oo}(\vec{k}_1, \vec{k}_2) \equiv -\frac{\sqrt{2}}{i\hbar \xi} \int d\vec{k} \phi_{ek} \int_{-\infty}^{\infty} dt' S_{ooe}(\vec{k}_1, \vec{k}_2, \vec{k}; t'). \quad (4.53)$$

In Eqs. (4.52, 4.53) $\xi$ is a normalization factor, chosen so that

$$\int d\vec{k}_1 d\vec{k}_2 |\phi_{oo}(\vec{k}_1, \vec{k}_2)|^2 = 1. \quad (4.54)$$

Recall from Eq. (4.39) that $|\alpha_P|^2$ is the number of pump polaritons; it is shown in Sec. 4.5 that for a weak pump $|\xi|^2$ is the pair production efficiency of the down-conversion process. Substituting Eqs. (4.34, 4.51) into Eq. (4.44), and using the fact that $A_{oo}$ and $A_P$ commute yields

$$|\psi_{out}\rangle = e^{A^{\dagger}_{oo} - A_{oo}} |\psi_{in}\rangle. \quad (4.55)$$

That the excitation of the pump mode is unchanged in this calculation under evolution from $|\psi_{in}\rangle$ to $|\psi_{out}\rangle$ reflects that the undepleted pump approximation is made in solving Eq. (4.48). In the limit that $|\alpha_P|^2$ is small enough that the probability of a pair being generated is small, the probability of multiple pairs being generated is negligible, and it is appropriate to use the linear expansion $\exp(A_{oo}^{\dagger} - A_{oo}) \simeq 1 + A_{oo}^{\dagger} - A_{oo}$. Making use of the fact that from Eqs. (4.34, 4.35) we have $A_{oo} |\psi_{in}\rangle = |0\rangle$, application of this linear expansion to Eq. (4.55) yields

$$|\psi_{out}\rangle = \left(1 + \frac{\alpha_P \xi}{\sqrt{2}} \int d\vec{k}_1 d\vec{k}_2 \phi_{oo}(\vec{k}_1, \vec{k}_2) c^{\dagger}_{ok_1} c^{\dagger}_{ok_2}\right) |\psi_{in}\rangle + ... \quad (4.56)$$
Chapter 4. Spontaneous parametric down-conversion

Were the polariton creation operators in the second term in the parentheses of Eq. (4.56) replaced with photon creation operators, that term would have the familiar form of a biphoton creation operator, with \( \phi_{oo}(k_1, k_2) \) being the biphoton wave function [83]. Since polariton operators appear here instead, we identify \( \phi_{oo}(k_1, k_2) \) as the “bipolariton wave function.”

4.4.2 Calculation of the bipolariton wave function

We next turn to the calculation of the bipolariton wave function \( \phi_{oo}(k_1, k_2) \) defined in Eq. (4.53). In this subsection we calculate the bipolariton wave function up to its normalization factor \( \xi \), which is calculated in the next subsection. We start by making some useful approximations. For components of the pump, we make use of approximations for the mode frequency, mode field amplitude, and polarization; for the down-converted fields we make no approximations here. Approximations for the down-converted fields are only involved via their use in the calculation of \( \xi \).

Recalling from after Eq. (4.38) that the pump pulse has a much smaller bandwidth than carrier frequency, it follows that we have \( \sqrt{\tilde{\omega}_{ek}/\omega_P} \simeq 1 \) for all \( k \) in the spatial bandwidth of the pump, i.e., for \( k_z \) within \( \kappa_z \) of \( k_P \). Then in expressions for \( \phi_{oo}(k_1, k_2) \), for factors that do not appear in the argument of an exponential we make the approximation \( \sqrt{\tilde{\omega}_{ek}} \simeq \sqrt{\omega_P} \). Similarly, we approximate that \( \zeta_{ek}/\zeta_{ek_P} \simeq 1 \) varies relatively little across the pump bandwidth and for these factors that do not appear in the argument of an exponential we take \( \zeta_{ek} \simeq \zeta_{ek_P} \equiv \zeta_P \).

In the exponentials approximations must be made more conservatively. For the time dependence \( \exp(-i\tilde{\omega}_{ek}t') \) of components of the pump we make use of a first-order Taylor expansion of the pump dispersion relation about the carrier frequency \( \omega_P \). We recall \( v_P = [\partial \tilde{\omega}_{ek}/\partial k_z]_{k_P} \) from Eq. (4.42) is the \( z \)-component of the pump group velocity. Then we have the Taylor expansion

\[
\tilde{\omega}_{ek} \simeq \omega_P + v_P (k_z - k_P).
\] (4.57)

In Eq. (4.57) we neglect the group velocity dispersion term. This is justified as long as during the transit time \( L/v_P \) only a negligible extra phase would be accumulated in the factor \( \exp(-i\tilde{\omega}_{ek}t') \) in Eq. (4.47) were the neglected term restored. This amounts to the condition

\[
\exp \left\{ -i \left[ \frac{\partial^2 \tilde{\omega}_{ek}}{\partial k^2_z} \right]_{k_P} \frac{\kappa_z^2 L}{v_P} \right\} \simeq 1; \quad (4.58)
\]
that (4.58) is satisfied follows immediately from the scaling condition (4.33).

Besides the neglect of the group velocity dispersion, in the Taylor expansion (4.57) we also neglect the terms involving derivatives in the transverse directions $x$ and $y$. For the terms containing first-order derivatives, this is justified as long as

$$\frac{\kappa_L L v_p}{v_p} \ll 1,$$

which holds trivially, and would hold even if $v_p / v_p$ were of unit order, since from Eq. (4.32) we have that $\kappa_L L \ll 1$. So this is a very good approximation for the system we consider here. Neglect of the terms containing second-order derivatives—those corresponding to diffraction—requires that

$$\left| \frac{\partial^2 \omega_{ek}}{\partial k^2_{\perp}} \right|_{kr} \frac{\kappa_L^2 L}{v_p} \ll 1.$$

If the scaling condition (4.33) is satisfied, it follows from comparison of (4.60) with (4.58) that the condition (4.60) will also be satisfied if we have

$$\left| \frac{[\partial^2 \omega_{ek} / \partial k^2_{\perp}]}{[\partial^2 \omega_{ek} / \partial k^2_{\parallel}]} \right|_{kr} \frac{\kappa_L^2 L}{\kappa_z^2} < 1.$$  

Since we have from (4.32) that $\kappa_{\perp} / \kappa_z \ll 1$, it follows that as long as $[\partial^2 \omega_{ek} / \partial k^2_{\perp}]_{kr}$ is not orders of magnitude smaller than $[\partial^2 \omega_{ek} / \partial k^2_{\parallel}]_{kr}$, that (4.60) is ensured by (4.32). This trivially holds for the specific system considered in Sec. 4.7. In systems in which (4.61) does not hold, (4.60) can still be satisfied directly if $[\partial^2 \omega_{ek} / \partial k^2_{\perp}]_{kr}$ is sufficiently well-behaved. Finally, we note that an argument similar to that leading to Eq. (4.61) can be constructed to justify the neglect of terms with mixed second-order derivatives.

This completes the justification of the Taylor expansion Eq. (4.57).

Now into Eq. (4.53) we substitute Eq. (4.11) for the mode fields. We make the approximations of Eqs. (4.41, 4.57) for the pump mode polarization and dispersion relation, as well as the approximations for $\tilde{\omega}_{ek}$ and $\zeta_{ek}$ in the text preceding Eq. (4.57). We also use that $\Gamma_{ij}^j (r) = \Gamma_{ij}^j$ for $z \in [-L/2, L/2]$, and that $\Gamma_{ij}^j (r)$ vanishes elsewhere. The resulting expression for $\phi_{oo} (k_1, k_2)$ involves integrals over $k$, $t'$, and $r$. In succession we perform the integrals over $x$, $y$, $k_x$, $k_y$, and $k_z$, which are all simple. The $x$- and $y$-integrals yield delta functions while the $k_x, y$, $z$-integrals involve Gaussians. At this point, we have

$$\phi_{oo} (k_1, k_2) = \frac{b_{k_1, k_2} \kappa_0^{1/2}}{\sqrt{2\pi \xi \kappa_L}} e^{-(k_{1x}+k_{2x})^2/\kappa_0^2} e^{-(k_{1y}+k_{2y})^2/\kappa_0^2}$$

$$\int_{-L/2}^{L/2} dt' \int_{-L/2}^{L/2} dz e^{i(k_{1z}+k_{2z})z} e^{-i(\omega_p-\omega_{ek_1}-\omega_{ek_2})t'} e^{-\kappa_0^2 (z-v_p t')^2 / 4},$$

(4.62)
where we define

\[ b_{k_1, k_2} \equiv i (2\pi)^{-9/4} \varepsilon_0^{1/2} h^{1/2} \zeta_{o k_1} \zeta_{o k_2} \zeta_P \sqrt{\omega_{o k_1} \omega_{o k_2} \omega_P} \]

\[ \zeta_{o k_1} \zeta_{o k_2} \left[ \Gamma_{ijz}^j \sin \theta + \Gamma_{ijz}^x \cos \theta \right]. \]

Next we perform the \( z \)-integral. To a very good approximation the integral is

\[ \int_{-\frac{L}{2}}^{\frac{L}{2}} dt' e^{i(k P - k_{1z} - k_{2z}) z - \kappa_z^2 (z-v_P t')^2/4} \approx \frac{2\sqrt{\pi}}{\kappa_z} e^{i(k P - k_{1z} - k_{2z}) v_P t'} e^{-(k P - k_{1z} - k_{2z})^2 / \kappa_z^2} \Theta \left( |t'| - \frac{L}{2v_P} \right) \]

where \( \Theta \) is the Heaviside step function. Note that \( \Theta \left( |t'| - L / (2v_P) \right) \) is just a “top hat” function with temporal full-width equal to the transit time of the pulse, \( L / v_P \).

The \( z \)-integral (4.64) can also be done exactly analytically, and the result is a sum of error functions of complex arguments; the difference from Eq. (4.64) is that the sharp edges of the Heaviside top hat function are smoothed out over the temporal width of the pulse, and this reflects that it takes a finite amount of time for the pulse to enter or exit the crystal. But since this entrance time is much less than the transit time (due to \( \kappa_z^{-1} \ll L \) (4.32)), it can be neglected.

It is clear from Eq. (4.64) that the \( z \)-integral has the effect of truncating the limits of the time integral, which is performed next. The relevant integral is

\[ \int_{-\frac{L}{2v_P}}^{\frac{L}{2v_P}} dt' e^{-i\Upsilon(k_1, k_2) v_P t'} = \frac{L}{v_P} \text{sinc} \left( \frac{\Upsilon (k_1, k_2) L}{2} \right), \]

where \( \text{sinc}(x) \equiv \sin(x) / x \) for nonzero \( x \) and \( \text{sinc}(0) \equiv 1 \), and where we define

\[ \Upsilon (k_1, k_2) \equiv \frac{1}{v_P} [\omega_P - \omega_{o k_1} - \omega_{o k_2} - v_P (k_P - k_{1z} - k_{2z})]. \]

Note the factor \( L / v_P \) multiplying the sinc on the right-hand side of Eq. (4.65) is the transit time, and recall \( v_P \) is the \( z \)-component of the pump group velocity.

The result of these efforts is the bipolariton wave function:

\[ \phi_{oo} (k_1, k_2) = \frac{\sqrt{2} b_{k_1, k_2} L}{\xi \kappa_z^{1/2} \kappa_\perp v_P} e^{-(k_{1x} + k_{2x})^2 / \kappa_\perp^2} e^{-(k_{1y} + k_{2y})^2 / \kappa_\perp^2} \]

\[ e^{-(k P - k_{1z} - k_{2z})^2 / \kappa_z^2} \text{sinc} \left( \frac{\Upsilon (k_1, k_2) L}{2} \right). \]

This completes the calculation of \( \phi_{oo} (k_1, k_2) \), except for the determination of its normalization factor \( \xi \), which is done in the following subsection.
The quantity $\Upsilon(k_1, k_2)$ in Eq. (4.66) can be written in an alternate form which is more intuitive and easier to use to compare the wave function (4.67) to 1D results in the literature. Collecting the terms from the first order Taylor expansion (4.57) of the pump into a factor of $\tilde{\omega}_{e(k_1+k_2)}$, we find

$$\Upsilon(k_1, k_2) = \frac{1}{v_p} \left[ \tilde{\omega}_{e(k_1+k_2)} - \tilde{\omega}_{ok_1} - \tilde{\omega}_{ok_2} \right]. \quad (4.68)$$

Had the transverse terms in the Taylor expansion for the pump been retained in Eq. (4.57), the above derivation of $\phi_{oo}(k_1, k_2)$ would have been trivially modified. The same result (4.67) is found, but with an extra term $v_{\perp} \cdot (k_1 + k_2)$ added inside the brackets of Eq. (4.66). Then once again collecting terms from the full first order Taylor expansion of the pump to identify $\tilde{\omega}_{e(k_1+k_2)}$, the expression in Eq. (4.68) is again obtained. Of course, as per the argument made after Eq. (4.57), the inclusion of the transverse part of the Taylor expansion leads to a negligible change in the bipolariton wave function.

We note here that for wave vectors $k_1$ and $k_2$ restricted to the positive $z$-axis, our result in Eq. (4.67) is consistent with recent work in the literature involving 1D propagation [80]. The only subtlety in making this check is that the scaling condition from Eq. (4.32) that the pulse is shorter than the crystal must be applied to the result of that recent work [80] before a direct comparison is made. Consistency with a 1D model calculation [84] of SPDC of a pump pulse is verified in the same way once the effect of dispersion on the normalization of the fields is neglected.

### 4.4.3 Normalization of the bipolariton wave function

Next we turn to the calculation of the bipolariton normalization factor $\xi$ (4.53). To do so we consider the generic integral

$$I \equiv \int d{k_1} d{k_2} h(k_1) |\phi_{oo}(k_1, k_2)|^2, \quad (4.69)$$

where $h(k_1)$ is a weighting function which we take to depend only on $|k_1|$. With the choice $h(k_1) = 1$, setting $I = 1$ gives Eq. (4.54), and therefore determines $\xi$. The reason

\[\text{In greater detail, the procedure is as follows. First we limit wave vectors to the positive z-axis, and take linear expansions of the dispersion relation about an arbitrary triad of phase- and frequency-matched points } \{(k, \tilde{\omega}_{ek}), (k_1, \tilde{\omega}_{ok_1}), (k_2, \tilde{\omega}_{ok_2})\}. \text{ At that level of expansion, we find the argument of the sinc function in Eq. (4.67) is equal to its counterpart in the literature. We then enforce the scaling assumption } \kappa^{-1} \ll L \text{ of Eq. (4.32) by taking the delta function limit of the sinc in both our expression (4.67) and in the expression in the literature. The resulting expressions are the same up to the overall amplitude, which is not expected to match given a 3D model of an inherently 3D process is being compared with a 1D model.}\]
for introducing the generic integral is that the choices \( h(k_1) = 1 \) and \( h(k_1) = \hbar \tilde{\omega}_{ok_1} \) are used to calculate pair production properties in Sec. 4.5.

In order to calculate \( I \) we first make a useful approximation for \( b_{k_1,k_2} \). That factor contains the down-converted mode polarizations, which can be simplified as follows. Writing \( k_j = k_{jz} \hat{e}_z + k_{jp} \hat{k}_j \) for \( j = 1, 2 \), Eq. (4.17) for the polarizations of the down-converted polaritons can be written in the form

\[
\hat{e}_{ok_j} = \frac{k_{jz} \sin \theta}{|\hat{k}_j \times \hat{q}|} \left( \hat{e}_y + \frac{k_{jp}}{k_{jz} \sin \theta} \hat{k}_j \times \hat{q} \right).
\] (4.70)

If the wave vectors \( k_1 \) and \( k_2 \) of a cogenerated pair of down-converted polaritons are close enough to the z-axis that for both \( j = 1, 2 \) we have

\[
\frac{k_{jp}}{k_{jz} \sin \theta} \ll 1,
\] (4.71)

then the expressions for the down-converted mode polarizations greatly simplify. Using (4.71) in Eq. (4.70), it follows that

\[
\hat{e}_{ok_1} \approx \hat{e}_{ok_2} \approx \hat{e}_y.
\] (4.72)

Substitution of Eq. (4.72) in Eq. (4.63) yields the following approximate form for \( b_{k_1,k_2} \):

\[
b_{k_1,k_2} \simeq i (2\pi)^{-9/4} \varepsilon_0^{1/2} \hbar^{1/2} \left[ \Gamma_{y'y'z'} \sin \theta + \Gamma_{y'y'x'} \cos \theta \right] \zeta_{ok_1} \zeta_{ok_2} \zeta_P \sqrt{\tilde{\omega}_{ok_1} \tilde{\omega}_{ok_2} \omega_P}.
\] (4.73)

With the use of Eq. (4.73) in Eq. (4.67), we obtain an approximate expression for \( \phi_{oo}(k_1,k_2) \) that has axial symmetry about the z-axis. Indeed, for very different signal and idler frequencies there exist pairs of perfectly phase- and frequency-matched down-converted fields such that one of the two down-converted mode polarizations is very different from \( \hat{e}_y \). However, we anticipate the result, to be seen in the numerical example of Sec. 4.7, that in our calculation whenever the small-angle approximation (4.71) does not hold, the amplitude of the down-conversion process is sufficiently small that the use of the axial symmetry approximation (4.73) does not significantly affect our results.

Returning to the evaluation of \( I \), we substitute Eq. (4.67) into Eq. (4.69), and use the approximation (4.73). The resulting expression contains six integrals over four peaked functions: two Gaussians of width \( \kappa_\perp \), a sinc of width \( L^{-1} \) and a Gaussian of width \( \kappa_z \). The plan is then to perform the integrals associated with the four peaked functions in an order from narrowest peaked function to least narrow, each time using the filter property

\[
\int dx f(x) g(x) \simeq \left[ \int dx f(x) \right] g(x_0),
\] (4.74)
which holds for \( f (x) \) which is peaked at \( x_0 \), as long as \( g (x) \) is slowly varying with respect to \( f (x) \). Here \( f (x) \) is termed the filter function.

From the scaling assumptions of Eq. (4.32) it follows that the exponentials involving \( \kappa_\perp \) are narrower than the other functions in the integrand of \( I \). So we first perform the \( k_{2x} \) and \( k_{2y} \) integrals. To do so, we take the exponential in \( 2 (k_{1x} + k_{2x})^2 / \kappa_\perp^2 \) to be a filter function and integrate over \( k_{2x} \). This is equivalent to taking the small, positive-\( \kappa_\perp \) delta function limit of the exponential in \( 2 (k_{1x} + k_{2x})^2 / \kappa_\perp^2 \), followed by the integral over \( k_{2x} \). Subsequently, the analogous process is followed to perform the integral over \( k_{2y} \).

Introducing a cylindrical coordinate system with axis \( \hat{e}_z \), we write \( \mathbf{k} = (k_\rho, \phi, k_z) \), and note that for \( o \)-waves, \( \tilde{\omega}_{\omega k} \) is independent of \( \phi \). In the arguments of functions like this in which the angle \( \phi \) is immaterial, in what follows we sometimes use the notation \( (k_\rho, k_z) \) to specify the relevant vector components. For example, here we write

\[
\omega_{\omega (k_\rho, k_z)} \equiv \omega_{\omega (k_\rho, \phi, k_z)}. \tag{4.75}
\]

We also introduce the vector \( \mathbf{K} = (k_{1x}, k_{1z}, k_{2z}) \), and define a single-argument function \( \Upsilon (\mathbf{K}) \) in terms of \( \Upsilon (\mathbf{k}_1, \mathbf{k}_2) \) according to

\[
\Upsilon (\mathbf{K}) \equiv \Upsilon (k_{1x}, \phi, k_{1z}) \Upsilon (k_{2x}, \phi, k_{2z}), \tag{4.76}
\]

with \( \mathbf{k}_1 (\mathbf{K}, \phi) = (k_{1x}, \phi, k_{1z}) \) and \( \mathbf{k}_2 (\mathbf{K}, \phi) = (k_{1x}, -\phi, k_{2z}) \). Like \( \Upsilon (\mathbf{k}_1, \mathbf{k}_2) \) (4.66), \( \Upsilon (\mathbf{K}) \) is \( \phi \)-independent.

Returning to evaluation of the integral \( I \), we convert to cylindrical coordinates and do the angular integral picking up \( 2\pi \). The result is

\[
I = \frac{2\pi^2 L^2}{|\xi|^2 \kappa_{\perp}^2 v_p^2} \int_0^{2\pi} dk_{1\rho} \int dk_{1z} dk_{2z} k_{1\rho} h (k_{1\rho}, k_{1z}) |b_{(k_{1\rho}, k_{1z}), (k_{1\rho}, k_{2z})}|^2 \left( \Upsilon (\mathbf{K}) \frac{L}{2} \right)^2.
\tag{4.77}
\]

In terms of the length scales of Eq. (4.32), the \( \text{sinc}^2 \) function in Eq. (4.77) has width \( \sim L^{-1} \) in \( \mathbf{K} \) in the direction parallel to \( \nabla_{\mathbf{K}} \Upsilon (\mathbf{K}) \). For any given \( \mathbf{K} \), in this direction the Gaussian has a width that is no narrower than its \( z \)-direction width, \( \kappa_z \), which according to Eq. (4.32) is much larger than \( L^{-1} \). So \( \text{sinc}^2 \) is taken to be a filter function.

As long as there is some material dispersion, \( \Upsilon (\mathbf{K}) \) vanishes on a 2D surface \( S_\Upsilon \) in \( \mathbf{K} \)-space. We then perform a change of variables, use the integral \( (L/2\pi) \int_{-\infty}^{\infty} \text{sinc}^2 (K L/2) dK = 1 \), and use the filter function property described in Eq. (4.74), to obtain the useful integral

\[
\int_{-\infty}^{\infty} dk_{2z} \int_{-\infty}^{\infty} dk_{1z} \int_0^{\infty} dk_{1\rho} \text{sinc}^2 (\Upsilon (\mathbf{K}) L/2) G (\mathbf{K}) = \frac{2\pi}{L} \int_{S_\Upsilon} d^2 K \frac{G (\mathbf{K})}{|\nabla_{\mathbf{K}} \Upsilon (\mathbf{K})|}. \tag{4.78}
\]
Here $G(K)$ is any function that varies slowly in $K$ over a distance $\sim L^{-1}$ in the direction normal to $S_\mathcal{T}$. We note here that if material dispersion were neglected in the calculation, $\Upsilon(K)$ would vanish on a 1D line segment instead of the 2D surface $S_\mathcal{T}$, and additional subtleties would arise.

After the integral (4.78) has been applied to $I$, the Gaussian in $(k_P - k_{1z} - k_{2z})/\kappa_z$ is the narrowest remaining function, i.e., compared with the variation of the Gaussian with respect to its arguments, both $h(k_{1\rho}, k_{1z})$ and $b(k_{1\rho}, k_{1z})(k_{1\rho}, k_{2z})$ vary slowly. This is established as follows. We only use $h(k_{1\rho}, k_{1z})$ that is either constant or is equal to $\hbar\omega_{ok_1}$. In the media with normal dispersion that we consider here, for those $k_1$ at which there is significant down-conversion—those with a $z$-component of order $k_P/2$, and with a smaller $\rho$-component than $z$-component—it follows from the scaling assumption $\kappa_z \ll k_P$ of (4.32), that $\hbar\omega_{ok_1}$ varies more slowly than the Gaussian. Given the approximate form (4.73) for $b_{k_1,k_2}$, its variation in $k_1$ and $k_2$ is dominated by the factor $\sqrt{\omega_{ok_1}\omega_{ok_2}}$, so it follows similarly that $b_{k_1,k_2}$ varies more slowly than the Gaussian with respect to its arguments.

Thus the Gaussian is taken to be a filter function. Anticipating the result of integration over this filter, we first evaluate $v_P |\nabla_K \Upsilon(K)|$ at points for which $k_{2z} = k_P - k_{1z}$; we call the result the three-wave group velocity walkoff $V(k_{1\rho}, k_{1z})$,

$$V(k_{1\rho}, k_{1z}) \equiv \left\{ (v_{oz}(k_{1\rho}, k_{1z}) - v_P)^2 + (v_{ox}(k_{1\rho}, k_P - k_{1z}) - v_P)^2 + (v_{op}(k_{1\rho}, k_{1z}) + v_{op}(k_{1\rho}, k_P - k_{1z}))^2 \right\}^{1/2},$$  

(4.79)

where we make use of the $o$-wave group velocity

$$v_o(k_{1\rho}, k_{1z}) = \frac{c}{n_o(\omega_o(k_{1\rho}, k_{1z})) + \omega_o(k_{1\rho}, k_{1z})n_o'(\omega_o(k_{1\rho}, k_{1z}))},$$  

(4.80)

and its respective $\rho$- and $z$-components $v_{op}$ and $v_{oz}$, and where we recall $v_P$ is the $z$-component of the pump group velocity.

The integral over the Gaussian filter, with the use of Eq. (4.79), then yields

$$I = \frac{2^{3/2}\pi^{7/2}L}{|\xi|^2 v_P^2} \int_C d^3K k_{1\rho} h(k_{1\rho}, k_{1z}) |b(k_{1\rho}, k_{1z})(k_{1\rho}, k_P - k_{1z})|^2 \frac{V(k_{1\rho}, k_{1z})}{v_P}.$$  

(4.81)

In Eq. (4.81), the integral is over the 1D curve $C$ in $(k_{1\rho}, k_{1z})$-space, which is a subspace of both $K$-space and $k_1$-space. The curve $C$ is the locus of points of perfect phase and
frequency matching, and we call it the “matching” curve; it is defined implicitly by

\[ \tilde{\omega}(k_1, k_{1z}) + \tilde{\omega}(k_1, k_P - k_{1z}) - \omega_P = 0, \quad (4.82) \]

where recall \( \omega_P = \tilde{\omega}_{ekp} \). Indeed it is seen that for any \((k_1, k_{1z})\) on \(C\), with \(k_{2z} = k_P - k_{1z}\), that the arguments of both the sinc\(^2\) and of the Gaussian in Eq. (4.77) vanish, indicating that this combination of variables is perfectly phase- and frequency-matched.

On the matching curve \(C\) in the system we consider, there is a unique value of \(k_1\) corresponding to any \(k_{1z}\). This can be seen if we make the replacement \(k_1 \rightarrow k_1 + \Delta\) in Eq. (4.82), then use that \(\tilde{\omega}_{ek}\) is monotonic in \(|k|\). Thus \(C\) can be parameterized by \(k_{1z}\). Then \(k_1\) is reinterpreted as an implicit function of \(k_{1z}\) defined by Eq. (4.82). And the integral \(\int_C d^1K\) is converted into an integral \(\int_{P(C)} dk_{1z}\), where \(P(C)\) is the projection of \(C\) onto the \(k_{1z}\) axis, i.e. it is the domain in \(k_{1z}\) over which some \(k_1\) satisfies Eq. (4.82).

This introduces a factor of the arc length \(\sqrt{1 + \left(\frac{dk_1}{dk_{1z}}\right)^2}\). Additionally, we finally specialize to a nonlinear crystal with trigonal symmetry point group \(3m\), of which BBO is an example [117][118]. In such a crystal, \(\Gamma'_{y'y'z'}\) vanishes due to a mirror plane normal to \(\hat{e}_x\). This gives the final result for \(I\),

\[
I = \frac{\hbar \varepsilon_0 \left(\Gamma'_{y'y'z'}\right)^2 \sin^2(\theta) L}{8\pi |\xi|^2 v_P^2} \int_{P(C)} dk_{1z} \sqrt{1 + \left(\frac{dk_1}{dk_{1z}}\right)^2 k_1 \epsilon(k_1, k_{1z}) F_D(k_{1z}, \omega_P) \tilde{\omega}_{ek}(k_1, k_{1z}) \tilde{\omega}_{ek}(k_1, k_P - k_{1z}) \omega_P} V(k_1, k_{1z}) / v_P,
\]

where here and in much of what follows we leave the \(k_{1z}\)-dependence of \(k_1\) implicit, and where we define a factor

\[
F_D(k_{1z}, \omega_P) \equiv \zeta_P \zeta_0^2 \xi_0^2 (k_1, k_{1z}) \xi_0^2 (k_1, k_P - k_{1z}).
\]

The factor \(F_D(k_{1z}, \omega_P)\) involves the refractive indices at the frequencies of the three interacting modes, but it also involves the frequency derivative of the refractive index. It is the contribution to \(I\) from the normalization conditions of the mode fields involved in the interaction.

It follows from the discussion after Eq. (4.69) that \(\xi\) is given by setting \(I = 1\) and
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\( h(k_{1\rho}, k_{1z}) = 1 \) in Eq. (4.83). This yields

\[
|\xi|^2 = \frac{\hbar \xi_0 \left( \Gamma'_y \gamma'_z \right)^2 \sin^2 (\theta) L}{8\pi v^2_p} \int d k_{1z} \sqrt{1 + \left( \frac{d k_{1\rho}}{d k_{1z}} \right)^2 k_{1\rho} F_D (k_{1z}, \omega_P) \tilde{\omega}_o(k_{1\rho}, k_{1z}) \tilde{\omega}_o(k_{1\rho}, k_{1z} - k_{1z}) \omega_P} V (k_{1\rho}, k_{1z}) / v^2_P.
\]

Substitution of Eq. (4.85) into Eq. (4.67) yields a complete specification of the bipolariton wave function \( \phi_{oo} (k_1, k_2) \).

4.5 Pair production properties

Having derived the generated state, we turn to the characterization of the pair production properties of SPDC. In this section and for the remainder of the chapter, we restrict the calculation to the limit in which \( |\alpha_P|^2 \) is small enough that the probability of multiple pairs being created is negligible. In this limit the asymptotic-out state is given by Eq. (4.56), which we reiterate:

\[
|\psi_{out}\rangle = \left( 1 + \frac{\alpha_P \xi}{\sqrt{2}} \right) \int d k_1 d k_2 \phi_{oo} (k_1, k_2) c^\dagger_{o k_1} c^\dagger_{o k_2} |\psi_{in}\rangle + ....
\]

Here we calculate both the expectation of the down-converted polariton number operator and its suitably smoothed distribution over wave vectors, and the expected down-converted field energy. We also show that in this limit \( |\xi|^2 \) is the pair production efficiency.

4.5.1 Number operator expectation and directional distribution

First we characterize the down-converted fields in terms of expectations of both the mode number operator \( N_{o k_1} \equiv c^\dagger_{o k_1} c_{o k_1} \) and the total down-converted polariton number operator \( N_o = \int d k_1 N_{o k_1} \).

To calculate \( \langle N_{o k_1} \rangle \), the expectation of \( N_{o k_1} \) in the asymptotic-out state, we use Eq. (4.56) for \( |\psi_{out}\rangle \). The expression for \( \langle N_{o k_1} \rangle \) is an integral over the expectation in the state \( |\psi_{in}\rangle \) of a product of six polariton ladder operators. To evaluate this expression we use the commutator (4.3), use the polariton exchange symmetry \( \phi_{oo} (k_1, k_2) = \phi_{oo} (k_2, k_1) \), use the normalization \( \langle \psi_{in} | \psi_{in} \rangle = 1 \) of the pump coherent state, and use the fact that
\(|\psi_{in}\rangle\) has no excitation in the o-wave states, i.e. \(c_{ak_1}|\psi_{in}\rangle = 0\). Also required is use of the identity
\[
\delta (k_2'' - k) \delta (k'_2 - k) = \delta (k_2'' - k'_2) \delta \left( k - \frac{1}{2} (k_2'' + k'_2) \right).
\]
(4.86)
The result is
\[
\langle N_{o k_1} \rangle \equiv \langle \psi_{out} | N_{ak_1} | \psi_{out} \rangle = 2 |\xi|^2 |\alpha_P|^2 \int dk_2 |\phi_{oo}(k_1, k_2)|^2.
\]
(4.87)
The total down-converted polariton number expectation \(\langle N_o \rangle\) is simply obtained from this expression for \(\langle N_{o k_1} \rangle\). Integrating Eq. (4.87) with respect to \(k_1\) and using the bipolariton wave function normalization condition (4.54) yields
\[
\langle N_o \rangle = 2 |\xi|^2 |\alpha_P|^2,
\]
(4.88)
which is both proportional to the pump polariton number \(|\alpha_P|^2\), and to the squared modulus of the bipolariton normalization factor \(\xi\), which is given by Eq. (4.85). It is clear from Eq. (4.88) that \(|\xi|^2\) is the expected number of down-converted polariton pairs generated per expected pump polariton. Thus \(|\xi|^2\) is the pair production efficiency of the down-conversion process, here in the low-\(|\alpha_P|^2\) limit in which \(|\psi_{out}\rangle\) can be taken to be given by Eq. (4.56).

For the purposes of analyzing the directional distribution of the expected polariton number, \(\langle N_{o k_1} \rangle\), we identify a “matching surface” \(S\) in \(k_1\)-space, related to the matching curve \(C\): It is the surface of revolution generated by rotating \(C\) in \(k_1\)-space about \(\hat{e}_z\), i.e. \(S\) is the space of wave vectors \(k_1\) such that \((k_{1,\rho}, k_{1,z})\) is on \(C\). Given any \(k_1\) on \(S\) in \(k_1\)-space, it is perfectly phase- and frequency-matched to a \(k_2\) given by the inversion of \(k_1\) through the point \((0, 0, k_P/2)\). Defining \(k_{1,\rho,typ}\) to be a typical radial component of a point on \(S\), we have that \(S\) has typical extents \(k_{1,\rho,typ}\), \(k_{1,\rho,typ}\), and \(k_P\) in the respective \(x\), \(y\), and \(z\) directions.

Now the quantity \(\langle N_{o k_1} \rangle\) given in Eq. (4.87) varies rapidly in \(k_1\) over distances comparable to the scale of \(S\) but has significant amplitude only for \(k_1\) on \(S\) or within \(\sim \kappa_z\) of it. This width is set by the Gaussian factor of \(|\phi_{oo}(k_1, k_2)|^2\) (4.67), which has standard deviation \(\kappa_z/2\). To obtain a coarser picture of the number distribution we introduce a smoothing volume \(\Lambda(k_1)\) as follows. Given a point \(k_1\) on \(S\), specify a small smoothing area \(A(k_1)\) on \(S\) containing \(k_1\). Then \(\Lambda(k_1)\) is the volume of translation generated by the displacement of \(A(k_1)\) along the line normal to \(S\) at \(k_1\), with this done a small distance
in either direction. The size of $\Lambda (k_1)$ is chosen to be a few times $\kappa_z$ in each direction; for example, with a full-width $3\kappa_z$, the Gaussian is contained out to 3 standard deviations from its peak.

We make use of this smoothing volume to introduce $\langle N_{k_1} \rangle$, a coarse-grained number operator expectation per unit area of $S$, which takes as its argument a point $k_1$. It is defined as the integral over a small volume $\Lambda (k_1)$ of $\langle N_{o k_1} \rangle$ per unit $k$-space area of $S$:

$$\langle N_{k_1} \rangle = \frac{1}{|A(k_1)|} \int_{\Lambda (k_1)} d^2 k_1' \langle N_{o k_1'} \rangle. \quad (4.89)$$

The size of the area $|A(k_1)|$ is given in terms of the respective extents $\Lambda_\phi$ and $\Lambda_z$ of $\Lambda (k_1)$ in the angular and $z$ directions,

$$|A(k_1)| = \Lambda_\phi \Lambda_z \sqrt{1 + \left(\frac{dk_1}{dk_1} \right)^2}. \quad (4.90)$$

Since $\Lambda (k_1)$ is wider than the widest filter function, in the evaluation of Eq. (4.89) the succession of filter functions used to obtain $I$ from $\phi_{oo}(k_1, k_2)$ can be used in the same way as was done to obtain Eq. (4.83) from Eq. (4.69). The process is described in detail in Appendix I. Employing this strategy, the expression for $\langle N_{k_1} \rangle$ reduces to

$$\langle N_{k_1} \rangle = \frac{\hbar \omega_P |\alpha_P|^2 \varepsilon_0 \left(1^y 1^y 1^z \right)^2 \sin^2 (\theta) L}{8\pi^2 v_P^2} \frac{F_D (k_{1z}, \omega_P) \tilde{\omega}_o(k_{1z}, k_{1z}) \tilde{\omega}_o(k_{1pz}, k_{1pz} - k_{1z})}{V (k_{1pz}, k_{1z}) / v_P}, \quad (4.91)$$

where we recall from after Eq. (4.82) that $k_{1pz}$ is a function of $k_{1z}$. Note that integrating Eq. (4.91) over $S$ and comparing the result with Eq. (4.88), it is verified that $\langle N_{k_1} \rangle$ satisfies the expected property

$$\int_S d^2 k_1 \langle N_{k_1} \rangle = \langle N_o \rangle. \quad (4.92)$$

At this point we make some observations about the result in Eq. (4.91) for $\langle N_{k_1} \rangle$. It is apparent from this equation that besides frequency matching and phase matching, the down-conversion rate into any given channel is largely determined by the three-field group velocity walkoff $V (k_{1pz}, k_{1z})$ from Eq. (4.79). This group velocity mismatch involves the addition in quadrature of three terms: the first two terms express $z$-component group velocity mismatches with the pump, with one term for each generated polariton; the third is the transverse part of the relative group velocity of the two down-converted polaritons,
or a transverse departure speed. The picture that arises is that down-conversion takes place spontaneously at a point in the crystal, and the amplitude of the process depends on the three-field walkoff $V(k_{1\rho}, k_{1z})$.

Note also the appearance in Eq. (4.91) of the factor $F_D(k_{1z}, \omega_P)$. While it has been long recognized that material dispersion affects phase matching curves, the appearance of $F_D(k_{1z}, \omega_P)$ in Eq. (4.91) indicates that each of the pump and two down-conversion modes contributes a mode field normalization factor, and these factors depend on the material dispersion.

This effect can be understood intuitively as follows. When a polariton state in a dispersive medium has energy $\hbar \omega$, that energy is shared between the medium and electromagnetic fields in a way that depends on both the linear response function and its frequency derivative, and this is seen explicitly in the expression (4.11) for the mode field. Thus the mapping between the energy of a quantum fluctuation or coherent state and its corresponding electromagnetic field amplitude depends on $n'(\omega)$. This survives in the expression for the nonlinear polarization, and in the final equation (4.91) for the down-conversion into a given channel.

### 4.5.2 Energy

Next we wish to calculate the expected (linear) energy in the down-converted fields. Defining $\mathcal{P}_o$ to be the projector onto the $o$-wave part of state space, we have

$$E_{SPDC} \equiv \langle \psi_{\text{out}} | \mathcal{P}_o H_L \mathcal{P}_o | \psi_{\text{out}} \rangle .$$

(4.93)

Using Eqs. (4.56, 4.52) then using $[\mathcal{P}_o, A_{oo}^\dagger] = 0$, it follows that

$$E_{SPDC} = \langle 0 | A_{oo} H_L A_{oo}^\dagger | 0 \rangle .$$

(4.94)

Next we substitute in Eq. (4.2) for $H_L$, and use the explicit form (4.52) for $A_{oo}$. Like the earlier calculation of $\langle N_{ok_1} \rangle$, here the result involves terms with six polariton creation or annihilation operators. Again, its simplification involves use of the commutator (4.3), along with the polariton permutation property $\phi_{oo}(k_1, k_2) = \phi_{oo}(k_2, k_1)$ and the identity (4.86). The result is $E_{SPDC} = 2 |\alpha_P|^2 |\xi|^2 \int dk_1 dk_2 \hbar \tilde{\omega}_{ok_1} |\phi_{oo}(k_1, k_2)|^2$. Recalling that $\tilde{\omega}_{ok_1} = \tilde{\omega}_{o(k_{1\rho}, k_{1z})}$ and substituting in $I$ from Eq. (4.83) with $\hbar(k_{1\rho}, k_{1z}) = \hbar \tilde{\omega}_{o(k_{1\rho}, k_{1z})}$.
yields

\[ E_{SPDC} = \frac{|\alpha_P|^2 \hbar \varepsilon_0 \left( \Gamma_2 y' z' \right) \sin^2 (\theta) L}{4\pi v_p^2} \int_{P(C)} dk_{1z} \sqrt{1 + \left( \frac{dk_{1z}}{dk_{1\rho}} \right)^2 k_{1\rho} \hbar F_D (k_{1z}, \omega_P) \tilde{\omega}_{o(k_{1z}, k_{1z})} \tilde{\omega}_{o(k_{1\rho}, k_P - k_{1z})} \omega_P} \frac{V (k_{1\rho}, k_{1z})}{v_P}. \] (4.95)

Eq. (4.95) gives the expected energy \( E_{SPDC} \) of the generated bipolariton state. Although it incorporates the full physics of the problem in 3-dimensions, it has been reduced to a well-behaved 1D integral that can be done numerically. No smoothing was done to obtain Eq. (4.95), but comparing it to Eq. (4.91) it can be written in the form

\[ E_{SPDC} = \int_S d^2 k_{1z} \hbar \tilde{\omega}_{o k_{1z}} \langle N_{k_{1z}} \rangle. \] (4.96)

So the explanation after Eq. (4.92) of the factors contributing to \( \langle N_{k_{1z}} \rangle \) applies here to the SPDC energy. Down-converted pairs are created in the neighborhood of the matching surface; the amplitude of generation into a down-conversion channel depends on the overlap of the matched triad of mode fields, which depends on material dispersion through the various \( \zeta \) factors in \( F_D (k_{1z}, \omega_P) \). And the three-wave group velocity walkoff \( V (k_{1\rho}, k_{1z}) \) determines the expected amount of energy transferred from the pump to the down-converted fields. We devote the next section to further interpretation of \( E_{SPDC} \).

### 4.6 Scaling properties of the down-converted energy

Here we recast Eq. (4.95) in a form that makes its scaling properties transparent. Note first that SPDC is different from other simple second-order nonlinear interactions in that it is nonresonant, i.e. rather than a single or countable set of wave-vectors being phase- and frequency-matched, the process is inherently broadband.

Still, the idea is to relate SPDC to the following general one-dimensional description of the interaction between a single triad of waves, mediated by a uniform, nondispersive second-order nonlinearity [112][80] in a crystal of length \( L \). Consider one wave to be an undepleted “pump” and the other two waves to initially have small amplitude, and consider the interaction to be coherent—that is, the waves are perfectly phase- and frequency-matched. Then the power generated in one of the weak waves is proportional to each of the powers in the other two waves, and is inversely proportional to the cross-sectional area of the interaction. The power transfer is also proportional to \( L^2 \), where
$L$ is the crystal length over which this coherent interaction takes place. The description is the same for nearly-matched triads: Associating a coherence length $\pi/|k_P - k_1 - k_2|$ with any given triad $(k_P, k_1, k_2)$, we have that for any triad with coherence length at least as long as $L$, power transfer is also proportional to $L^2$.

By contrast, here in Eq. (4.95) for an undepleted pump pulse of finite energy we find that $E_{SPDC}$ is proportional to $L$ rather than $L^2$. This is indicative of the fact that SPDC is broadband; while for any given matched triad the power transfer is proportional to $L^2$, the phase-matching spatial bandwidth $\kappa_w$, (i.e., the magnitude of the interval of triads with coherence length $\geq L$) is inversely proportional to $L$. Note that in the equations leading up to Eq. (4.95) that $E_{SPDC}$ is indeed proportional to $L^2$, except for a factor of $L^{-1}$ due to integration over the $\text{sinc}^2$ function in the direction normal to $S_\Upsilon$ in Eq. (4.78). This suggests that the phase-matching spatial bandwidth is associated with the $\text{sinc}^2$ function.

To identify a phase-matching spatial bandwidth, we consider a typical matched triad of wave vectors such that $k_1$ is on $S$ and $k_2$ is its complement with $k_1$ and $k_2$ mutually matched to $k_P$. This corresponds to some $K$ (q.v. after Eq. (4.77)) on $S_\Upsilon$, which we recall is the surface on which the argument of the $\text{sinc}^2$ vanishes. We take $k_1\rho \ll k_1z$ and let $k_{1z} \approx k_{2z} \approx k_{DC}$, where $k_{DC} \equiv k_P/2$ is a typical down-conversion wave vector magnitude; it will be seen in the numerical calculation of Sec. 4.7 that these are typical relationships between the components of $K$ at points at which down-conversion is significant. Focusing on the argument of the $\text{sinc}^2$, we next take a linear expansion in $K$ of $\Upsilon(K)$, and write

$$\Upsilon(K + \kappa) \simeq \kappa \cdot \nabla_K \Upsilon(K).$$

The $\text{sinc}^2$ factor falls off most quickly in the direction of the gradient $\nabla_K \Upsilon(K)$. We recall from before Eq. (4.79) that on $S$ the quantity $v_P |\nabla_K \Upsilon(K)|$ is just the three-wave group velocity walkoff, and we denote by $v_w$ a typical value of this walkoff speed. Accordingly, considered for $\kappa$ parallel to $\nabla_K \Upsilon(K)$ we write $\Upsilon(K + \kappa) \approx \kappa v_w / v_P$. Using this typical value as an estimate for the argument, the full-width of the $\text{sinc}^2$ function between its centralmost zeros is used to define a phase-matching spatial bandwidth:

$$\kappa_w \equiv \frac{4\pi v_P}{v_w L}.$$ 

Note that if $V(k_{1\rho}, k_{1z})$ varies by more than an order of magnitude over the part of $S$ on which the bulk of SPDC occurs, then it is less appropriate to define a single phase-matching spatial bandwidth. Instead of making the replacement $V(k_{1\rho}, k_{1z}) \rightarrow v_w$ in the
argument leading to Eq. (4.98), a separate phase-matching spatial bandwidth parameter can be associated with each point on $S$. However, it is seen in the example calculation of Sec. 4.7 that for our system a single estimate for $v_w$ is representative of the walkoff speed in the problem.

Next we identify a typical down-converted polariton frequency, $\omega_{DC} \equiv \omega_P/2$, and an energy associated with quantum fluctuations, $E_{fl,DC} \equiv \frac{1}{2}\hbar \omega_{DC}$. We recall from Eq. (4.39) that the pump energy is $E_P \simeq \hbar \omega_P |\alpha_P|^2$, and recall from after Eq. (4.82) that $k_{1p,typ}$ is a typical value for $k_{1p}$. Then as a rough estimate of the surface area of $S$ we take $S \equiv 2\pi k_{1p,typ} k_{DC}$. We identify a typical normalization factor for the generated fields, $\zeta_0 \equiv \zeta_0(k_\omega(\omega_P/2))$. And anticipating that the dispersion factor $F_D(k_{1z}, \omega_P)$ does not vary too much with $k_{1z}$ we define a simpler factor that is independent of $k_{1z}$ but retains dependence on the pump frequency. Overloading notation (q.v. Eq. (4.84)) we define

$$ F_D(\omega_P) \equiv \zeta_0^2 k_{DC}^4, \quad (4.99) $$

where recall $\zeta_0^2 = \zeta_{ek_P}^2$ and $\zeta_0 \equiv \zeta_0(k_\omega(\omega_P/2))$. We then identify a parameter with units of energy density associated with the properties of the nonlinear crystal system (note $\omega_{DC}^2 / (k_{DC} v_P^2) \simeq 1$):

$$ U_{sys} \equiv \left[ \frac{1}{4\pi} \varepsilon_0 \left( \Gamma_{zz'}^{y'y''} \right)^2 \sin^2 (\theta) F_D(\omega_P) \frac{\omega_{DC}^2}{k_{DC}^2 v_P^2} \right]^{-1}. \quad (4.100) $$

Identifying $\lambda_{DC} \equiv 2\pi / k_{DC}$, the expected energy can then be written as

$$ E_{SPDC} = J E_P E_{fl,DC} U_{sys}^{-1}(S\kappa_w) \frac{L^2}{\lambda_{DC}^2}, \quad (4.101) $$

where

$$ J \equiv \int_{P(C)} dk_{1z} k_{DC}^2 \sqrt{1 + \left( \frac{dk_{1z}}{dk_{1z}} \right)^2 \frac{\tilde{\omega}_{0(k_{1p},k_{1z})} \tilde{\omega}_{0(k_{1p},k_{1z})} - \omega_{DC}^2}{2\omega_{DC}^2} \frac{k_{1p}}{k_{1p,typ}} \frac{1}{V(k_{1p}, k_{1z})}} \quad (4.102) $$

is a unitless integral of order 1, with $V(k_{1p}, k_{1z}) \equiv V(k_{1p}, k_{1z}) / v_w$.

In terms of the description of coherent interaction at the beginning of this section, Eq. (4.101) suggests a narrative of the SPDC process: A down-conversion “signal” polariton fluctuation interacts coherently with the pump field to generate an idler. Those triads that contribute significantly to the SPDC energy, i.e. those with a coherence length at
least as long as the nonlinear crystal, are associated with a region of \( k \)-space that can
roughly be visualized as the locus of points within a tolerance \( \kappa_w \) of the matching surface \( S \); the associated volume in \( k \)-space is \( (S\kappa_w) \).

Note that the physical area of the pump pulse, \( \propto \kappa_{-2} \), is irrelevant to the amount of
down-converted energy produced, so focusing of a pump pulse has no effect on \( E_{SPDC} \). Even the pump pulse length, \( \propto \kappa_{-1} z \), does not appear in the final result, although it is
involved in setting the relative length scales in the problem in Eq. (4.32).

Besides its contribution to the shape of the matching curve \( C \), the overall contribution
of dispersion to the down-converted energy is seen Eq. (4.101). Except for the unitless
integral \( J \), the material dispersion appears in Eq. (4.101) via \( U_{sys} \) defined in Eq. (4.100),
so that \( E_{SPDC} \) is proportional to \( F_D(\omega_P) \).

### 4.7 Numerical examples: BBO and AlGaAs

In this section we apply our results in detail to a sample BBO crystal, and estimate the
effect of material dispersion on the SPDC energy generated in AlGaAs structures.

BBO (\( \beta \)-barium borate) is a standard negative uniaxial nonlinear crystal used in
current type-I SPDC experiments [119]. It has trigonal \( 3m \) symmetry [118], and is
transparent and phase-matchable for wavelengths as short as 0.2 \( \mu \)m. Its ordinary and
extraordinary indices of refraction are each given approximately by a Sellmeier formula,
and these are each good fits for free-space wavelengths from 1 \( \mu \)m down to 205 nm [120]. We use the Sellmeier formulae for wavelengths shorter than 1 \( \mu \)m; for longer wavelengths
dispersion is small and we employ a smoothly-matched linear extrapolation of \( n(\omega) \).

Compared with tabulated experimental index measurements [118], this fit matches all
datapoints within a relative error of \(< 5 \times 10^{-4} \).

For the detailed calculation we take the pump carrier frequency to be 3.53 eV, which
corresponds to a line of an Ar-ion laser, a common pumping device. We are interested in a
system in which it is possible to have phase and frequency matching for the generation of
two fields near \( \omega_P/2 \). This is possible if \( n_e(\omega_P, \theta) = n_o(\omega_P/2) + \delta \) for any sufficiently small
positive \( \delta \), which implies a small positive \( k_{1,2,3}P \) for phase matching at \( k_{1z} = k_{2z} = k_P/2 \).

Although the choice \( \delta = 0 \) would correspond to collinear degenerate phase matching, it
represents a point of measure zero in parameter space, so to simulate the parameters
of real experiments we take a nonzero but small value of \( \delta \). Specifically we take \( \delta = 1.0 \times 10^{-4} \), which corresponds to the optic axis tilt angle \( \theta = 33.56^\circ \).
Fig. 4.3 gives the indices of refraction of BBO. The upper and lower curves display the respective ordinary and extraordinary indices. The middle (magenta) curve shows $n_e(\omega, 33.56^\circ)$, the flatter (pink) curve shows $n_o(\omega/2)$, and the grey vertical line indicates the pump frequency $\omega_P$; so $|\delta|$ is the vertical distance between the two middle curves at their intersections with the grey line.

The pump pulse is taken to have full-width at half-maximum (FWHM) duration $100 f_P$ fs, with $f_P$ a unitless free parameter of order 1; this choice corresponds to typical experimental scenarios [121]. The corresponding FWHM spatial length of the pulse in the $z$-direction is $18.0 f_P \mu m$, and this implies a spatial bandwidth of $\kappa_z = 3.03 \times 10^{-2} f_P^{-1}$ eV/ℏc; for reference we note that $1 \text{ eV}/\hbar c = 5.067 \mu m^{-1}$. The scaling choices $\kappa_z^{-1} \ll L$ from (4.32) and $R_D \ll \kappa_z^{-2} L^{-1}$ from (4.33) respectively set lower and upper bounds on the crystal length for which the theory is valid. Together, these conditions imply for a $3.53 \text{ eV}$ pump that

$$7 f_P \mu m \ll L \ll 6 f_P^2 \text{ mm}. \quad (4.103)$$

Thus for $f_P = 1$ the condition (4.103) is satisfied in a standard $L = 0.5 \text{ mm}$ crystal, and we take this to be the length of the crystal in this detailed example. We note though that for $f_P = 1$, a $0.5 \text{ mm}$ crystal is found to satisfy the scaling conditions (4.32) and (4.33) for any pump carrier frequency up to $5.5 \text{ eV}$.

In Fig. 4.4 is shown the matching curve $C$, which is expressed as $k_{1\rho}(k_{1z})$, as defined implicitly by Eq. (4.82). The grey vertical lines indicate $k_{1z} = 0$ and $k_{1z} = k_P$. Note that
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Figure 4.4: Phase- and frequency-matching curve $C$ for BBO in the geometry of Fig. 4.1. The (violet) circles overlaid above $C$ have areas respectively proportional to $\langle N_k \rangle$. The (cyan) circles below $C$ have areas respectively proportional to the linear density analogue of $\langle N_k \rangle$.

$C$ has reflection symmetry about the dashed vertical midline $k_{1z} = k_P/2$. There are three extrema, and we use $E$ to denote this set of extrema. The wave vectors of both members of a perfectly matched pair of down-converted polaritons can be read from this graph: Given a $k_1 = (k_{1\rho}, \phi, k_{1z})$ with $k_{1\rho}$ given in terms of $k_{1z}$ by Fig. 4.4, its matched pair is $k_2 = (k_{1\rho}, -\phi, k_P - k_{1z})$. That is, the $\rho$- and $z$-components of the $k_2$ corresponding to some $k_1$ are given by the reflection of $k_1$ in the dashed vertical midline of Fig. 4.4. Observe that for a perfectly matched pair we have that $k_1$ is in $E$ iff $k_2$ is in $E$.

It is apparent from Fig. 4.4 that in addition to phase matching at frequencies near $\omega_P/2$, phase matching also occurs for $k_1$ and $k_2$ corresponding to very different frequencies, and at a wide range of relative angles from nearly parallel (copropagating) to anti-parallel (counterpropagating).

Anticipating Fig. 4.5, in Fig. 4.4 the (violet) circles overlaid just above the curve $C$ have areas relatively proportional to $\langle N_k \rangle$, the coarse polariton number per unit area of the matching surface $S = C \otimes \varphi$ defined after Eq. (4.88). The (cyan) circles overlaid just below $C$ in Fig. 4.4 have areas in relative proportion to the integral of $\langle N_k \rangle$ over $\varphi$, so they are the number per unit length along $C$. From the upper set of circles it is apparent that SPDC has a broad bandwidth with the greatest concentration of $\langle N_k \rangle$. 
Figure 4.5: For points on \( C \), shown is \( \langle N_{k_1} \rangle \), a coarse-grained number operator expectation per unit area of \( S = C \otimes \varphi \). The vertical axis unit is \( 10^{-11} |\alpha_P|^2 (\chi_2)^2 \mu m^2 \).

being at \( k_{1z} = k_{2z} = k_P/2 \); from the lower set of circles it is apparent that after doing the angular integral over the full circle of radius \( 2\pi k_{1\rho} \) in \( k \)-space that although the peak area density of \( \langle N_k \rangle \) is at \( k_{1z} = k_{2z} = k_P/2 \), the peak of the number of bipolaritons is at either side of that region.

It is also seen from Fig. 4.4 that the small-angle approximation of (4.71) holds most strongly at the midpoint of \( C \), and does not hold from the left and right extrema outwards. However, 74% of the contribution to the down-converted polariton number expectation \( \langle N_o \rangle \) is from points at which for both \( j = 1, 2 \) the left-hand side (LHS) of (4.71) is less than 1/3, and 87% of the contribution to \( \langle N_o \rangle \) is from points at which the LHS of (4.71) is less than 1/2. This justifies the use of the axial symmetry approximation (4.73).

The magnitude of \( \langle N_k \rangle \) is plotted fully in Fig. 4.5. The expected number of generated polaritons is proportional to \( |\alpha_P|^2 \), the expected number of input pump polaritons. It is also proportional to the squared magnitude of the nonlinear response, \( (\chi_2)^2 \), which in turn depends on \( \chi_2^{y'y'z'} \) via Eq. (4.6). Now what is usually tabulated is \( d_{31} = \chi_2^{y'y'z'}/2 \), and its empirical value is not very well-known; estimates vary by an order of magnitude \[118\][65][122]. However, they suggest that \( \chi_2^{y'y'z'} \) is on the order of 1 pm/V, so we write \( \chi_2^{y'y'z'} = \chi_2 \) pm/V, and leave \( \chi_2 \) as a unitless parameter of order 1. Thus in Fig. 4.5, \( \langle N_k \rangle \) is quoted in units of \( 10^{-11} |\alpha_P|^2 (\chi_2)^2 \mu m^2 \).

For points \( k_1 \) on \( C \), Fig. 4.6 shows the group velocity of a down-converted polariton as a function of \( k_{1z} \), that is, the function shown is \( v_o(k_{1\rho}(k_{1z}), k_{1z}) \) from Eq. (4.80); the group velocity of the matching \( k_2 \) polariton is given by the reflection of this plot.
Chapter 4. Spontaneous parametric down-conversion

Figure 4.6: Group velocity of a down-converted polariton as a function of $k_{1z}$ for points on $C$. The uppermost curve (red) is $v_o$. The $\rho$- and $z$- components $v_{op}$ and $v_{oz}$ are also shown (both orange) as a function of $k_{1z}$.

in the vertical dashed line at $k_P/2$. The horizontal (blue) dashed line indicates $v_P$, the $z$-component of the pump group velocity. The uppermost curve (red) is $v_o$. The $\rho$- and $z$- components $v_{op}$ and $v_{oz}$ are also shown (both orange) as a function of $k_{1z}$, with $v_{oz}$ being the curve that merges with $v_o$ at large $k_{1z}$. Note that $v_{oz}$ and $v_P$ cross at two points on the interval $k_{1z} \in [0, k_P]$, and that these points nearly mirror each other under reflection in the vertical dashed line at $k_P/2$. These crossings are a topological feature that occur in BBO regardless of the pump frequency; we use $T$ to denote the set of wave vectors that correspond to these points.

In Fig. 4.7 we plot $1/V(k_{1\rho}, k_{1z})$, the reciprocal of the three-wave group velocity walkoff from Eq. (4.79), as a function of $k_{1z}$ for points on $C$. The SPDC energy generated is proportional to $V^{-1}$. It is clear from Fig. 4.7 that the group velocity walkoff suppresses SPDC for significantly different $\mathbf{k}_1$ and $\mathbf{k}_2$. Also shown is $1/V(\mathbf{z})(k_{1\rho}, k_{1z})$, which is the reciprocal group velocity walkoff that would be obtained from Eq. (4.79) were the transverse group velocity walkoff term neglected. The peaks of $V(\mathbf{z})^{-1}$ are finite and asymmetric, but each reaches $10^3$.

These peaks are a striking feature, and are robust, in that they appear in this system for any pump frequency at which BBO is transparent. More generally, they arise not just in this system but in systems with a wide class of $C$ curves. They are due to the
interplay of a symmetry property with a topological property, and can be explained as follows. Consider a perfectly matched pair of wave vectors \( k_1 \) and \( k_2 = k_P - k_1 \) which correspond to a pair of cogenerated polaritons. Let \( k_1 \) be an extremum, i.e. a member of \( \mathcal{E} \); it follows from the discussion after Eq. (4.103) that \( k_2 \) is also in \( \mathcal{E} \). Now substitute the parameterization \( k_{1\rho} = k_{1\rho}(k_{1z}) \) into Eq. (4.82), take the total \( k_{1z} \)-derivative \( d/dk_{1z} \) of the resulting equation, and use that \( dk_{1\rho}/dk_{1z} \) vanishes for points in \( \mathcal{E} \). From the result it is seen that

\[
\mathbf{k}_1 \in \mathcal{E} \implies v_{oz}(k_{1\rho}, k_{1z}) = v_{oz}(k_{2\rho}, k_{2z}),
\]

where \( \implies \) denotes implication. Given any \( \mathbf{k}_1 \) at an extremum of \( \mathcal{C} \), and given its matched \( \mathbf{k}_2 \), the \( z \)-components of the group velocities are identical. This does not hold for the transverse components of the group velocities. Now were \( \mathbf{k}_1 \) a member of \( \mathcal{T} \) as well as \( \mathcal{E} \), it would follow that all three polaritons in the matched triad would have identical group velocity \( z \)-components. This does not occur here, but for a \( \mathbf{k}_1 \) that is either the leftmost or rightmost extremum of \( \mathcal{C} \), it is seen that \( \mathbf{k}_1 \) is close to a point in \( \mathcal{T} \). Consequently, we have \( v_P \approx v_{oz}(k_{1\rho}, k_{1z}) = v_{oz}(k_{2\rho}, k_{2z}) \), thus there is little longitudinal walkoff and \( V_{(z)}^{-1} \) is large\(^2\).

In reality, this is checked because the generated polaritons walk off from one another in the transverse direction. However, this effect is only captured in the expressions for

\(^2\)For lower pump frequencies, e.g. 2 eV, in addition to the leftmost and rightmost extrema, the extremum at the midpoint of \( \mathcal{C} \) is also sufficiently near a point in \( \mathcal{T} \) that \( V_{(z)}^{-1} \) deviates significantly from \( V^{-1} \).
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Figure 4.8: Dispersion normalization factor $F_D(k_{1z}, \omega_P)$ as a function of $k_{1z}$ for points on $\mathcal{C}$ (blue), shown with $\overline{F}_D(k_{1z}, \omega_P)$ (violet), which neglects the contribution of $n'$ to mode normalization.

The generated energy and bipolariton number if the full group velocity walkoff factor is used, including the transverse component.

In Fig. 4.8 as the lower curve (blue) we plot the dispersion normalization factor $F_D(k_{1z}, \omega_P)$ defined in Eq. (4.84), for points on $\mathcal{C}$. The accompanying upper curve (violet) is $\overline{F}_D(k_{1z}, \omega_P)$, which is just $F_D(k_{1z}, \omega_P)$ but with the terms containing $n'$ neglected. For this particular system, correct normalization involving the $n'$ terms makes a difference: $\overline{F}_D(k_{1z}, \omega_P)$ differs from $F_D(k_{1z}, \omega_P)$ by between 10% and 14% on $\mathcal{C}$. Note that these factors individually vary by less than 2% on $\mathcal{C}$, so they are well-estimated respectively by the wave-vector-independent values $\overline{F}_D(\omega_P)$ and $F_D(\omega_P)$ defined according to Eq. (4.99).

The factors $F_D(\omega_P)$ and $\overline{F}_D(\omega_P)$ are plotted as the respective lower (blue) and upper (violet) curves in Fig. 4.9. Recall these factors are global estimates of the effect of dispersion on the SPDC energy, as a function of the pump frequency. The relative error of $\overline{F}_D(\omega_P)$ with respect to $F_D(\omega_P)$ ranges from 7% to 47% over the domain of pump frequencies shown, which reaches the upper edge of the transparency range for BBO. The relative error increases with pump frequency, which is as expected, since dispersion in the indices of refraction also increases over this range of frequencies. And the dispersion normalization effect is significant for pump frequencies near the band edge.

We next turn to the evaluation of $E_{SPDC}$ and of the various physical scaling parameters which appear in Eq. (4.101) for $E_{SPDC}$. First consider the phase-matching spatial bandwidth $\kappa_w$. We have $v_P = 0.564c$, and take $v_w = 0.0383c$, which is the $z$-component
of the group velocity difference between the pump and an o-wave at $\omega_P/2$; the transverse component of the walkoff is not significant at this point, so this choice of $v_w$ is smaller than the value of $V$ at the midpoint of $\mathcal{C}$ by only 5%. In general, however, the transverse walkoff must be included; for example, for a pump at $2.0$ eV with $\delta = 10^{-4}$, were $v_w$ set in this way it would be smaller than the value of $V$ at the midpoint of $\mathcal{C}$ by 45%. Then $\kappa_w$ is in the end obtained by substituting $v_w$, $v_P$ and $L$ into Eq. (4.98), and doing so we obtain $\kappa_w = 0.0729 \text{eV}/\hbar c$. Next take $k_{1p,typ}$ to be $0.212 \text{eV}/\hbar c$, which is seen from Fig. 4.4 to be a typical value. We have $k_{DC} = k_P/2 = 2.94 \text{eV}/\hbar c$, which corresponds to $\lambda_{DC} = 2\pi/k_{DC} = 422 \text{nm}$. Then using the definition of $S$ after Eq. (4.98) we find $S = 11.5 \text{(eV}/\hbar c)^3$. The pump energy is $E_P = (3.53 \text{eV})|\alpha_P|^2$. We take $\hbar \omega_{DC} = \hbar \omega_P/2 = 1.76 \text{eV}$, and take $E_{fl,DC}$ to be $\hbar \omega_{DC}/2 = 0.882 \text{eV}$. The integrand in Eq. (4.102) is well-behaved, and numerical integration yields $J = 0.401$. Since we are neglecting dispersion in the nonlinear response, to construct $\Gamma_{ijl}^2$ from $\chi_{ijl}^2$ in Eq. (4.6), we use the indices $n_o(\omega_P/2)$ and $n_e(\omega_P)$; thus the denominator of the right-hand side of Eq. (4.6) contains $n_o^4(\omega_P/2)n_e^2(\omega_P)$. Making this substitution in Eq. (4.100) yields $U_{sys} = 2.91 \times 10^{14} \bar{\chi}_2^{-2} \text{eV}^4 \hbar^{-3} c^{-3}$. Substituting all of the above values into Eq. (4.101), we finally obtain $E_{SPDC} = 1.72 \times 10^{-9} |\alpha_P|^2 \bar{\chi}_2 \text{eV}$, where we recall that $\bar{\chi}_2$ is a unitless parameter of order 1, and $|\alpha_P|^2$ is the expected number of pump polaritons.

Although this calculation was made for $\delta = 1.0 \times 10^{-4}$, the qualitative results of this particular example are not sensitive to the precise value of $\delta$. Were $\delta$ decreased to zero, in Fig. 4.4 the $k_{1p}$-value of the extremum at the midpoint of $\mathcal{C}$ would monotonically decrease.
to zero, and at $\delta = 0$ this extremum would become a cusp. However, the corresponding plot of $\langle N_k \rangle$ in Fig. 4.5 would not change significantly with the decrease of $\delta$. This is explained as follows. From Eq. (4.91), $\langle N_k \rangle$ is inversely proportional to $V(k_{1\rho}, k_{1z})$, which in turn depends on $\delta$. For the parameters of this example calculation, for $(k_{1\rho}, k_{1z})$ at the midpoint of $\mathcal{C}$, a decrease in $\delta$ towards zero corresponds to a decrease towards zero of the transverse part of $V(k_{1\rho}, k_{1z})$, but to only a small change in the longitudinal part of $V(k_{1\rho}, k_{1z})$. It can be seen from Fig. 4.7 that at the midpoint of $\mathcal{C}$, $V(z)(k_{1\rho}, k_{1z})$ is a good approximation for $V(k_{1\rho}, k_{1z})$. Since $V(k_{1\rho}, k_{1z})$ at that midpoint is not sensitive to its transverse part, it follows that $\langle N_k \rangle$ is not significantly changed by a decrease in $\delta$. By contrast, were the parameters of the example such that $V$ at the midpoint of $\mathcal{C}$ were sensitive to the transverse part of the three-wave walkoff, then a decrease in $\delta$ would appreciably affect $\langle N_k \rangle$.

Considering our final result for $E_{SPDC}$, we note that were the transverse component of the group velocity walkoff neglected in this calculation, $E_{SPDC}$ would be calculated to be $>100$ times the correct value. Similarly, were the effect of dispersion on the normalization of the modes is neglected, i.e., if terms containing $n'$ were set to zero in the various $\zeta$ factors, then the value obtained for $E_{SPDC}$ would be too large by 11%. This is a modest correction in the particular system considered here; for a system with a pump nearer to the band gap, the effect would be much larger. For example, for a 5.5 eV pump with $\delta = 1 \times 10^{-4}$, neglect of dispersion in the $\zeta$ factors leads to a value of $E_{SPDC}$ that is too large by 35%.

The effect would be larger still in a system that, at a typical pump carrier frequency, is more dispersive than BBO. Indeed, the dispersion factor $F_D(\omega_P)$ defined in Eq. (4.99) is general, and applies to SPDC in any material or structured medium. To obtain a sense of the size of the correction that can arise in common media, we step back from the specific BBO system considered so far. Precise calculations of the SPDC energy are of most interest for structured media designed to maximize the conversion efficiency, including waveguides [77], microring resonator systems, photonic crystals [79][65], and other nanostructures. We consider $F_D(\omega_P)$ then for Al$_x$Ga$_{1-x}$As, which is a semiconductor commonly used in the fabrication of functional nanostructures. It is also commonly used at frequencies near the electronic band gap where dispersion is significant, and it is a material for which very good optical constant data is available [123]. Although Al$_x$Ga$_{1-x}$As has cubic crystal symmetry and therefore does not produce SPDC in bulk, it has a large $\chi_2$ compared with BBO [65], it can be used for down-conversion in nanostructures, and it
has been suggested as a material for SPDC in photonic crystals [65]. Indeed, a detailed calculation for an Al$_x$Ga$_{1-x}$As waveguide has recently been carried out [80] using the normalization condition for the linear modes taken here [1].

For an isotropic crystal, Eq. (4.99) is clearly replaced by

\[
F_D(\omega_P) \equiv \left[ \frac{1}{n^2(\omega_P)} + \omega_P \frac{n'(\omega_P)}{n^3(\omega_P)} \right]^{-1} \left[ \frac{1}{n^2(\omega_P/2)} + \frac{\omega_P n'(\omega_P/2)}{2 n^3(\omega_P/2)} \right]^{-2}. \tag{4.105}
\]

Of course, $F_D(\omega_P)$ contains no information about the mode profile, which is an important factor in structured media, but we expect that $F_D(\omega_P)$ is useful for estimating the effect on SPDC of the contribution of material dispersion to the mode normalization. In Fig. 4.10 we plot $F_D(\omega_P)$ and for comparison the quantity $F_D(\omega_P) \equiv n^2(\omega_P) n^4(\omega_P/2)$, which is the dispersion factor that would arise were the terms in $F_D(\omega_P)$ containing $n'$ neglected. Here the doping fraction is taken for specificity to be $x = 0.176$, at which level of doping Al$_x$Ga$_{1-x}$As is a direct-gap semiconductor (as it is for all $x < 0.4$); the results are similar for other direct-gap semiconductors, since they all have considerable dispersion near the band edge. For a pump close to the band edge, use of $F_D(\omega_P)$ leads to a significant overestimate of the SPDC energy, which we recall is proportional to $F_D(\omega_P)$. The factor $F_D(\omega_P)$ is an overestimate of $F_D(\omega_P)$ by $> 350\%$ for a pump close to the band edge; similarly $F_D(\omega_P)$ overestimates $F_D(\omega_P)$ by $> 50\%$ if the pump is $< 4\%$ detuned from the band edge, by $> 31\%$ if the pump is $< 10\%$ detuned from the band edge, and by $> 20\%$ if the pump is $< 20\%$ detuned from the band edge.

![Figure 4.10: For Al$_x$Ga$_{1-x}$As with $x = 0.176$, shown are $F_D(\omega_P)$ (blue) and $F_D(\omega_P)$ (violet).]
4.8 Discussion

In this chapter I have presented a rigorous calculation of spontaneous parametric down-conversion. The calculation keeps much of the underlying physics: It is based on a Hamiltonian formulation of the electromagnetic field in dispersive media that fully accounts for material dispersion, and it captures the full 3D physics including the three-wave group velocity walkoff. Approximations are made in a controlled way. In addition to characterizing the 3D phase- and frequency-matching surface, the calculation gives both the number operator distribution over wave vectors and the expected energy, including their absolute magnitudes. It also gives the scaling behavior of the SPDC energy.

Even for a narrowband input pulse, I have shown that dispersion typically has a significant effect beyond its effect on defining the phase matching curves, and can have a large effect on the total down-converted energy. This is especially so if the pump carrier frequency is close to the band gap. For such a system, a calculation that fully captures the effect of dispersion on phase and frequency matching, but neglects the explicit contribution of $n'(\omega)$ to the normalization of the linear modes, leads to an overestimate of the conversion energy by as much as $> 40\%$ in BBO and $> 300\%$ in $\text{Al}_{0.176}\text{Ga}_{0.824}\text{As}$.

I identified a fully 3D, three-wave group velocity walkoff that is inversely proportional to the SPDC energy produced, and I showed that naïve approximations using a 1D group velocity walkoff can yield extremely wrong results, even if the pump and down-converted fields all propagate at only a small angle from the $z$-axis. This phenomenon is due to the interplay of a topological property with a symmetry property of the matching curve $\mathcal{C}$, and occurs in BBO for any pump carrier frequency below the BBO band gap. In our specific example calculation in BBO, neglect of the transverse walkoff would yield a result for $E_{\text{SPDC}}$ that is too large by two orders of magnitude.

Finally, the fact that the three-wave group velocity walkoff is a key determinant of the SPDC energy suggests there is much scope for tailoring dispersion curves to enhance the conversion efficiency using structured media.
Chapter 5

Conclusion

This thesis presents a Hamiltonian formulation of dispersive and absorptive structured (inhomogeneous) media, and its application to two problems of current interest: propagation in structured media and spontaneous parametric down-conversion. In this concluding section I summarize and contextualize the contributions of Ch. 2–4, and then point to future applications of the work presented here.

At the theoretical core of the thesis is Ch. 2, in which I presented a Hamiltonian formulation of dispersive and absorptive structured media explicitly obeying the Kramers-Kronig relations. The method focuses on the photonic component of the polariton modes of the system. The medium is represented using an intuitive model, and given an arbitrary space- and frequency-dependent dielectric response function, the model and canonical description of the modes of the coupled medium and electromagnetic field can be immediately found. In the final equations specifying the photonic component of the polariton mode field, the dielectric function is the only information about the medium that appears. Throughout Ch. 2, I explicitly treated transparent and absorptive frequency regimes, which have qualitatively different polariton characteristics.

The transparent regime supports a discrete polariton spectrum, and the polariton dispersion relation is just the dispersion relation of the electromagnetic modes of the dispersive system. Each polariton mode consists of its dispersive electromagnetic component augmented by the associated medium excitation component, and the electromagnetic components are obtained from the usual dispersive Maxwell master equation.

In the absorptive regime, I obtained a continuous spectrum of polaritons, even for a single wave vector, but showed that it is possible to identify effective broadened polariton bands. Furthermore, I showed that the electromagnetic component of each polariton
mode obeys the familiar nonabsorptive Maxwell master equation, but with an effective
dielectric function parameterized by a quantity termed the resonant oscillator amplitude.

Having established the formalism that is the core of the thesis, in Ch. 3–4 I pre-
sented applications of that theory. In Ch. 3, I presented a formulation of electromagnetic
propagation in structured media with arbitrary dispersive and absorptive linear response
consistent with the Kramers-Kronig relations.

I showed that those states that represent an arbitrary classical-like beam incident on a
quiescent absorptive and dispersive sample constitute a subset of the polariton coherent
states. States in this subset are the dispersive/absorptive analog of photon coherent
states, and I formally treated their generation via a classical source polarization, but
then hid these details from the user by allowing specification of an initial state within
this subset, so the user can naturally consider pulses with this formalism without having
to deal with the details of state generation.

In the transparent regime the dynamics are formulated in terms of a Schrödinger
equation satisfied by a polariton effective field, with emphasis on the photonic component
of the polariton modes of the system. In the absorptive regime fields separate into
a fluctuating part described in terms of vacuum polariton operators and modes, and a
non-fluctuating part represented by a polariton effective field with Schrödinger dynamics.
The coefficients in these dynamical Schrödinger equations are the propagation parameters
that capture the various physical effects in the problem: modal and material dispersion,
absorption, and diffraction.

The transparent regime propagation parameters are easily calculated. In the absorp-
tive regime, I showed that the complex propagation parameters are hidden in—and are
easily retrievable from—the same generalized dispersion surfaces typically used while
neglecting absorption; only a deformation of the cutting surfaces is needed.

As an example, complex propagation parameters (loss, group velocity, dispersion
and diffraction) were calculated as a function of carrier wave vector for an optical pulse
traversing a 1D Au/MgF$_2$ metallodielectric stack, using the full dispersive and absorptive
$\varepsilon(r, \omega)$. These propagation parameter curves were used to elucidate the various roles of
Bragg scattering, (electronic) interband absorption and field expulsion on propagation
in that sample. In calculating the propagation parameters I also showed that the Drude
model is inadequate for even determining the first order dynamics of a pulse in that
particular sample.

Further, I both corrected the result in the literature for the Beer’s law decay rate
in structured media, and provided a more general result applicable to structured media with causal absorptive response.

In Ch. 4 I applied the formalism of Ch. 2 to a purely quantum mechanical problem in optics: spontaneous parametric down-conversion. The calculation is fully 3D, and material dispersion is fully accounted for in the Hamiltonian formulation. I characterized the 3D phase- and frequency-matching surface, captured the full physics of the three-wave group velocity walkoff in 3D, and characterized the scaling behavior of the SPDC energy. I also rigorously derived expressions—including the absolute magnitudes—both for the down-converted state and for the number operator distribution over wave vectors.

Dispersion affects the normalization of the polariton modes of the system, and calculations of the down-conversion efficiency that neglect this can be off by 100% or more for common uniaxial or semiconductor media regardless of geometry if the pump is near the band edge.

Furthermore, due to the interplay of a topological property with a symmetry property of the phase- and frequency-matching curve, I showed that even when the down-converted polaritons are generated into a narrow forward cone, neglect of the transverse walkoff can lead to an overestimate of the SPDC energy by orders of magnitude.

Having summarized the contributions of this thesis, I turn to characterizing this work in the context of the literature. Besides the polariton-mode based approach presented in Ch. 2, there are other Hamiltonian formulations of structured dispersive and absorptive media, and most of these fall into two categories: noise-current methods and auxiliary field methods.

In noise-current methods [54][55][44] the medium is introduced as phenomenological noise currents in the Maxwell equations, and the noise current operators are the canonical operators of the theory. The Green function plays a central role in calculations. These methods apply naturally to problems in which the physics of the medium excitations is of primary interest, and the electromagnetic fields are of interest insofar as they mediate interactions between medium excitations; we call these medium-photon-medium (MPM) problems. Noise-current methods are also well-applied to problems in which the Green function is the standard tool with which practitioners represent states of the electromagnetic field. However, in these methods the electromagnetic fields are obtained only indirectly from the noise current operators via the Green function. Since the electromagnetic fields play a secondary role, noise-current methods are less-well suited to photon-medium-photon (PMP) problems, i.e. those that are most intuitively understood
as interactions between electromagnetic field states, mediated by the medium.

A second approach is the auxiliary field method [45][56][46][47]. In this method, the usual electromagnetic fields are augmented by two additional fields, and a modified set of Maxwell equations is postulated for their joint dynamics. This formulation has been shown to be formally equivalent to noise-current methods [58], but is less intuitive so I do not directly compare with it.

My approach falls into a third category: polariton-mode methods. Instead of noise-current operators, polariton operators are primary. Instead of expressing the electromagnetic fields in terms of Green functions acting on noise operators, I express these fields using an expansion in polariton operators and mode fields. Instead of solving for electromagnetic field states by using Green function methods, I focus directly on mode fields and the dispersion relation; I determine these latter quantities using the usual Maxwell master equation for dispersive, nonabsorptive media, but with an effective material response depending on the resonant oscillator amplitude.

Unlike noise-current methods, my approach is naturally suited to PMP problems because it focuses primarily on the photonic component of the polariton modes. Furthermore, since the modes and dispersion relation are central, my approach applies especially naturally to structured media for which these concepts are a natural way of expressing the fields in the nondispersive, nonabsorptive limit. And since most work in the literature on structured media is formulated in terms of the modes and band structure, my approach connects more directly to that literature than do noise-current methods. Most computational methods and available codes for structured media also work in terms of these concepts, and it is a further advantage over noise-current methods that these codes and numerical methods can be easily leveraged to calculate the polariton modes.

Besides the formulation presented in Ch. 2 of this thesis, there exist other approaches in the literature that fall into the category of polariton mode methods. Two such approaches are applicable to structured media, but do not involve the dispersion relation, Maxwell master equation, effective response, or resonant oscillator amplitude. They rely instead on Green function methods, and thus are more akin to noise current methods [59][51]. One provides the microscopic justification for noise-current approaches [51]. More closely comparable to my approach is the early work of Huttner and Barnett [43][52], and work that followed it [57]. However, these are respectively limited to uniform media and to a dielectric half-space, and historically attempts to extend them to structured media led to formulations that rely on Green function methods [51].
It should be noted that the applications of Ch. 3–4 of this thesis are PMP problems, i.e. they belong to a class of problems for which the formalism of Ch. 2 is well-suited. These problems have not been treated in the literature using other Hamiltonian formulations of dispersive and absorptive structured media, but they can be contextualized in terms of other adjacent work.

Much of the work in Ch. 3 focuses on classical propagation in absorptive and dispersive structured media. In comparison with the literature, it provides a dynamical equation with coefficients that are easily calculated, despite the multitude of physical effects included in its derivation: full dispersion and absorption based on empirical optical constants, scattering off interfaces of structured media, all aspects of the 3D physics, ability to handle metals, handling of wave packet states rather than continuous wave (CW) states, quantum fluctuations if desired, etc.

By contrast, there is no other formalism that simultaneously includes all of these effects and provides a dynamical equation with the propagation constants in terms of mode structure even up to first order. For example, envelope function approaches [2] have included material dispersion and absorption only at the perturbative level. Previous formulations of photon effective fields [3][75] easily handle modal dispersion and diffraction to higher orders, but not material dispersion or absorption.

Similarly, consider pulse propagation in metallodielectrics. These are difficult systems on which to perform numerical calculations, both due to the presence of significant dispersion, and because skin depths are small relative to the dimensions of the metallic and dielectric constituents. Band structure calculations have been done for a 1D metallodielectric stack using a Drude model for the metal [15], and transmission calculations have been done using empirical optical constants [110]. Other recent work has involved absorption for CW fields at a single frequency only [12][13], or has used a Drude model [109]; I showed in Ch. 3 that the Drude model is inadequate to even obtain the lowest order dynamics in the metallodielectric studied there. In the context of this literature, only using the results of Ch. 3 is it possible to understand pulse propagation, and to do it based on the full empirical response function of the metal. As discussed earlier in this section, even the literature result for the Beer’s law decay rate is replaced by the work done in Ch. 3.

Like the work of Ch. 3, the work in Ch. 4 is beyond work in the literature on SPDC in terms of the amount of essential physics included in the derivation. Even the nondispersive limit of the 3D calculation done here does not yet exist in the literature to my
knowledge. By performing the derivation at the level of generality done in Ch. 4 I identified and characterized SPDC features and phenomena not found in work in the literature. The literature work has typically used over-simplistic approximations, usually has not fully included dispersion, and is often quasi-1D at some level, and only by performing the derivation at the greater level of generality done in Ch. 4 is it possible to even qualitatively obtain some of the results there. Work on SPDC in the literature has focused on other issues, and applying the more general approach of Ch. 4 to those issues should be fruitful.

More broadly, the work done in this thesis suggests a wide range of avenues for future investigations. Many of these stem from the application chapters, i.e. Ch. 3 and Ch. 4.

In Ch. 3 I did not explicitly consider the generation and propagation of few-polariton states and squeezed states; this could be done and is of increasing interest. The formalism could be used to study spatial suppression or enhancement of fluctuations in structured media. The approach of Ch. 3 could also be used to classify the propagation characteristics of a range of particular dispersive and absorptive structures, in terms of propagation parameters that include pulse dispersion and diffraction. It could be used to design structures for particular dispersion or absorption properties. Generally it would be fruitful to apply it to further study of pulse propagation in metallodielectrics and to plasmon-polariton propagation. It would also be interesting to apply the approach to nonlinear classical optics studies. This is especially the case because the metallodielectrics it treats have much potential as nonlinear materials due to their high material nonlinearities and their ability to strongly concentrate fields.

I showed in Ch. 4 that three-wave group velocity walkoff is a key determinant of the down-conversion efficiency in typical nonlinear crystals, and this suggests that there is much scope for tailoring dispersion curves to enhance the conversion efficiency using structured media.

Although the calculation performed in Ch. 4 was done for type-I phase matching, the method can be adapted to type-II phase matching as well. And since the formulation leverages the photonic component of the polariton modes of the system, it is well-suited to calculations of SPDC in structured media, for which new designs of resonant structures have the potential to greatly enhance the conversion efficiency.

Additionally, it would be useful to improve beyond perturbation theory and the undepleted pump approximation, to further explore the role of dispersion, to modify the derivation for application to CW input fields, and to calculate the SPDC energy and
Chapter 5. Conclusion

Statistics for a number of common SPDC crystals following the approach taken for BBO in Ch. 4.

Besides its use for SPDC, perturbative nonlinearity could be introduced to the formalism of Ch. 2 for detailed studies of other nonlinear quantum optical effects in dispersive media. In highly dispersive media, the usually-neglected term $T_D$ (see Eq. (3.15)) in the normalization condition for the linear modes can contribute significantly to the nonlinear polarization: Based on typical parameters for propagation in electromagnetically induced transparency (EIT) atomic vapor cells, the contribution of the extra dispersion term $T_D$ can be larger than that of $T_0$ [124].

Similarly in Al$_{0.176}$Ga$_{0.824}$As, for which very precise optical constant data is available below the electronic band gap [123], the ratio $T_D$ to $T_0$ is positive in frequency domains with both normal dispersion and positive $\varepsilon(\omega)$; it is already as high as 10.0% at 1.16 eV, but is 28.3% at 1.55 eV, 100% at 1.64 eV and is 140% at 1.65 eV. That is, the dispersion term $T_D$ is significant (10%) for $\leq 31\%$ detuning from the gap, but actually dominates for $\leq 2.8\%$ detuning from the gap.

The formalism of Ch. 2 also invites future work. My approach is well-suited to considering the effect of material dispersion and absorption on photonic band gaps in particular systems of interest. Such a question is naturally posed in terms of the band gap of a nondispersive/nonabsorptive limiting photonic crystal being closed or altered by the material dispersion. Since my approach references a nominal crystal, the mechanism for closing and altering gaps can easily be explored within my framework.

The model of the medium described in Ch. 2 can also be extended. It does not include the effect of propagation of the medium fields, which would model a spatially dispersive material. However, since the method introduces a physically intuitive model Hamiltonian for these fields, they could be made to propagate by augmenting the Hamiltonian by a suitable kinetic term. Similarly, the theory does not currently allow for nontrivial magnetic response, and again this could be included if a suitable magnetic coupling tensor were introduced, and if an extra magnetic coupling term containing that tensor were added to the Hamiltonian.
Appendix A

List of symbols

Table A.1: Symbols used in Chapter 2

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D(r,t)$</td>
<td>Displacement field</td>
</tr>
<tr>
<td>$H(r,t)$</td>
<td>Magnetic H-field</td>
</tr>
<tr>
<td>$E(r,t)$</td>
<td>Electric field</td>
</tr>
<tr>
<td>$B(r,t)$</td>
<td>Magnetic B-field</td>
</tr>
<tr>
<td>$P(r,t)$</td>
<td>Polarization field</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>Permittivity of free space</td>
</tr>
<tr>
<td>$\mu_0$</td>
<td>Permeability of free space</td>
</tr>
<tr>
<td>$\Gamma^0_{ij}(r)$</td>
<td>Nominal dielectric response</td>
</tr>
<tr>
<td>$n(r)$</td>
<td>Nominal index of refraction</td>
</tr>
<tr>
<td>$\Gamma^t_{ij}(r,t)$</td>
<td>Dielectric response function</td>
</tr>
<tr>
<td>$\delta_{ij}$</td>
<td>Kronecker delta</td>
</tr>
<tr>
<td>$\mathcal{H}$</td>
<td>Hamiltonian</td>
</tr>
<tr>
<td>$H_{em}$</td>
<td>Bare electromagnetic Hamiltonian</td>
</tr>
<tr>
<td>$H_{med}$</td>
<td>Medium Hamiltonian</td>
</tr>
<tr>
<td>$H_{int}$</td>
<td>Interaction Hamiltonian that couples electromagnetic fields and medium</td>
</tr>
<tr>
<td>$\varepsilon^{ij}$</td>
<td>Levi-Civita symbol ($\varepsilon^{123} = -\varepsilon^{213} = 1$)</td>
</tr>
<tr>
<td>$\Lambda^{ij}(r,\Omega)$</td>
<td>Coupling tensor</td>
</tr>
<tr>
<td>$\Omega_g, \omega_g$</td>
<td>Minimum cutoff frequency for absorption</td>
</tr>
</tbody>
</table>

continued overleaf...
Appendix A. List of Symbols

$X_\Omega (r, t)$ Medium oscillator field

$X_\Omega (r)$ Medium oscillator field, time dependence kept implicit

$\Pi_\Omega (r, t)$ Medium oscillator conjugate momentum field

$\Pi_\Omega (r)$ Medium oscillator conjugate momentum field, time dependence kept implicit

$\mathcal{V}_\Omega$ Region over which the medium has nonzero absorption at frequency $\Omega$

$\hat{\zeta}_j (r, \Omega)$ Local principal axes of coupling tensor

$\lambda, \lambda'$ Components in basis of the local principal axes

$i,j$ Cartesian components

$\psi_\Omega (r)$ Medium field operator, time dependence left implicit

$X_\Omega^0 (r)$ Nondriven part of $X_\Omega (r)$

$\Gamma (r, \omega)$ Fourier transform of $\Gamma (r, t)$

$P$ Cauchy principal value

$n (r)$ Nominal index profile

$H_0$ Hamiltonian of nominal system

$H_\Delta$ Counter-term Hamiltonian

$m$ Mode label

$u, k$ Mode sub-labels: discrete mode label $u$ and wave vector $k$

$D_m (r), B_m (r)$ Mode fields of nominal system

$\omega_m$ Frequency of mode $m$ of nominal system

$\bar{m}$ Label of mode that is physical conjugate of $m$

$V$ Normalization region

$V$ Volume of $V$

$C_m^{(1)}$ Nominal displacement mode amplitude

$C_m^{(2)}$ Nominal magnetic mode amplitude

$a_m$ Nominal mode amplitude

$\Gamma_m^{(0)}$ See Eq. 2.51 mode coupling constant

$\Lambda_{\lambda}^m (r, \Omega)$ Mode coupling constant; see Eq. 2.56

$H_{LP}$ Longitudinal polarization polariton Hamiltonian

$H_{TP}$ Transverse polarization polariton Hamiltonian

$s_{lm}$ Longitudinal polarization polariton operator

$c_m$ Transparent regime transverse polarization polariton operator

continued overleaf...
Appendix A. List of symbols

$c_{\Omega m}$ Absorptive regime transverse polarization polariton operator

$\rho_{\Omega m}^{r} (r)$ Expansion coefficient of LP polariton

$\phi_{\Omega m}^{r} (r)$ Scalar “potential” field for LP polariton

$f_{\lambda}^{\Omega} (r)$ See Eq. 2.74

$S_{\Omega}$ Boundary of $V_{\Omega}$

$\alpha_{n}^{\Omega}, \beta_{n}^{\Omega m}$ Expansion coefficients for absorptive regime TP polariton operator in terms of nominal electromagnetic mode operators; see Eq. 2.76

$\alpha_{\Omega'}^{\Omega m} (r), \beta_{\Omega'}^{\Omega m} (r)$ Expansion coefficients for absorptive regime TP polariton operator in terms of medium field operators; see Eq. 2.76

$Z_{\Omega m}$ Resonant oscillator amplitude; See Eq. 2.84

$Z$ Resonant oscillator amplitude, considered as independent variable

$\gamma_{n}^{\Omega m}$ See Eq. 2.85

$\gamma_{\Omega'}^{\Omega m} (r)$ See Eq. 2.87

$\alpha_{n}^{m}, \beta_{n}^{m}$ Expansion coefficients for transparent regime TP polariton operator in terms of nominal electromagnetic mode operators; see Eq. 2.94

$\alpha_{\Omega'}^{m} (r), \beta_{\Omega'}^{m} (r)$ Expansion coefficients for transparent regime TP polariton operator in terms of medium field operators; see Eq. 2.94

$\gamma_{n}^{m}$ See Eq. 2.102

$\tilde{D}_{m} (r), \tilde{B}_{m} (r)$ Dispersive mode fields

$\tilde{\omega}_{m}$ Frequency of dispersive mode

$\Gamma' (r, \tilde{\omega}_{m})$ Material dispersion tensor

$\Gamma (r, \Omega; Z)$ Effective dielectric response function

$P_{T} (r, t)$ Transverse part of polarization field

$P_{L} (r, t)$ Longitudinal part of polarization field

$P_{m} (r)$ Transparent regime polarization mode field

$\tilde{P}_{\Omega m} (r)$ Transverse part of absorptive regime polarization mode field

$\tilde{P}_{\Omega m}^{(L)} (r)$ Longitudinal part of absorptive regime polarization mode field

$E_{T} (r, t)$ Transverse part of electric field

$E_{L} (r, t)$ Longitudinal part of electric field

$\tilde{E}_{m} (r)$ Transparent regime electric mode field

continued overleaf...
\[
\begin{align*}
\tilde{E}_{\Omega m} & \quad \text{Transverse part of absorptive regime electric mode field} \\
\tilde{E}_{\Omega m}^{(LP)} (r) & \quad \text{Longitudinal part of absorptive regime electric mode field} \\
D_\infty & \quad \text{Projection of } V \text{ onto its dimensions of infinite extent} \\
F & \quad \text{Projection of } V \text{ onto its dimensions of finite extent} \\
C_\infty & \quad \text{See Eq. 2.145} \\
\delta^d (k) & \quad d\text{-dimensional Dirac delta function} \\
\varepsilon (r, \omega) & \quad \text{Dielectric function} \\
\varepsilon (r, \omega; Z) & \quad \text{Effective dielectric function} \\
\tilde{d}_{\Omega k} (r), \tilde{b}_{\Omega k} (r) & \quad \text{Periodic part of Bloch mode field in transparent regime} \\
\tilde{d}_{\Omega m} (r), \tilde{b}_{\Omega m} (r) & \quad \text{Periodic part of Bloch mode field in absorptive regime} \\
V_{\omega m k} (t) & \quad \text{See Eq. 2.172}
\end{align*}
\]

Table A.2: Symbols first used in Chapter 3

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>(H_{\text{sys}})</td>
<td>System Hamiltonian excluding driving field</td>
</tr>
<tr>
<td>(H_{\text{drive}})</td>
<td>Driving Hamiltonian</td>
</tr>
<tr>
<td>(P_s (r, t))</td>
<td>Source polarization field associated with (H_{\text{drive}})</td>
</tr>
<tr>
<td>(P_{\text{extra}} (r, t))</td>
<td>Extra polarization beyond that in undriven Maxwell equations</td>
</tr>
<tr>
<td>(\Omega)</td>
<td>Frequency, typically in the absorptive regime</td>
</tr>
<tr>
<td>(T (r, \tilde{\omega}_m))</td>
<td>Normalization factor isotropic tensor</td>
</tr>
<tr>
<td>(v_p (r, \omega))</td>
<td>Local phase velocity</td>
</tr>
<tr>
<td>(v_g (r, \omega))</td>
<td>Local group velocity</td>
</tr>
<tr>
<td>(\Omega_m (Z))</td>
<td>Frequency dependent on the resonant oscillator amplitude</td>
</tr>
<tr>
<td>(\overline{\Omega}_m)</td>
<td>Central frequency of continuum band (m); see Eq. 3.21</td>
</tr>
<tr>
<td>(\overline{\Omega})</td>
<td>Central frequency of continuum band; label (m) left implicit</td>
</tr>
<tr>
<td>(L_{\Omega m})</td>
<td>See Eq. 3.27</td>
</tr>
<tr>
<td>(\Omega_m^{(i)})</td>
<td>Pole number (i) of (L_{\Omega m})</td>
</tr>
<tr>
<td>(C_m)</td>
<td>Normalization constant for definition of (L_{\Omega m}) in Eq. 3.27</td>
</tr>
<tr>
<td>(D_{\Omega m} (r), B_{\Omega m} (r))</td>
<td>Mode fields rescaled for use in propagation; see Eq. 3.28</td>
</tr>
<tr>
<td>(E_{\Omega m} (r))</td>
<td>Electric mode field rescaled for use in propagation; see Eq. 3.28</td>
</tr>
</tbody>
</table>

continued overleaf...
Appendix A. List of Symbols

ℜ  Real line
D  Reciprocal space unit cell, e.g. first Brillouin zone
|Φ⟩  Arbitrary absorptive regime one-polariton state
b† (A, t)  Creation operator for |Φ⟩, above
|0⟩  Vacuum state
A_{αm}  Expansion coefficient for b† (A, t), above
B† (λ, A, t)  Coherent polariton state creation operator
τ_s  Earliest turn-on time for driving polarization field
α  1) When used as an index, this is a generic mode label: α = m and α = (Ω, m) in the transparent and absorptive regimes respectively
2) When not used as an index, α is the Beer coefficient
c^0_α (t)  Fluctuating part of polariton operator
c^0_m (t)  Transparent regime polariton fluctuation operator
c^0_{1m} (t)  Absorptive regime polariton fluctuation operator
 τ_α (t)  Expectation part of polariton operator
ω_α  Refers to Ω in absorptive regime or ˜ω_m in transparent regime
g_u (r, t)  Polariton effective field (either transparent or absorptive regime)
^k  Central wave vector around which g_u (r, t) is built
^Π (r − r′)  See Eq. 3.52
^ω^{(1)i}_{uk}  Transparent regime group velocity; see Eq. 3.55
^ω^{(2)ij}_{uk}  Transparent regime group velocity dispersion; see Eq. 3.56
c_m (t)  Composite polariton operator (absorptive regime only)
φ_{Ωm}  See Eq. 3.61
ν_m  See Eq. 3.62
τ_m, c^0_m  Respectively, expected and fluctuating parts of composite polariton operator
D (r, t), D^0 (r, t)  Respective expected and fluctuating parts of displacement field
τ_{αk} (t)  Non-fluctuating part of composite polariton operator, made slow-varying
f_u (r, t)  Non-fluctuation part of polariton effective field (absorptive regime only)
γ_{αk} (Ω)  See Eq. 3.75

continued overleaf...
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma^{(n)}_{u k} (\Omega)$</td>
<td>Taylor coefficient in wave vector expansion of $\gamma_{u k} (\Omega)$; See Eq. 3.76</td>
</tr>
<tr>
<td>$Y^{(n)}_u$</td>
<td>See Eq. 3.78</td>
</tr>
<tr>
<td>$\bar{f} (\mathbf{r}, t)$</td>
<td>See Eq. 3.81</td>
</tr>
<tr>
<td>$\eta$</td>
<td>Small parameter</td>
</tr>
<tr>
<td>$G$</td>
<td>Multiple scales function corresponding to $\bar{g}_u (\mathbf{r}, t)$</td>
</tr>
<tr>
<td>$d$</td>
<td>Multiple scales fundamental distance scale</td>
</tr>
<tr>
<td>$\tau_{cw}$</td>
<td>Multiple scales fundamental time scale</td>
</tr>
<tr>
<td>$\tilde{C}^{(n,l)}_m$</td>
<td>Multiple scales coefficient</td>
</tr>
<tr>
<td>$\tau_{de}$</td>
<td>Decay time</td>
</tr>
<tr>
<td>$\tau_{pr}$</td>
<td>Propagation time</td>
</tr>
<tr>
<td>$\mathcal{O}(x)$</td>
<td>Big-O notation</td>
</tr>
<tr>
<td>$w^{(n)}_{u k}$</td>
<td>Propagation parameter $n$; see Eqs. 3.92, 3.93, 3.94</td>
</tr>
<tr>
<td>$C^{(n,l)}_m$</td>
<td>See Eq. 3.95</td>
</tr>
<tr>
<td>$Z'<em>{u k}, Z''</em>{u k}$</td>
<td>Frequency derivatives of resonant oscillator amplitude evaluated at band center</td>
</tr>
<tr>
<td>$\Omega_{\pm}^{\pm}_{u k}$</td>
<td>See Eq. 3.100</td>
</tr>
<tr>
<td>$\Omega_{u k}$</td>
<td>$Z$ Derivative of frequency at band center where $Z = 0$</td>
</tr>
<tr>
<td>$k (\omega, Z)$</td>
<td>See Eq. 3.107</td>
</tr>
<tr>
<td>$d_1, d_2$</td>
<td>Thicknesses of medium 1 and 2</td>
</tr>
<tr>
<td>$a$</td>
<td>Unit cell spatial size</td>
</tr>
<tr>
<td>$k_1 (\omega, Z)$</td>
<td>Local wave vector in medium 1; see Eq. 3.108</td>
</tr>
<tr>
<td>$\kappa_2 (\omega, Z)$</td>
<td>$i \times$ local wave vector in medium 2; see Eq. 3.109</td>
</tr>
<tr>
<td>$\varepsilon_{1,2} (\omega, Z)$</td>
<td>Effective dielectric function in medium 1,2</td>
</tr>
<tr>
<td>$V_{cell}$</td>
<td>Unit cell spatial volume</td>
</tr>
<tr>
<td>$\varepsilon_d$</td>
<td>Drude model dielectric function</td>
</tr>
<tr>
<td>$\tilde{v}_p (\mathbf{r}, \Omega)$</td>
<td>Effective local phase velocity; see after Eq. 3.114</td>
</tr>
<tr>
<td>$\tilde{v}_g (\mathbf{r}, \Omega)$</td>
<td>Effective local group velocity; see after Eq. 3.114</td>
</tr>
<tr>
<td>$\tilde{\varepsilon}^{-1} (\mathbf{r}, \Omega)$</td>
<td>Effective local dielectric function; see after Eq. 3.115</td>
</tr>
<tr>
<td>$T_0$</td>
<td>Nondispersion part of normalization integral</td>
</tr>
<tr>
<td>$T_D$</td>
<td>Dispersion part of normalization integral</td>
</tr>
</tbody>
</table>
Appendix A. List of symbols

Table A.3: Symbols first used in Chapter 4

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_L$</td>
<td>Linear Hamiltonian</td>
</tr>
<tr>
<td>$H_{NL}$</td>
<td>Nonlinear Hamiltonian</td>
</tr>
<tr>
<td>$\Gamma_{ijl}^2 (r)$</td>
<td>Second-order nonlinear response to $D$ field</td>
</tr>
<tr>
<td>$P_2^r (r, t)$</td>
<td>Nonlinear polarization</td>
</tr>
<tr>
<td>$\chi_{ijl}^2 (r)$</td>
<td>Second-order nonlinear response to $E$ field</td>
</tr>
<tr>
<td>$\hat{e}_{uk}$</td>
<td>Mode polarization unit vector</td>
</tr>
<tr>
<td>$\zeta_{uk}$</td>
<td>Normalization factor; see Eq. 4.12</td>
</tr>
<tr>
<td>$\zeta_P$</td>
<td>Normalization factor of pump carrier wave</td>
</tr>
<tr>
<td>$n_u' (\omega)$</td>
<td>Frequency derivative of principal axis index component $u$</td>
</tr>
<tr>
<td>$k_u$</td>
<td>Wave vector considered as a function of frequency for mode $u$</td>
</tr>
<tr>
<td>$u = {o, e}$</td>
<td>Respectively ordinary- and extraordinary-wave mode label</td>
</tr>
<tr>
<td>$n_o (\omega)$</td>
<td>Ordinary index of refraction</td>
</tr>
<tr>
<td>$n_e (\omega)$</td>
<td>Extraordinary index of refraction</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Angle between wave vector and optic axis</td>
</tr>
<tr>
<td>$\hat{q}$</td>
<td>Optic axis of the nonlinear crystal</td>
</tr>
<tr>
<td>$n_e (\omega; \theta)$</td>
<td>Extraordinary index for $k$ at arbitrary angle</td>
</tr>
<tr>
<td>$U (t', t)$</td>
<td>Interaction picture propagator</td>
</tr>
<tr>
<td>$U_S (t', t)$</td>
<td>Schrödinger picture propagator</td>
</tr>
<tr>
<td>$t_0, t_1$</td>
<td>Points in time before and respectively after $t = 0$</td>
</tr>
<tr>
<td>$</td>
<td>\psi_{in}\rangle$</td>
</tr>
<tr>
<td>$</td>
<td>\psi_{out}\rangle$</td>
</tr>
<tr>
<td>$O$</td>
<td>Arbitrary Schrödinger picture operator</td>
</tr>
<tr>
<td>$\hat{O} (t)$</td>
<td>See Eq. 4.28</td>
</tr>
<tr>
<td>$\hat{H}_{NL} (t)$</td>
<td>See Eq. 4.31</td>
</tr>
<tr>
<td>${x, y, z}$</td>
<td>Lab frame</td>
</tr>
<tr>
<td>${x', y', z'}$</td>
<td>Crystal frame</td>
</tr>
<tr>
<td>$L$</td>
<td>Crystal length</td>
</tr>
<tr>
<td>$v_P$</td>
<td>$z$-component of pump group velocity</td>
</tr>
<tr>
<td>$R_D$</td>
<td>Magnitude of ratio of the group velocity dispersion to the group velocity, all taken in the $z$-direction</td>
</tr>
</tbody>
</table>

continued overleaf...
\( \hat{e}_s \)  
Unit vector in the \( s \) direction

\( \omega_p \)  
Pump carrier frequency

\( \mathbf{k}_p \)  
Pump carrier wave vector

\( \kappa_z \)  
Pump spatial band width in \( z \)-direction

\( \kappa_\perp \)  
Pump spatial band width in the perpendicular directions \( x, y \)

\( A^\dagger_p \)  
Pump single-polariton creation operator

\( \alpha_p \)  
\( |\alpha_p|^2 \) is expected number of pump polaritons

\( \phi_{\mathbf{k}_p} \)  
Pump Gaussian beam profile

\( \mathcal{A}_p (t_0) \)  
See Eq. 4.45

\( S_{\text{ooe}} (\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}; t) \)  
See Eq. 4.47

\( \mathbf{k} \)  
Wave vector for pump

\( \mathbf{k}_1, \mathbf{k}_2 \)  
Wave vectors of down-converted polaritons

\( \epsilon_{\mathbf{k}_1} \)  
Schrödinger picture polariton operator

\( \mathfrak{S}_{\mathbf{k}_1} (t) \)  
Polariton operator with time dependence of Eq. 4.28

\( A^\dagger_\mathbf{f} \)  
Single bipolariton creation operator

\( \phi_{\mathbf{oo}} (\mathbf{k}_1, \mathbf{k}_2) \)  
Bipolariton wave function

\( \xi \)  
Bipolariton wave function normalization factor; also the quantum efficiency of type-I SPDC

\( \mathbf{v}_{P\perp} \)  
Transverse part of pump group velocity

\( s_{\text{disp}} \)  
See Eq. 4.43

\( \hat{\mathbf{e}}_p \)  
Polarization of pump carrier wave

\( \mathbf{b}_{\mathbf{k}_1, \mathbf{k}_2} \)  
See Eq. 4.63

\( \Theta (x) \)  
Heaviside function

\( \Upsilon (\mathbf{k}_1, \mathbf{k}_2) \)  
See Eq. 4.66

\( I \)  
Generic integral over bipolariton wave function density; see 4.69

\( h (\mathbf{k}_1) \)  
Arbitrary weighting function; see generic integral 4.69

\( \mathbf{k} = (k_\rho, \phi, k_z) \)  
Cylindrical coordinate system

\( \omega_o(k_\rho, \phi, k_z), \omega_o(k_\rho, k_z) \)  
k-dependence respectively with and without dependence on polar angle

\( \mathbf{K} = (k_{1\rho}, k_{1z}, k_{2z}) \)  
See before Eq. 4.76

\( \Upsilon (\mathbf{K}) \)  
See Eq. 4.76

\( \mathcal{S}_\gamma \)  
2D surface in \( \mathbf{K} \)-space on which \( \Upsilon (\mathbf{K}) \) vanishes

\( V (k_{1\rho}, k_{1z}) \)  
3-wave group velocity mismatch

continued overleaf...
Appendix A. List of Symbols

\[ v_o (k_{1\rho}, k_{1z}) \] \hspace{1cm} Ordinary wave group velocity

\[ v_{oz} (k_{1\rho}, k_{1z}) \] \hspace{1cm} \( z \)-component of ordinary wave group velocity

\[ v_{\rho \rho} (k_{1\rho}, k_{1z}) \] \hspace{1cm} Magnitude of transverse component of o-wave group velocity

\[ C \] \hspace{1cm} 1D phase- and frequency-matching curve in \((k_{1\rho}, k_{1z})\)-space, defined by Eq. 4.82

\[ \varphi \] \hspace{1cm} The interval \([0, 2\pi)\) corresponding to possible values of the polar angle \( \phi \)

\[ S \] \hspace{1cm} 2D phase- and frequency-matching curve in \( k_1 \)-space; \( S \equiv C \otimes \varphi \)

\[ P(C) \] \hspace{1cm} Projection of \( C \) onto the \( k_{1z} \) axis

\[ k_{1\rho} (k_{1z}) \] \hspace{1cm} \( k_{1\rho} \) as a function of \( k_{1z} \) for points on \( C \)

\[ N_{ok_1} \] \hspace{1cm} Polariton number operator for given wave vector

\[ N_o \] \hspace{1cm} Total down-converted polariton number

\[ P_o \] \hspace{1cm} Projector onto o-wave subspace

\[ \langle N_{ok_1} \rangle \] \hspace{1cm} Coarse-grained number operator expectation

\[ \Lambda (k_1) \] \hspace{1cm} Smoothing volume for \( \langle N_{ok_1} \rangle \)

\[ \mathcal{A}(k_1) \] \hspace{1cm} Smoothing area on \( S \) for \( \langle N_{ok_1} \rangle \)

\[ \Lambda_\phi \] \hspace{1cm} Angular direction width of \( \Lambda (k_1) \)

\[ \Lambda_z \] \hspace{1cm} \( z \)-direction width of \( \Lambda (k_1) \)

\[ E_{SPDC} \] \hspace{1cm} Expected energy of SPDC-generated fields

\[ F_D (k_{1z}, \omega_P) \] \hspace{1cm} Factor encapsulating effect on SPDC efficiency of field normalization including dispersion, as a function of down-converted polariton wave vector

\[ \overline{F_D} (k_{1z}, \omega_P) \] \hspace{1cm} \( F_D (k_{1z}, \omega_P) \) after neglecting terms containing \( n'_u(\omega) \)

\[ k_{DC} \] \hspace{1cm} Typical amplitude of down-converted polariton wave vector

\[ \omega_{DC} \] \hspace{1cm} Typical frequency of down-converted polariton

\[ \lambda_{DC} \] \hspace{1cm} Typical wavelength of down-converted polariton

\[ v_w \] \hspace{1cm} Typical value of the 3-wave group velocity mismatch \( V (k_{1\rho}, k_{1z}) \)

\[ t_{tr} \] \hspace{1cm} Transit time of the pump pulse through the crystal

\[ E_{fl,DC} \] \hspace{1cm} Fluctuation energy: half the energy of a polariton at frequency \( \omega_{DC} \)

\[ k_{1\rho,typ} \] \hspace{1cm} Typical transverse component of down-converted polariton wave vector

\[ E_P \] \hspace{1cm} Pump energy

continued overleaf...
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Appendix B

Canonical commutators for the polaritons

Here we impose canonical commutation relations on the polariton operators, and obtain normalization conditions on the photonic component mode fields.

B.0.1 Polaritons in the absorptive regime

Making use of the expansion (2.76) in the commutation relation (2.66), we obtain

\[
\left[ \alpha_\Omega m, \alpha_{\Omega'}^{\dagger} m' \right] = \sum_n \left( \alpha_\Omega m \alpha_{\Omega'} m' - \beta_\Omega m \beta_{\Omega'} m' \right) + \int_{\Omega_s} d\Omega'' \int_{\mathcal{V}_{\Omega''}} d\mathbf{r} \alpha_{\Omega''}^{\dagger} \mathbf{r} \cdot \alpha_{\Omega'} \mathbf{r} - \beta_{\Omega''} \mathbf{r} \cdot \beta_{\Omega'} \mathbf{r}. \tag{B.1}
\]

Substitution of (2.83) and (2.85) in (B.1) yields a useful intermediate result for the electromagnetic part:

\[
\sum_n \left( \alpha_\Omega m \alpha_{\Omega'}^{\dagger} m' - \beta_\Omega m \beta_{\Omega'}^{\dagger} m' \right) = \sum_{n,n'} \gamma_{n'n'} \left( \frac{h\Omega + h\Omega'}{2\hbar \omega_{n'}} \right) \delta_{n'n} \gamma_n^{\Omega m}. \tag{B.2}
\]

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Substitution of (2.84) and (2.86) into (B.1) gives
\[
\int_{\Omega_0}^{\infty} d\Omega' \int_{V_{\Omega'}} dr \, \mathbf{a}^{\Omega m}_{\Omega'} (r) \cdot \mathbf{a}^{\Omega' m' \dagger}_{\Omega'} (r) - \mathbf{a}^{\Omega m}_{\Omega'} (r) \cdot \mathbf{b}^{\Omega' m' \dagger}_{\Omega'} (r) \tag{B.3}
\]
\[
= \int_{\Omega_0}^{\infty} d\Omega' \int_{V_{\Omega'}} dr \sum_{n,n'} \gamma_{n' m' \dagger}^{\Omega' \Omega} (r, \Omega') \left[ \left( P \frac{1}{\hbar \Omega' - \hbar \Omega''} + Z_{\Omega m} \delta (\hbar \Omega' - \hbar \Omega'') \right) \right.
\]
\[
\left. \left( P \frac{1}{\hbar \Omega' - \hbar \Omega''} + Z_{\Omega' m'} \delta (\hbar \Omega' - \hbar \Omega'') \right) - \frac{1}{\hbar \Omega' - \hbar \Omega''} \frac{1}{\hbar \Omega' + \hbar \Omega''} \right] \cdot \mathbf{A}_n (r, \Omega') \gamma_{n m}^{\Omega m}.
\tag{B.4}
\]
Care must be taken in expanding the factors inside the square brackets. Combining the two principal parts involves an extra contribution from the double pole for \( \Omega = \Omega' \). This is resolved with the identity [98]
\[
\left( \frac{P}{x' - y} \right) \left( \frac{P}{x - y} \right) = \left( \frac{P}{x' - x} \right) \left( \frac{P}{x - y} - \frac{P}{x' - y} \right) + \pi^2 \delta (x' - y) \delta (x - y). \tag{B.5}
\]
Combination of the two delta functions requires use of the identity
\[
\delta (x - y) \delta (x' - y) = \delta (x - x') \delta \left( y - \frac{1}{2} (x + x') \right). \tag{B.6}
\]
The use of (B.5), (B.6) and more straightforward identities simplifies (B.3); combining this result with (B.2) in (B.1) leads to
\[
\left[ c_{\Omega m}, c_{\Omega' m'} \dagger \right] = \sum_{n,n'} \gamma_{n' m' \dagger}^{\Omega' \Omega} \left\{ \left[ \frac{(\hbar \Omega)^2}{2 \hbar \omega_n^\Omega} \delta_{n'n} + (\Delta \text{Re} \Gamma)^\Omega_{nn'} - \frac{Z_{\Omega m}^{\Omega m}}{\pi} (\text{Im} \Gamma)^\Omega_{nn'} \right. \right.
\]
\[
\left. \left. - \left[ \frac{(\hbar \Omega)^2}{2 \hbar \omega_n^\Omega} \delta_{n'n} + (\Delta \text{Re} \Gamma)^\Omega_{nn'} - \frac{Z_{\Omega m}^{\Omega m'}}{\pi} (\text{Im} \Gamma)^\Omega_{nn'} \right] \right) \right\} \gamma_{n m}^{\Omega m}.
\tag{B.7}
\]
Application of the eigenvalue equation (2.90) twice gives
\[
\left[ c_{\Omega m}, c_{\Omega' m'} \dagger \right] = \delta (\hbar \Omega - \hbar \Omega') \left( \frac{\pi^2 + Z_{\Omega m} Z_{\Omega m'}}{\pi} \right) \sum_{n,n'} \gamma_{n' m' \dagger}^{\Omega' \Omega} (\text{Im} \Gamma)^\Omega_{nn'} \gamma_{n m}^{\Omega m}. \tag{B.8}
\]
The summand can be simplified by using the orthogonality condition for the generalized Hermitian eigenvalue problem (2.90), which is
\[
\sum_{n,n'} \gamma_{n' m' \dagger}^{\Omega' \Omega} (\text{Im} \Gamma)^\Omega_{nn'} \gamma_{n m}^{\Omega m} = \xi_{\Omega m} \delta_{mm'}. \tag{B.9}
\]
for some normalization factor $\xi_{\Omega m}$. Consistency of (B.8) with (2.66) then specifies $\xi_{\Omega m}$, so that the canonical commutation relation (2.66) is obtained if the amplitudes are normalized according to (2.93). In terms of the mode fields $\tilde{D}_{\Omega m}(r)$ and $\tilde{B}_{\Omega m}(r)$, defined in (2.118, 2.119), the normalization condition becomes (2.126, 2.127).

We note here that the absorptive regime TP polariton operators commute with the LP polariton operators:

$$\left[ c_{\Omega m}, s_{q' m'}^\dagger \right] = 0. \quad \text{(B.10)}$$

This is determined by substitution of the expansions (2.69) and (2.76) into the left hand side of (B.10), use of the commutation relation (2.17), use of the expression (2.84) for the medium field coefficients in terms of the nominal system coefficients for the TP polaritons, and the longitudinality of the polarization of the LP modes expressed in (2.70). Recall that the expression (2.84) ensures suppression of the homogeneous solution to (2.80).

### B.0.2 Polaritons in the transparent regime

Expanding $c_m$ using (2.94) in the commutation relation (2.65) gives

$$\left[ c_m, c_{m'}^\dagger \right] = T_{mm'}^{em} + T_{mm'}^{med}, \quad \text{(B.11)}$$

where

$$T_{mm'}^{em} \equiv \sum_n \left( \alpha_n^m \alpha_{n'}^{m' \dagger} - \beta_n^m \beta_{n'}^{m' \dagger} \right), \quad \text{(B.12)}$$

$$T_{mm'}^{med} \equiv \int_{\Omega_s} d\Omega'' \int_{\Omega_{\Omega''}} d\Omega' \int_V d^3r \alpha_{\Omega''}^{m'}(r) \cdot \alpha_{\Omega''}^{m' \dagger}(r) - \beta_{\Omega''}^{m'}(r) \cdot \beta_{\Omega''}^{m' \dagger}(r), \quad \text{(B.13)}$$

give the electromagnetic and medium contributions respectively. The electromagnetic contribution can be expressed in terms of the dispersive electromagnetic mode fields as follows: we write the $\alpha_n^m$ and $\beta_n^m$ in terms of $\gamma_n^m$ using (2.102); we then factor the result and make use of the normalization condition for the nominal modes (2.44) and apply the system (2.103) as well, in the form

$$\sum_{n,n'} \gamma_n^{m*} G_{n'n}(\tilde{\omega}_m) \gamma_n^m = 0, \quad \text{(B.14)}$$

where we recall the definition of $G_{n'n}(\tilde{\omega}_m)$ in the text after (2.105). Identifying the dispersive modes defined in (2.107), the result is

$$T_{mm'}^{em} = \frac{1}{\varepsilon_0} \int_V d^3r \tilde{D}_m^{is}(r) \left[ \frac{1}{\hbar \omega_m} (\delta_{ij} - \Gamma_{ij}(r, \tilde{\omega}_m)) + \frac{1}{\hbar \omega_{m'}'} (\delta_{ij} - \Gamma_{ij}(r, \tilde{\omega}_{m'}')) \right] \tilde{D}_{m'}(r). \quad \text{(B.15)}$$
The medium contribution to the commutator in (B.11) is related to the electromagnetic contribution by substituting (2.101) into (B.13), yielding

\[ T_{mn}^{\text{med}} = \int_{\Omega''}^{\infty} d\Omega'' \int_{V_{\Omega''}} dr \sum_{n,n'} \gamma_{n'}^{m'} \Lambda_{n'}^{*} (r, \Omega'') \left[ \frac{1}{\hbar \omega_{n} - \hbar \Omega''} - \frac{1}{\hbar \omega_{m} + \hbar \Omega''} \right] \cdot \Lambda_{n} (r, \Omega'') \gamma_{n}^{m} \tag{B.16} \]

For \( m \neq m' \), the rest of the calculation of the commutator (B.11) mirrors that for the continuous polariton spectrum; the electromagnetic and medium contributions are combined, and the system (2.103) is identified twice, yielding\(^1\)

\[ [c_{m}, c_{m'}^{\dagger}] = 0; \quad m \neq m'. \tag{B.17} \]

For \( m = m' \), the partial fraction expansion cannot be used due to the double pole. Instead, taking \( m = m' \), (B.16) is simplified by using the frequency derivative of the Kramers-Kronig relation (2.30). Then the medium contribution is found to be

\[ T_{mn}^{\text{med}} = \frac{1}{\varepsilon_{0}} \int_{V} dr \tilde{D}_{m}^{*} (r) \cdot \frac{1}{\hbar} \Gamma' (r, \tilde{\omega}_{m}) \cdot \tilde{D}_{m} (r), \tag{B.18} \]

where we recall the definition of the material dispersion \( \Gamma' (r, \tilde{\omega}_{m}) \) in (2.115). With the requirement (2.65), Equation (B.11), along with Eqs. (B.18) and (B.15), gives the normalization conditions for the electromagnetic mode fields as (2.113, 2.114). The normalization condition (2.114) for \( \tilde{B}_{m} (r) \) was obtained from the normalization condition (2.113) for \( \tilde{D}_{m} (r) \) by using (2.38) and (2.39) as well as partial integration. Equation (2.114) can be written in an alternative form that is sometimes easier to use:

\[ \frac{1}{\mu_{0}} \int_{V} dr \tilde{B}_{m}^{*} (r) \cdot \tilde{B}_{m} (r) = \frac{1}{\varepsilon_{0}} \int_{V} dr \tilde{D}_{m}^{*} (r) \cdot [1 - \Gamma (r, \tilde{\omega}_{m})] \cdot \tilde{D}_{m} (r). \tag{B.19} \]

We note that the transparent regime TP polariton operators commute with the LP polariton operators, as stated in (2.106). This is determined by following the same line of reasoning as leads to (B.10).

\(^1\)A similar technique can be used to verify that all commutators between \( c_{m} \) and \( c_{\Omega' m'}^{\dagger} \) vanish.
Appendix C

Consistency conditions

Two useful identities for the transformation coefficients can be obtained by substituting the expressions (2.130) and (2.131) into the commutation relations (2.47) and (2.17). Respectively, these give

$$\delta_{nn'} = \sum_m \int d\Omega \left( \alpha_n^{\Omega m *} \alpha_n^{\Omega m} - \beta_n^{\Omega m * \beta_n^{\Omega m}} \right) + \sum_m (\alpha_n^{m *} \alpha_n^m - \beta_n^m \beta_n^m), \quad (C.1)$$

and

$$\delta_{ij} \delta (\Omega - \Omega') \delta (r - r') = \sum_m \left\{ \int_{\Omega_0}^\infty d\Omega'' \alpha_{\Omega''}^{\Omega'' m*} (r) \alpha_{\Omega'' j}^{\Omega'' m} (r') - \beta_{\Omega''}^{\Omega'' m*} (r) \beta_{\Omega'' j}^{\Omega'' m} (r') \right\} \quad (C.2)$$

$$+ \alpha_{\Omega i}^m (r) \alpha_{\Omega j}^m (r') - \beta_{\Omega i}^m (r) \beta_{\Omega j}^m (r')$$

$$+ \rho_{\Omega i}^{m*} (r) \rho_{\Omega j}^{m} (r') \right\},$$

where here we refer to components of $\alpha_{\Omega i}^{\Omega m} (r)$ by writing, e.g., $\alpha_{\Omega i}^{\Omega m} (r)$, and similarly for $\beta_{\Omega i}^{\Omega m} (r)$. 
Appendix D

Field generation issues

In this appendix we consider systems with modes labeled \((u, k)\) for continuous \(k\), and establish three things. First, we show that there exists an infinite set of \(P_s(r, t)\) to generate any desired initial condition \(\tau_{uk}(0)\) in the transparent regime, and similarly to generate any desired initial \(\bar{\tau}_{uk}(0)\) in the absorptive regime; in doing so we identify in terms of a simple expression a non-exhaustive set of specific initial conditions \(\gamma_{\Omega uk}(0)\) that can be generated via \(P_s(r, t)\). Second, we establish that an infinite set of \(P_s(r, t)\) exist for the generation of any initial effective field \(g_u(r,0)\) in the transparent or absorptive regime. Third, we establish that the quantity \(\bar{f}(r, t)\) that appears in Eq. (3.80) vanishes for \(t \geq 0\), so that for nonnegative times the RHS of Eq. (3.91) is indeed zero.

We begin with the absorptive regime, and consider a non-exhaustive parameterization of the set of possible driving polarizations \(P_s(r, \Omega)\) (q.v. Eq. (3.47)). The idea is to define a set that is readily analyzed and yet is sufficiently comprehensive to be able to generate any \(\tau_{uk}(0)\). For this set of \(P_s(r, \Omega)\) we take the form

\[
P_s(r, \Omega) = \sum_u \int d\Omega A_{\Omega uk} \text{Im} \Gamma(r, \Omega) \cdot D_{\Omega uk}(r),
\]

where \(\Omega > 0\), and for \(\Omega < 0\) we take \(P_s(r, \Omega) = P_s(r, -\Omega)^*\), so that \(P_s(r, t)\) is real. In Eq. (D.1) the \(A_{\Omega uk}\) are the coefficients of the parameterization, which have Fourier transforms \(A_{uk}(t)\). We limit ourselves to those \(A_{\Omega uk}\) that satisfy three additional conditions which we note must be independently satisfied for each \((u, k)\): (1) \(A_{\Omega uk}\) is sufficiently differentiable in \(\Omega\), and negligible outside some \((u, k)\)-dependent frequency domain \(D_{uk}^A\) containing \(\Omega_{uk}\); (2) \(A_{uk}(t)\) vanishes for times \(t \geq 0\), except for perhaps a negligible tail extending into the \(t \geq 0\) region; (3) \(D_{uk}^A\) is small enough that \(\text{Im} \Gamma(r, \Omega) \cdot D_{\Omega uk}(r)\) is well-described by a truncated Taylor series in frequency on \(D_{uk}^A\).
The restriction to the neighborhood of $\Omega_{\Omega k}$ in Condition (1) ensures sufficient electromagnetic excitation. In what follows we show that Condition (2) ensures that $P_s(r, t)$ subsides before $t = 0$. Condition (3) is very mild if $\text{Im} \Gamma (r, \Omega)$ is not too rapidly varying over the bandwidth of $A_{\Omega k}$, in light of the phase choice made for $D_{\Omega k}(r)$ after Eq. (3.28).

We then impose these three conditions on Eq. (D.1) and derive the set of $c_{\Omega k}$ that correspond to the set of $P_s(r, \Omega)$ we consider here. Comparing Eq. (3.46) with Eq. (3.47), we have for $t \geq -\tau_s$ that

$$c_{\Omega k}(t) = -\frac{i}{\hbar} e^{-i\Omega t} \int dr \tilde{D}_{\Omega k}(r) \cdot P_s(r, \Omega) + \zeta, \quad (D.2)$$

where

$$\zeta \equiv \frac{1}{\hbar} e^{-i\Omega t} \int_0^\infty dt' e^{+i\Omega t'} \int dr \tilde{D}_{\Omega k}(r) \cdot P_s(r, t'). \quad (D.3)$$

Eq. (D.2) is simplified by substituting in Eq. (D.1). Making use of Eq. (3.28), Eq. (3.25), and Eq. (3.30), we obtain

$$c_{\Omega k}(t) = i \hbar^{-1/2} L_{\Omega k}^* A_{\Omega k} e^{-i\Omega t} + \zeta. \quad (D.4)$$

Next we show $\zeta$ to be negligible at any time after $P_s(r, t)$ becomes negligible. We begin by taking the Fourier transform of (D.1), which yields

$$P_s(r, t) = \sum_u \int d\Omega \int_{\Omega_g} d\Omega \, A_{\Omega k} \text{Im} \Gamma (r, \Omega) \cdot D_{\Omega k}(r) e^{-i\Omega t} + c.c.. \quad (D.5)$$

For each $(u, k)$, and in both the explicitly-written term and in the $c.c.$ term, the frequency integral in Eq. (D.5) is of the form Eq. (F.1) of Appendix F, with $P_s(r, t)$, $A_{\Omega k}$, $\text{Im} \Gamma (r, \Omega) \cdot D_{\Omega k}(r)$, and $D_A^A$ respectively corresponding to the $E(t)$, $A(\Omega)$, $X(\Omega)$ and $D_A$ defined in that appendix. The domain $D_X$ of Appendix F corresponds here to the frequency range over which there is absorption, i.e. that over which $\text{Im} \Gamma (r, \Omega)$ is nonzero, and from Condition (3) it is clear that for all $(u, k)$, the corresponding $D_A$ is contained in $D_X$. It then follows from Eq. (F.13) that $P_s(r, t)$ goes to zero very rapidly for $t \geq 0$. Substituting this result into the equation (D.3) for $\zeta$ it follows trivially that $\zeta$ is negligible for $t \geq 0$. Then we have

$$\tilde{c}_{\Omega k}(t) \simeq i \hbar^{-1/2} L_{\Omega k}^* A_{\Omega k} e^{-i\Omega t}, \quad t \geq 0. \quad (D.6)$$

The correspondence between the set of $A_{\Omega k}$ satisfying Conditions (1-3) and the set of $\tilde{c}_{\Omega k}(0)$ they generate is given by Eq. (D.6) with $t = 0$. 


Next we establish the set of expected composite polariton operators that can be generated this way. Substituting Eq. (D.6) into Eq. (3.60) using Eq. (3.64), we have
\[ \tilde{c}_{\Omega_{0}k}(0) = i\hbar^{-1/2}\nu_{\Omega_{0}k}^{-1/2} \int d\Omega |L_{\Omega_{0}k}|^2 A_{\Omega_{0}k}. \] (D.7)

Observe from Eq. (D.7) that given a desired \( \tilde{c}_{\Omega_{0}k}(0) \), an \( A_{\Omega_{0}k} \) that generates it can be generated from any \( \tilde{A}_{\Omega_{0}k} \) satisfying Conditions (1-3) by using the formula
\[ A_{\Omega_{0}k} = \frac{\tilde{A}_{\Omega_{0}k}\tilde{c}_{\Omega_{0}k}(0)}{i\hbar^{-1/2}\nu_{\Omega_{0}k}^{-1/2} \int d\Omega |L_{\Omega_{0}k}|^2 \tilde{A}_{\Omega_{0}k}}. \] (D.8)

The structure of Eq. (D.8) is such that \( A_{\Omega_{0}k} \) is obtained from \( \tilde{A}_{\Omega_{0}k} \) via separate normalization at each \( k \) of \( \tilde{A}_{\Omega_{0}k} \). Since Conditions (1-3) are satisfied independently at each \( k \), it follows that since \( \tilde{A}_{\Omega_{0}k} \) satisfies Conditions (1-3), so does \( A_{\Omega_{0}k} \). Thus we have shown by construction that any \( \tilde{c}_{\Omega_{0}k}(0) \) can be generated by a \( P_{s} \) of the form (D.1) satisfying Conditions (1-3).

The set of initial absorptive-regime effective fields \( \tilde{g}_{u}(r,0) \) that can be generated by a \( P_{s} \) of the form (D.1) subject to Conditions (1-3) trivially follows from this result. Since \( \tilde{g}_{u}(r,0) \) is proportional to a spatial Fourier transform of \( \tilde{c}_{\Omega_{0}k}(0) \), it follows that given a desired \( \tilde{g}_{u}(r,0) \), the corresponding \( \tilde{c}_{\Omega_{0}k}(0) \) is found via inverse Fourier transformation; then a set of coefficients \( A_{\Omega_{0}k} \) that generates \( \tilde{g}_{u}(r,0) \) is found using Eq. (D.8). Thus it is clear that any smooth function \( \tilde{g}_{u}(r,0) \) can be generated by some \( A_{\Omega_{0}k} \) satisfying (1-3), as long as the \( k \)-space Fourier spectrum of \( \tilde{g}_{u}(r,0) \) has support on \( D \), which is the first Brillouin zone when the system is periodic. This mild limitation is a consequence of the choice to restrict to one band \( u \) the content of any given wave packet. Under a mild approximation we also allow a negligible tail of the spatial Fourier transform of \( \tilde{g}_{u}(r,0) \) to extend outside of \( D \) without any impact on the validity of Eq. (3.91).

It is also clear from Eqs. (D.7, D.8), and from the discussion that follows those equations, that there is in fact an infinite set of \( P_{s}(r,t) \) that can generate any given initial \( \tilde{c}_{\Omega_{0}k}(0) \) or \( \tilde{g}_{u}(r,0) \). This follows because the frequency dependence of \( A_{\Omega_{0}k} \) is integrated out in Eq. (D.7), so that Eq. (D.8) gives a many-to-one mapping from \( A_{\Omega_{0}k} \) to \( \tilde{g}_{u}(r,0) \). This is perhaps unsurprising, since there are many ways to drive even a single simple harmonic oscillator to a given initial point in phase space. However, here there is an additional complication: A given \( \tilde{g}_{u}(r,0) \) corresponds to an infinite set of possible \( \tilde{c}_{\Omega_{0}k}(0) \), so \( \tilde{g}_{u}(r,0) \) (and similarly, \( \tilde{c}_{\Omega_{0}k}(0) \)) actually underdetermines the state of the system. This is a manifestation of the fact that the system is dispersive, so that the time
derivative of any state depends on that state’s entire evolution history. This dependency degenerates for states in the transparent regime, but in the absorptive regime it means that $\mathcal{g}_u (r, t)$ depends on $\mathcal{g}_u (r, t')$ for all previous times $t'$. In differential form this is seen in that Eq. (3.80) is not first order in time in $\mathcal{g}_u (r, t)$, but requires higher temporal derivatives. However, within the multiple scales approximation of Section 3.4, as long as the scaling assumptions (3.87, 3.88) are met and the solution can be taken to be of the form (3.82), the evolution of $\mathcal{g}_u (r, t)$ is given approximately by the initial value problem Eq. (3.91), and additional information about which specific $P_s (r, t)$ is used to formally generate $\mathcal{g}_u (r, 0)$ makes no difference to the dynamics in that approximation.

Finally, as an aid to intuition we note here that to linear order of $Z_{\Omega m}$ in $\Omega$ about $\Omega$, the time-dependent version of Eq. (D.7) gives that $\mathcal{r}_{uk} (t)$ (or similarly, each $k$-component of $\mathcal{g}_u (r, t)$) is a temporal convolution of exponential decay with a source $A_{uk} (t)$.

Next we turn to the transparent regime, for which an analogous argument can be used to demonstrate that both $\mathcal{c}_{uk} (0)$, and the non-fluctuating part of the transparent regime effective field $\mathcal{g}_u (r, 0)$, can be set to any initial value. The driving polarization is taken to be of the form

$$P_s (r, t) = \sum_{u} \int_{D} d\mathbf{k} A_{uk} (t) T (r, \mathbf{\omega}_{uk}) \cdot \mathcal{D}_{uk} (r) + c.c.,$$  \hspace{1cm} (D.9)

for some $A_{uk} (t)$ with a Fourier transform supported on positive frequencies; overloading notation we use $A_{uk} (\omega)$ to denote this Fourier transform of $A_{uk} (t)$. Instead of the Conditions (1-3) of the absorptive regime, states in the set we consider here are taken to satisfy only a single condition that is an analogue of (2): We require that $A_{uk} (t)$ vanishes for times $t \geq 0$, except for perhaps a negligible tail extending into the $t \geq 0$ region.

Using Eq. (D.9) in Eq. (3.47), then making use of Eq. (3.31) we find

$$\mathcal{r}_{uk} (t) = e^{-i\omega_{uk} t} \frac{i\omega_{uk}}{2} A_{uk} (\mathbf{\omega}_{uk}).$$  \hspace{1cm} (D.10)

From Eq. (D.10) it is clearly possible to generate any $\mathcal{r}_{uk} (0)$. Here a given initial condition $\mathcal{r}_{uk} (0)$ can be generated by any of an infinity of possible driving fields $P_s (r, t)$ such that $A_{uk} (\mathbf{\omega}_{uk})$ satisfies Eq. (D.10). Substituting Eq. (D.10) into the definition after Eq. (3.58) for $\mathcal{g}_u (r, t)$ we find

$$\mathcal{g}_u (r, 0) = (2\pi)^{-3/2} \int_{D} d\mathbf{k} \frac{i\mathbf{\omega}_{uk}}{2} A_{uk} (\mathbf{\omega}_{uk}) e^{i(k-\mathbf{\bar{k}}) \cdot \mathbf{r}}.$$  \hspace{1cm} (D.11)

From the Fourier transform of Eq. (D.11) it is seen that it is possible to find some $A_{uk} (\mathbf{\omega}_{uk})$ so as to generate any desired $\mathcal{g}_u (r, 0)$ with $k$-space Fourier spectrum supported
on \( D \). We make a mild approximation and allow a negligible tail of the spatial Fourier transform of \( g_u(r, t) \) outside of \( D \). In contrast with the absorptive regime, here in Eq. (D.11) there is a one-to-one mapping between the \( \tilde{e}_{uk}(0) \) and the \( \tilde{g}_u(r, 0) \) they construct, so that in the transparent regime \( g_u(r, 0) \) uniquely determines the state of the system, even though the system is dispersive.

Up to this point in this appendix we have discussed independently generating absorptive-regime and transparent-regime states, such that a state is generated in one of these two regimes, and the component from the other regime of Hilbert space is left in its vacuum state. We note here that by superposing driving fields it is possible to generate states in both regimes simultaneously.

We close this appendix by showing that \( \tilde{f}(r, t) \) defined in Eq. (3.81) vanishes for all \( t \geq 0 \). To do this we note that

\[
I \equiv \int \frac{d\Omega}{2\pi} L_{\Omega}^* A_{\Omega} e^{-i\Omega t} \quad \text{(D.12)}
\]

can be written using the convolution theorem as

\[
I = \int dt' L_{uk}^* (t - t') A_{uk} (t') \quad \text{(D.13)}
\]

where \( L_{uk}^* (t) \) is the Fourier transform of \( L_{\Omega}^* A_{\Omega} \). The poles of \( L_{\Omega}^* A_{\Omega} \) are in the UHP and \( A_{uk} (t) \) is negligible for \( t \geq 0 \). It then follows from the result of Appendix H that \( I = 0 \).

Finally, substituting (D.6) into Eq. (3.81) and making use of the fact that \( I \) vanishes for \( t \geq 0 \), we obtain

\[
\tilde{f}(r, t) = 0 \text{ for } t \geq 0. \quad \text{(D.14)}
\]

It can now be seen why Eq. (3.27) gives for \( L_{\Omega} A_{\Omega} \) the single good choice for the phase in \( \Omega \): This choice ensures the argument leading to Eq. (D.14) holds, so that Eq. (3.80) is transparently a homogeneous equation for \( t > 0 \).
Appendix E

Approximation of Z as low order polynomial in frequency

In practice, we will only be interested in $Z_{Ωm}$ up to its first one or two derivatives in frequency, with $Z'_m$ and $Z''_m$ calculated either numerically or using the perturbation theory of Appendix G. Accordingly in this appendix we consider $Z_{Ωm}$ to be approximated by a first or second order polynomial in frequency, and obtain the resultant expressions for the various quantities $L_{Ωm}$, $ϕ_{Ωm}$, $C_m$, $ν_m$. In what follows, we append to approximate variables the superscript $(n)$ to label the order of polynomial approximation of $Z_{Ωm}$ to which they correspond. We recall also that $Ω$ is often written in the place of $Ω_m$ for the sake of brevity, and recall from Eqs. (3.97, 3.98) that $Z'_m$ and $Z''_m$ are frequency derivatives of $Z_{Ωm}$ at the band center $Ω$.

E.0.3 Linear approximation for Z

Consider expanding $Z_{Ωm}$ to first order in $Ω$. Since $Z_{Ωm} = 0$, the Taylor expansion truncated at 1st order is

$$Z_{Ωm}^{(1)} = Z'_m (Ω - Ω) .$$  \hspace{1cm} (E.1)

It follows immediately that

$$Ω_{m}^{(1)} = Ω_m - \frac{iπ}{|Z'_m|} ,$$  \hspace{1cm} (E.2)

$$C_{m}^{(1)} = \frac{|\text{Im} Ω_m^{(1)}|}{\sqrt{π}} ,$$  \hspace{1cm} (E.3)

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$$L^{(1)}_{\Omega m} = \left| \Im \Omega^{(1)}_m \right| \frac{1}{\sqrt{\pi}} \frac{1}{\Omega - \Omega^{(1)}_m}.$$  \hfill (E.4)

$$\nu^{(1)}_m = \left| \Im \Omega^{(1)}_m \right|.$$  \hfill (E.5)

$$\phi^{(1)}_{\Omega m} = \sqrt{\left| \Im \Omega^{(1)}_m \right|} \frac{1}{\pi} \frac{1}{\Omega - \Omega^{(1)}_m}.$$  \hfill (E.6)

$$\int_{-\infty}^{\infty} d\Omega \left| \phi^{(1)}_{\Omega m} \right|^2 = 1.$$  \hfill (E.7)

In obtaining Eq. (E.5) from (E.4), we assume that \( \left| \Im \Omega^{(1)}_m \right| \ll \left| \Re \Omega^{(1)}_m - \Omega_q \right| \) so that the lower limit of integration in (3.62) can be extended to \(-\infty\), i.e. \( \nu^{(1)}_m \simeq \int_{-\infty}^{\infty} d\Omega \left| L^{(1)}_{\Omega m} \right|^2 \).

The Schrödinger equation (3.91) can be written simply at this level of approximation. We keep terms up to \( O(\eta) \), and consider times \( t > 0 \). At this order, we obtain the dynamics of Eq. (3.103); using Eqs. (3.92, 3.93, 3.85, 3.77, 3.75, E.4, E.2) the coefficients are found to be given by Eqs. (3.104, 3.105).

Recalling that \( \overline{g}_u (r, t) \) was defined in Eq. (3.73) with a factor of \( \exp (i\Omega_u k t) \) to make it slowly varying, and recalling from Eq. (E.2) that \( \Re \Omega^{(1)}_u = \overline{\Omega}_u k \), we see from Eq. (3.103) that to lowest order, \( \Omega^{(1)}_u k \) is the complex carrier wave frequency which includes decay due to absorption. From the real part of Eq. (3.105) we see that \( \left[ \partial_k \overline{\Omega}_u k \right] \) is the group velocity’s real part. The imaginary part of Eq. (3.105) represents an imaginary part of the group velocity.

E.0.4 Quadratic approximation for $Z$

Now consider expanding \( Z_{\Omega m} \) to second order in \( \Omega \). Since \( Z_{\Omega m} = 0 \), the Taylor expansion truncated at 2nd order is

$$Z_{\Omega m} = Z'_m (\Omega - \overline{\Omega}) + \frac{1}{2} Z''_m (\Omega - \overline{\Omega})^2.$$  \hfill (E.8)

Substituting this into the expression for \( |L_{\Omega m}|^2 \), and factoring the difference of squares yields

$$|L_{\Omega m}|^2 = \frac{\sqrt{\pi}}{i\pi + Z'_m (\Omega - \overline{\Omega}) + \frac{1}{2} Z''_m (\Omega - \overline{\Omega})^2} \times c.c.$$  \hfill (E.9)

Applying the quadratic formula to the denominator and simplifying gives the pole locations \( \Omega^{\pm} \) given in Eq. (3.100). The poles of the c.c. factor of Eq. (E.9) are just the complex conjugates of the poles given by Eq. (3.100). In total \( |L_{\Omega m}|^2 \) then has 4 poles.
Appendix E. Polynomial approximation for \( Z \)

in this approximation: \( \Omega^+, \Omega^-, (\Omega^+)^* \) and \( (\Omega^-)^* \). We take the two poles in the LHP and label them \( \Omega_m^{(1)} \) and \( \Omega_m^{(2)} \), with \( \Omega_m^{(1)} \) chosen to match the root found in the linear approximation in the limit \( Z_m'' \to 0 \).

For a class of systems, typically where dispersion and absorption are not too large, it holds that

\[
|Z_m''| < < |Z_m'|^2.
\]  
(E.10)

Making use of Eq. (E.10) the binomial approximation can be used, and the roots are found to be

\[
\Omega_m^+ \simeq \frac{\Omega_m}{Z_m'} - \frac{i\pi}{Z_m''},
\]  
(E.11)

\[
\Omega_m^- \simeq \left(1 - \frac{2Z_m'}{\Omega_m Z_m''}\right) \Omega_m + \frac{i\pi}{Z_m''}.
\]  
(E.12)

Note that one of these roots is in the UHP and one is in the LHP. From these two roots and the complex conjugate roots it is clear from the prescription after Eq. (3.26) that the correct choice of the \( \Omega_m^{(i)} \) is

\[
\Omega_m^{(1)} \equiv \begin{cases} 
\Omega_m^+; Z_m' > 0, \\
(\Omega_m^+)^*; Z_m' < 0,
\end{cases}
\]  
(E.13)

\[
\Omega_m^{(2)} \equiv \begin{cases} 
(\Omega_m^-)^*; Z_m' > 0, \\
\Omega_m^-; Z_m' < 0.
\end{cases}
\]  
(E.14)

The first root obtained is just \( \Omega_m^{(1)} \), the root obtained in Eq. (E.2) by taking the linear approximation for (E.8). As expected, it is in the LHP. The second root \( \Omega_m^- \) is in the UHP, and its real part differs fractionally by a factor of \( 2Z_m' / (\Omega_m Z_m'') \). It follows that

\[
C_m^{(2)} = \frac{2\sqrt{\pi}}{|Z_m''|},
\]  
(E.15)

\[
L_{\Omega m}^{(2)} = \frac{2\sqrt{\pi}}{|Z_m''|} \frac{1}{(\Omega - \Omega_m^{(1)})(\Omega - \Omega_m^{(2)})}.
\]  
(E.16)

Very often, it holds that \( \Omega_m^{(1)} \) and \( \Omega_m^{(2)} \) are in the right-half plane such that \( |\text{Im} \Omega_m^{(i)}| \ll |\text{Re} \Omega_m^{(i)} - \Omega_g| \) for \( i = 1, 2 \). We assume this to be the case in the calculation of \( \nu_m^{(2)} \). Then the lower limit of integration in (3.62) can be extended to \(-\infty\), i.e. \( \nu_m^{(2)} \simeq \int_{-\infty}^{\infty} d\Omega \left| L_{\Omega m}^{(2)} \right|^2 \). The contour of integration can be closed in either the upper half plane or the lower half.
Appendix E. Polynomial approximation for $Z$

plane. The result is

$$
\nu_m^{(2)} = \left( \frac{2\pi}{Z''_m} \right)^2 \frac{\left| \text{Im} \Omega_m^{(1)} \right| + \left| \text{Im} \Omega_m^{(2)} \right|}{\text{Im} \Omega_m^{(1)} \text{Im} \Omega_m^{(2)} \left| \Omega_m^{(1)} - \Omega_m^{(2)*} \right|^2}.
$$

(E.17)

As a check, we substitute into Eq. (E.17) the Eqs. (E.13,E.14) under the approximation (E.10). In the limit $Z''_m \to 0$ we might think we should recover the expression for $\nu_m^{(1)}$; however we obtain twice $\nu_m^{(1)}$. This is a pathological property of the approximation: While the quadratic $Z\Omega_m$ approximation is more accurate than the linear $Z\Omega_m$ approximation for frequencies near $\Omega_m$, the quadratic approximation introduces a second pole at $\Omega_m^{(2)}$ and therefore a second peak on the real axis at $\text{Re} \Omega_m^{(2)}$. Integrating over frequency picks up the contribution from this pole. This can be remedied by either calculating $\nu_m$ exactly, using $\nu_m^{(1)}$ exclusively, or if the peaks associated with $\Omega_m^{(2)}$ and $\Omega_m^{(1)}$ are well-spaced on the real axis as is typically the case in propagation problems, then the contribution associated with the $\Omega_m^{(2)}$ peak could be removed by a suitable further approximation. We note though that the actual value of $\nu_m$ affects only the fluctuating (superscript 0) fields, and does not affect any physical result relating to the nonfluctuating (overbar) fields.

For completeness, we write

$$
\phi_{\Omega_m}^{(2)} = \frac{L_{\Omega_m}^{(2)}}{\sqrt{\nu_m^{(2)}}},
$$

(E.18)

and note that

$$
\int_{-\infty}^{\infty} d\Omega \left| \phi_{\Omega_m}^{(2)} \right|^2 = 1.
$$

(E.19)
Appendix F

Asymptotic series for convolution integral

In this appendix we demonstrate some key properties of an integral that is ubiquitous in the derivation and application of the results of Ch. 3. The integral is

\[ E(t) = \int_{\mathcal{D}_X} \frac{d\omega}{2\pi} X(\omega) A(\omega) e^{-i\omega t}, \]  

where \( X(\omega) \) and \( A(\omega) \) are the functions with the following properties:

a) \( A(\omega) \) is \( n \)-times differentiable and smooth, and sufficiently negligible everywhere except over a frequency domain \( \mathcal{D}_A \) containing frequency \( \overline{\omega} \);

b) \( X(\omega) \) has compact support on frequency domain \( \mathcal{D}_X \) containing \( \mathcal{D}_A \), and \( X(\omega) \) is \( n \)-times differentiable, smooth, and slowly varying on \( \mathcal{D}_A \) (but not necessarily on \( \mathcal{D}_X \)).

A schematic illustration of typical \( X(\omega) \), \( A(\omega) \), \( \mathcal{D}_X \) and \( \mathcal{D}_A \) is given in Fig. F.1.

We define the truncated Taylor series of order \( n \) as

\[ X_n(\omega) \equiv \sum_{l=0}^{n} \frac{1}{l!} (\omega - \overline{\omega})^l X^{(l)}(\overline{\omega}). \]  

where \( X^{(l)}(\overline{\omega}) \) is the \( l \)th derivative of \( X(\omega) \) at \( \overline{\omega} \). We also define the difference

\[ \Delta_n(\omega) \equiv X(\omega) - X_n(\omega). \]  

Then

\[ E(t) = I(t) + I_{\Delta}(t), \]  

where

\[ I(t) = \int_{\mathcal{D}_X} \frac{d\omega}{2\pi} X_n(\omega) A(\omega) e^{-i\omega t}. \]
and

\[ I_\Delta (t) = \int_{D_A} \frac{d\omega}{2\pi} \Delta_n (\omega) A (\omega) e^{-i\omega t}. \quad (F.6) \]

We first show \( I_\Delta (t) \) to be small:

\[ |I_\Delta (t)| \leq \int_{D_X} \frac{d\omega}{2\pi} \left| \Delta_n (\omega) A (\omega) e^{-i\omega t} \right| \]
\[ \leq W_X \max_\omega |\Delta_n (\omega) A (\omega)|, \quad (F.7) \]

where \( W_X \) is the frequency width of the domain \( D_X \).

Intuitively, as long as \( X (\omega) \) is sufficiently slowly varying on \( D_A \) to be well-approximated by a Taylor series truncated at order \( n \), we have that \( \Delta_n (\omega) \), the error in the \( n \)th order partial Taylor series for \( X (\omega) \), is negligible on \( D_A \). And we are considering an \( A (\omega) \) that is negligible outside of \( D_A \). It follows that the product \( |\Delta_n (\omega) A (\omega)| \) is negligible everywhere, and we have \( I_\Delta (t) \approx 0 \), so that

\[ E (t) \approx I (t). \quad (F.9) \]

To simplify \( I (t) \), we change the variable of integration to \( \omega - \overline{\omega} \). We then notice that \( i\partial_t = \omega \) under the integral, and identify the Fourier transform of \( A (t) \). We obtain

\[ I (t) = e^{-i\overline{\omega}t} \left[ \sum_{j=0}^{n} X^{(j)} (\overline{\omega}) (i\partial_t)^j A (t) \right]. \quad (F.10) \]

Consider for example an \( A (t) \) that is a Gaussian function \( G (t) \). The \( n \)th derivative of a Gaussian is

\[ (\partial_t)^n G (t) = P_n (t) G (t), \quad (F.11) \]
where $P_n(t)$ is a polynomial in $t$ of degree $n$. Using this in (F.10) yields

$$I(t) = e^{-i\omega t} P_n(t) G(t)$$  \hspace{1cm} (F.12)

for some $P_n$. The Gaussian completely dominates the polynomial, so for $t$ large enough $A(t)$ becomes negligible; at worst $I(t)$ (and therefore also $E(t)$) becomes negligible a very short time later.

Of use in Ch. 3 is the special case in which $A(t)$ is negligible for $t \geq 0$. It then follows that

$$E(t) \simeq 0 \text{ for } t \geq 0. \hspace{1cm} (F.13)$$

Another special case of interest is that where $X(\omega)$ and $A(\omega)$ both have Fourier transforms. It then follows from the convolution theorem applied to (F.1) that

$$\int_{-\infty}^{\infty} dt' X(t-t') A(t') = e^{-i\omega t} \left[ \sum_{j=0}^{n} X^{(j)}(\omega) (i\partial_t)^j A(t) \right] + I_\Delta(t)$$  \hspace{1cm} (F.14)

$$\simeq e^{-i\omega t} \left[ \sum_{j=0}^{n} X^{(j)}(\omega) (i\partial_t)^j A(t) \right].$$

since we have shown $I_\Delta(t)$ to be negligible.

Note that it was important that we considered a truncated Taylor expansion and an error term here [125], to avoid the divergences associated with asymptotic series. We also note that results similar to those in this appendix can be obtained when $X(\omega)$ does not have compact support but $X(\omega)$ or $A(\omega)$ drops off sufficiently quickly as $|\omega| \to \infty$. 


Appendix G

Perturbation theory for the resonant oscillator amplitude

Here we give an approach for obtaining the $\Omega$-dependence of $Z_{\Omega m}$ near $\bar{\Omega}$, and specifically calculate an expression for the frequency derivative $Z'_m$ defined in Eq. (3.97). Consider a frequency $\Omega$ shifted a small amount $\Delta$ from the band center $\bar{\Omega}$ of some mode $m$, i.e., $\Omega = \bar{\Omega} + \Delta$. At this shifted frequency the effective response function is

$$
\Gamma \left( r, \bar{\Omega} + \Delta, Z_{\bar{\Omega}+\Delta,m} \right) = \text{Re} \left( \Gamma (r, \bar{\Omega}) \right) + \Delta \left[ \text{Re} \left( \Gamma' (r, \bar{\Omega}) \right) - \frac{Z'_m}{\pi} \text{Im} \left( \Gamma (r, \bar{\Omega}) \right) \right] + \mathcal{O} (\Delta^2). \tag{G.1}
$$

The effective Maxwell master equation of Eqs. 3.19,3.20 can be written as the Hermitian eigenvalue problem

$$(\mathcal{H} + \Delta V) B_{\Omega m} (r) = s_m B_{\Omega m} (r), \tag{G.2}$$

with

$$
\mathcal{H} \equiv \nabla \times \left[ 1 - \text{Re} \left( \Gamma (r, \bar{\Omega}) \right) \right] \cdot \nabla \times , \tag{G.3}
$$

$$
V \equiv \nabla \times \left[ - \text{Re} \left( \Gamma' (r, \bar{\Omega}) \right) + \frac{Z'_m}{\pi} \text{Im} \left( \Gamma (r, \bar{\Omega}) \right) \right] \cdot \nabla \times , \tag{G.4}
$$

$$
s_m \equiv \frac{\Omega^2}{\varepsilon_0}, \tag{G.5}
$$

with $B_{\Omega m} (r)$ being the polariton mode magnetic field normalized analogous to Eq. (3.28) for $D_{\Omega m} (r)$, so that

$$
B_{\Omega m} (r) \equiv \hbar^{-1/2} L_{\Omega m}^{-1} \tilde{B}_{\Omega m} (r), \tag{G.6}
$$

which implies the normalization

$$
\frac{1}{\varepsilon_0} \frac{1}{\Omega^2 \mu_0^2} \int_V d\vec{r} \nabla \times B_{\Omega m}^* (r) \cdot \text{Im} \left( \Gamma (r, \Omega) \right) \cdot \nabla \times B_{\Omega m'} (r) = \delta_{m m'}. \tag{G.7}
$$
Appendix G. Perturbation theory for Z

We introduce for later use “complementary modes” which comprise the full set of eigenmodes of $\mathcal{H}$. Since $\mathcal{H}$ is the effective Maxwell operator for a particular frequency and $Z$ pair $(\Omega, 0)$, we have that the full set of modes of $\mathcal{H}$ are not polariton modes of the system, but are actually an artificial set of modes, except for the distinguished mode $m$ corresponding to frequency $\Omega$ which is the polariton band center mode. We denote complementary modes built from $\Omega_m$ as $\hat{B}_n (r; \Omega_m)$. They satisfy

$$\mathcal{H} \hat{B}_n (r; \Omega_m) = \check{s}_n (\Omega_m) \hat{B}_n (r; \Omega_m)$$ (G.8)

and since $\mathcal{H}$ is Hermitian, as long as they are nondegenerate in eigenvalue $\check{s}_n (\Omega_m)$, we have that these modes are orthogonal with

$$\int_V d\mathbf{r} \hat{B}_n^* (\mathbf{r}; \Omega_m) \cdot \hat{B}_{n'} (\mathbf{r}; \Omega_m) = \nu_{\Omega_m} \delta_{nn'},$$ (G.9)

where we have chosen the normalization constant

$$\nu_{\Omega_m} = \int_V d\mathbf{r} B_{\Omega_m}^* (\mathbf{r}) \cdot B_{\Omega_m} (\mathbf{r}),$$ (G.10)

which ensures that the distinguished complementary mode $m$ is equal to the polariton mode field at the band center:

$$\hat{B}_m (\mathbf{r}; \Omega_m) = B_{\Omega_m} (\mathbf{r}).$$ (G.11)

Next we consider a perturbation expansion of Eq. (G.2). We write the eigenvalue and eigenmode as expansions in $\Delta$, which we write here up to first order in $\Delta$.

$$s_m = \bar{s}_m + \Delta s_m^{(1)},$$ (G.12)

$$B_{\Omega_m} (\mathbf{r}) = B_{\Omega_m}^{(1)} (\mathbf{r}) + \Delta B_{\Omega_m}^{(1)} (\mathbf{r}).$$ (G.13)

Given that $s_m$ is the known function of frequency given in Eq. (G.5), it is trivially determined that $\bar{s}_m = \Omega^2 / c^2$ and $s_m^{(1)} = 2 \Omega / c^2$. We will use this later: our approach is to follow standard perturbation theory to obtain the eigenvalue shift $s_m^{(1)}$ in terms of the potential $V$, but then to use our preknowledge of the value of $s_m^{(1)}$ to solve the inverse problem of finding $V$, which specifically fixes its one parameter $Z_m'$.

At the cost of foregoing normalization of the $B_{\Omega_m} (\mathbf{r})$ fields we insist that the field $B_{\Omega_m}^{(1)} (\mathbf{r})$ is orthogonal to $B_{\Omega_m} (\mathbf{r})$ in Eq. (G.13). Then $B_{\Omega_m}^{(1)} (\mathbf{r})$ can be expanded in the complementary modes according to

$$B_{\Omega_m}^{(1)} (\mathbf{r}) = \sum_{m' \neq m} \theta_{\Omega_m}^m \hat{B}_{m'} (\mathbf{r}; \Omega_m),$$ (G.14)
for some coefficients $\theta_{\Pi m'}$.

Substituting the expansions from Eqs. (G.12,G.13) into Eq. (G.2) and separating into separate an equation at each order in $\Delta$, we have at zeroth order

$$H_{\Omega m}(r) = \mathcal{H}_{\Omega m}(r) \tag{G.15}$$

which is just the Maxwell master equation for the band center polariton mode. At first order in $\Delta$ is

$$H_{\Omega m}^{(1)}(r) + V_{\Omega m}(r) = \mathcal{H}_{\Omega m}^{(1)}(r) + s_m^{(1)}B_{\Omega m}(r). \tag{G.16}$$

Operating with $\int_V d\mathbf{r} \hat{B}_{\Omega m}^*(r; \Omega) \cdot$, and using Eqs. (G.14,G.8,G.9) we obtain the result for the first order eigenvalue shift of Eq. (G.2),

$$s_m^{(1)} = \frac{\int_V d\mathbf{r} B_{\Omega m}(r) \cdot V_{\Omega m}(r)}{\int_V d\mathbf{r} B_{\Omega m}^*(r) \cdot B_{\Pi m}(r)}. \tag{G.17}$$

We next substitute into this that $s_m^{(1)} = \frac{2\Omega}{c^2}$, then use the Maxwell equations $-i\Omega\mu_0 \mathbf{D}_{\Pi m}(r) = \nabla \times \mathbf{B}_{\Pi m}(r)$ and $\mathbf{B}_{\Pi m}(r) = -i \frac{1}{\epsilon_0 c} \nabla \times \left[(1 - \text{Re} \Gamma(r, \Omega)) \cdot \mathbf{D}_{\Pi m}(r)\right]$, use that $\Gamma^{ij} = \Gamma^{ji}$ and integrate by parts. To ensure the result for $Z'_m$ is independent of the normalization of the $D$ fields, we also refrain from using Eq. (3.29) to simplify the result. Rearranging we obtain

$$Z'_m = 2 \frac{\int_V d\mathbf{r} D_{\Pi m}^*(r) \cdot \left(1 - \text{Re} \Gamma(r, \Omega) + \frac{\Omega}{2} \text{Re} \Gamma'(r, \Omega)\right) \cdot \mathbf{D}_{\Pi m}(r)}{\int_V d\mathbf{r} D_{\Pi m}^*(r) \cdot \text{Im} \Gamma(r, \Omega) \cdot \mathbf{D}_{\Pi m}(r)}, \tag{G.18}$$

which is independent of the normalization of $D_{\Pi m}(r)$. Trivially, in periodic media the domains of integration in Eq. (G.18) can be replaced by the unit cell, and the mode fields can be replaced by the periodic-part Bloch mode fields if desired.

The approach taken here can be used to obtain $Z''_m$ as well, by continuing the the above procedure up to second order in $\Delta$. Frequency derivatives of polariton mode fields can also be obtained this way, which can be useful in evaluating equations like Eq. (3.74).
Appendix H

A convolution integral result

Here we prove the following mathematical result: given a convolution integral

\[ j(t) = \int dt' f(t - t') h(t'), \quad (H.1) \]

if the Fourier transform of \( f(t) \) (3.1), denoted \( F(\omega) \), has no poles in the LHP and is well-behaved at infinity, and \( h(t) \) vanishes for \( t \geq 0 \), then \( j(t) \) vanishes for \( t \geq 0 \).

The proof is as follows. Since \( F(\omega) \) has no poles in the LHP, for \( t > 0 \) we can take its Fourier transform to obtain \( f(t) \) via a contour integral that is closed with a semicircle at infinity in a half-plane in which the integrand falls off quickly enough to satisfy Jordan’s lemma. For \( t > 0 \) the LHP is such a half-plane. No poles are enclosed, so the integral on the real axis vanishes, and we have \( f(t) = 0 \) for all \( t > 0 \). And we assume \( F(\omega) \) falls off sufficiently at infinity that for \( t = 0 \) the integral over the LHP semicircle at infinity vanishes, so that similarly we have \( f(t) = 0 \) for \( t = 0 \).

Since we then have that \( f(t) \) vanishes for all \( t \geq 0 \), in (H.1) \( f(t) \) plays the role of a purely acausal response function. Since the “source” \( h(t) \) has support on the negative real axis, it follows that under pure acausal response \( j(t) \) vanishes for \( t \geq 0 \).
Appendix I

Integration over the smoothing volume

This appendix gives a detailed account of the derivation of Eq. (4.91). We begin by substituting Eq. (4.87) into Eq. (4.89) to obtain

$$\langle N_{k_1} \rangle = 2 |\xi|^2 |\alpha_P|^2 \frac{1}{A(k_1)} \int_{\Lambda(k_1)} dk'_1 \int dk_2 |\phi_{oo}(k'_1, k_2)|^2. \quad (I.1)$$

This contains the integral

$$\tilde{I} \equiv \int_{\Lambda(k_1)} dk'_1 \int dk_2 |\phi_{oo}(k'_1, k_2)|^2, \quad (I.2)$$

which is similar to the expression $I \equiv \int dk'_1 dk_2 |\phi_{oo}(k'_1, k_2)|^2$, which is obtained by setting $h(k_1) = 1$ in Eq. (4.69). The difference between $\tilde{I}$ and $I$ is the domain of the $k'_1$ integral.

To evaluate $\tilde{I}$ we first perform the $k_{2x}$ and $k_{2y}$ integrals, which are identical to those done to obtain Eq. (4.83) from Eq. (4.69). Next is the $\phi$-integral: Instead of picking up $2\pi$, we pick up $\Lambda_{\kappa_{zp}}$, which is the angle subtended by $\Lambda(k_1)$. The $k_1$ integral has limited domains in two remaining (local) directions: $\hat{e}_{par}$, which is defined to be parallel to $C$ in $(k_{1P}, k_{1z})$-space; and $\hat{e}_{perp}$, which is defined to be perpendicular to $C$ in $(k_{1P}, k_{1z})$-space. The sinc and remaining Gaussian integral are performed in succession. Only points that lie in the neighborhood $C$ yield non-negligible contributions after these two integrals have been done, since these filter functions are both non-negligible only in the neighborhood of $C$.

Note from Eq. (4.67) that $|\phi_{oo}(k_1, k_2)|^2$ is non-negligible for $k_1$ only within a width of order $\kappa_z$ about the matching surface, and for $k_2$ only within $\kappa_z$ of its matching surface.
We make two observations. First, in the direction $\hat{\mathbf{e}}_{\text{perp}}$, it is seen from Eq. (4.67) that $|\phi_{oo}(\mathbf{k}_1, \mathbf{k}_2)|^2$ vanishes for arguments more than a few times $\kappa_z$ distant from $C$. Since the extent of the smoothing volume in this direction large enough that the weight of $|\phi_{oo}(\mathbf{k}_1, \mathbf{k}_2)|^2$ is fully contained in this direction, we can freely extend the limits of integration in the direction $\hat{\mathbf{e}}_{\text{perp}}$ so that they match the limits used in the calculation of $I$.

Second, note that in the direction $\hat{\mathbf{e}}_{\text{par}}$ that $|\phi_{oo}(\mathbf{k}_1, \mathbf{k}_2)|^2$ varies negligibly over the domain of integration. This follows from the fact that all filter functions have vanishing arguments for any point on $C$, so the rapid variation of the filter functions is therefore perpendicular to $C$. Since our expression for $\tilde{I}$ has the same limits of integration as the expression for $I$ in the directions in which all of the filter functions vary, therefore the sinc and Gaussian filter integrals yield the same results as they do for $I$.

After these sinc and Gaussian filter integrals have been performed, $\tilde{I}$ has a single remaining integral in $K$, parallel to $C$. This integral has a domain of integration that is a few times $\kappa_z$ in size, but the integrand varies negligibly over that domain. It is performed using the simple result $(b - a)^{-1} \int_a^b dx g(x) \simeq g(x_0)$, which holds for $g(x)$ that varies negligibly over the domain of integration, requiring only that $x_0 \in [a, b]$. This yields the final result for $\langle N_{k_1} \rangle$, Eq. (4.91).
Bibliography


[43] B. Huttner and S. M. Barnett, Phys. Rev. A 46, 4306 (1992), note that Huttner and Barnett’s expressions for the quantized transverse fields are implicit in earlier work on the Casimir interaction between absorptive and dispersive dielectric half-spaces, if that work is applied to a uniform medium [126].


[92] L. Landau and E. Lifshitz, Electrodynamics of Continuous Media (Pergamon, 1960), 1st ed., p. 258; the mathematical core of the proof is given elsewhere [127].


