Coherent Control of Photocurrent in Bulk Semiconductors

by Alain Haché

A thesis submitted in conformity with the requirements for the degree of Doctor of Philosophy

Graduate Department of Physics
University of Toronto

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Abstract

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Alain Haché

A thesis submitted for the degree of Doctor of Philosophy,
Department of Physics, University of Toronto, 1997.

Coherent control of photocurrent via interband transitions in bulk semiconductors is proposed and demonstrated in GaAs. A Fermi's Golden Rule approach is used to calculate the overall transition rate for simultaneous single- and two-photon excitation in direct band gap semiconductors using phase-related beams at frequencies $\omega$ and $2\omega$, with $\hbar\omega < E_g < 2\hbar\omega$, where $E_g$ is the band gap energy. The calculation is carried out to obtain the current generation rate as a function of the beams' intensity, polarization and frequency; numerical results are given for GaAs.

Coherent control of photocurrent is demonstrated using picosecond and femtosecond laser pulses on GaAs at room temperature. By integrating the current in a metal-semiconductor-metal (MSM) device, peak current densities as high as 20 mA/$\mu$m$^2$ are measured using phase-related 100 fs pulses at 1550 and 775 nm from an optical parametric generator. Carrier densities of $\sim 10^{18}$ cm$^{-3}$ are achieved during the process, but carrier-carrier scattering does not seem to significantly affect the current decay time. Non-degenerate coherent control is also demonstrated with three beams of different frequency such that $\omega_1 + \omega_2 = \omega_3$ and $\hbar\omega_1 \neq \hbar\omega_2 < E_g < \hbar\omega_3$. Low-temperature grown GaAs is found to be more suitable for current integration than normally-grown GaAs because the former has a shorter carrier lifetime which prevents the MSM device from discharging during the time between pulses.
Acknowledgments

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<tr>
<th>Symbol</th>
<th>Meaning</th>
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<tr>
<td>$\alpha$</td>
<td>linear absorption coefficient at $2\omega$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>two-photon absorption coefficient at $\omega$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>absorption parameter</td>
</tr>
<tr>
<td>$\delta$</td>
<td>phase mismatch parameter</td>
</tr>
<tr>
<td>$\Delta\delta$</td>
<td>phase shift between $\omega$ and $2\omega$ after one pass through glass window</td>
</tr>
<tr>
<td>$\Delta v_g$</td>
<td>group velocity mismatch between $\omega$ and $2\omega$</td>
</tr>
<tr>
<td>$\Delta \omega$</td>
<td>phase difference between the $\omega$ and $2\omega$ beams</td>
</tr>
<tr>
<td>$\Delta k$</td>
<td>wavevector mismatch</td>
</tr>
<tr>
<td>$\Delta n$</td>
<td>index of refraction mismatch between $\omega$ and $2\omega$</td>
</tr>
<tr>
<td>$\vec{\eta}_{ie(n)}$</td>
<td>tensor of the photocurrent injection</td>
</tr>
<tr>
<td>$\theta$</td>
<td>incidence angle of the beams on the glass window</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>wavelength</td>
</tr>
<tr>
<td>$\mu_e$</td>
<td>electron mobility</td>
</tr>
<tr>
<td>$\mu_{e1}$</td>
<td>dipole matrix element for s- and p-like states</td>
</tr>
<tr>
<td>$\xi_{1,2}$</td>
<td>tensor for carrier injection rate with single- and two-photon transitions</td>
</tr>
<tr>
<td>$\rho$</td>
<td>carrier density</td>
</tr>
<tr>
<td>$\tau_{\omega,2\omega}$</td>
<td>duration of the $\omega$ and $2\omega$ pulses</td>
</tr>
<tr>
<td>$\tau_c$</td>
<td>phenomenological photocurrent decay time</td>
</tr>
<tr>
<td>$\tau_{LO}$</td>
<td>electron-LO-phonon effective scattering time</td>
</tr>
<tr>
<td>$\tau_p$</td>
<td>general pulse duration</td>
</tr>
<tr>
<td>$\nu_e$</td>
<td>electron speed</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>angular separation between the $\omega$ and $2\omega$ beams</td>
</tr>
<tr>
<td>$\phi_{\omega,2\omega}$</td>
<td>phase of the optical beam at $\omega$ and $2\omega$</td>
</tr>
<tr>
<td>$\chi^{(n)}$</td>
<td>$n^{th}$-order susceptibility</td>
</tr>
<tr>
<td>$A$</td>
<td>non-radiative recombination coefficient</td>
</tr>
<tr>
<td>$A(t)$</td>
<td>vector potential</td>
</tr>
<tr>
<td>$B$</td>
<td>radiative recombination coefficient</td>
</tr>
<tr>
<td>$b$</td>
<td>frequency chirp coefficient</td>
</tr>
<tr>
<td>$c_{eu,k}$</td>
<td>coefficient of the linear superposition of states</td>
</tr>
<tr>
<td>$C$</td>
<td>capacitance</td>
</tr>
<tr>
<td>$C_{load}$</td>
<td>load capacitance across the MSM device</td>
</tr>
<tr>
<td>$C_{MSM}$</td>
<td>MSM device internal capacitance</td>
</tr>
<tr>
<td>Symbol</td>
<td>Meaning</td>
</tr>
<tr>
<td>--------</td>
<td>---------</td>
</tr>
<tr>
<td>$D$</td>
<td>pulse broadening coefficient</td>
</tr>
<tr>
<td>$d_{\text{eff}}$</td>
<td>effective nonlinear coefficient for second harmonic generation</td>
</tr>
<tr>
<td>$E, E$</td>
<td>complex and real electric field amplitude</td>
</tr>
<tr>
<td>$E_g$</td>
<td>semiconductor band gap energy</td>
</tr>
<tr>
<td>$E_{\text{LO}}$</td>
<td>LO-phonon energy</td>
</tr>
<tr>
<td>$f_{s,p}(r)$</td>
<td>radial functions of s- and p-like states of the electron</td>
</tr>
<tr>
<td>$H(t)$</td>
<td>Hamiltonian</td>
</tr>
<tr>
<td>$I_{\omega, 2\omega}$</td>
<td>irradiance of the $\omega$ and $2\omega$ beams</td>
</tr>
<tr>
<td>$J_{e,h}$</td>
<td>contribution of electrons and holes to the current density</td>
</tr>
<tr>
<td>$J^I, J^I$</td>
<td>current density injection rate</td>
</tr>
<tr>
<td>$k$</td>
<td>wavevector</td>
</tr>
<tr>
<td>$L'$</td>
<td>inductance per unit length</td>
</tr>
<tr>
<td>$l'$</td>
<td>length parameter for second harmonic generation</td>
</tr>
<tr>
<td>$L_g$</td>
<td>glass plate thickness</td>
</tr>
<tr>
<td>$M_{ji}^{(1,2)}$</td>
<td>dipole matrix element for one- and two-photon transitions</td>
</tr>
<tr>
<td>$M_{\text{vuk}}$</td>
<td>tensor for two-photon transition</td>
</tr>
<tr>
<td>$m_e$</td>
<td>electron effective mass</td>
</tr>
<tr>
<td>$m_{hh}$</td>
<td>heavy hole effective mass</td>
</tr>
<tr>
<td>$m_{lh}$</td>
<td>light hole effective mass</td>
</tr>
<tr>
<td>$m_o$</td>
<td>rest mass of the electron</td>
</tr>
<tr>
<td>$N_{\text{vuk}}$</td>
<td>tensor for single photon transition</td>
</tr>
<tr>
<td>$n_{\omega, 2\omega}$</td>
<td>index of refraction of GaAs at $\omega$ and $2\omega$</td>
</tr>
<tr>
<td>$n_{p,\omega, 2\omega}$</td>
<td>index of refraction of glass plate at $\omega$ and $2\omega$</td>
</tr>
<tr>
<td>$P$</td>
<td>complex amplitude of the polarization</td>
</tr>
<tr>
<td>$P_{\omega, 2\omega}$</td>
<td>average power of the $\omega$ and $2\omega$ beams</td>
</tr>
<tr>
<td>$p$</td>
<td>momentum operator</td>
</tr>
<tr>
<td>$q$</td>
<td>charge accumulated on the MSM electrodes</td>
</tr>
<tr>
<td>$R$</td>
<td>surface reflectance of the semiconductor</td>
</tr>
<tr>
<td>$R_{\text{LO}}$</td>
<td>electron-LO-phonon scattering rate</td>
</tr>
<tr>
<td>$R_{\text{MSM}}$</td>
<td>MSM device internal resistance</td>
</tr>
<tr>
<td>$R_{\text{MSM}}(t)$</td>
<td>MSM device internal resistance</td>
</tr>
<tr>
<td>$R_l$</td>
<td>load resistance across the MSM device</td>
</tr>
<tr>
<td>$R_{\text{load}}$</td>
<td>load resistance across the MSM device</td>
</tr>
<tr>
<td>$T$</td>
<td>laser pulse period</td>
</tr>
<tr>
<td>$t_d$</td>
<td>time delay between the $\omega$ and $2\omega$ pulses</td>
</tr>
<tr>
<td>$v_{ij}$</td>
<td>matrix elements of the velocity operator</td>
</tr>
<tr>
<td>$W_{ij}$</td>
<td>transition rate from state $i$ to state $j$</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

1.1 Overview

The advent of lasers producing high peak power pulses with short duration, and the development of techniques for ‘tailoring’ optical electric fields has opened up new areas in photochemistry and photophysics. In particular, the use of optical phase to control physical and chemical processes has attracted a great deal of attention during the last decade. Unlike most light/matter interactions where the important parameters of the optical beam are the frequency and the irradiance, coherent control processes offer the additional possibility of phase-sensitivity, making them an appealing concept both from a fundamental and technological point of view.

Coherent control processes can be understood in terms of interfering quantum mechanical pathways. When two or more optical fields couple the same initial and final states of a system, as illustrated in Fig. 1-1a, an interference between the transition amplitudes can occur because an electron in the lower level can reach the upper level via two pathways. By controlling the relative phase of the beams, the overall transition rate can be modified. The classical analog to this quantum mechanical effect is the famous Young’s double slit experiment (see Fig. 1-1b). Light emitted from a source is presented with two possible paths leading to a point A on a distant screen. As a result, an inter-
Figure 1-1: a) Schematic diagram of electron eigenstates coupled by electric fields at frequencies $\omega$ and $2\omega$; b) the classical analog: the Young’s double slit experiment.

Interference pattern appears on the screen with a spatial profile depending on the length of each path. Unlike this rather simple picture, however, coherent control in matter is even richer in possibilities since electrons excited to the upper energy level can have access to states of different parity, spin, angular momentum, etc.

One area where coherent control is particularly interesting, but relatively unexplored, is photocurrent generation in semiconductors. Today, nearly all electronic technology is based on controlling the magnitude and direction of electric currents. The conventional method for controlling currents in semiconductors is to apply an electric bias or to provide some structural asymmetry, e.g., through potential wells and barriers. In this thesis, the author shows how optical coherence effects can be used to generate and control the directionality of photocurrents in semiconductors. In essence, quantum mechanical interference between pathways coupling the same initial (valence) and final (conduction) states leads to an optically induced asymmetrical distribution of free carriers in momentum-space, as illustrated in Fig. 1-2. For instance, the overall rate of interband transitions...
Figure 1-2: Schematic band diagram of a bulk semiconductor with electrons excited into preferential states in momentum-space. The resulting distribution of carriers creates a net current flow.

induced by simultaneous one- and two-photon excitation in a crystal can be different for two states with anti-parallel wavevectors. The resulting anisotropic distribution of carriers in the conduction band creates a net current flow that can be controlled by adjusting the relative phase of the beams. This phenomenon can also be understood in terms of interfering wave-functions. Indeed, single- and two-photon transitions promote electrons to states of different parity. These symmetric and antisymmetric wave-functions can in turn interfere constructively in one spatial direction, and destructively in the other direction, depending on the optical phase. Before going into the details of photocurrent control – the main object of this thesis – we will review coherent control from a historical perspective.

The interaction between strong optical fields and atomic media was investigated as early as 1967 by Gurevich and Khronopulo [1], and Manykin and Alfanas’ev [2]. It was shown theoretically that the absorbance could be altered by changing the relative phase
of two beams which excite electrons via two or more allowed transition pathways. The enhancement of multiphoton ionization was first demonstrated in xenon gas in 1980 by Miller et al. [3] using 440 nm light from a dye laser, and was further described theoretically by Jackson et al. [4]. In this process, the creation of higher optical harmonics during the beam propagation provides a new transition channel for electrons which interferes with the original excitation pathway. This concept of interfering harmonics was later exploited by Chen et al. to actively control photoionization in mercury by adjusting the relative phase between a fundamental beam $\omega$ and its third harmonic $3\omega$ (554 and 135 nm respectively) [5]. Using three- and five-photon transitions between bound and continuum states, a transition probability which varied as $\sin(3\phi_\omega - \phi_{3\omega})$ was observed, where $\phi$ denotes the phase of each beam. Subsequently, experiments of a similar nature were carried out with other atomic gases such as krypton [6], rubidium [7] and barium [8]. Experiments with rubidium showed directional photoionization via simultaneous single- and two-photon excitation. By coherently populating electronic states with even (two photons) and odd (one photon) parity, constructive and destructive interference of the wave-functions leads to photoelectrons being emitted in a preferential direction.

The idea of coherent control has been extended to molecules with the intention of manipulating chemical reactions. One of the first attempts to do so made use of a pump-probe arrangement [9]: wave packets of excited electrons were prepared with one pulse, then excited into another state by a second pulse after a chosen interval of time. A different approach consists of simultaneously using two pulses to populate a coherent, degenerate state corresponding to a linear combination of the possible reaction products [10][11]. Adjusting the relative phase of the two beams enhances one product channel and decreases the other. For example, this process has been demonstrated in HD$^+$ for the simplest chemical reaction, photodissociation [12]. With two beams at frequency $\omega$ and $2\omega$ coupling a bound and repulsive state, molecules of HD$^+$ were dissociated into H+D$^+$ or D+H$^+$ in a ratio depending on the relative phase of the optical fields. Phase-controlled photodissociation has also been observed in diatomic molecules such as H$_2^+$, HI and Na$_2$.
[13][14][15], as has photoionization of HCl [16].

Even though the great majority of the systems studied so far are atomic and molecular, a growing interest has been shown in crystalline solids. Traditionally, solids have been considered as marginal candidates for coherent control experiments because of the continuum of available states in the valence and conduction bands and the short electronic dephasing time associated with them. Nonetheless, the idea of producing directional photocurrents by ionizing impurity states in semiconductors [20] or through photogalvanic effect in conducting media [21] via phase-related optical beams was investigated theoretically. Coherent excitation has also been used to explain laser-induced second harmonic generation in doped glass optical fibers [17,18,19]. In this case, trapping of photoelectrons emitted asymmetrically creates a space charge and a DC electric field which permits second harmonic generation.

In 1989, Zel'dovich et al. [18] and Baranova et al. [22] demonstrated directional photoemission from Sb-Cs photocathodes using two phase-related harmonics of a laser beam. The rate of photoejection of electrons from the electrode of a photomultiplier varied with the phase of the beams. Our research group proposed using interband transitions in bulk semiconductors to achieve directional photocurrents [23][24], an effect also investigated theoretically by Khurgin [25]. Dupont et al. [26] were able to demonstrate directional ionization from GaAs/GaAlAs multiple quantum wells at 82 K. One- and two-photon transitions with the first and second harmonic of a pulsed CO₂ laser (10.6 and 5.3 μm respectively, and 100 ns time duration) were used to promote electrons from bound states to the continuum, thereby creating a directional photocurrent. However, the process is 'atomic-like' and similar to the photoionization experiment in the sense that electrons are excited from a discrete level to the continuum. We claimed that coherent control in semiconductors using interband transitions in bulk semiconductors would also yield controllable photocurrents even though both the initial and final states lie in the continuum. A first-principles theory for the effect was offered [27] and experimental confirmation was carried out later in bulk, unbiased GaAs at room temperature [28].
1.2 Photocurrent control in semiconductors

Each coherent control process mentioned above has unique characteristics in terms of the physics and the experimental technique used to observe it. In the case of photocurrent control in bulk solids, there are three points that need to be addressed: 1) the continuum of available states, 2) the carrier momentum relaxation time and 3) the current generation and measurement technique.

Unlike quantum wells and atomic and molecular systems where electrons occupy discrete levels of energy, bulk solids possess electronic bands with a continuum of states. This raises the question of whether the same states would be accessible via single- and multi-photon transitions, since the selection rules for both are not identical in general. For instance, in some materials, it is known that there are excitonic states accessible through two-photon transitions but forbidden with one-photon transitions [29]. In order to interfere, the two excitation pathways should have access to the same final states. However, states in the valence and conduction bands are made of hybridized sp\(^3\) orbitals with no defined parity, and therefore can be coupled with any multi-photon excitation. Since laser beams in practice have a finite frequency bandwidth, a number of different states will be coupled, but interference is still possible provided that the fields are phase-related.

The momentum relaxation time \(\tau\) of the injected carriers is important for two reasons: first, it determines the saturation of the photocurrent (giving a maximum value \(\approx J \cdot \tau\) for constant illumination, where \(J\) is the current injection rate), and second, it indicates how long the current remains after the exciting fields are turned off. However, there is no one-to-one relationship between carrier momentum relaxation time and the overall current decay time. As pointed out by Auston [30], the photocurrent lifetime may be much longer than the carrier scattering time because only certain collision events will cause the current to decay. For instance, electron-electron interactions preserve the total momentum as well as the current, whereas phonon and impurity scattering do not. Also, even in the case of phonon interactions, many scattering events may be needed for the
momentum of the carriers to relax.

Last, but not least, detecting the coherently generated photocurrent presents a serious experimental challenge. Multi-photon transitions are rather inefficient processes and require the use of short, high peak intensity laser pulses. For instance, the mode-locked Ti:sapphire laser can generate a train of pulses each with ~100 fs duration. For such brief illumination, coherent photocurrents in semiconductors would rise and decay on a time scale of less than a picosecond. Even the most advanced electronic device would fail to measure such brief current bursts. One alternative method, which the author has made use of in these experiments, is to integrate the current instead of time-resolving it. A metal-semiconductor-metal (MSM) structure allowed for the monitoring of the space-charge accumulation as a function of the phase of the fields.

1.3 Thesis outline

Following this introduction, a theoretical treatment of coherent control of photocurrent in semiconductors is presented along with an experimental demonstration in bulk GaAs. In chapter 2, the theory, starting from a transition rate perspective using Fermi's Golden Rule, is discussed. The current injection rate is calculated from first principles and numerical results are given for GaAs. The injection rate is connected to the optical electric fields via a fourth-rank tensor. A phenomenological model is laid out to describe the current dynamics and the influence of carrier concentration, momentum relaxation time and circuit parameters. The impact of optical dispersion and pulse chirping on the photocurrent measurement are discussed. Chapter 3 includes the important experimental and technical aspects of the coherent control measurement: laser sources, MSM devices, sample characteristics and beam wave-front analysis. The experimental results are presented in chapter 4. Photocurrent control is demonstrated in GaAs grown at low temperature (LT-GaAs) using pairs of 1 ps pulses from an optical parametric oscillator (OPO), as well as with 100 fs pulses from an optical parametric amplifier (OPA). Po-
larization effects and power dependence of the current are investigated. The results of a similar experiment with normally-grown GaAs using the OPA are later described, with an analysis of power dependence and a comparison with theory. Non-degenerate coherent control using three beams of different frequencies is also demonstrated in LT-GaAs. The effect of pulse chirping on the current integration efficiency is investigated and attempts at measuring currents in silicon are discussed. Conclusions appear in chapter 5.
Chapter 2

Theory

This chapter presents a Fermi’s Golden Rule approach for interband transitions to model the generation of asymmetrical distributions of carriers in momentum-space in the presence of two excitation beams at $\omega$ and $2\omega$. In a more rigorous treatment, a first-principles calculation of the tensor which describes the current generation response to the fields is carried out and numerical results are given for GaAs. The dynamics of the current after pulse excitation are discussed from a phenomenological point of view, taking into account the influence of carrier-carrier, phonon-carrier and carrier-impurity scattering, and effects of current self-inductance, electromagnetic radiation and free carrier absorption.

A model is developed specifically for the experimental technique used in these experiments to predict the charge accumulation on a MSM structure as a function of beam, device and material parameters. The importance of each parameter relative to the current integration efficiency determines the most suitable type of material, experimental geometry and laser source to be used. Finally, optical effects associated with the use of finite bandwidth laser pulses are discussed. Pulse broadening, frequency chirp, pulse delays and refractive index mismatch in the semiconductor are treated through consideration of first- and second-order dispersion effects.

The calculations and analysis to follow in this and the next chapters assume certain values for the material parameters of GaAs and LT-GaAs. Table 2.1 lists the values used
Table 2.1: Values for important parameters of GaAs and LT-GaAs at 300 K

throughout this thesis. Parameters of LT-GaAs for which no unique reference is available have been assumed to be the same as for GaAs.

GaAs is a direct band gap semiconductor with zinc-blende crystalline structure. Indirect transitions are allowed from the \( \Gamma \) valence band edge to the L- and X-points of the reciprocal lattice (see Fig. 2-1). But the transition energies are 1.71 and 1.90 eV respectively, larger than the 1.6 eV maximum photon energy used in our experiments. Furthermore, the split-off band (not shown in the diagram) is situated just below the heavy and light hole valence bands and would require a minimum energy of 1.77 eV to allow a single-photon transition. We can therefore restrict the analysis to direct transitions from heavy- and light-hole bands to the conduction band edge.

### 2.1 A simple model for current injection

The easiest and perhaps most natural way of understanding how photocurrent control can occur in direct band gap semiconductors is to consider interband transition rates. In this treatment we consider one valence and one conduction band (labeled with \( v \) and \( c \)
Figure 2-1: Schematic diagram of the band structure of GaAs at 300 K near the fundamental energy gap.

respectively) coupled with a dual frequency electric field:

\[ \mathbf{E}(t) = E_\omega e^{i(\omega t + \phi_\omega)} + E_{2\omega} e^{i(2\omega t + \phi_{2\omega})} + c.c. \]  \hspace{1cm} (2.1)

where \( E_\omega, E_{2\omega}, \phi_\omega \) and \( \phi_{2\omega} \) are the complex electric field amplitudes and phases for each frequency. The frequency \( \omega \) is chosen so that \( \hbar \omega < E_g < 2\hbar \omega \), where \( E_g \) is the bandgap of the material. Since at this point we are only interested in a demonstration of principle, we consider only local effects and the spatial dependence of the electric fields is not taken into account. In the presence of a weak perturbation, the Hamiltonian of the system including the interaction with the electromagnetic field can be separated into a time-independent and a time-varying part

\[ H(t) = V(r) + \frac{1}{2m_0} (p - e\mathbf{A}(t))^2 = V(r) + \frac{\mathbf{p}^2}{2m_0} + \frac{e^2}{2m_0} A^2(t) - \frac{e}{m_0} \mathbf{A}(t) \cdot \mathbf{p} \]  \hspace{1cm} (2.2)
in SI units and where $V(r)$ is a periodic electrostatic potential, $m_0$ is the rest mass of the electron. $A$ is the vector potential of the electric field (defined by $E(t) = -\partial A(t)/\partial t$) and $p$ is the momentum operator. The potential $A(t)$ can be separated into its frequency components, $A_{\omega}e^{-i(\omega t + \phi_{\omega})}$ and $A_{2\omega}e^{-i(2\omega t + \phi_{2\omega})}$, where $A_{\omega,2\omega}$ are complex and are assumed to have parallel orientation in this case. The second term in $A^2(t)$ is not of interest here since it only alters the phase of the wavefunction on which it operates. The last term is used to obtain the transition probability, calculated using the standard perturbation theory. The transition rate $W_{v,c}$ from the valence to the conduction band is obtained from Fermi’s Golden Rule:

$$W_{v,c}(k) = 2\pi\hbar^{-1} \sum_{\nu} |M_{v\nu}^{(1)}(k, 2\omega) + M_{v\nu}^{(2)}(k, \omega)|^2 \delta(\omega_{\nu}(k) - 2\omega)$$  \hspace{1cm} (2.3)

where the summation is carried out over all possible valence and conduction states and where $\omega_{\nu}(k) = \omega_r(k) - \omega_i(k)$, with $\omega_r(k)$ and $\omega_i(k)$ the dispersion relations for each band. The dipole matrix elements for one- and two-photon transitions, $M_{v\nu}^{(1)}(k)$ and $M_{v\nu}^{(2)}(k)$, are obtained using Eq. 2.2:

$$M_{v\nu}^{(1)}(k, 2\omega) = \frac{e}{m_0} e^{-i(2\omega t + \phi_{2\omega})} A_{2\omega} \cdot p_{\nu}$$  \hspace{1cm} (2.4)

$$M_{v\nu}^{(2)}(k, \omega) = \sum_a \frac{M_{v\alpha}^{(1)}(k, \omega) M_{\alpha\nu}^{(1)}(k, \omega)}{\hbar(\omega(k)_{\nu} - \omega)}$$

$$= \frac{e^2}{\hbar m_0^2(\omega_{\nu}(k) - \omega)} e^{-i(2\omega t - 2\phi_{2\omega})} \sum_a A_{\nu} \cdot p_{\alpha} A_{\alpha} \cdot p_{\varepsilon}$$  \hspace{1cm} (2.5)

where the summation is performed over all intermediate states $a$. In GaAs, to a first order approximation, the valence states are 'p-like' and conduction states (away from the band edge) are a mixture of 's-like' and 'p-like' states. Indeed, the Bloch wave-functions can be written

$$\Psi_{v,\mathbf{k}}(r) = e^{i\mathbf{k} \cdot \mathbf{r}} f_{p}(r)$$  \hspace{1cm} (2.7)
in the valence band and
\[
\Psi_{c,k}(r) = e^{ik \cdot r} (f_s(r) + (k \cdot r) f_p(r))
\]  \hspace{1cm} (2.8)

in the conduction band. where \( r = |r| \). The unit cell functions \( f_s(r) \) and \( f_p(r) \) have radial symmetry and characterize the s- and p-states respectively. As a result,
\[
p_{cv} = \int \Psi_{c,k}^*(r)p \Psi_{v,k}(r) dr^3 = -i \hbar \int \Psi_{c,k}^*(r) \nabla \Psi_{v,k}(r) dr^3
\]  \hspace{1cm} (2.9)
\[
= -i \hbar \int dr^3 \left[ (k \cdot r) \hat{r} f_p^*(r) \frac{df_p(r)}{dr} \right] = k \mu_{cv}
\]  \hspace{1cm} (2.10)

with \( \hat{r} \) the unit vector oriented along \( r \). Since \( p_{cv} \parallel k \) and \( A \parallel E \), the product \( A_2 \cdot p_{cv} \) can be written in terms of \( E_2 \cos \theta \), where \( \theta = \hat{E} \cdot \hat{k} \). Similarly, \( A_2 \cdot p_{ca} A_2 \cdot p_{av} \) is found to be proportional to \( E_2^2 \cos^2 \theta \). Before going further in calculating \( W_{cv}(k) \), let us consider Eq. 2.5 again. The summation involves many possible pathways for two-photon transitions, but as pointed out by Wherrett [42], the dominant contribution to the two-photon interband transition in direct band gap semiconductors is \( v \rightarrow c \rightarrow c \) all with the same \( k \), i.e. the second step is a self-transition within the conduction band. The corresponding dipole matrix element is therefore \( M_{cv}^{(2)}(k, \omega) \propto M_{cc}^{(1)}(k, \omega) M_{cv}^{(1)}(k, \omega) \). However, whether we choose to include all combinations or not, the conclusions we will draw from this calculation remain unchanged.

Expansion of the square modulus in Eq. 2.3 leads to three terms:
\[
M_{cv}^{(1)}(k, 2\omega) M_{cv}^{(1)*}(k, 2\omega) = \left( \frac{e^2 k^2 A_2^2 \mu_{cv}}{m_0} \right)^2 \cos^2 \theta,
\]  \hspace{1cm} (2.11)
\[
M_{cv}^{(2)}(k, \omega) M_{cv}^{(2)*}(k, \omega) = \left( \frac{e^2 k^2 A_2^2}{m_0^2 \hbar (\omega_{cv}(k) - \omega)} \sum_a \mu_{ca} \mu_{av} \right)^2 \cos^4 \theta,
\]  \hspace{1cm} (2.12)

and
\[
M_{cv}^{(1)}(k, 2\omega) M_{cv}^{(2)*}(k, \omega) + M_{cv}^{(1)*}(k, 2\omega) M_{cv}^{(2)}(k, \omega)
\]  \hspace{1cm} (2.13)
\[ = \left( \frac{2e^3 A_{2\omega} A_{\omega}^2}{m_0^2 \hbar (\omega_{\text{r},(k)} - \omega)} \mu_{cu} \sum_a \mu_{ca} \mu_{av} \right) \sin(2\phi_\omega - \phi_{2\omega}) \cos^3 \theta. \] (2.14)

The first and second terms correspond to single- and two-photon excitation respectively whereas the third one can be regarded as an 'interference' term. The important feature to be considered here is the dependence on \(\cos^3 \theta\) leading to a transition rate with polar asymmetry, i.e. carriers with anti-parallel wavevectors do not have the same transition probability. Furthermore, the third term varies with \(\Delta \phi = 2\phi_\omega - \phi_{2\omega}\), which means that the transition rate at a given \(k\) can be phase-controlled. By adjusting \(\Delta \phi\) one can therefore modify the ratio between the populations of free carriers with wavevector \(k_\alpha\) (or \(\theta = \theta_\alpha\)) and \(-k_\alpha\) (or \(\theta = \theta_\alpha + \pi\)). Integration over \(\theta\) leads to an asymmetric distribution of electrons and holes in momentum-space and the presence of a net photocurrent controllable with the phase of the beams.

This simple model does not give any details of the current injection rate and its dependence on crystal properties, but it reveals the possibility of creating currents by interfering transition pathways. In the next section, coherent control is considered at a deeper level for the case of direct band gap semiconductors with a realistic band structure.

### 2.2 First-principles calculation of current injection

A more complete understanding of coherent control in semiconductors requires a full quantum mechanical treatment of the effect. We can understand this step by returning to the analogy with the double slit experiment. A first look at Young's experiment tells us that interference occurs at the screen since two paths are taken by each photon emitted at the source. This observation states that there will be an interference pattern occurring on the screen but it does not say *how exactly* it will appear. In order to determine the shape of the interference pattern, a detailed calculation is necessary, based on the spectral width of the light source, the position of the source, slits and screen, the width and separation of the slits, wave diffraction through the apertures, etc. There is
no real middle step between understanding the interference phenomenon and knowing the shape of the pattern. Similarly, a clear picture of the photocurrent coherent control can only be obtained through a detailed calculation of the interaction of the \( \omega \) and \( 2\omega \) beams with the semiconductor. Although the effect has been described in the framework of the susceptibility formalism [43] – the common language of nonlinear optics – this theoretical treatment was also made at a very fundamental level, recalculating the diverging susceptibility in the length \((\mathbf{r} \cdot \mathbf{E})\) instead of the velocity \((\mathbf{p} \cdot \mathbf{A})\) gauge.

Our goal is to determine the expression relating the current injection to the optical fields in a similar fashion as the third order DC polarization \( \mathbf{P}_{DC} \) is connected to the fields via the nonlinear susceptibility: \( \mathbf{P}_{DC} = \chi^{(3)}(0; \omega, \omega, -2\omega)\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{E}(-2\omega) \). Most of the following analytical and numerical calculations were carried out by R. Atanasov, J. L. P. Hughes and J. E. Sipe (see ref. [27]); the author was involved in the calculation of interband dipole moments for GaAs.

In a manner similar to that in the previous section, perturbation theory is applied to a valence and a conduction band coupled with optical fields at \( \omega \) and \( 2\omega \). In the presence of a classical electromagnetic field, we look for states of the form

\[
|\Psi(t)\rangle = c_0(t) |0\rangle + c_{cv,k}(t) |cv,k\rangle
\]

where the coefficients \( c_0(t) \) and \( c_{cv,k}(t) \) are determined from perturbation theory: the usual minimal coupling Hamiltonian is used in the long wavelength limit and many-body effects are neglected. The ket \(|0\rangle\) is the ground state of the crystal (filled valence band) and \(|cv,k\rangle\) is an excited state, i.e. an electron-hole pair with wavevector \( \mathbf{k} \). From the transition probability \( |c_{cv,k}(t)|^2 \), the carrier density is given by

\[
\rho(t) = \frac{1}{V} \sum_{cv,k} |c_{cv,k}(t)|^2
\]

(2.15)

where \( V \) is the volume over which the summation is made. Since we are interested in the current and not the carrier density, we make use of the current density operator to
calculate $J = J_e + J_h$, where

$$J_e = -\frac{e}{V} \sum_{\nu, \nu', k} |c_{\nu, k}(t)|^2 v_{\nu, \nu'}(k)$$

for electrons and

$$J_h = \frac{e}{V} \sum_{\nu, \nu', k} |c_{\nu, k}(t)|^2 v_{\nu, \nu'}(k)$$

for holes. The matrix elements $v_{\nu, \nu'}$ of the velocity operator can be understood as the expectation value of the velocity of the particle. We can therefore see the analogy with $J = q \rho v$, the general expression for the current density produced by moving particles of charge $q$.

To calculate $|c_{\nu, k}(t)|^2$, we suppose the vector potential associated with the electric field is

$$A(t) = \sum_n A(n\omega) \exp(-i n \omega t); n = \pm 1, \pm 2.$$

where $A(n\omega)$ is the complex amplitude at frequency $n\omega$. The Hamiltonian for the crystal and the interaction with the optical field can be expressed as

$$H = H_0 + H'(t) + V(t),$$

where

$$H_0 = \sum_{c, \nu, k} \hbar \omega_c(k) a_{\nu, c, k}^\dagger a_{\nu, c, k} - \sum_{\nu, \nu', k} \hbar \omega_{\nu, \nu'}(k) b_{\nu, \nu', k}^\dagger b_{\nu, \nu', k}.$$

$$H'(t) = \frac{e^2}{2m_0} A^2(t),$$

$$V(t) = a_A(t) \left\{ \sum_{c, c', k} v_{cc'}(k) a_{c, c', k}^\dagger a_{c', k}(t) - \sum_{\nu, \nu', k} v_{\nu, \nu'}(k) b_{\nu, \nu', k}^\dagger b_{\nu, k}(t) \right\}$$

and where the annihilation and creation operators are $a$ and $a^\dagger$ for electrons and $b$ and $b^\dagger$ for holes. The time-dependent terms, $H'(t)$ and $V(t)$, are obtained from expanding the
interaction Hamiltonian \((p - eA(t))^2/2m_0\). The generation rate of electron-hole pairs is determined by \(V(t)\) and from perturbation theory:

\[
\dot{c}_{cv,k}(t) = \langle cvk|V(t)|\Phi(t)\rangle
\]

with solutions of the form

\[
c_{cv,k}(t) = -i \frac{\exp[i(\omega_{cv}(k) - 2\omega)] - 1}{\omega_{cv}(k) - 2\omega} \{N_{cv,k}(2\omega) : E(2\omega) + M_{cv,k}(\omega) : E(\omega)E(\omega)\}.
\]

where

\[
N_{cv,k}(2\omega) = \frac{-e}{2\omega \hbar} v_{cv}(k),
\]

\[
M_{cv,k}(\omega) = \frac{i e^2}{\hbar^2 \omega^2} \left\{ \sum_{\nu} \frac{\nu_{\nu\nu}(k)v_{\nu\nu}(k)}{\omega_{\nu\nu}(k) - \omega} - \sum_{\nu'} \frac{v_{\nu\nu}(k)v_{\nu\nu}(k)}{\omega_{\nu\nu}(k) - \omega} \right\}.
\]

The distribution of injected carriers in \(k\)-space is asymmetric because \(|c_{cv,k}|^2 \neq |c_{cv,-k}|^2\). This follows from the form of \(|c_{cv,k}|^2\), which is the sum of terms \(\hat{A} : E(2\omega)E(-2\omega)\), \(\hat{B} : E(\omega)E(\omega)E(-\omega)E(-\omega)\), and \(\hat{C}v_{cv}(k) : E(-2\omega)E(\omega)E(\omega)\), where tensors \(\hat{A}, \hat{B}\) and \(\hat{C}\) are even in \(k\). It is the last of these terms that is odd in \(k\) and leads to the asymmetry of \(|c_{cv,k}|^2\). resulting in a net current density injection rate \(\mathbf{J}_I\). This applies to both centrosymmetric and non-centrosymmetric crystals. To calculate \(\mathbf{J}_I\), Eqs. 2.16 and 2.17 can be combined with Eq. 2.24 to give

\[
\mathbf{J}_I^{(\mathbf{c})} = \tilde{\eta}^{(\mathbf{c})}(2\omega) : E(-\omega)E(-\omega)E(2\omega) + \ldots
\]

and the purely imaginary tensors \(\tilde{\eta}^{(\mathbf{c})}\) are given by

\[
\tilde{\eta}^{(\mathbf{c})}(2\omega) = (+1)i \frac{8\pi e^4}{\hbar^3} \sum_{\nu\nu,\alpha\alpha} \int \frac{d\mathbf{k}}{4\pi^2} \delta(\omega_{\nu\nu}(k) - 2\omega) \frac{v_{\nu\nu}(k)v_{\nu\nu}(k)}{\omega_{\nu\nu}(k)} \frac{v_{\alpha\alpha}(k)v_{\alpha\alpha}(k)}{\omega_{\alpha\alpha}(k)} - \omega_{\alpha\alpha}(k)
\]

where the subscript \(\alpha\) denotes a conduction or valence band. We have neglected correction
Figure 2-2: Tensor components of the total current generation tensor \( \eta \) as a function of the fundamental beam photon energy (\( \hbar \omega \)) for a complete band calculation.

Terms that can arise in the absence of a center of inversion. Since \( E(-\omega) = E^*(\omega) \) and \( E(2\omega) \) are complex amplitudes, the resulting \( \mathbf{J}_{\text{eff}}(\hbar) \) are clearly sensitive to the relative phase of the two beams. Numerical results for the total \( \eta = \eta_e + \eta_h \) are plotted in Fig. 2-2 for different combination of polarization orientation. According to these results, the peak current density that can be reached using existing pulsed laser sources is quite high. For example, using \( \eta_{xxxx} = 20 \text{ s}^{-2}\text{mCV}^{-3} \) and irradiances of 1 GWcm\(^{-2}\) and 100 MWcm\(^{-2}\) at 1550 and 775 nm (achievable with commercial parametric generators), the generation rate of current density would be \( 10^{20} \text{ As}^{-1}\text{cm}^{-2} \), leading to a current of \( 10^7 \text{Acm}^{-2} \) within 100 fs.
Since we often refer to single- and two-photon excitation carrier generation in relation to coherent control, it is useful calculate the full expressions for each process. Eq. 2.15 includes the contribution of both fields to the carrier density, but one could in fact consider them as separate sources of free carrier generation:

\[ \rho^{(1)} = \hat{\xi}_1(2\omega) : E(-2\omega)E(2\omega), \]
\[ \rho^{(2)} = \hat{\xi}_2(\omega) : E(-\omega)E(-\omega)E(\omega)E(\omega), \]

where we have also neglected a set of correction terms that arise in the absence of a center of inversion. The tensors \( \hat{\xi}_1 \) and \( \hat{\xi}_2 \) relate to the linear and two-photon absorption coefficients according to \( \alpha \sim |\hat{\xi}_1(2\omega)| \) and \( \beta \sim |\hat{\xi}_2(\omega)| \). The full expressions for each tensor are given by

\[ \hat{\xi}_1(2\omega) = \frac{2\pi e^2}{\hbar^2} \sum_{\alpha, \alpha'} \int \frac{dk}{4\pi^3} \delta(\omega_{\nu\alpha}(k) - 2\omega) \frac{v_{\nu\alpha}(k) v_{\nu\alpha}(k)}{\omega_{\nu\alpha}(k)}. \]
\[ \hat{\xi}_2(\omega) = \frac{32\pi e^4}{\hbar^4} \sum_{\alpha, \alpha', \alpha''} \int \frac{dk}{4\pi^3} \delta(\omega_{\nu\alpha}(k) - 2\omega) \frac{\{v_{\alpha\nu}(k) v_{\nu\alpha}(k)\} \{v_{\alpha'\nu}(k) v_{\nu\alpha'}(k)\}}{\omega_{\alpha\nu}(k) (\omega_{\nu\alpha}(k) - \omega_{\alpha\nu}(k)) (\omega_{\nu\alpha}(k) - \omega_{\alpha'\nu}(k))}. \]

The calculation for the current injection rate could be extended to the nondegenerate case involving beams of different frequencies such that \( \omega_1 + \omega_2 = \omega_3 \), with \( \hbar \omega_1, \hbar \omega_2 < E_g < \hbar \omega_3 \). Continuity requires that all elements of the nonlinear tensor \( \hat{n}_{e(h)} \) be the same when \( \omega_1 \approx \omega_2 \), as suggested by other studies on nondegenerate two-photon absorption in semiconductors [44].

Dephasing effects have been neglected since scattering typically occurs on a much longer time scale (\( \sim 100 \) fs) than the interband transition time (a few optical cycles). Also, the calculation does not take into account other transitions such as excitation to midgap impurity states or excitonic transitions, each of which may enhance the current injection.
2.3 Photocurrent dynamics

In this section the dynamics of free carriers on time scales of femtoseconds to nanoseconds are discussed. These considerations are necessary for understanding how current integration can occur in an MSM structure and how efficient it can be. On short time scales, current relaxation via carrier scattering, e-h plasma polarization relaxation and current self-inductance must also be considered. On a time scales >10^{-12} s, radiative and non-radiative recombination and diffusion of electrons and holes, Schottky barrier effects and space charge relaxation become important.

2.3.1 Current relaxation

The current relaxation time $\tau_r$ is an important parameter to consider because it determines the maximum current density obtainable with a given pulse intensity and duration. Phenomenologically, it enters the current rate equation according to

$$\dot{J} = J' - J/\tau_r.$$  

(2.33)

where $J'$ is the rate of injected current via coherent control given by Eq. 2.27. In general, $\tau_r$ will be dependent on the carrier density and the details of the interaction of carriers with phonons, impurities, and defects. Scattering rates are also different for electrons, heavy holes and light holes; we therefore must take into account the different contributions to the current for each quasi-particle. Electrons and holes will make different contributions because of their different effective masses and instantaneous velocities. From the relations $J = \rho ev$ and $v = \frac{1}{\hbar} \nabla_k \epsilon = \hbar k/m_e$ in the parabolic band approximation ($\epsilon$ is the electron energy), the total photocurrent density can be expressed as

$$J = \frac{e\hbar}{V} \left( -\frac{1}{m_e} \sum_i k_{e,i} + \frac{1}{m_h} \sum_i k_{h,i} \right),$$  

(2.34)
Figure 2-3: Particle speed versus excitation energy in GaAs for electrons excited from heavy and light hole band ($e_{hh}$ and $e_{lh}$) and their hole counterparts ($hh$ and $lh$) near the $\Gamma$-point.

where the summation is made over all free electrons and holes in a volume $V$. In order to conserve momentum, an electron-hole pair must satisfy the condition $k_e + k_h = 0$: electrons and holes will therefore have velocities in opposite directions.

Plotted in Fig. 2-3 are the heavy holes and light hole ($hh$ and $lh$) and electron ($e$) speeds in GaAs at the $\Gamma$-point as a function of photon energy. The electron speed in the conduction band is given by

$$v_e = \left[\frac{2\hbar \omega - E_g}{m_e(1 + m_e/m_h)}\right]^{1/2}$$

while the speed of the corresponding holes is obtained from momentum conservation $v_h m_h = v_e m_e$. Electrons in Fig. 2-3 are distinguished by whether they are associated with light or heavy holes. Since electrons and light holes have similar effective masses, they contribute more to the current than $e$-$hh$ pairs. However, in GaAs, the number of
e-lh pairs near the band edge is smaller than e-hh pairs by about a factor of 3 because of the lower density of states. At 1.6 eV excitation energy, 80% of the injected photocurrent is carried by the electrons. Therefore, for the sake of simplicity, only the electron contribution to the current will be considered for the remainder of this thesis.

The current relaxation time depends on both the rate of inelastic scattering of carriers from phonons, defects, and impurities, and that of elastic scattering between carriers of different effective mass. Current decay is not expected to depend on e-e, hh-hh or lh-lh interactions, since momentum conservation preserves the sum of velocities and therefore the current. On the other hand, e-hh collisions will have the most significant impact of all particle interactions on current decay, but the cross section for this process is typically significantly smaller than for e-e and e-lh interactions. These scattering mechanisms have been calculated using several methods [38]. At room temperature, the calculated e-e scattering rate in GaAs is between 0.4 to 60 ps$^{-1}$ for carrier densities of $\rho = 10^{15}$ to $10^{19}\text{cm}^{-3}$ and the e-lh scattering rate 0.05 to 20 ps$^{-1}$ for $\rho = 10^{17}$ to $10^{19}\text{cm}^{-3}$. The corresponding numbers for e-hh interaction are 0.1 to 2 ps$^{-1}$ for $\rho = 10^{17}$ to $10^{19}\text{cm}^{-3}$. The rate of bare LO-phonon absorption/emission by electrons is 4.5 ps$^{-1}$ and independent of density from $10^{14}$ to $10^{18}\text{cm}^{-3}$. Experimental measurements [37] of e-e and e-LO-phonon scattering times were found to be in agreement with these theoretical calculations.

In high purity GaAs, interactions between quasi-particles of different mass would therefore be the dominant cause for current relaxation at high carrier densities ($\rho > 10^{18}\text{cm}^{-3}$), whereas low density regimes would be dominated by LO-phonon scattering. In LT-GaAs at room temperature, Ralph et al. [39] measured the electron momentum relaxation time to be 185±50 fs for $\rho \sim 10^{15}\text{ cm}^{-3}$, or a scattering rate of 5.5 ps$^{-1}$, in agreement with theory [45]. It is therefore expected that LO-phonon and impurity scattering will dominate the relaxation process in this material for carrier densities up to $\sim 10^{18}\text{cm}^{-3}$.

LO-phonon emission and absorption rates in GaAs by electrons excited above the band gap have been calculated and measured by a number of groups [37][46][47]. At
room temperature, the combined emission/absorption rate in GaAs is $8 \text{ ps}^{-1}$ for electrons with 150 meV excess energy (this corresponds to a two-photon transition at 1.55 $\mu$m) [46]. This interaction is strongly directional, i.e. the scattering probability has an angular FWHM of the order of $1^\circ$ [47]. As a result, each scattering event alters the magnitude of $k$ while preserving its directionality, and the process continues until carriers relax to the band edge. On average, many LO-phonon scattering events will be necessary when the carrier excess energy is large. As a rough approximation, the effective phonon scattering time $\tau_{LO}$ for an electron with excess energies $\Delta E >> E_{LO}$ (the LO-phonon energy $E_{LO} \sim 35$ meV in GaAs) would be

$$
\tau_{LO} \sim \frac{\Delta E}{R_{LO} E_{LO}}
$$

(2.36)

where $R_{LO}$ is the LO-phonon scattering rate. Of course, this expression is not valid if $\Delta E < E_{LO}$ in which case $\tau_{LO} = 1/R_{LO}$.

The current decay time $\tau_c$ is related to the various scattering mechanisms according to

$$
\frac{1}{\tau_c} = \sum_i \frac{1}{\tau_i}
$$

(2.37)

where the $\tau_i$'s are the phonon, impurity and carrier-carrier effective scattering times.

### 2.3.2 The effect current-induced magnetic fields, free carrier absorption and band filling

Time-varying electrical currents are a source of magnetic fields in Maxwell’s equations. In certain geometries, current-induced magnetic fields have important consequences when currents have fast rise-times. Although unidirectional currents have no self-inductance associated with them, the magnetic field thereby created can attract or repel charges moving along the same axis, an effect called ‘magnetic pinching’. The highest peak irradiances reached in the experiments discussed in this thesis are $I_0 = 10 \text{ GW cm}^{-2}$
and $I_{2\omega} = 2 \text{ GW cm}^{-2}$, giving a maximum rate of current injection of the order of $10^{19} \text{ As}^{-1} \text{ cm}^{-2}$ (obtained from Eq. 2.27 and including the Fresnel reflection at the surface of GaAs). Therefore, over a period of 100 fs, the current density can rise to about $10^6 \text{ A cm}^{-2}$. Assuming a uniform current density with a circular cross-section of 50 $\mu \text{m}^2$, the corresponding magnetic field at the edge of this area is about 0.05 T. The cyclotron period for a free electron with an effective mass of 0.067 $m_0$ moving at a speed of $10^6 \text{ ms}^{-1}$ perpendicular to the field is 50 ps, which is much longer than the typical scattering time of the electron ($\sim 100$ fs). In other words, the change in velocity of the electrons on a time scale of 100 fs is negligible. It is therefore expected that magnetic effects, including self-inductance, will not play a major role in altering the photocurrent in these experiments.

Free carrier absorption is also an issue to be considered in these experiments. The reflectivity and absorbance of the electron plasma would increase dramatically if the plasma frequency were to exceed the optical frequency. However, at the highest carrier density reached in the experiments discussed in this thesis ($7 \times 10^{18} \text{ cm}^{-3}$), the plasma frequency is $5.6 \times 10^{14} \text{ s}^{-1}$ compared to $1.2 \times 10^{15} \text{ s}^{-1}$, the frequency of the optical beam at 1.55 $\mu \text{m}$. The corresponding change in reflectivity is only 0.2%. Furthermore, plasma dispersion cannot account for any significant change in the relative phase of the beams over the absorption depth of the $2\omega$ beam ($< 1 \mu \text{m}$). Plasma effects will therefore be neglected in the following discussion and results analysis.

Band filling in semiconductors can also change the indices of refraction, but for carrier densities of the order of $10^{18} \text{ cm}^{-3}$, the expected change in reflectivity at 1.55 $\mu \text{m}$ is only about 1% [48].

### 2.4 Model for current integration

The photocurrent detection technique is described in detail in Chapter 3. This method measures the space charge created by coherent injection of current across an MSM device.
oxide layer

\[ E^\omega, E^{2\omega} \]

electrode

GaAs

\[ \circlearrowleft \rightarrow \text{ballistic mean free path} \]

\[ \circlearrowleft \rightarrow \text{back drift} \]

Figure 2-4: Cross-sectional view of one electrode in the MSM device. Electrons move ballistically underneath the electrode then slowly drift in the presence of an internal bias until they are trapped or recombine with holes. A similar effect occurs for holes near the other electrode.

In essence, each pair of pulses at \( \omega \) and \( 2\omega \) incident on the sample produces a current burst. The resulting potential difference across the electrodes is measured in a steady-state regime, thereby giving information on the integrated current rather than on its time evolution. The reader may refer to Fig. 3-5 for a diagram of the beams and the MSM.

The simplest model for the mechanism of current integration assumes that the electrons in the middle region move and cross the semiconductor/metal interface. As a result, a potential difference appears across the electrodes according the accumulated charge and the device capacitance. However, electron transport through the interface is not straightforward because of the Schottky potential barrier. The MSM structure is effectively a Schottky diode[46]; the rectifying properties of the device are caused by an electrostatic potential barrier at the boundary due to a difference in electronic affinity between the metal and the semiconductor. The barrier height is determined in part by the electron affinity mismatch. Studies have shown that the Au-GaAs interface barrier is 0.5 eV for p-doped GaAs and 0.9 eV for n-doped GaAs, with or without an intermediate oxide layer [49][50].
If electrons were indeed allowed to cross the interface, the effect of the Schottky barrier would be to decrease the current integration efficiency because of the finite tunnelling probability of the electrons and holes. Electrons can easily tunnel through the thin (typically ~5 nm) oxide layer, but the Schottky barrier can extend to about a micrometer.

Since the MSM electrodes and the crystal are not in a 'sandwich-like' geometry and the charge carriers are moving parallel to the sample surface, it is reasonable to suppose that most of them never cross the Schottky boundary but instead are trapped underneath the metallic electrodes. This mechanism would also lead to an electrostatic potential difference across the electrodes proportional to the space charge created under the electrodes. Such an accumulation can occur if the electron mean free path (distance travelled during the effective momentum relaxation time) is greater than the mean back drift path due to the external electric field, as illustrated in Fig. 2-4. In GaAs, the ballistic mean free path is of the order of 70 nm for electrons and holes, whereas the back drift is of the order of 70 pm for 1 ps and 70 nm for 1 ns with an electric field of 100 Vm\(^{-1}\) (typical electric field across the electrodes during the experiment). Carrier trapping times in LT- and regular GaAs are on the order of 1 ps and >10 ns respectively. However, the presence of a high density of surface states created during metal deposition [46] could make the surface carrier lifetime much shorter than 10 ns in normal GaAs.

The equivalent circuit diagram for the MSM and voltage measurement apparatus is illustrated in Fig. 2-5. The voltmeter is in parallel with the MSM, and each has a characteristic resistance and a capacitance. If all the current generated were dissipated through the load resistance of the voltmeter \(R_l\), one could directly extract the average current \(\langle I \rangle = \langle V/R_l \rangle = \langle V \rangle/R_l\) and the peak current \(\sim \langle I \rangle \cdot T/\tau_p\) where \(\tau_p\) is the pulse duration and \(T\) the pulse period. However, because of internal discharge through the sample, only a small fraction of the current flows through the external circuit. High carrier densities after illumination in the semiconductor reduce the resistance significantly, which must be taken into account in the calculation.

In this model, current integration is described in terms of a charge \(q\) accumulated on a
Figure 2-5: Equivalent circuit diagram of the MSM device and voltmeter. Both capacitances and the load resistance are constant but the MSM internal resistance varies with time according to the free carrier density.
Figure 2-6: Evolution of the charge $q$ on the MSM capacitor. Each pair of pulses causes a charge $q_0$ to accumulate on the capacitor. Pulses are synchronised to arrive at $t = 0$ and at every time interval $T$ thereafter.

capacitor comprised of the MSM and voltmeter. The internal and load capacitors $C_{MSM}$ and $C_{load}$ in parallel make up for a total capacitance $C$. The resistors $R_{MSM}(t)$ and $R_{load}$ in parallel provide two discharge channels for the electrons and a total resistance $R(t)$.

We consider a source of current $I(t)$ charging the capacitors during illumination with a periodicity such that $I(t) = I(t + T)$. Between and during pulse excitation, discharge occurs at a rate given by $R(t)C$: the rate equation for $q$ of is therefore

$$q = -q/R(t)C + I(t).$$

(2.38)

$I(t)$ is determined from Eq. 2.33, which yields

$$\dot{I}(t) = \dot{j}^I(t)S - I(t)/\tau_c,$$

(2.39)

where $S$ is the current cross-sectional area.
\( R(t) \) relates to the time-varying carrier density according to

\[
R(t) = \frac{d}{S \epsilon \mu_e \rho(t)},
\]

with \( d \) the electrode spacing. The rate equation for the free carrier density is

\[
\dot{\rho}(t) = -A \rho(t) - B \rho^2(t) + \dot{\rho}'(t),
\]

where \( \dot{\rho}' \) is the combined rate of thermal and optical electron-hole pair generation. \( A \) is the non-radiative and \( B \) the radiative recombination coefficient. Carrier diffusion has been excluded since it is not expected to be an important factor compared to carrier recombination and trapping. The diffusion length can be expressed as \( L = (2D_T)^{1/2} \)
and, with \( D \sim 20 \text{ cm}^2\text{s}^{-1} \) in GaAs, is of the order of a few micrometers for \( T \sim 10 \text{ ns} \). Because the MSM structures are of the same length scale, diffusion will not play an important role in determining the resistance of the device. With no illumination, the carrier density reaches a steady value \( \rho_{th} \) due to thermal excitation. The rate equation for the charge can therefore be written in the form

\[
\dot{q}(t) = -q(t) \left( \frac{1}{R_{MSM}(t)} + \frac{1}{R_{load}} \right) + I(t),
\]

with the steady-state boundary condition

\[
q(t) = q(t + T).
\]

Fig. 2-6 sketches the expected time evolution of \( q \). In general, the average charge \( \langle q \rangle \) on the capacitor is obtained from numerically solving Eqs. 2.41, 2.42 and 2.43, giving an average signal

\[
\langle V \rangle = \frac{\langle q \rangle}{C} = \frac{1}{TC} \int_0^T q(t) dt.
\]

For certain conditions of laser illumination, some useful approximations can be made.
For example, if the pulse duration is much shorter than the free carrier density lifetime, one can assume that the carrier density starts from an initial value \( \rho_o = \rho_\omega + \rho_{2\omega} \) (\( \rho_\omega \) and \( \rho_{2\omega} \) are the contributions of each pulse to the carrier density) and then decays according to Eq. 2.41. This equation can be solved exactly, but this is not necessary if \( \rho_o >> \rho_{th} \), in which case the following solution can be derived:

\[
\rho(t) = \frac{A e^{-\lambda t}}{1 + B \rho_o(1 - e^{-\lambda t})}.
\]  

(2.45)

The expressions for \( \rho_\omega \) and \( \rho_{2\omega} \) are:

\[
\rho_\omega = \frac{(1 - R)^2 I_{2\omega} \tau_{2\omega}^2}{2\hbar\omega}
\]

(2.46)

\[
\rho_{2\omega} = \frac{(1 - R) I_{2\omega} \tau_{2\omega} \alpha}{2\hbar\omega}
\]

(2.47)

where \( \tau_{\omega,2\omega} \) and \( I_{\omega,2\omega} \) are the pulse durations and peak irradiances, and \( R \) is the reflectivity of the semiconductor surface (\( \sim 30\% \) in GaAs for each beam). The square-pulse approximation can also be used to calculate the amount of charge \( q_o \) deposited on the electrodes by each current burst:

\[
q_o = J^\ell S \tau^2 \left( \frac{\tau_p}{\tau_c} + e^{-\tau_p/\tau_c} - 1 \right).
\]

(2.48)

This approximation allows one to solve Eq. 2.42 without the term \( I(t) \).

2.5 Optical effects

The theoretical description of photocurrent injection in semiconductors presented in Section 2.2 assumed coherent, monochromatic plane waves. This assumption is made in order to simplify the Fermi's Golden Rule calculation for the transition amplitudes leading to the current injection rate. In practice, however, sizeable currents are generated only by the high peak irradiances that ultrashort laser pulses provide. These pulses have
a minimum frequency bandwidth determined by their duration. Furthermore, dispersion within the experimental apparatus and the semiconductor cannot be ignored. Propagation through media such as lenses, windows, polarizers and wave-plates introduces pulse broadening, chirping and temporal walk-off between the $\omega$ and $2\omega$ pulses. In phasesensitive experiments, these effects can be crucial, especially with pulse durations $\leq 1$ ps. The large frequency difference between $\omega$ and $2\omega$ also has important consequences on the carrier injection rate, since $2\hbar\omega$ lies above and $\hbar\omega$ lies below the semiconductor bandgap.

As a result, a large refractive index mismatch may cause the relative phase of the beams to vary by a full cycle within only a few micrometers.

### 2.5.1 Frequency doubling in nonlinear crystals

Second harmonic generation of ultrashort laser pulses is generally influenced by dispersion. For monochromatic beams, collinear phase matching requires that $n(\omega) = n(2\omega)$. When a finite bandwidth is involved this condition extends, in a first order approximation, to

$$\frac{dn(\omega)}{d\omega} - \frac{dn(2\omega)}{d\omega} = 0.$$  \hspace{1cm} (2.49)

Here the derivatives are associated with the group velocity of each pulse. Consequently, in the presence of unequal group velocities, phase matching will not be satisfied for the whole pulse spectrum and the $2\omega$ pulse time envelope and spectrum will be affected. Quantifying these effects analytically is a complicated problem involving solving nonlinear coupled-wave equations for propagating pulses, but simplification is possible in some cases.

One can assume that the pump pulse remains undepleted throughout the interaction, an acceptable approximation for the low conversion efficiency in our experiments (less than 10%). One can then identify two regimes for the parametric process [51]: the first when the interaction length $l$ (or crystal length, whichever is shorter) is smaller than $l'$,
and the second when \( l > l' \), where \( l' \) is the walk-off length defined as

\[
l' = \left( \left| \Delta v_g^{-1} \right| \Delta \omega \right)^{-1}. \tag{2.50}
\]

\( \Delta \omega \) is the frequency bandwidth and \( \Delta v_g^{-1} \) is the inverse group velocity mismatch given by

\[
\Delta v_g^{-1} = \frac{1}{c} \left( n(2\omega) - n(\omega) + \omega \frac{dn(2\omega)}{d\omega} - \omega \frac{dn(\omega)}{d\omega} \right). \tag{2.51}
\]

The length \( l' \) corresponds to the distance over which the relative phase of the pulses changes by 1 radian due to dispersion. When \( l < l' \) the frequency doubling process takes place almost the same way as under conditions of perfect group velocity matching: the full spectrum is phase matched (i.e. \( \Delta \omega_2 = 2\Delta \omega_1 \)), the pulses are overlapped in time (no group delay) and \( r_{2\omega} = \tau_\omega / \sqrt{2} \). Since phase matching is satisfied, the phase relationship between \( \omega \) and \( 2\omega \) is preserved across the pulse even in the presence of a frequency chirp.

In situations where \( l > l' \), the pulse time delay at the exit of the crystal can be shown to be

\[
t_d \approx \frac{l}{2\Delta v_g}. \tag{2.52}
\]

As a result of this time delay, the \( 2\omega \) pulse spreads from a minimum time duration of \( \tau_\omega / \sqrt{2} \) to

\[
r_{2\omega} \approx \left( \frac{\tau^2}{2} + \left( \frac{l}{\Delta v_g} \right)^2 \right)^{1/2}. \tag{2.53}
\]

The filtered spectrum of the \( 2\omega \) pulse can be calculated from the phase matching bandwidth of the crystal. For a type I interaction (say, \( e + e \rightarrow o \) as with the BBO crystal used in the experiment for frequency doubling), the bandwidth is given by [52]

\[
\Delta \omega = \frac{(2.4 \times 10^{-10} \text{ms}) \omega}{l |\partial n^e(\omega, \theta)/\partial \omega - \partial n^o(2\omega)/\partial (2\omega)|} \tag{2.54}
\]

For an \( o + o \rightarrow e \) process, the indices associated with \( e \) and \( o \) are interchanged. Figure 2-7 shows the inverse group velocity mismatch (\( \Delta v_g^{-1} \)) in BBO (\( \beta \)-BaB\(_2\)O\(_4\)) type I (curve
1a) for $\lambda = 0.5$ to 2.0 $\mu m$. The curve crosses zero near 2.0 $\mu m$, making it the crystal of choice for frequency doubling in this region of the spectrum. Near 1.55 $\mu m$ the group delay is $\approx 50$ fs/mm: dispersive effects should therefore be taken into account for pulses $< 100$ fs and crystal lengths $> 1$ mm. In our experiment, a 0.6 mm-thick BBO crystal is used with 100 fs pulses and a 1.5 mm-thick KTP (KTiOPO$_4$) crystal is used with 1 ps pulses, so that pulse dispersion is minimized.

2.5.2 Pulse broadening

As the two pulses exit the doubling crystal and propagate through other optical elements, pulse broadening and temporal walk-off take place. One can show that the broadened pulse duration is

$$\Delta \tau(z) \approx (D \Delta \lambda) z$$

where $z$ is the distance propagated, $\Delta \lambda$ is the spectral width and $D$ is the broadening coefficient given by

$$D = \frac{\lambda}{c} \left( \frac{\partial^2 n}{\partial \lambda^2} \right).$$

The effect of $D$ — either compressing or broadening the pulse — depends on the sign of the frequency chirp. For $D > 0$ (normally dispersive medium), a positively chirped pulse will experience increasing chirp and duration, but will start compressing if $D < 0$. The converse situation applies to a pulse with negative chirp. In optical materials of interest here, a transition from normal to anomalous dispersion occurs somewhere between 1.3 and 1.5 $\mu m$. A typical value of $D$ in that spectral region is $\leq 10^{-4}$ s m$^{-2}$. With such a broadening coefficient, 50% stretching occurs within a few millimeters for a 100 fs pulse, or several centimeters in the case of a 1 ps pulse.

Figure 2-7 shows calculated values of $D$ in BBO (ordinary axis), BK7 (common Schott glass) and quartz (ordinary axis), some of the materials used in the experiments presented in this thesis.
Figure 2-7: Pulse broadening parameter $D$ and inverse group velocity mismatch $\Delta v_g^{-1}$ between $\omega$ and $2\omega$ for BBO ordinary axis (a and A), BK7 (b and B) and quartz ordinary axis (c and C). The sign of $D$ is chosen so that a positive value will broaden a positively chirped pulse; positive $\Delta v_g^{-1}$ corresponds to a $2\omega$ pulse travelling faster than an $\omega$ pulse. Results for the extraordinary axis of BBO do not differ significantly.
2.5.3 Current injection with chirped and delayed pulses

To a first approximation, a frequency chirp in a pulse can be expressed by a linear term such that [53]:

$$\omega(t) = \omega_0 + 2bt.$$  \hfill (2.57)

with $\omega_0$ the center frequency and $b$ the chirp parameter. The relationship of $b$ to the pulse duration $\Delta\tau$ and frequency bandwidth $\Delta\omega$ is given by

$$b = \frac{2\ln 2}{\tau^2} \left[ \left( \frac{\Delta\tau \Delta\omega}{4 \ln 2} \right)^2 - 1 \right]^{1/2}.$$  \hfill (2.58)

Strictly speaking, this expression applies only to Gaussian pulses, and changes slightly for a sech$^2$ time envelope. Hence, if $b_\omega$ and $b_{2\omega}$ are the chirp parameters for $\omega$ and $2\omega$ and the pulses are delayed relative to each other by a time $t_d$, the phase difference as a function of time will be

$$\Delta\phi(t) = \phi_\omega(t + t_d) - \phi_{2\omega}(t) = 2b_\omega(t + t_d)^2 - 2b_{2\omega}t^2.$$  \hfill (2.59)

When pulse broadening is relatively small, one can assume that $b_{2\omega} \approx 2b_\omega$. since $\omega$ and $2\omega$ are phase-related. In this case, Eq. 2.59 becomes

$$\Delta\phi(t) = 2b_\omega(2t_dt + t_d^2).$$  \hfill (2.60)

Therefore, a general criterion for chirping to be an important factor in the current injection rate is that:

$$4b_\omega t_dt_\omega > 1.$$  \hfill (2.61)

This corresponds to a phase variation greater than 1 radian within the pulse duration. Note that $\Delta\phi$ remains constant if $t_d = 0$. 

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The rate equation for the current injection can be written

\[ j' = \eta |E_\omega|^2 |E_{2\omega}| \sin(\Delta \phi) - \frac{J}{\tau_c}, \quad (2.62) \]

which becomes

\[ j'(b_\omega, t_d) = 2^{5/2} \eta (\mu_0 / \varepsilon_0)^{3/4} n_\omega n_{2\omega}^{1/2} I_\omega(t) I_{2\omega}^2(t - t_d) \cos(4b_\omega t_d) - \frac{J}{\tau_c}. \quad (2.63) \]

where \( \eta \) is the proper tensor element of \( \tilde{n} \). Any delay between the pulses will then reduce the amount of current injection because of non-ideal pulse overlap and reduce the integrated current because of variation in \( \Delta \phi \). Fig. 2-8 shows the time dependence of the current density as a function of time delay for moderately chirped pulses (\( \Delta \tau \Delta \omega / 2\pi = 0.66 \)). Curve (a) is not centered at zero because of the assumed finite current relaxation time. As the delay increases, the current alternates and quickly averages to zero. Indeed, as shown in Fig. 2-9, the integrated current for chirped and delayed pulses (curve b and c) drops considerably faster than for transform-limited pulses (curve a).

### 2.5.4 Effect of semiconductor refractive index mismatch

Another dispersion-related effect that can appear within the semiconductor is spatial variation of the phase due to a refractive index mismatch. As the beams propagate, the injected current can change direction as the relative phase varies:

\[ \Delta \phi(z) = \frac{4\omega}{c} (n_{2\omega} - n_\omega) z = \frac{4\omega \Delta n}{c} z, \quad (2.64) \]

where \( z \) is the distance along the propagation axis. On the other hand, the pulse amplitude is being attenuated according to

\[ \frac{dI_\omega}{dz} = -3I_{2\omega}^2 \quad (2.65) \]
Figure 2-8: Time evolution of the current density for delay 0 (a), 0.5 (b) and 1 (c) in units of the pulse time duration \( t_\omega = \tau_{2\omega} \). The scattering time is \( t_\omega/2 \) and \( \Delta t \Delta \omega/2\pi = 0.66 \).

Figure 2-9: Integrated current density as a function of time delay for time-bandwidth products of 0.44 (a), 0.66 (b) and 0.88 (c). The carrier scattering time is assumed to be \( t_\omega/2 \).
and
\[
\frac{dI_2}{dz} = -\alpha I_2. \quad (2.66)
\]

The averaged current injection is therefore given by
\[
j_{\text{avg}} \propto \int_0^d I_\omega(z) I_2^{1/2}(z) \sin(\Delta \phi(z) + \phi_o) dz, \quad (2.67)
\]

where \( d \) is the depth over which the current can be collected, or that in which one of the beams is absorbed, whichever is smaller. The quantity \( \phi_o \) is the relative phase of the beams at \( z = 0 \). The effect of \( \Delta \phi(z) \) is therefore to reduce the overall current integration efficiency. One can show that the ratio between the current integration efficiency for \( \Delta n \neq 0 \) and that for \( \Delta n = 0 \) is
\[
\frac{\gamma}{(\gamma^2 + \delta^2)^{1/2}},
\]

where \( \gamma = 3I_\omega + \frac{1}{2} \alpha \) and \( \delta = \frac{k \omega}{\rho} \Delta n \). Using the parameter values listed in Table 2.1 and \( I_\omega = 1 \) GW cm\(^{-2}\), one obtains a ratio of 0.26.
Chapter 3

Apparatus and Experimental Techniques

3.1 Overview

The experimental techniques and the apparatus used to generate and detect coherent photocurrents in bulk GaAs are discussed in this chapter. Phase-sensitive experiments such as this one require the proper choice of optical source, beam phase properties, semiconductor characteristics and current detection geometry, each of which will be described in detail. As illustrated in Fig. 3-1, the general experimental layout consists of four parts: the source of ultrashort light pulses at $\omega$ and $2\omega$, a means for controlling the relative phase of the beams, the current generation/detection device and the beam wave front analysis apparatus.

Pico- and femtosecond pulses were used in order to obtain high enough electric fields to produce sizeable currents while having a pulse energy below the damage threshold of the material. Optical parametric sources were found to be appropriate for this purpose because tunable radiation is also required to adjust the photon energy to an appropriate value relative to the semiconductor bandgap ($\hbar\omega < E_g < 2\hbar\omega$). These optical sources are described in the next section. Measurements were also carried out for the nondegenerate
Figure 3-1: Schematic diagram of the experimental apparatus. An optical source produces phase-related beams with tunable wavelengths; the relative phase of the pulses is controlled while the photocurrent is measured. The beam quality is monitored using a phase front analysis device.

Condition with beams at frequency $\omega_1, \omega_2$ and $\omega_3 = \omega_1 + \omega_2$ such that $\hbar \omega_1, \hbar \omega_2 < E_g < \hbar \omega_3$ using the same optical sources and similar apparatus. The importance of phase uniformity is discussed as well as the technique used for beam phase front analysis. As in any phase-sensitive experiment, good knowledge of the temporal and spatial phase of the beams is paramount. MSM structures were used to integrate the current produced via the coherent process. Since carrier density lifetime is an important parameter to consider, two samples of GaAs were prepared with different carrier recombination times: the first is GaAs grown at low temperature (~1 ps lifetime) and the other is regular GaAs (>10 ns lifetime).
Figure 3-2: Diagram of the externally pumped optical parametric oscillator. The KTP-based resonator is angle-tuned and produces signal pulses of 1 ps duration with up to 200 mW average power at 82 MHz repetition rate. The output coupler (OC) has a transmittance of 2%. A piezoelectric transducer (PZT) is used for fine adjustment of the cavity length.
Figure 3-3: Diagram of the femtosecond optical parametric amplifier (Coherent OPA 9400) converted to produce infrared signal and idler pulses. White light continuum is generated inside a sapphire crystal (S); part of the pump beam is polarization-rotated by 90 degrees through a half-wave plate (W); dichroic mirrors D1 and D2 allow the pump and white light to be mixed in the BBO crystal and then separated for double-pass amplification. Arrows represent variable delays.

3.2 Optical sources

Tunable optical parametric sources producing ultrashort light pulses were used in these experiments. The main differences between the two sources used – an optical parametric oscillator (OPO) and an optical parametric amplifier (OPA) – are the repetition rate, the pulse duration and pulse energy. The synchronously-pumped OPO has a high repetition rate which facilitates current integration because the shorter pulse period gives less time for the MSM to discharge. However it delivers less peak power for the same average power than the lower repetition rate OPA. Either source may therefore be more appropriate for specific MSM devices and substrate characteristics.
3.2.1 Synchronously pumped optical parametric oscillator

The source of picosecond pulses used for the experiments is a synchronously pumped optical parametric oscillator (OPO) [54] similar to a femtosecond version built and characterized earlier in the author's laboratory [55][56]. The system consists of an argon-ion laser (Spectra Physics BeamLok) pumping a continuous wave, actively mode-locked titanium-doped sapphire laser (Ti:S) (Spectra Physics Tsunami) producing 1.8 ps FWHM pulses near 790 nm and at a repetition rate of 82 MHz. The latter laser is used as an external pump source for the OPO and delivers up to 2.3 W of average power between 720 and 850 nm in a near TEM\textsubscript{00} mode.

The OPO is singly resonant (signal beam only), uses KTP as a parametric gain medium and is angle-tunable (Fig. 3-2). The crystal is 7 mm-thick with AR coating at 1.55 μm on each face and cut at θ = 52° (φ = 0°). Broadband dielectric mirrors are used for the high reflectors and the 2% output coupler. In order to maximize conversion efficiency, phase matching is achieved in the x-z plane of the crystal. This reduces the walk-off angle between the pump and the signal beams, therefore preserving good overlap throughout the crystal. Since the direction of the beam energy flow (Poynting vector) in an anisotropic medium is not always parallel to the beam wavevector, phase matching does not necessarily imply collinear propagation. Angle compensation with non-collinear pumping geometry ensures beam overlap for a distance of a few millimeters. The OPO signal is tunable from 1.44 to 1.64 μm with an average power up to 200 mW for pump power of 2.2 W (the pump threshold is 800 mW). Potential tunability extends from 1.38 to 1.85 μm with additional mirror sets. Intracavity loss through the mirrors and at the crystal surfaces is estimated to be 1.5%. The idler beam exits the crystal at an angle relative to the signal beam and is not used. The signal beam profile is close to a TEM\textsubscript{00} mode.

Autocorrelation measurements of the signal pulse duration gave a typical FWHM of 1.1 ps. The spectral FWHM of 3.5 nm gives a time-bandwidth product of 0.48, close to the Fourier transform limit of 0.44 for Gaussian pulses.
3.2.2 Optical parametric amplifier

A second set of experiments was carried out using a femtosecond optical parametric amplifier (OPA) (Coherent Model 9400). The author modified this system - originally designed to produce tunable light in the visible region - into a near-infrared source tunable from 1.3 to 2.1 μm for the signal and idler beams. The system is constituted as follows. A cw passively mode-locked Ti:S laser (Coherent Mira 900) seeds a regenerative amplifier (Coherent RegA 9000) which brings the 100 fs pulses from a repetition rate of 76 MHz down to 250 kHz, with a 500-fold increase in peak power. The Mira laser wavelength is tunable from 750 to 850 nm and yields an average power of up to 1.1 W.

Fig. 3-3 shows a diagram of the OPA: the pump beam passes through a 1:3 beam splitter. The weaker beam is focussed onto a sapphire crystal to achieve white light continuum generation, while the other beam is polarization-rotated by 90°. The two beams are then recombined and focussed onto a BBO crystal to produce infrared light via down-conversion. Infrared light is created from difference frequency mixing between the strong pump and the white light. A second pass improves the conversion efficiency, yielding typical average powers of 30-50 mW for the signal and idler beams combined for a pump power of 1 W.

Because of the spatial structure of the white-light continuum and the double pass arrangement of the OPA source, the beams are of much lower quality than that from the OPO, and they generally present some structure unless care is specifically taken to improve them. In general, it is possible to obtain close to TEM00 modes by minor tweaking, but this usually entails sacrificing some power. The best conversion efficiency occurs in a regime where the pump beam is close to self-focussing in the BBO, but it is desirable to stay away from that condition in order to prevent beam fragmentation.

Autocorrelation measurements gave values for the signal pulse duration of 110 to 180 fs FWHM (assuming a Gaussian pulse shape), depending on the pumping conditions. The regenerative amplifier pulse duration has a tendency to vary on a daily basis because of its high sensitivity to the seed pulse bandwidth. Typical signal pulse bandwidths are 30
nm. consistently giving chirped pulses with \(0.6 \leq \Delta \tau \Delta \omega / 2\pi \leq 0.9\).

### 3.3 Photocurrent measurement techniques

Detecting the photocurrent constituted the greatest experimental challenge. Indeed, for electron scattering times comparable to or smaller than the laser pulse duration (~100 fs), the injected current is expected to last only as long as the pulse itself, i.e. a few hundred femtoseconds. Since measuring currents on a sub-picosecond time scale is beyond the reach of conventional electronics, the only viable option was current integration.

Although time-resolution of photocurrent should be possible on a longer time scale, several earlier unsuccessful attempts at using high intensity nanosecond pulses showed that sample damage occurs well before a current can be measured. Experiments with a Q-switched YAG (10 ns and 100 mJ per pulse) and an infrared parametric converter (4 ns, 1550 nm and 1 mJ per pulse) gave no measurable signal. Early attempts at measuring integrated currents were also made using superlattices of asymmetric quantum wells [24]. Radiative recombination of electron-hole pairs in wells of different width lead to two distinguishable peaks in the photoluminescence spectrum. It was hoped that coherent generation of photocurrent between the wells would lead to an asymmetric probability for the electrons to recombine in either well, therefore leaving a signature in the luminescence spectrum. However, the weakness of two-photon absorption in the middle layer and the difficulty of aligning the electric field of the beams along the growth direction caused the author to seek better alternatives for current detection.

Detection of photocurrent was made possible using MSM devices in which the current is generated in the middle gap and the charge carriers are trapped inside or beneath the metal electrodes. Pulses coming in at a high repetition rate allow space charge to accumulate which is measured in form of a steady-state potential difference across the electrodes.

The experimental setup involving the OPO is shown in Fig. 3–4. The second harmonic
Figure 3-4: Diagram of the experimental setup for the coherent control of photocurrent using the picosecond OPO. Double-pass through a BK7 glass window is used to vary the phase of the two beams. M1 and M2 are curved mirrors with focal lengths of 10 and 2.5 cm respectively.
is obtained in a 0.6 mm-thick BBO crystal via type I phase matching \((\sigma + \sigma \rightarrow e)\). Gold-coated reflective focusing optics (M1 and M2) are used beyond this point in order to obtain good phase front quality. A BK7 glass plate (1.16-mm thick) is placed in the path of the beams and is rotated to control the phase of the beams; a double-pass geometry ensures steady beam positioning for different angles of incidence. The polarization of the \(\omega\) beam alone is then rotated by 90° using a half-wave plate at 1.55 \(\mu\)m (full-wave plate at 0.775 \(\mu\)m). Lock-in amplification is used to measure modulation of the MSM signal with a beam chopping frequency of about 400 Hz. Fig. 3-5 sketches the MSM electrodes with the focused beams. The same experimental layout was used for some of the measurements taken with the OPA. For the demonstration of coherent control in regular GaAs, rotation of the glass plate was automated using a stepper motor. An analog/digital card (National Instrument, model AT-MIO-16) was used to send data directly to a PC computer. This improvement significantly increased the data acquisition rate so that multiple scans and averaging could be done.

After the above setup with the femtosecond OPA source was used for a few experiments, an improvement was made on the technique of phase control by using a Michelson interferometer. As shown in Fig. 3-6, a dichroic mirror (D) splits the beams and a piezoelectric transducer (PZT) is used to control the phase of the beams by adjusting the length of one arm. This technique allows for background-free measurements with rapid phase scan. Instead of using lock-in amplification with a beam chopper, detection is made by modulating the phase with the PZT. A 35 mV square wave voltage is applied to the piezo controller (Physik Instrumente, High Voltage Power Supply P-263) using a function generator (Stanford Research Systems, model DS345) to give a displacement amplitude of \(\lambda/4\) (or a total of \(\lambda/2\) path change). Such a displacement makes the phase \(\Delta\phi\) vary by half a cycle and allows the signal extrema to be measured in a single sweep. A modulation frequency of 85 Hz was found to be optimum for this purpose. Although the PZT displacement may not be perfectly linear at that frequency, the signal periodicity would be preserved. Another advantage of using an interferometer for controlling the
Figure 3-5: View of the excitation beams and the MSM electrodes. The focal spot size is made larger than the gap size so as to expose the edges of the electrodes. The sample is positioned using a micrometer to maximize the signal and minimize the background.
Figure 3-6: Cascaded frequency doubling incorporated into the experimental setup to monitor the beams' phase front quality. The $2\omega$ beam is projected onto a screen after being generated in the BBO and KTP crystals. A dichroic mirror (D) splits the $\omega$ and $2\omega$ beams while one arm of a Michelson interferometer is adjusted to control the phase using a piezoelectric transducer (PZT).

phase is that the time delay between the pulses can be adjusted, eliminating the need for reflective optics, which were used to prevent pulse temporal walk-off caused by group velocity dispersion.

Non-degenerate coherent control of photocurrent was also demonstrated using the OPA setup of Fig. 3-6, with the exception that the positions of the BBO and KTP crystals are switched. A thicker (1.5 mm) KTP is used in a type II phase matching geometry ($o(signal) + e(idler) \rightarrow o(pump)$) to create sum-frequency light at 0.794 $\mu$m. The spectrum of this light was measured using an optical multi-channel array (EG&G PARC silicon detector, model 1455). Temporal and spatial overlap of the three pulses were obtained via sum-frequency mixing in BBO ($e(signal) + o(pump) \rightarrow o(sum-frequency)$). Average powers of the beams were measured with a Newport Optical Power Meter, Model 835, with silicon and germanium photodiodes.
3.4 Samples

The experimental results presented in the next chapter are based on the two samples described below. They are the only two types of semiconductors for which the author has been able to measure coherent photocurrents.

Sample #1

The first sample consists of an epilayer of GaAs grown at low temperature (hereafter referred to as LT-GaAs). The undoped 1.0 μm-thick layer was epitaxially grown at 200°C with (100) orientation onto a regular GaAs substrate with a 80 nm-thick AlAs buffer layer in between. The non-stoichiometric material has 2% excess arsenic which leaves a high density of defects after annealing (600 °C for 10 min.). Carrier lifetime of ~ 1 ps [57] and high resistivity (> 10^6 Ω cm) [58] arise as a result of the midgap states made available by these defects.

The buffer layer allowed for lift-off of the epilayer from the substrate using an acid etching technique. Since AlAs is dissolved by a hydrogen fluoride (HF) solution at a much faster rate than GaAs, the thin film could be removed from the substrate and then van der Waals-bonded onto a sapphire substrate. Absorption spectroscopy was then carried out (Bomem FT-Raman Spectrometer) to determine the layer’s exact thickness.

On another identical substrate, several pairs of 200×250 μm gold electrodes of thickness 170 nm and with gaps of 5, 10, 25 and 50 μm were deposited on the sample using photolithography. No particular care was taken to produce ohmic contacts, resulting in dark resistances across the electrodes varying from 14 MΩ to 4.5 GΩ for the 5 and 50 micron gaps respectively. This corresponds to an estimated resistivity for the material of about 10^8 Ωm. I-V curves for these samples show deviations from linear behaviour for biases exceeding a few volts, as shown in Fig. 3-7. Wire bonding was done to connect the metal pads to the sample holder. Fig. 3-8 shows the electrode pattern of the MSM device.

Sample #2

Similar photolithography was carried out for an undoped, high mobility, normally-
Figure 3-7: Current-voltage for the 50 μm LT-GaAs MSM electrodes. The solid curve is the best fit with a third-order polynomial.

Figure 3-8: Electrode pattern of the MSM sample obtained via photolithography.
grown GaAs substrate with (100) orientation. Identical electrodes with non-ohmic contacts were deposited on the 350 μm-thick layer. The electron-hole recombination time in such a material is typically $> 10$ ns for carrier densities below $10^{18} \text{cm}^{-3}$. The resistance across the gaps varies from $10^6 - 10^9 \ \Omega$.

### 3.5 Beam wave front analysis

Since the current injected into the semiconductor depends on $\Delta \phi = 2\phi_\omega - \phi_{2\omega}$, it is very important to preserve the beam phase front quality in this experiment. Spatial variations of $\Delta \phi$ across the beams would quickly average the current injection to zero and this is especially true if measurements are made over the entire area.

If phase matching is satisfied, the value of $\Delta \phi$ inside and at the exit of the doubling crystal is $\pi/2$ across the whole beam. The phase is fixed because the second harmonic beam starts with zero intensity and its phase is determined by the initial value of $\phi_\omega$. Therefore, phase distortions in a non-ideal mode for the $\omega$ beam are not of great concern. However, after the beam travels through and reflects from various optical elements in the apparatus, the phase front can experience severe distortions. High flatness optics are generally used in coherent control experiments to prevent this from occurring ($\lambda/5$ surface flatness or better).

Cascaded frequency doubling constitutes a good diagnostic tool to determine the beams' relative phase. It consists of generating second harmonic light in one crystal and then mixing the $\omega$ and $2\omega$ beams in a second crystal. This technique was used by Chudinov et al. to measure the phase of two harmonically-related beams [59]. The overall conversion efficiency of $\omega$ into $2\omega$ depends on the value of $\Delta \phi$ when the waves enter the second crystal. As a result, any relative phase non-uniformity will become apparent in the far field of the $2\omega$ beam. This process can be understood by considering
Figure 3-9: Second harmonic electric field amplitude versus distance of propagation in the second nonlinear crystal of the cascaded frequency doubling. Perfect phase matching is assumed. The two opposite cases, $\Delta \phi = \pi/2$ and $\Delta \phi = -\pi/2$, correspond respectively to a continued amplification of the $2\omega$ beam and conversion back into an $\omega$ beam.

The coupled-wave equation for frequency doubling[60]

$$\frac{dE_{2\omega}}{dz} = \frac{16\pi \omega^2 d_{\text{eff}}}{c^2 k_{2\omega}} E_{2\omega}^2 e^{i\Delta k z}$$  \hspace{1cm} (3.1)

with $d_{\text{eff}}$ the effective nonlinear coefficient for second harmonic generation; $k_{2\omega}$ is the wavevector for $2\omega$ and $\Delta k = 2k_\omega - k_{2\omega}$. The electric fields $E_\omega$ and $E_{2\omega}$ are complex. In the undepleted pump approximation ($|E_\omega|$ constant), Eq. 3.1 reduces to

$$\frac{d |E_{2\omega}|}{dz} = -C |E_\omega^2| \sin(\Delta k z - \Delta \phi)$$  \hspace{1cm} (3.2)

where $C$ is a real, positive constant. In other words, if the gain is negative, $2\omega$ is converted back into $\omega$ and $E_{2\omega}$ decreases to zero, and increases from that point. Fig. 3-9 illustrates two extreme situations, with $\Delta \phi = \pm \pi/2$.
As a result of phase variations, one can notice brighter and darker fringes appearing in the far field of the beams. The contrast ratio between the fringes is maximum when $E_{2\omega} = 0$ for $\Delta \phi = -\pi/2$ at the exit of the second crystal, a condition which is satisfied if $d_1 |E_\omega| l_1 = d_2 |E_{2\omega}| l_2$, where $l$ is the crystal length and the labels 1 and 2 refer to the first and second nonlinear crystal. The observed fringe pattern therefore reflects the current injection profile across the beam area.

The fringe pattern is extremely sensitive to the relative directionality of both beams. Expressed in terms of the angle between the beams $\Phi$ and the distance to the center of their overlap $r$, the phase $\Delta \phi$ takes the form

$$\Delta \phi(r) = \frac{8\pi r}{\lambda} \sin \Phi$$

where $\lambda$ is the fundamental wavelength. The fringe spacing is therefore

$$s = \frac{\lambda}{4 \sin \Phi}$$

For example, if $\Phi = 1^\circ$ and $\lambda = 1.55 \mu m$, we obtain $s = 22 \mu m$, a width smaller than a typical focal spot size.

Fig. 3-6 shows the experimental setup for phase front diagnostics. The cascaded frequency doubling device is incorporated in the coherent control apparatus for quick diagnostics of the beam quality. One of the two mirrors of the interferometer and the dichroic beam splitter (D) are used for alignment of the two beams. Approximate spatial and temporal overlap is first obtained by sum frequency generation in KTP. The process is of type II with $\omega(e) + 2\omega(e) \rightarrow 3\omega(o)$. It is convenient to use KTP since the same crystal can be used to produce third harmonic and cascaded doubling, with phase matching angles of $\theta = 49^\circ$ and $53^\circ$ respectively for $1.55 \mu m$. This technique generally does not provide adequate parallelism, i.e. no interference pattern can be observed. The KTP crystal is then rotated to produce SH from the fundamental and fine adjustments of the mirrors are made until the fringes appear. The procedure is repeated until good
spatial overlap (good third harmonic efficiency) and parallelism (good fringe pattern) are obtained. Fine phase control is achieved using a piezoelectric transducer (displacement of 30 nm/V). The SH beam is projected in the far field onto a screen and inspected using an infrared viewer (FJW Find-R-Scope).

Fig. 3-10a shows the OPA $2\omega$ beam profile as incident on the MSM sample, and produced via type I conversion in BBO. whereas Fig. 3-10b shows the same beam after passing through a second doubling crystal (KTP, type II, $o + e \rightarrow o$). A $500 \times 500$ silicon CCD camera (Spiricon Beam Profiler) was used for these measurements. In this case, beam alignment was done carefully to produce only two visible fringes over the whole focal spot size ($\sim 50 \mu m$). Since the angle of incidence of the beams on the KTP crystal is $\sim 1^\circ$, the internal angle difference between the beams is negligible. However it could become significant for angles $> 5^\circ$. The fringe pattern can be seen moving perpendicular to the fringe orientation as $\Delta \phi$ is scanned with the PZT. Depending on the size of the electrode gap being used, it desirable to obtain at most a few interference cycles in order to measure any injected current on the MSM to prevent the current from averaging to zero.
Chapter 4

Results and discussion

This chapter gives the results of coherent control experiments performed in bulk GaAs at room temperature. This semiconductor was chosen because its band structure allows direct transitions with light in the near infrared region of the spectrum, appropriate for our optical sources. The basic demonstration of photocurrent control in LT-GaAs and normally-grown GaAs using picosecond and femtosecond optical pulses is shown, as well as non-degenerate coherent control in LT-GaAs. Two potential artifacts to the coherent control signal – cascaded frequency doubling and third-order optical rectification – are ruled out through measurements of second harmonic generation, polarization and optical power dependence. Other aspects are investigated, such as the polarization dependence of the current injection, the effect of pulse chirping and the influence of sample and beam parameters. Attempts to generate and control photocurrents in bulk silicon are also briefly discussed.

4.1 Photocurrent coherent control in LT-GaAs

4.1.1 Experiments with picosecond pulses

The author first performed measurements of coherent control on the LT-GaAs sample (sample #1) using a picosecond OPO and the experimental setup shown in Fig. 3–4. Two-
Figure 4-1: Induced steady-state voltage on a 25 \( \mu \)m gap LT-GaAs MSM in the presence of the \( \omega \) beam (circles), the \( 2\omega \) beam (triangles), and both beams (squares) from the OPO as a function of angle of incidence of the beams on the glass plate. Except for the squares, the size of the data points is representative of the experimental error.

Photon excitation is achieved using an \( \omega \) pulse at 1.55 \( \mu \)m and single-photon excitation using \( 2\omega \) at 0.775 \( \mu \)m, with average powers of 150 mW and 40 \( \mu \)W respectively. The corresponding peak irradiances at the sample surface are 30 MW cm\(^{-2}\) and 9 kW cm\(^{-2}\) producing contributions to an initial carrier density of \( \rho_\omega = 2 \times 10^{13} \) and \( \rho_{2\omega} = 6 \times 10^{14} \) cm\(^{-3}\). The polarization of each beam is oriented across the MSM gap (along the x-crystallographic axis) so as to make use of the largest tensor element, \( \eta_{zzz} \). The beams are individually attenuated using Schott RG1000 filters (\( 2\omega \) beam) and a dielectric high reflector or Schott KG3 filters (\( \omega \) beam) to verify the contribution of each to the MSM photoresponse.

Fig. 4-1 shows the voltage across the MSM as a function of the incidence angle of the beams on the glass window. When the beams are individually incident on the sample, the voltage is practically constant; when the two are simultaneously present.
Figure 4-2: Coherent control signal from a 5 μm MSM with LT-GaAs versus Δφ. The constant background (offset) has been removed and the data fitted to a sine function using reported values of the indices of refraction for BK7.
voltage modulation is observed. Because the photoresponse of the MSM depends on the position of the beams, all three curves have different background levels which slowly drift with the angle of incidence. In an ideal situation this would not happen, but a slight misalignment causes the beams to shift as the window is rotated. The changing amplitude of the modulated curve is also partly related to beam positioning and to OPO power fluctuations during the time taken to complete one scan (4 to 5 min.).

Fig. 4-1 represents raw data as they are measured with the lock-in amplifier. They present a variable periodicity which is expected from the dispersive property of a rotated flat window. It is necessary to take into account the plate's thickness $l_p$ and indices of refraction $n_{p,\omega}$ and $n_{p,2\omega}$ in order to extract the phase difference between the $\omega$ and $2\omega$ beams. In an isotropic medium such as glass, the phase difference $\Delta \delta$ as a function of angle of incidence $\theta$ is given by

$$\Delta \delta = \frac{2l_p\omega}{c} \left[ \frac{n_{p,\omega}}{\cos \theta_\omega} - \frac{n_{p,2\omega}}{\cos \theta_{2\omega}} - \sin (\tan \theta_\omega - \tan \theta_{2\omega}) \right]$$  (4.1)

for a single pass. The internal angle of propagation of the beams, $\theta_\omega$ and $\theta_{2\omega}$, are related to the angle of incidence $\theta$ according to

$$\sin \theta_{\omega,2\omega} = \frac{1}{n_{p,\omega,2\omega}} \sin \theta.$$  (4.2)

We see that at normal incidence ($\theta = 0$), the derivative $\frac{d\Delta \delta}{d\theta}$ is zero but increases steadily thereafter. We used Eq. 4.1 to plot again the voltage as a function of the real phase difference $2\phi_\omega - \phi_{2\omega}$ for the data taken from the 5 $\mu$m gap MSM device (Fig. 4-2). In this figure, the constant background removed and the data are fitted to a sine curve using the measured thickness of the plate and indices of refraction of BK7 glass: $n_{p,\omega} = 1.5013$ and $n_{p,2\omega} = 1.5118$ [61]. Even though the fit is highly sensitive to the value of these parameters, they always lie within the experimental error of the thickness and the uncertainty of $n_{p,\omega,2\omega}$ due to the laser bandwidth.

Fig. 4-3 shows coherent control signal from various electrode pairs on the LT-GaAs
Figure 4-3: Coherent current control signal from various pairs of electrodes on the LT-GaAs sample: MSM gap sizes are 5 μm (crosses), 25 μm (circles) and 50 μm (squares). Experimental errors are represented by size of the data points.

...sample and for conditions of illumination similar to that used for Fig. 4-1. It is interesting to note that the phase at \( \theta = 0^\circ \) for each curve is different. Using Eq. 4.1, one can find the initial phase to be 70°, -11° and -45° for the 5, 25 and 50 μm gaps respectively. This is due to the fact that the signal depends on the phase of the beams at the very edge of the electrodes, since only the free carriers within a mean free path or so of the boundary will be collected. Therefore, any phase front non-uniformity will make the signal initial phase to be different for each gap width. For the same reason, the curve for the 5 μm gap appears smoother, since the smaller gap is exposing a fairly narrow and uniform area of the beam. The MSM geometry makes the photocurrent integration intrinsically anisotropic; it should be sensitive only to the \( x \)-component of \( \vec{J} \) because charges need to separate in a direction perpendicular to the gap in order for a voltage to be measured. As expected, no signal modulation is measured when the polarization of the beams are aligned parallel to electrode edges, as shown in Fig. 4-1.

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Figure 4-4: Coherent control signal on a 25 \( \mu \text{m} \) MSM with LT-GaAs for polarizations across (dots) and parallel (squares) to the gap. Background has been removed.
Figure 4-5: Voltage modulation amplitude versus fundamental beam average power $P_{\omega}$. Since the beam is attenuated before the BBO doubling crystal, the $2\omega$ beam power varies quadratically with $P_{\omega}$. The solid line is a fit of the form $y = ax^n$, with $n = 1.6 \pm 0.2$.

To verify the nonlinear dependence of the coherent control signal on the beam power, the modulation amplitude was measured as a function of the fundamental beam average power $P_{\omega}$ before the doubling crystal. From a simple analysis, one would expect the dependence to be quadratic. Indeed, since the current injection is $\dot{J} \propto |E_{\omega}|^2 |E_{2\omega}|$ and $|E_{2\omega}| \propto |E_{\omega}|^2$ from the parametric up-conversion, then $\dot{J} \propto |E_{\omega}|^4 \propto P_{\omega}^2$. However, as shown in Fig.4-5, the signal amplitude on a 25 $\mu$m MSM device is proportional to $P_{\omega}^{1.6\pm0.2}$ for $40 < P_{\omega} < 150$ mW. Although the range of power is not large (below 35 mW the signal becomes difficult to measure), the error in the exponent is not greater than 0.2. To explain this discrepancy, it could be argued that the measured signal reflects the average space charge and not directly the magnitude of the current generated. Indeed, the creation of space charge involves additional effects such as carrier recombination and displacement in the presence of an electric field. Nevertheless, numerical calculations based on the model of Section 2.4 also predict that with the high repetition rate of the OPO and the
short carrier lifetime of LT-GaAs, the dependence of the coherent control on \( P_\omega \) would follow closely a second power law. Plotted in Fig. 4-6a is the calculated signal amplitude from a 10 \( \mu \)m gap MSM versus the average power incident on the sample. At lower \( P_\omega \), the power dependence is exactly quadratic, but saturation occurs as power increases. Over the range of power \( 40 < P_\omega < 150 \) mW, the dependance of the voltage is \( P_\omega^{1.9} \). The model explains this effect in terms of an increased conductivity in the sample causing the MSM to discharge faster and making it a less efficient integrator. It is not possible at this point in time to determine whether the slight disagreement between theory and experiment comes from additional physical effects not included in the model, or simply from the uncertainty in the values of the parameters used in the model. Some parameters, such as the linear and two-photon absorption coefficients, influence the calculated power dependence. A better knowledge of the material and device parameters would therefore be needed to solve that question.

We now estimate the peak current density generated in the experimental results of Fig. 4-1. Theory (Eq. 2.27) predicts a peak current density as high as 50 \( \mu \)A\( \mu \)m\(^{-2}\), using \( \eta_{xxx} = 20 \text{s}^{-2}\text{mCV}^{-3} \), \( \tau_c = 100 \text{fs} \), \( I_\omega = 30 \text{MWcm}^{-2} \) and \( I_{2\omega} = 9 \text{kWcm}^{-2} \). However, the curve in Fig. 4-1 presents a voltage modulation of 200 \( \mu \)V. This bias across the load resistance of the lock-in amplifier corresponds to an average current of 2 pA or, with a pulse duration of 1 ps at 82 MHz repetition rate, a peak current of approximately 20 nA (1 nA\( \mu \)m\(^{-2}\) peak current density, assuming \( \alpha = 1.5 \times 10^6 \text{m}^{-1} \) and 75 \( \mu \)m beam focal spot size). However, this value represents the most conservative estimate since most of the current does not pass through the voltmeter but is dissipated internally. In order to evaluate the total current generated coherently, a calculation must be carried out based on the model presented in Section 2.4. Using the values listed in Table 2.1 for the parameters for LT-GaAs, the estimated real peak current density is 100 nA\( \mu \)m\(^{-2}\). In other words, according to the integration model, 99% of the current coherently generated is dissipated inside the MSM.
Figure 4-6: Calculated voltage on an LT-GaAs sample versus the fundamental beam average power. Curve a) uses the coherent control source of current whereas curve b) assumes third-order optical rectification. From the fitting of the two curves to a function of the form $y = x^n$ using the least standard error method, exponent values of $n = 1.9$ and $3.7$ are obtained for each case. The values of the parameters used are: 1 ps pulse duration, 82 MHz repetition rate, 75 μm spot size, 10 μm gap size, 1 ps carrier lifetime and 100 fs current relaxation time. The relationship $P_{2\omega} \propto P_{\omega}^2$ is assumed.
4.1.2 Experiments with femtosecond pulses

Coherent control was also demonstrated in LT-GaAs using the femtosecond OPA system to study this process under different conditions of illumination. The main difference between using this source and the OPO is that the lower repetition rate makes current integration on an MSM device less efficient: space charge relaxation between two successive pulses occurs for a longer period of time. On the other hand, the OPA delivers pulses with a peak power $10^3$ times larger than the ps OPO, making it a better source to inject current.

The experimental setup is illustrated in Fig. 3-6. In this experiment, the polarizations of the $\omega$ and $2\omega$ beams are again made parallel. The pulse durations are about 130 and 100 fs and the wavelengths are 1.55 and 0.775 $\mu$m respectively. The repetition rate of the OPA is 250 kHz and the peak irradiances are $I_{\omega} = 10$ GWcm$^{-2}$ ($P_{\omega} = 9.5$ mW) and $I_{2\omega} = 2$ GWcm$^{-2}$ ($P_{2\omega} = 1.8$ mW). As the mirror mounted on the piezoelectric transducer is moved, a clean sinusoidal voltage is measured across the MSM electrodes (10 $\mu$m gap). In this experimental arrangement, the signal appears without any background since the lock-in amplifier is sensitive only to the phase-dependent signal.

Control over the $2\omega$ beam power alone is achieved via slight rotation of the BBO doubling crystal in the plane of phase matching. In order to make sure that adjusting the crystal does not affect the phase profile of the beams (and therefore alter the signal by itself), monitoring of the wavefront is done simultaneously to verify that the fringe pattern remains unchanged over the full range of rotation (a few degrees).

Fig. 4-7 shows the signal amplitude on LT-GaAs as a function of $P_{2\omega}$, with $P_{\omega}$ constant. The interesting feature is the sharp rise and slower fall of the signal curve as $P_{2\omega}$ increases, a behavior which agrees qualitatively with the theoretical model, represented by the scaled solid curve. Once again, the calculation uses the values for the material parameters listed in Table 2.1 along with the above mentioned beam and MSM device parameters. The decrease in the signal once $P_{2\omega}$ exceeds a certain value is expected from the increased conductivity of the semiconductor. Indeed, one can understand the current
integration mechanism in terms of two competing processes. On the one hand, the current injection rate increases with the beam irradiance, and along with it the amount of charge per pulse deposited on the MSM electrodes. On the other hand, the carrier density also increases, thereby lowering the MSM device resistance and allowing more internal discharge to take place. Whereas the former process varies according to $J \sim I_1^{1/2}$, the carrier density (and the material conductivity) increases according to $\rho_2 \sim I_2$. As a result, there will be an optimal level of irradiance beyond which internal discharge takes over and current integration efficiency decreases.

In order to match the data points, the theoretical curve of Fig. 4-7 was scaled down by a factor of about $10^2$. As in the experiment with picosecond pulses, the discrepancy between experimental results and theory may arise from an inaccurate estimate of some of the material and device parameters. Among others, the carrier lifetime and scattering time have a dramatic effect on the calculated signal amplitude. In these calculations, they
were taken to be 1 ps and 180 fs respectively, based on published results for other samples grown and prepared in a similar fashion. However, in LT-GaAs, the carrier lifetime (and possibly the scattering time) is known to depend strongly on several factors, such as the amount of excess arsenic and the anneal and growth temperatures [41].

At the maximum signal amplitude of 700 μV, the average current drawn by the lock-in amplifier is 7 pA. This establishes the most conservative estimate of the peak current density at 5 μAμm⁻². In a similar fashion as in the previous section, we can obtain a better estimate based on the model for current integration. A value 20 mAμm⁻² is obtained based on the material and sample parameters. Although this may sound large, one must keep in mind that the current burst only lasts for ~100 fs. Also, it is still below the fundamental current density limit of $J = \rho e v_c = 120$ mAμm⁻², assuming $\rho = 7 \times 10^{17}$ cm⁻³ (the carrier density for $P_{2\omega} = 200$ μW) and $v_c = 1 \times 10^6$ ms⁻¹.

Contribution to the injected carrier densities at the highest power levels are $1 \times 10^{17}$ and $7 \times 10^{18}$ cm⁻³ for the $\omega$ and $2\omega$ pulse respectively. We can see that even at these high carrier densities, electron and hole scattering does not have a major impact on current damping because elastic collisions between particles of similar effective mass tend to conserve the current. If this were not so, the signal would decrease to zero much more rapidly and diverge from the model. Although e-hh collisions could contribute to attenuating the current, their scattering rate is relatively low, as pointed out in section 2.3.1.

### 4.2 Demonstration of coherent control in GaAs

Experiments with the sample made of normally-grown GaAs (sample #2) showed that coherent control is possible in this material, although much more difficult than with LT-GaAs. In this experiment, phase control and voltage measurement were automated using a stepper motor to rotate the BK7 glass plate, which allowed for many different power levels, beam position and electrode sets to be tested. The experiment was performed with
the femtosecond OPA and an experimental setup similar to the one shown in Fig. 3-1. Fig. 4-8 shows the voltage produced across a 10 μm MSM device versus Δφ. A slowly varying background was first removed from the data, then fitting to a sine function was done based on the refractive index of the glass plate. The fit shows fairly good agreement with the data, at least in its periodicity. Results similar to those indicated in Fig. 4-8 were obtained on a few occasions but it was in general very difficult to reproduce the data. Slight motion of the beams caused the signal to vanish. Once again, this is explained in terms of the MSM discharge effect. It is indeed expected that the longer recombination time (> 10 ns) of electron-hole pairs in regular GaAs would make current integration less efficient, since after pulse excitation, the material remains highly conductive for a longer period of time. Furthermore, it was found from the model that space charge relaxation and current injection compete with each other in such a way that the overall signal is largest at low irradiance. Indeed, the above results were only obtained with $P_\omega \sim 0.5$ mW and $P_\omega \sim 10$ μW (600 and 10 MW cm$^{-2}$ peak irradiance respectively). High sensitivity to the beam power has contributed in making the data noisier than with LT-GaAs, since reflection off the glass window varies with the angle of incidence. Also, sensitivity to the beam position could be due to a contact between the metal and the substrate of lesser quality than that of the LT-GaAs sample. In the latter material, excess arsenic desorbs during annealing and the many voids and defects thereby created may cause the metal to permeate the semiconductor better. Because of the experimental difficulties encountered, it was not possible to make any other studies with the GaAs sample, such as power dependence and polarization effects. However, it is clear from these measurements that, although coherent control has been observed, normally-grown GaAs is not the ideal material for detecting the current using the present integration technique.

Another sample of regular GaAs with ohmic contacts between the gold pads and the substrate was prepared. Ion implantation in the regions covered by the electrodes was performed before metal deposition in order to ensure good penetration of conductive material in the substrate. It was hoped that a better electrical contact would lead to
Figure 4-8: Coherent control signal on a $10 \, \mu m$ gap MSM device made from GaAs versus the phase of the $\omega$ and $2\omega$ beams. The solid curve is a sine function obtained using accepted values for the index of refraction of BK7 glass.
a higher carrier collection efficiency. However, coherent control measurements with this sample were similar to the ones with the previous GaAs sample, i.e. small and noisy signals (< 10 µV) with high sensitivity to the beam positioning.

4.3 MSM photoresponse and artifacts to the coherent control signal

When considering possible artifacts to the coherent control signal, one must look for effects that depend on \( E_\omega, E_{2\omega} \) and the phase \( \Delta \phi = 2\phi_\omega - \phi_\omega \). In this section, two candidates that satisfy this condition are discussed, namely cascaded frequency doubling and optical rectification.

4.3.1 Cascaded second harmonic generation (SHG)

Any light incident near the MSM device produces an electric bias across the electrodes. The photovoltage originates from free carriers being generated near the Schottky barrier formed at the metal/substrate interface. This effect cannot be avoided, but positioning the beams in the middle of the gap can help minimize it. Both the \( \omega \) and \( 2\omega \) beams produce by themselves a photoresponse of similar magnitude, typically ranging from µV to a few mV, depending on the sample gap size and the beam power.

It is important to be sure that there is no second harmonic generation within the sample. Otherwise, reabsorption of this light would lead to a phase-varying carrier density and a photovoltage which mimics the coherent control signal. As pointed out in section 3.5, two successive second harmonic generation processes lead to an overall conversion efficiency which depends on \( \sin \Delta \phi \), i.e. the value of \( \Delta \phi \) just before the second crystal determines whether the \( 2\omega \) beam will be enhanced or converted back into an \( \omega \) beam. SHG can be quite efficient in semiconductors, even without proper phase-matching. For instance, \( \chi^{(2)}_{xyz}(2\omega, -\omega, -\omega) \) in GaAs is approximately 150 larger than that of most optical nonlinear crystals (such as KTP, KDP or BBO). Since the conversion efficiency is
proportional to $d_{eff}^2$ (the effective nonlinear coefficient), as much conversion could occur within 1 $\mu$m of GaAs as in a few millimeters of BBO, for the same pump intensity. Phase matching is not important in this case because the coherence length (distance over which $\Delta \phi$ varies by $\pi$) in GaAs is about the same length; with 1.55 and 0.775 $\mu$m, the index mismatch is 0.33 [33], giving a coherence length of $\lambda/4\Delta n \sim 1.2$ $\mu$m.

In GaAs, the only non-zero tensor elements for SHG are those of the form $d_{xyz}$ and permutations. These are not involved in the beam geometry employed in our experiment since at normal incidence, the beams have no electric field component in the z-direction of the (100) sample. Nevertheless, because of the magnitude of $d_{xyz}$, the author thought it important to make an experimental verification.

Several tests, both with the OPO and the OPA systems, have been performed to rule out cascaded frequency doubling in the coherent control experiments. Perhaps the strongest evidence of the absence of this artifact are the polarization results of Fig. 4-4. These results show the signal decreasing virtually to zero as the polarizations of the beams are aligned parallel to instead of across the electrodes gap. This suggests that no significant SHG is taking place since an equal amount of carriers would be created for both polarization orientations. Further evidence was obtained by the fact that no periodic modulation in the $2\omega$ average power was detected via cascaded frequency doubling using a 1 $\mu$m film of LT-GaAs instead of the MSM detector.

Additional verifications were made with the OPA system because of the high peak irradiance achieved with this optical source. One way of determining the importance of cascaded $\chi^{(2)}$ is to observe the appearance of fringes through a thin GaAs film, a technique similar to the one discussed in Section 3.5. The setup used for this purpose is identical to the one shown in Fig. 3-6, except that the MSM sample was replaced by a thin GaAs sample. The two beams were first aligned to produce a clean fringe pattern through KTP. The 765 nm SH beam was projected onto a distant screen in the far field and viewed with an infrared viewer. A thin epilayer ($\sim 1 \mu$m thick) of LT-GaAs bonded on sapphire was then placed immediately after the KTP and the fringes were
still visible. This confirmed that any surface roughness of the layer did not scramble the fringe pattern. The beams were then focussed onto the film and projected onto a screen in the far field. No evidence of fringe formation was observed, either visually with the viewer or by mean of a photodetector. The latter measurement was used in combination with a lock-in amplifier and phase modulation with the interferometer, establishing that any modulations in $I_{2\omega}$ would be less than the signal noise (2%). In order to produce the measured signal modulations, at least a comparable amount of second harmonic should be produced in the thin film as in the BBO crystal. Indeed, generating $\sim 500 \, \mu V$ photovoltage requires $P_{2\omega}$ to be of the order of 1 mW. The detector was positioned so as to be exposed to only part of the $2\omega$ beam in the far field so as to measure local variations. Several incidence angles were used, but always with no observable modulation. The same procedure was repeated and identical results were obtained with a 2 $\mu m$ film of GaAs.

In a second set of experiments, a more direct approach was taken by looking at bare SHG in the film. With an absorption coefficient of $1.5 \times 10^6 m^{-1}$ at 775 nm (absorption depth 0.3 $\mu m$), any significant SH produced inside the semiconductor should be measurable. However, focussing a 1.55 $\mu m$. 10 mW average power, fundamental beam onto the LT-GaAs film (peak irradiance $>10 \, GW cm^{-2}$), no SH was measured within the detector’s accuracy limit (50 nW). Similar results, independent of incidence angle, were obtained with the 2 $\mu m$ GaAs film.

We conclude from these tests that no significant cascaded $\chi^{(2)}$ occurs in our experiment, i.e., not sufficient to account for the modulation observed in the signal.

### 4.3.2 Phase-sensitive optical rectification

In addition to internal sources of electric fields, light induced DC fields must also be considered, especially if they are phase-sensitive. Second-order optical rectification via $\chi^{(2)}(0; \omega, -\omega)$ is a well-known source of DC electric field, but since it has no dependence on the phase of the beams, it cannot affect the experiment other than adding some constant background to the signal. On the other hand, the nonlinear tensor $\chi^{(3)}(0; \omega, \omega, -2\omega)$ is
associated with a polarization density varying as \( \sin(2\phi_\omega - \phi_\omega) \). This effect has been considered by Khurgin for the generation of fast electric signals \[25\].

Although Aversa and Sipe \[43\] have established a connection between coherent control of photocurrent and the nonlinear susceptibility \( \chi^{(3)}(0; \omega, \omega, -2\omega) \), the photocurrent injection cannot be understood simply in terms of free charges driven by a DC electric field. Starting from the relation

\[
\mathbf{J} = \frac{\partial^2 \mathbf{P}}{\partial t^2} = (-i\omega_\Sigma)^2 \chi^{(3)}(\omega_\Sigma; \omega_1, \omega_2, \omega_3) : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \mathbf{E}(\omega_3) 
\]

(4.3)

with \( \omega_\Sigma = \omega_1 + \omega_2 + \omega_3 \), one can show that the current injection tensor \( \hat{n}_{c(A)} \) is indeed related to a divergent part of \( \chi^{(3)}(\omega_\Sigma; \omega_1, \omega_2, \omega_3) \) as \( \omega_\Sigma \to 0 \). Indeed, expanding \( \chi^{(3)}(\omega_\Sigma; \omega_1, \omega_2, \omega_3) \) to second order in powers of \( \omega_\Sigma \) one finds

\[
(-i\omega_\Sigma)^2 \chi^{(3)}(\omega_\Sigma; \omega_1, \omega_2, \omega_3) = \nu^{(3)}(\omega_1, \omega_2, \omega_3) - i\omega_\Sigma \sigma^{(3)}(\omega_1, \omega_2, \omega_3) - \omega_\Sigma^2 \psi^{(3)}(\omega_1, \omega_2, \omega_3) 
\]

(4.4)

where \( \psi^{(3)} \) is finite as \( \omega_\Sigma \to 0 \). Thus, \( \nu^{(3)} \) and \( \sigma^{(3)} \) characterize the divergence of \( \chi^{(3)} \) as \( \omega_\Sigma \to 0 \), and the current injection tensor \( \hat{n}_{c(A)} \) is related to the fourth-rank tensor \( \nu^{(3)} \).

However, optical rectification, as it is usually understood, cannot give rise to a coherent control effect. Whereas coherent injection of photocurrent produces carriers which have preferential momentum, a DC field simply adds a drift velocity to a population of carriers evenly distributed in momentum space. The fundamental difference between the two processes is made evident by the fact that the dependence of the current on the optical fields is different for each case. Optical rectification of the third order creates a static electric field with a magnitude given by

\[
|E_{DC}| = \frac{\chi^{(3)}(0; \omega, \omega, -2\omega)}{\chi^{(1)}(0)} E_\omega^2 E_{2\omega} 
\]

(4.5)

where \( \chi^{(1)}(0) \) is the linear susceptibility at \( \omega = 0 \) which is equal to \( n^2 - 1 \), with \( n \) the
From the relation \( J = \rho ev \) one finds, neglecting the contribution of the holes:

\[
\dot{J}_{DC} = \dot{\rho} ev + \rho e\dot{v} = \frac{\rho(t)e^2|E_{DC}|}{m_e}
\]  

(4.6)

where \( v_0 = 0 \) is taken to be the initial drift velocity of the excited carriers. Since \( |E_{DC}| \sim |E_\omega|^2 |E_{2\omega}| \) and \( |E_{2\omega}| \sim |E_\omega|^2 \) from the parametric up-conversion, then the current injection rate would vary as \( \dot{J}_{DC} \sim |E_\omega|^4 \) instead of following the second power law as in the case of coherent injection. Power dependence is therefore a good criterion for identifying the origin of the measured photocurrent.

It is interesting to calculate the expected signal with the assumption that the current originates from a DC electric field. The model for the current integration predicts a voltage amplitude varying with \( P_2^{3,7} \). Fig. 4-6b shows the calculated voltage across a 10 \( \mu m \) MSM device using a value of \( 8 \times 10^{-20} \) m\(^2\)V\(^{-2}\). Reported values for the third-order tensor elements of GaAs range from 1 to \( 15 \times 10^{-20} \) m\(^2\)V\(^{-2}\) [62][63] when \( \omega \) absorption is involved (below bandgap radiation). However, since \( 2\hbar \omega \) is above the band gap, we can expect some enhancement due to real transitions. For instance, calculations for GaAs by Huang and Ching [64] showed that \( |\chi^{(3)}_{1111}(\omega; \omega, \omega, -\omega)| \) increases by a factor of \( \sim 2 \) as \( \hbar \omega \) exceeds the bandgap. Another study of second order optical rectification showed that \( |\chi^{(2)}(0; \omega, -\omega)| \) is enhanced by two orders of magnitude when \( \hbar \omega \) is tuned above bandgap [65]. In all cases however, the current thereby generated would still be at least \( 10^3 \) smaller than with the coherent injection process.

### 4.4 Non-degenerate coherent control in LT-GaAs

Section 2.2 discussed the possibility of extending coherent control to a case involving three phase-related beams with photons of different energy, provided the condition \( \omega_1 = \omega_2 + \omega_3 \) is satisfied. It was pointed out that the non-degenerate case should yield approximately the same tensor elements as the degenerate case when \( \omega_2 \approx \omega_3 \).

Experimentally, the general approach to measuring the current is similar to the degen-
erate case, with the exception that it involves three laser beams. One obvious possibility was to use the signal, idler and pump beams of the OPA (or OPO) system. However, to control, joint collinearly and overlap in time three beams exiting the OPA system with different orientations and time delays is not straightforward, since they have different wavelengths and polarization states. Instead, the author chose the simpler solution of re-creating the pump beam via sum-frequency generation from the signal and idler waves. In this way, the three beams are collinear, provided the signal and idler beams are themselves collinear; time overlap is automatically satisfied. The signal and idler beams can be made collinear with some careful alignment of the OPA, and the pulses are overlapped in time because of their wavelength proximity and the small dispersion within the apparatus.

Experimental confirmation for non-degenerate coherent control was made in LT-GaAs using the OPA source. The signal (1.51 μm) and the idler (1.70 μm) beams are mixed to produce sum-frequency (0.794 μm). Average output powers are 4.2 mW, 2.4 mW and 900 μW for the signal, idler and sum frequency respectively. The carrier densities due to two- and single-photon absorption are $2 \times 10^{16}$ and $2 \times 10^{18}$ cm$^{-3}$ respectively.

Fig. 4-9 shows the resulting voltage across the MSM electrodes versus the displacement of the mirror mounted on a PZT driver in one arm of the interferometer (see Fig. 3-6). The displacement is calculated from the piezo device specifications: a motion of 30 μm for the full range voltages provided by the controller box (1000 V), or 30 nm/V. Several cycles could be obtained with a periodicity matching the expected value to within 5%. Verification that the coherent control signal truly involved the three beams being present was made by inserting an RG1000 (OD 5) filter in the arm with the signal and idler and a KG3 (OD 3) filter in the other arm of the interferometer. Each filter ensures that the beams coming out of the interferometer travelled through one arm only. In each case the signal modulation is still obtained – although decreased by a factor of $\sim 2$ – which proves that the effect is not merely due to linear interference caused by a beam travelling through both arms.
Figure 4-9: Non-degenerate coherent control signal on a 10 µm LT-GaAs MSM with pulse wavelengths of 1.70, 1.51 and 0.794 µm. A sine curve is fitted to the data.
This demonstration of coherent control of photocurrent using three beams of different frequencies indicates that the effect is not dependent on having mid-gap state resonances since in that case two different states would be accessed.

4.5 Polarization dependence of the current integration

Experiments with the OPO system showed a well-behaved dependence of the coherent control signal on the polarization state of the exciting beams. When the polarizations of the $\omega$ and $2\omega$ beams were oriented across the gap, the signal is a maximum, but it vanishes when one or both polarizations are rotated by $90^\circ$. With the OPA system however, the situation is different, even with an identical sample, wavelengths and beam parameters.

It was not possible to obtain a well-defined polarization state with the dual wavelength wave plate because of the large bandwidth associated with femtosecond pulses. The spectral widths of 30 nm for the $\omega$ and 15 nm for the $2\omega$ beams are much larger than the waveplate tolerance of about 4 nm. Hence, linearly polarized light is transformed into elliptically or circularly-polarized light, depending on the center wavelength. As a result, there is always a component of the electric field in the x-direction (\perp to the MSM gap) and the coherent control signal never reaches zero. This is not true however for the OPO beam which has a spectral width of $\sim$3.5 nm.

Under such conditions, analysis of the polarization dependence requires a more thorough investigation of the current injection; all nonzero tensor elements of $\tilde{\eta}_{\varepsilon(h)}$ must be included. The resulting equation for the total current injection rate along $x$ becomes

\[ \dot{J}_x = 2 \left[ \eta_{xxx} E_x^\omega E_x^{2\omega} \sin \Delta \phi_1 + \eta_{xxy} E_y^\omega E_y^{2\omega} \sin \Delta \phi_2 + \eta_{xyy} E_x^\omega E_y^{2\omega} \sin \Delta \phi_3 \right] \]

(4.7)

where the phases are defined as $\Delta \phi_1 = \phi_{2\omega,x} - 2\phi_{\omega,x}$, $\Delta \phi_2 = \phi_{2\omega,x} - 2\phi_{\omega,y}$ and $\Delta \phi_3 =$
\( \phi_{2\omega,y} - \phi_{\omega,x} - \phi_{\omega,y} \), with \( x \) and \( y \) referring to the components of the electric field (normal incidence is assumed).

As a means to comparing experiment with theory, the \( x \)- and \( y \)-components of the electric fields of the \( \omega \) and \( 2\omega \) beams were measured for different waveplate orientation, along with the corresponding coherent control signal amplitude. The polarization state of each beam was analyzed after it passed through the waveplate by measuring the transmittance through a broadband polarizing beam splitter cube. Figure 4-10 shows the transmitted power for horizontal polarization as a function of waveplate angle. We see that the waveplate behaves like a half-waveplate at \( \omega \) but does not act as a full-waveplate at \( 2\omega \). Indeed, up to 10\% of the total power can be measured in the \( x \)-direction, in which case the polarization is elliptical. Analysis with narrow band light (Coherent Mira laser operating in a continuous wave regime) from 750 to 800 nm revealed that the waveplate periodically acts as a full waveplate with regions of elliptical polarization in between.

The solid and dashed curves show the best fits for functions of the form

\[
|E_x|^2 \sim \sin^2(a\theta + b) + c \tag{4.8}
\]

\[
|E_x^2\omega|^2 \sim \sin^2(d\theta + e) \cos^2(d\theta + f) \tag{4.9}
\]

where \( a \), \( f \) are fitting parameters. Eq. 4.8 and 4.9 are characteristic of linearly and elliptically polarized beams respectively. Each function gives a satisfactory fit with a standard error of less than 0.04.

The electric fields of the beams along \( y \) are obtained from the relationship

\[
E_y^{\omega,2\omega} = \left( |E_y^{\omega,2\omega}|^2 - |E_x^{\omega,2\omega}(\theta)|^2 \right)^{1/2} \tag{4.10}
\]

where \( |E_y^{\omega,2\omega}| \) are the electric field amplitudes found from the beams' average power.

In order to fit data with Eq. 4.7, one must know the values of the phases \( \Delta\phi_{1,3} \) (\( \Delta\phi_2 \) is not considered since the tensor element \( \eta_{ixyz} \) in GaAs is negligible at the wavelengths
Figure 4-10: Data and fitting curves of the transmitted power of the ω (dashed curve) and 2ω beams (solid curve) through a polarizing beam splitter cube as a function of waveplate angle. The x-component is parallel to the table plane and across the MSM gap. Pulse wavelengths are 1582 and 791 nm with spectral widths of 15 and 30 nm respectively.
Figure 4-11: Coherent control signal amplitude (squares) from a 10 μm gap LT-GaAs MSM device as a function of waveplate orientation. The solid curve is a theoretical fit involving the $\eta_{xxx}$ and $\eta_{xyy}$ tensor elements based on measurements of the $x$ and $y$ electric field components of both beams. Discontinuities in the fitting curve are due to taking the absolute value of $J_x$ in Eq. 4.7.

Because the quartz plate acts as a half-waveplate at $\omega$, the phase difference between $E_x^{\omega}$ and $E_y^{\omega}$ is constant at $\pi$. We can extract an approximate value of $\Delta = 25^\circ$ for the phase difference between $E_x^{2\omega}$ and $E_y^{2\omega}$ based on the results of Fig. 4-10. We can now simplify $\Delta \phi_1 = \phi_{2\omega,x} - 2\phi_{\omega,x}$ and $\Delta \phi_3 = \phi_{2\omega,x} - \phi_{\omega,x} - \pi - \Delta$. The signal amplitude is obtained when the phases are adjusted in such a way that $J$ is maximized and can be expressed as \( A^2 + B^2 + 2AB \cos \Delta \) with $A = \eta_{xxx} E_x^{\omega} E_x^{2\omega}$ and $B = \eta_{xyy} E_x^{\omega} E_y^{\omega} E_y^{2\omega}$. Finally, the ratio $\eta_{xxx}/\eta_{xyy}$ is chosen to be 1.7, as suggested by the calculation of Fig. 2-2.

The results plotted in Fig. 4-11 shows a signal amplitude with $180^\circ$ periodicity. The fit is not perfect but exhibits the proper trend and, interestingly, is fairly insensitive to the phase $\Delta$. In addition to experimental error in the signal amplitude measurement, another source of discrepancy between theory and experiment may be the uncertainty in
the strength of the electric field components. Indeed, no combination of tensor element ratio and value of $\Delta$ can produce a fitted curve with the two side peaks larger than the two center peaks, which suggests that only differing values of electric field components could appreciably change the shape of the curve.

4.6 Effects of pulse chirping

In section 2.5.3, the effect of pulse chirping on the current injection rate was calculated. When a pair of chirped pulses are delayed relative to each other, the appearance of a time-varying $\Delta\phi$ causes $\mathbf{j}^I$ to oscillate reducing the overall current density. The phase variation increases with chirping and time delay (Fourier-transform-limited pulses would not produce this effect).

Demonstration of the influence of chirp on the current integration was done using the OPA system and the LT-GaAs sample. Chirped pulses at $\lambda_1 = 1.53 \, \mu m$ and $\lambda_2 = 0.765 \, \mu m$ were used with an average power of about 5 mW and 500 $\mu W$ respectively. First, the pulse durations were determined by measuring their cross correlation trace via sum-frequency generation in KTP ($\omega + 2\omega \rightarrow 3\omega$). As for any cross correlation involving $\chi^{(2)}$, the relationship between the duration of Gaussian pulses and the correlation width is given by

$$e^{-at^2} \otimes e^{-b(t-t_d)^2} \rightarrow e^{-abt_d^2}$$

(4.11)

where $a$ and $b$ are the related to the pulses durations and $\otimes$ is the correlation symbol. Consequently,

$$\tau_{cc}^2 = \tau_\omega^2 + \tau_{2\omega}^2$$

(4.12)

where $\tau_{cc}$, $\tau_\omega$, and $\tau_{2\omega}$ are the FWHM of the cross-correlation trace and the $\omega$ and $2\omega$ pulse envelopes. Note that sum-frequency generation is not sensitive to the relative phase of the input pulses or their chirp since the resulting nonlinear polarization starts with
Figure 4-12: Coherent control signal and cross correlation versus time delay between the \( \omega \) and \( 2\omega \) pulses for: (a) expected coherent control signal with bandwidth-limited pulses \( (\Delta T \Delta \omega / 2\pi = 0.44) \), (b) cross-correlation signal via sum-frequency generation and (c) experimentally measured coherent control signal. The FWHM for each curve are 172.145 and 65 fs respectively.

Zero intensity and therefore its phase is optimized. Fig. 4-12 b shows a cross-correlation FWHM of 145 fs, giving values of \( T_\omega = 114 \) fs and \( T_{2\omega} = 91 \) fs assuming \( T_{2\omega} = T_\omega / \sqrt{2} \). Calculation of \( T_{2\omega} \) takes into account a spectral bandwidth of 16 nm producing 11 fs of pulse broadening through the quartz waveplate and the 3.8 mm-thick lens. Pulse broadening for the \( \omega \) beam \( (\Delta \lambda = 32 \) nm) is negligible due to the low dispersion of glass and quartz at 1.53 \( \mu \)m. From the values of \( T_\omega \) and \( T_{2\omega} \), the width of the coherent control signal amplitude is expected to be \( \sim 170 \) fs, assuming Fourier-transform-limited pulses (curve a); however, this value is about 3 times larger than the measured width of 65 fs (curve c). This result is consistent with a time-bandwidth product of about 0.9, according to calculations of Section 2.5.3.

In an independent set of measurements, the effect of stretching one of the optical pulses was investigated. Pulses with \( T_\omega = 97 \) fs and \( T_{2\omega} = 80 \) fs duration were first used.
Figure 4-13: Gaussian functions representing the measured cross-correlation traces between the $\omega$ and $2\omega$ pulses (curve a and b with 155 and 125 fs FWHM respectively) and the corresponding coherent control signal amplitude as a function of time delay (curve c and d): for details, see text.

as determined from a cross-correlation width of 125 fs (curve a). The corresponding coherent control signal curve width is $94\pm2$ fs (curve c). When a 9.5 mm-thick KZF-2 (Schott glass) flat window was inserted in the path of the $2\omega$ beam (in one arm of the interferometer), the width of the cross-correlation trace was increased to 155 fs (curve b), suggesting a new pulse duration $\tau_{2\omega} = 121$ fs (41 fs total pulse stretching). As a result of this increased chirp, the width of the coherent control signal amplitude is reduced to $79\pm2$ fs (curve d). If one ignores pulse chirping, a larger width would be expected since one of the pulses is now much longer. In contrast, a reduction by $15 \pm 4$ fs is observed. These experiments clearly show the negative impact of the combination of chirp and time delay on the current-integration efficiency.
4.7 Device parameters and efficiency of current integration

The model for current integration has been successful in predicting the qualitative behavior of the integrated current as a function of beam power. A great number of computations have been carried out in order to determine which parameters most influence the current-integration efficiency. Three classes of parameters can be identified: 1) material, 2) device and 3) beam parameters. All the material parameters (carrier scattering and lifetime, single and two-photon absorption coefficients) have an important impact. Of all the sample parameters, gap size is the most important; capacitance and dark resistance do not have a strong influence. Beam profile, pulse duration, peak irradiance and repetition rate are important.

For beams with uniform irradiance, the voltage is expected to increase linearly with gap size because the voltage is proportional to the space charge separation. For Gaussian beam profiles, however, the average voltage \( \langle V \rangle \) should vary with the gap size \( d \) according to

\[
\langle V(d) \rangle \sim d \exp \left( -4 \ln 2 \frac{d^2}{s^2} \right)
\]

where \( s \) is the spot diameter of the \( \omega \) beam (assumed to be larger than that of the \( 2\omega \) beam by a factor of \( \sqrt{2} \)). This equation is obtained by considering that the irradiance at the edge of each electrode drops exponentially with radial distance, i.e. \( I(r) \sim \exp(-r^2/s^2) \).

Figure 4-14 shows the signal amplitude from different electrode pairs on the LT-GaAs sample for identical conditions of beam illumination with the OPA. The average power is 4.5 mW and 375 \( \mu \)W for the \( \omega \) and \( 2\omega \) beams respectively. The dashed line is the calculated amplitude (scaled to match the first data point) whereas the solid curve is a fit using Eq. 4.13. A beam spot diameter of 66 \( \mu \)m gives the best result.

Equation 4.13 suggests that the optimum condition for current integration occurs when the ratio \( s/d = \sqrt{\ln 2} \sim 2.35 \). However, provided that the ratio remains optimized,
smaller gaps will give larger voltages. Indeed, although the voltage is proportional to $d$, the current injection is inversely proportional to $s^3$.

4.8 Preliminary measurements with silicon sample

Since silicon is the semiconductor of choice in the electronic industry, the author attempted to demonstrate current control in this material. A sample with semi-insulating silicon (doping concentration $\sim 10^{18} \text{cm}^{-3}$) oriented along a [100] direction was prepared with photolithography in an identical fashion as sample #1 and #2 of GaAs. The OPA signal wavelength of 1.55 $\mu\text{m}$ and its second harmonic are such that the condition $\hbar \omega < E_g < 2\hbar \omega$ is satisfied ($E_g = 1.11$ eV in silicon). Using the experimental setup of Fig. 3-6, a wide range of power levels and polarization states (parallel and perpendicular to the gap) were tested with no detectable modulation. The average power for the $2\omega$
beam varied from a few $\mu$W to 1 mW and for the $\omega$ beam, tens of $\mu$W to several mW. Using long time averaging, it was established that if any signal existed, it would be smaller than the 0.5 $\mu$V accuracy limit of the lock-in amplifier. More accurate measurements were not possible due to the presence of a large photovoltaic background ($\sim$ 4.5 mV at the highest power level) which increases the sensitivity to beam positioning.

The absence of signal in bulk silicon was not unexpected, considering the relatively long carrier lifetime ($> 1 \mu$s). Also, since silicon is an indirect band gap semiconductor, phonon transitions may have a strong influence on the current injection. Wave-function dephasing at the intermediate state could help destroy the interference between the transition pathways.
Chapter 5

Conclusions and future work

Coherent control of photocurrent has been investigated both theoretically and experimentally in bulk GaAs at room temperature. Phase-controlled currents were observed in both regular and LT-GaAs using ultrashort pulses at 1550 and 775 nm. This chapter summarizes the results of this thesis and suggests potential areas for investigation in the future.

The theory of current control was discussed both from a phenomenological and a rigorous, purely quantum mechanical point of view. The overall probability for interband direct transition in a solid with simultaneous single- and two-photon excitation is intrinsically asymmetric in $\mathbf{k}$. The probability of promoting electrons from the valence to the conduction band may be greater for wavevector $+\mathbf{k}$ than for $-\mathbf{k}$, or vice versa, depending on the relative phase of the beams. A first-principles calculation was carried out to establish the connection between the resulting current injection rate $J^I$ and the electric fields of the beams. Results for GaAs suggest the use of the largest tensor element $\eta_{zzzz}$.

Using metal-semiconductor-metal (MSM) devices to integrate the current, coherent control was demonstrated in GaAs using 100 fs and 1 ps fundamental and second harmonic pulses from two optical parametric sources: an optical parametric oscillator (OPO) and a parametric amplifier (OPA). Peak current densities were estimated to be as high as 20 mA $\mu$m$^{-2}$ with free carrier densities up to $7 \times 10^{18}$ cm$^{-3}$. In the experiment with the
OPO, the dependence of the voltage across the MSM device on the fundamental beam average power is $V \sim P_{\text{av}}^{1.8\pm0.2}$, which is close, but not exactly as expected from a detailed calculation based on a model for current integration ($V \sim P_{\text{av}}^{1.9}$). The polarization dependence of the signal rules out the possibility that cascaded frequency doubling is responsible for the effect: a voltage modulation is only measured when both beams were polarized across the MSM electrodes. LT-GaAs was found to be more appropriate than GaAs for photocurrent integration because a shorter carrier lifetime facilitates the accumulation of space-charge through carrier trapping. Non-degenerate coherent control was also observed using phase-related beams with wavelengths of 1.70, 1.51 and 0.794 μm. The presence of a signal of similar magnitude as with the degenerate case confirms that the coherent control effect is general and does not depend on the availability of intermediate states.

The effect of pulse frequency chirping on the current injection was also investigated. Increasing the chirp of one of the pulses increased the sensitivity of the coherent control signal to the pulse delay because of variations in the relative phase of the fields within the pulse envelope. Pulses with a time-bandwidth product of 0.9 gave a cross-correlation width nearly one-third of that expected from bandwidth-limited pulses. It is therefore important to take into account frequency chirp when the two pulses are delayed relative to each other. Experiments with various sets of MSM electrodes revealed that resistance and capacitance have little impact on the efficiency of current collection. However, electrode spacing is important and the optimum condition is when the Gaussian beam spot diameter is $\sim 2.8$ times larger than the electrode gap.

The author does hope that his work will have triggered enough interest for others to pursue studies on photocurrent coherent control, since many details have yet to be clarified. Throughout this thesis, limitations in the understanding of the coherent control process have been made evident. Many approximations were made in the calculation of the nonlinear tensor for the current injection. For instance, the effect of carrier dephasing and intermediate (midgap) states have been neglected. Excitonic transitions may lead
Figure 3-1: Time evolution of the current density for a pair of 100 fs bandwidth-limited pulses and chirped pulses with 50 fs delay. A current relaxation time of 20 fs is assumed.

to some interesting enhancement effects on the photocurrent injection. Also, the tensor elements for non-degenerate coherent control have yet to be calculated. Experimentally, some of our knowledge of the photocurrent dynamics remains superficial. We can only speculate on the exact mechanism for carrier trapping, since it may involve a combination of various factors: surface states, defects, Schottky barrier, arsenic anti-sites (bulk traps), etc. Alternative methods of measuring the photocurrent may shed more light on this rather complicated problem.

On the more applied side, the author thinks that a promising avenue for coherent current injection is the generation of phase-controlled terahertz radiation. Time-varying currents are a source of electromagnetic radiation. On the one hand, a current of short time duration could potentially create radiation in the THz spectrum. On the other hand, since the directionality of the current can be changed within the pulse duration, higher frequencies may be reached. By using pairs of chirped and delayed pulses, the THz spectral content may be varied, as illustrated in Fig. 5-1. Eventually, current density
profiles may be temporally 'tailored' at will by using more than one pair of pulses. For example, a first set of pulses producing a current in a given direction may be cancelled out by a second set, dephased by π relative to the first one. This may lead to a current burst of a duration less than the pulses themselves.
Bibliography


[61] Schott Optical Glass [Schott Mfg., USA].


