Direct Evidence for LO Phonon Reabsorption by Free Holes in GaAs Quantum Wells

by

Peter Brockmann

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

Direct Evidence for LO Phnon Reabsorption by Free Holes in GaAs Quantum Wells

Peter Brockmann

A thesis submitted for the degree of Doctor Of Philosophy.
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The influence of an electron-hole plasma with a temperature close to that of the ambient lattice (T≈5K) on the population lifetime of non-equilibrium LO phonons created by the relaxation of hot carriers is studied in a GaAs quantum well sample that was designed to allow one to clearly separate LO phonon emission from absorption processes. For the experiments a novel multiple-beam Raman scattering technique was developed that allows one to separately control the LO phonon generation and the electron-hole plasma injection process. The lifetime of non-equilibrium, small-wavevector LO phonons is found to decrease from 4.5 ps to 1.9 ps as the density of the plasma is increased to ~3×10^{11} \text{ cm}^{-2} by adjusting the intensity of an infrared laser tuned close to the lowest subband gap of the quantum well. This result represents the first direct experimental evidence from the lattice perspective for longitudinal optical (LO) phonon reabsorption by free carriers.
# Symbol Glossary

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Meaning</th>
<th>Section</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>1.1</td>
</tr>
<tr>
<td>$\vec{k}$</td>
<td>wavevector</td>
<td>1.1</td>
</tr>
<tr>
<td>$\vec{p}$</td>
<td>momentum</td>
<td>1.1</td>
</tr>
<tr>
<td>$z$</td>
<td>growth direction</td>
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</tr>
<tr>
<td>$\vec{k}_l, \vec{q}_l$</td>
<td>wavevector, in-plane component</td>
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<tr>
<td>$\omega_{LO}$</td>
<td>LO phonon frequency</td>
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<tr>
<td>$\omega_{AS}$</td>
<td>anti-Stokes frequency</td>
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<td>conduction subbands</td>
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<tr>
<td>$v_0, v_1$</td>
<td>valence subbands</td>
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<tr>
<td>$E_F$</td>
<td>Fermi energy</td>
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### Chapter 2

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<td>effective LO phonon lifetime</td>
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<td>$R_{em}, R_{abs}$</td>
<td>LO phonon emission and absorption rates</td>
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<tr>
<td>$n_{q_l}$</td>
<td>LO phonon occupation number</td>
<td>2.1</td>
</tr>
<tr>
<td>$\hbar$</td>
<td>Planck’s constant divided by $2\pi$</td>
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</tr>
<tr>
<td>$\tau_{anh}$</td>
<td>anharmonic decay time constant</td>
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<tr>
<td>$G_{KK'}^{se}$</td>
<td>unscreened Green’s function</td>
<td>2.2</td>
</tr>
<tr>
<td>$G_{KK'}^{sc}$</td>
<td>screened Green’s function</td>
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Chapter 2 (continued)

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<td>LO phonon self energy</td>
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<tr>
<td>$\Sigma_{LO}$</td>
<td>LO phonon self energy</td>
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<tr>
<td>$M_{LO}^{hf}$</td>
<td>matrix element for hole-LO phonon scattering</td>
<td>2.3</td>
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<tr>
<td>$P_{01}, P_{10}$</td>
<td>2D polarizability function</td>
<td>2.3</td>
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<tr>
<td>$\epsilon_{0101}$</td>
<td>2D dielectric function</td>
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<td>$f_0, f_1$</td>
<td>Fermi-Dirac occupation factors</td>
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<td>$E_0, E_1$</td>
<td>free hole energies</td>
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<tr>
<td>$V_{0101}$</td>
<td>Coulomb matrix element</td>
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<tr>
<td>$e^\prime$</td>
<td>electron charge</td>
<td>2.3</td>
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<tr>
<td>$\epsilon_\infty, \epsilon_1$</td>
<td>high frequency dielectric constant</td>
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<tr>
<td>$\epsilon(q, \omega)$</td>
<td>dielectric function</td>
<td>2.3</td>
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<td>$V_{int}, V_{ext}$</td>
<td>internal, external potential</td>
<td>2.3</td>
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<tr>
<td>A, B</td>
<td>quantum well and barrier materials</td>
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<tr>
<td>$V_b$</td>
<td>barrier potential</td>
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<td>d</td>
<td>well width</td>
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<tr>
<td>a</td>
<td>well width divided by two</td>
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<td>$\Gamma_6, \Gamma_7, \Gamma_8$</td>
<td>band valleys</td>
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<tr>
<td>$E_{\Gamma}$</td>
<td>bandgap energy</td>
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<td>$\Delta$</td>
<td>split-off band energy offset</td>
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<td>n</td>
<td>band index</td>
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<td>Bloch wavefunction</td>
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<td>lattice periodic part of $\Psi_{n,k_{AB}}^{A,B}$</td>
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<tr>
<td>$\Omega$</td>
<td>normalization factor</td>
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<tr>
<td>J</td>
<td>total angular momentum</td>
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<tr>
<td>m</td>
<td>projection of J onto z-axis</td>
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<tr>
<td>$\Omega_{n,k_{AB}}$</td>
<td>spinor basis functions</td>
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<td>X, Y, Z</td>
<td>p-orbital-like wavefunctions</td>
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<tr>
<td>$H$</td>
<td>Luttinger-Kohn Hamiltonian</td>
<td>2.4</td>
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<tr>
<td>$P, Q, R, S$</td>
<td>elements of $H$</td>
<td>2.4</td>
</tr>
<tr>
<td>$\gamma_1, \gamma_2, \gamma_3$</td>
<td>Luttinger parameters</td>
<td>2.4</td>
</tr>
<tr>
<td>m</td>
<td>free electron mass</td>
<td>2.4</td>
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<tr>
<td>$k_r, k_\theta, k_z$</td>
<td>wavevector components</td>
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<tr>
<td>Symbol</td>
<td>Meaning</td>
<td>Section</td>
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<tr>
<td>--------</td>
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<tr>
<td>( p )</td>
<td>parity quantum number</td>
<td>2.1</td>
</tr>
<tr>
<td>( F_p )</td>
<td>hole envelope spinor</td>
<td>2.1</td>
</tr>
<tr>
<td>( \epsilon_A, \epsilon_B )</td>
<td>dielectric constants of well and barrier materials</td>
<td>2.5</td>
</tr>
<tr>
<td>( \nu )</td>
<td>label for phonon modes</td>
<td>2.5</td>
</tr>
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<td>( \mathcal{P}_I, \mathcal{P}_S, \mathcal{P}_A, \mathcal{P}<em>C, \mathcal{P}</em>\nu, \mathcal{P}_H )</td>
<td>phonon mode polarizabilities</td>
<td>2.5</td>
</tr>
<tr>
<td>( \alpha_\infty )</td>
<td>coupling function</td>
<td>2.5</td>
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<tr>
<td>( f_{\nu}^\alpha )</td>
<td>form factor</td>
<td>2.5</td>
</tr>
<tr>
<td>( \eta_{\nu}^{\alpha,\beta} )</td>
<td>interaction function</td>
<td>2.5</td>
</tr>
<tr>
<td>( \epsilon_0 )</td>
<td>static dielectric constant</td>
<td>2.5</td>
</tr>
<tr>
<td>( \mathcal{I} )</td>
<td>polarizability integral</td>
<td>2.5</td>
</tr>
<tr>
<td>( k_F )</td>
<td>Fermi wavevector</td>
<td>2.5</td>
</tr>
</tbody>
</table>

**Chapter 3**

| \( \lambda \) | wavelength | 3.2 |
| \( P_1, P_2 \) | polarizers | 3.2 |
| \( L_1, L_2 \) | lenses | 3.2 |
| \( \Delta t \) | time delay | 3.2 |
| \( l, l_0 \) | optical path difference | 3.2 |
| \( c \) | speed of light | 3.2 |
| \( \lambda_L \) | laser wavelength | 3.2 |
| \( \phi_1, \phi_2 \) | angles with respect to sample normal | 3.2 |

**Chapter 4**

| \( n_{PL} \) | electron hole plasma density | 4.1 |
| \( \epsilon_{el}^{\prime} \) | dielectric function, electrons only | 4.2 |
| \( \epsilon_{hole}^{\prime} \) | dielectric function, holes only | 4.2 |
| \( \Gamma_{n_{PL}=0} \) | bare scattering rate | 4.2 |
| \( \Gamma_{IVB} \) | intervalence band scattering rate | 4.2 |
| \( \sigma_{AS}, \sigma_S \) | cross sections for anti-Stokes and Stokes scattering | 4.2 |
| \( \omega_{in} \) | Raman probe frequency | 4.2 |
### Acronym and Abbreviation Glossary

<table>
<thead>
<tr>
<th>Acronym or Abbreviation</th>
<th>Meaning</th>
</tr>
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<tbody>
<tr>
<td>LO</td>
<td>longitudinal optical</td>
</tr>
<tr>
<td>TRROD</td>
<td>time-resolved Raman/optical doping</td>
</tr>
<tr>
<td>AC</td>
<td>acoustic phonon</td>
</tr>
<tr>
<td>III-V</td>
<td>group III and V of the periodic table of elements</td>
</tr>
<tr>
<td>2D</td>
<td>two-dimensional</td>
</tr>
<tr>
<td>RPA</td>
<td>Random Phase Approximation</td>
</tr>
<tr>
<td>hh, lh, so</td>
<td>heavy hole, light hole, split-off hole</td>
</tr>
<tr>
<td>ps</td>
<td>picoseconds</td>
</tr>
<tr>
<td>FWHM</td>
<td>full width at half maximum</td>
</tr>
<tr>
<td>NRC</td>
<td>National Research Council of Canada</td>
</tr>
<tr>
<td>Nd:YAG</td>
<td>Nd:Yttrium Aluminum Garnet</td>
</tr>
<tr>
<td>LBO</td>
<td>Lithium Triborate</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge Coupled Device</td>
</tr>
<tr>
<td>PI</td>
<td>photoluminescence</td>
</tr>
<tr>
<td>Ti:Sapphire</td>
<td>Ti:Al$_2$O$_3$</td>
</tr>
<tr>
<td>CPPM</td>
<td>couple plasmon phonon mode</td>
</tr>
<tr>
<td>SPE</td>
<td>single particle excitations</td>
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</table>
Chapter 1

Introduction

1.1 Overview

Technology based on opto-electronic devices has made significant advances in the last few years. The telecommunications field especially has seen an explosive growth in the capability of transmitting data at ever increasing rates. One reason for this development is the introduction of high bandwidth fiber optic data transmission lines. The other reason is the advance in technology of the opto-electronic semiconductor devices that convert electronic signals into the light that is transmitted down the fiber and convert it back into electronic signals at the receiving end.

The speed of these devices is largely determined by the dynamics of the electrons and holes that are usually injected under high electric field conditions. This injection can take place electrically (e.g. in semiconductor lasers) or optically (e.g. in detectors). The carriers thus have a large excess kinetic energy with respect to the valence and conduction band minima. This kinetic energy corresponds to a temperature $T$ that is higher than that of the lattice. Hence these carriers are commonly referred to as “hot carriers” in the literature. It is the energy relaxation of these hot carriers that is crucial for the performance of many electronic and opto-electronic devices. The interaction of hot carriers with the semiconductor crystal lattice is of central importance to the relaxation process and is the main topic of this thesis.
The knowledge of hot carrier relaxation in \textit{bulk} semiconductors is based on a large body of work performed over the last four decades. A collection of excellent reviews of the hot carrier relaxation field can be found in the recent book by Shah \cite{1}. In the 1950's and 1960's, transport measurements were very successful in identifying the main contributions to the energy loss processes of hot carriers \cite{2}. These experiments measured properties that are related to the entire carrier distribution function. Lasers offered new degrees of freedom in the investigation of carrier relaxation processes and began to be used for these purposes in the 1970's. Optical absorption and luminescence experiments can directly measure the distribution functions of electrons and holes. In addition, pulsed lasers enabled scientists to map out changes in the distribution functions on pico- and femtosecond timescales through time-resolved luminescence and absorption measurements. Another class of optical experiments, time-resolved and \textit{cw} Raman spectroscopy, added new information by probing relaxation processes from the lattice perspective. The experiments described in this thesis are based on a Raman scattering technique.

Even though it is now understood that the carriers ultimately lose their energy through excitation of the lattice, there exists only indirect knowledge about an important aspect of the free carrier-lattice interaction. This issue that has attracted considerable attention is the experimental observation of a reduced cooling rate of hot carriers with increasing carrier density in polar semiconductors. For the polar III-V semiconductors, that are now used in most existing opto-electronic devices, it is well known that hot carriers are strongly coupled to the lattice via the electric field associated with long-wavelength longitudinal optical lattice vibrations (LO phonons). In the early days of the hot carrier relaxation field there was a debate whether the reduced cooling rate was due to screening of the free carrier-LO phonon interaction \cite{3} or reabsorption of LO phonons by the free carriers ("hot phonon effect") \cite{4}. The current understanding is based on several detailed microscopic models for carrier-LO phonon interaction that have been used to model experimental results that measured the reduced cooling rate in terms of the temporal evolution of the (macroscopic) temperature of the carrier distributions alone.
The results of these studies indicated that there is strong theoretical evidence that the hot phonon effect is responsible for the reduced cooling rate, although direct microscopic evidence from the lattice perspective is still lacking.

If it would be possible to experimentally isolate the LO phonon reabsorption processes that lead to a reheating of free carriers, the hot phonon effect should manifest itself as a reduction in the LO phonon lifetime. This thesis presents this evidence by providing a detailed experimental [5, 6] and theoretical study [7] of the microscopic free carrier-LO phonon interaction from the lattice perspective. By taking advantage of the ability to “engineer” the band structure in quantum wells, a sample was designed that allows for the separation of LO phonon generation and absorption processes. The influence of free carriers on the LO phonon dynamics was systematically measured in this sample by using a novel experimental approach developed by the author based on a time-resolved Raman scattering technique. As a result of these measurements the author was able to clearly demonstrate a significant reduction of the LO phonon lifetime with increasing carrier density which is a clear indication of efficient reabsorption of LO phonons by free carriers. Due to the specific bandstructure of the sample used in the experiments, intersubband transitions of free holes could be isolated as the re-absorption mechanism. In addition, the Raman spectra obtained clearly show evidence of anti-screening effects that manifest themselves as a blue-shift of the LO phonon energy caused by interaction with a free electron-hole plasma. In order to explain the observed LO phonon reabsorption, the author developed a model for calculating free hole contributions to the LO phonon lifetime. This model, based on a calculation of the LO phonon self energy, explicitly takes the complicated valence band structure and screening effects due to many-body interactions between holes and LO phonons into account. A comparison of the theoretical results with the data shows that the model can reproduce the salient features of the carrier density dependence of the LO phonon lifetime and clearly shows the importance of screening effects on the results.

In the following discussion the author presents a brief review of experiments that have addressed the dynamics of LO phonons from the lattice perspective. The first direct measurement of the LO phonon dynamics with picosecond resolution
was performed in bulk GaAs with a time-resolved Raman scattering experiment by von der Linde et al. [8], who determined the LO phonon lifetime in GaAs to be 7 ps at $T = 77$ K. In recent years the time-resolved Raman scattering technique has also been used to obtain detailed information on the LO phonon dynamics in semiconductor quantum wells. Measurements of the LO phonon lifetime in GaAs quantum wells have given values between 5 ps and 8 ps at $T = 10$ K [9, 10, 11]. The reason for this variation in lifetime is not known presently. All of the above experiments were carried out at a fixed carrier density and thus gave only limited information on the contributions of free carriers to the measured lifetime of the phonons. Since the experiments were carried out in the low carrier density limit, the observed lifetime was attributed to decay into acoustic phonons. Kash et al. were the first to investigate the interplay of free carrier and LO phonon dynamics from the lattice perspective in more detail. In a time-resolved Raman study of the dependence of the LO phonon lifetime on the hot plasma density in bulk GaAs they observed a reduction of the lifetime with increasing carrier density [12]. It was suggested that this result was due to reabsorption of LO phonons by intervalence band transitions of free holes. The interpretation of the experimental results is complicated by the fact that both the hot carrier and the hot LO phonon population scale with the incident laser intensity. In the same publication the authors support their interpretation by an observed reduction of the LO phonon lifetime in p-doped bulk GaAs. Nevertheless, Kash and Tsang suggested further studies to clarify and clearly isolate the contributions of intervalence band transitions to LO phonon reabsorption [53].

The experimental technique presented in this thesis overcomes the limitations associated with the above experiments by specifically designing an experimental situation that is closely related to the hot phonon effect but allows for the unambiguous isolation of LO phonon reabsorption. The basic idea of this technique is to use an infrared laser to create an electron-hole plasma of variable and well-controlled density in a GaAs quantum well sample. This plasma is near the ambient temperature of the lattice ($T = 5$ K); hence it does not directly contribute to the LO phonon generation. It can therefore be used to isolate the density-dependent
effects these carriers have on the dynamics of hot LO phonons generated by the relaxation of a small and fixed population of hot carriers that are injected by a visible laser. The LO phonon dynamics is monitored using Raman scattering. This time-resolved Raman/optical doping (TRROD) technique in conjunction with a specifically designed quantum well bandstructure permits a clear demonstration of LO phonon absorption effects by intervalence band transitions of free holes from the lattice perspective. Two advantages of the TRROD technique over that of the similar experiment by Kash et al. in a p-doped GaAs sample are the fact that there are no contributions to the LO phonon decay by scattering with doping-induced impurities, and the fact that a wide range of carrier densities can be investigated in one sample. Previous Raman scattering experiments in p-doped bulk GaAs have shown that the Raman spectra can differ qualitatively for different dopants [13].

From a theoretical perspective, calculations of the hot phonon effect in GaAs quantum wells have focussed on modelling the temporal evolution of the electron temperature and energy loss rate as measured in time-resolved luminescence and absorption measurements [15, 16]. These calculations are based on a solution of the time-dependent coupled Boltzmann equations for the single-particle distribution functions of carriers and phonons. Common features of these calculations are that screening due to many-body interactions is included phenomenologically in the static limit and the valence band structure is approximated by a parabolic band model. The calculations by Pötz and Kocevar [16] are restricted to a single subband for electrons and holes and the "method of moments" is used for solving the equations. Goodnick and Lugli [15] use a multi-subband model and a "Monte Carlo" approach. The above calculations specifically address the influence of the hot plasma density on the electron energy loss rate. These models are closely related to the situation in the TRROD experiment but differ in that the TRROD technique measures the influence of a variable density electron-hole plasma with a temperature close to that of the lattice ("cold plasma") on the dynamics of a fixed population of hot LO phonons. This specific situation has not been addressed in the literature to date. Thus the author had to develop a model to calculate the contribution of intervalence band transitions to the LO phonon lifetime. This
model is based on a calculation of the imaginary part of the LO phonon self energy. The strong coupling of free carriers and LO phonons in GaAs requires the inclusion of screening effects in the calculation. The model includes \textit{dynamic} screening effects and thus goes beyond the static screening limit. To obtain a realistic model for the experimental results, the author went beyond the parabolic band model for the valence band structure and included a $\vec{k} \cdot \vec{p}$ band structure model in both density of states and matrix elements. Using this approach, the author could achieve good semi-quantitative agreement with the observed reduction in the LO phonon lifetime.

Even though the TRROD experiments and the calculations were performed for a GaAs quantum well, it is expected that similar experiments in bulk GaAs would lead to results that are qualitatively the same. The results of this thesis thus give further support to the suggestion of Kash \textit{et al.} that intervalence band transitions play an important role in the reabsorption of hot LO phonons in bulk GaAs.

\subsection*{1.2 Details of Free Carrier-LO Phonon Interactions in GaAs Quantum Wells}

The experimental technique used in this thesis uses a quantum well sample with growth parameters chosen specifically to isolate LO phonon reabsorption processes. Quantum wells consist of very thin semiconductor layers (e.g., GaAs with a thickness on the order of 100 Å) that are sandwiched between a semiconductor with larger band gap (e.g. AlGaAs). In this way, quasi-two dimensional "quantum confinement" of the electrons and holes is achieved in the growth direction, leading to a splitting of the bulk energy levels into electronic subbands that can be controlled by choosing a particular well width and band gap of the barriers. Quantum wells share many relaxation features of the bulk structures but display some additional effects due to the two-dimensional confinement. These are reviewed in this section. This thesis specifically deals with carrier relaxation in GaAs/AlGaAs quantum wells, but the physical processes discussed in this section are common to
any quantum wells that incorporate polar semiconductor materials. LO phonons interact with the free carriers by the short range deformation potential interaction and the long range, polar Fröhlich interaction [1]. This thesis focusses on the Fröhlich interaction which strongly favours interactions with small wavevector LO phonons and dominates the carrier relaxation process in polar materials. There are two types of carrier transitions that are important in the emission and absorption of small wavevector phonons (see Fig. 1.1): 1) Intrasubband transitions, i.e. transitions where initial and final state of the carrier are in the same subband and 2) Intersubband transitions where initial and final states are in different subbands. Due to the layered structure of quantum wells, the translational symmetry of the semiconductor is broken in the growth (z) direction. Hence there is no momentum conservation for carrier motion along the z-direction and the usual three dimensional wavevector is not a good quantum number. However, the motion in the plane of the quantum well (i.e., perpendicular to the growth direction) is unaffected by the confinement and the in-plane component of the wavevector is conserved. In the of the thesis this component of the wavevector is denoted \( \vec{k}_\parallel \) for the free carriers and \( \vec{q}_\parallel \) for phonons. The quantum confinement not only affects electrons and holes but also the phonons. The bulk acoustic phonons are zone-folded at the edges of the Brillouin "mini-zone" defined by the inverse of the period of a (multiple) quantum well and the bulk LO phonons split into bulk-like confined modes and surface-like interface modes. Each type of mode has a different interaction strength with the free carriers and thus in general the details of the free carrier LO phonon interaction are very complicated. In this thesis LO phonon confinement effects are considered only in terms of parity selection rules for free hole-LO phonon interactions and a bulk LO phonon model is used for the strength of the interaction.

The small \( \vec{q}_\parallel \) LO phonons generated by the scattering processes can be reabsorbed by free carriers or decay by anharmonic lattice interactions into two zone boundary acoustic phonons (Fig. 1.1). The latter process is the one that ultimately removes energy from the coupled free carrier-LO phonon system. Decay into acoustic phonons by anharmonic interactions typically occurs at a lower rate than LO
Figure 1.1: Schematic overview of free carrier-phonon interactions in polar semiconductors and experimental methods for investigating them. 1) Intrasubband scattering process, 2) intersubband scattering process, 3) anharmonic decay into acoustic (AC) phonons.
phonon generation. As a consequence, there is a buildup of a nonequilibrium ("hot") LO phonon population that can reheat the free carriers by reabsorption [4] that manifests itself in the hot phonon effect.

1.3 Time-resolved Raman Scattering from Optically Doped Semiconductors

The experimental technique presented in this thesis is an extension of the time-resolved resonant Raman scattering technique using a backscattering geometry[17]. It was developed to isolate LO phonon reabsorption processes and thus allows for an unambiguous measurement of free hole contributions to the LO phonon lifetime.

Raman scattering (or inelastic light scattering) is a powerful technique for the direct measurement of the population of elementary excitations (e.g. phonons and plasmons) in semiconductor samples [18] since the ratio of the intensities of the Stokes and anti-Stokes Raman lines are directly related (except for resonance effects) to the occupation number of the corresponding excitation. Time-resolved Raman scattering thus allows for a measurement of the temporal evolution of occupation numbers. A further advantage of the Raman scattering technique is its selectivity in energy and wavevector of the elementary excitation. This in principle makes a measurement of relaxation processes at different points in the Brillouin zone possible. However, for the Raman backscattering technique used in the TRROD experiments $q_\parallel$ is restricted to values near the center of the Brillouin zone. In quantum well samples the quantum well only constitutes a very small fraction of the total sample volume. Due to this small filling factor, it is imperative in most experiments to use resonant Raman scattering. In order to resonantly enhance the Raman signal, the wavelength of the incoming or scattered light can be tuned to an electronic transition of the sample under consideration. This resonant enhancement typically increases the signal by several orders of magnitude but can lead to a breakdown in the momentum selection rules.

The TRROD technique can be described with the aid of Fig. 1.2: at time $t = \tau_1$ a cold electron-hole plasma is injected into a GaAs quantum well sample by
Figure 1.2: Schematic overview of TRROD technique
a pulse from laser 1. Laser 1 is tuned close to the fundamental gap of the sample. Thus the excess energy of the electrons and holes is small. At \( t = \tau_2 \), electrons and holes with a large amount of excess energy ("hot carriers") are injected into the sample using a pulse from laser 2. The fixed time delay \( \tau_2 - \tau_1 \) between the pulses of laser 1 and laser 2 is set to be larger than the pulse duration of laser 1, but much shorter than the recombination time of the electron-hole pairs injected at the band gap. Subsequently at \( t = \tau_3 \) a second pulse from laser 2 is used to monitor the population of hot LO phonons created during the carrier relaxation process of the hot electrons and holes by measuring the anti-Stokes Raman scattering signal (at \( \omega_{AS} \)). The experiment is repeated at various delays \( \tau_3 - \tau_2 \). In this way the temporal evolution of the LO phonon population under the influence of the cold electron-hole plasma can be determined.

It is important to keep the hot carrier injection levels low, in order to minimize hot phonon reabsorption effects within the cooling hot plasma. By keeping the hot carrier and Raman probe injection levels constant and varying the density of the cold plasma, any change in the phonon lifetime is then due to the cold plasma. This technique of combining time-resolved Raman scattering with optical doping thus makes it possible to isolate LO phonon reabsorption by the cold plasma from generation processes and introduces a way to systematically study free carrier effects on the LO phonon lifetime. In addition to the lifetime effects, this method also provides access to the excitation spectra of the sample which contain direct information on the renormalization of coupled plasmon-phonon modes and their role in the carrier relaxation process.

In order to isolate the influence of valence band transitions on the LO phonon lifetime by using the TRROD technique, a GaAs/AlGaAs quantum well sample with a specific subband structure was chosen for the studies presented in this thesis. The following section outlines the criteria for choosing this particular structure. Details of the sample parameters are given in Chapter 3.

The energy spacings between the subbands are chosen in such a way that the spacings between two conduction band levels (\( c_1 \) and \( c_2 \), see Fig. 1.3) and two valence band levels (\( v_0 \) and \( v_1 \)) are close to the LO phonon energy. As a consequence
Figure 1.3: Schematic diagram of model used in reabsorption calculations

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there exist allowed emission and absorption processes for small wavevector LO phonons between these levels in the course of the relaxation of optically injected hot carriers. The spacing between $c_0$ and $c_1$ is less than the LO phonon energy and as a consequence no reabsorption of LO phonons by the cold electrons can take place between these two bands. If the density of the cold electron-hole plasma in the TRROD experiment is much larger than that of the hot plasma but the Fermi energy, $E_F$, for the electrons is less than the $c_0$-$c_1$ spacing, reabsorption of LO phonons by free carriers is dominated by $v_0$-$v_1$ intervalence band transitions. Reabsorption of cooled hot electrons from $c_1$ to $c_2$ can be neglected due to the low hot carrier densities involved.

1.4 Calculations of Free Carrier Effects on the LO Phonon Lifetime

The two contributions to the LO phonon lifetime measured in the TRROD experiment described in the previous section are anharmonic decay into acoustic phonons and absorption by intersubband transitions of free holes. The calculations in this thesis focus on the contributions to the LO phonon lifetime by free holes. To compare the experimental results with the calculations, the anharmonic decay rate is included phenomenologically by using the inverse of the measured LO phonon lifetime with no cold plasma present.

The simplest approach for calculating the free hole contributions to the LO phonon lifetime is to use a Fermi's golden rule based analysis. This single-particle approach is not expected to give good agreement with the experimental results due to the strong interaction of the small wavevector LO phonons probed in the experiment with the cold electron-hole plasma.

These interaction effects can be taken into account by calculating the LO phonon lifetime using a many-body calculation that takes screening by the free carriers into account self-consistently. The approach taken by the author is to calculate the LO phonon lifetime through the LO phonon self energy. The self energy of a (quasi-) particle incorporates all interactions of this particle with the
other excitations of the system. The real part of the self energy gives the energy renormalization of the particle due to the many-body interactions and the imaginary part of the self energy is directly related to the lifetime of the particle. Self energy calculations have been successfully used in the past for modelling hot carrier relaxation processes in GaAs quantum wells (see, e.g., Ref. [19]), but have so far focussed on calculations of electron self energies. In this thesis the author derives a formalism for evaluating the contributions of \( \nu_0-\nu_1 \) intersubband transitions to the imaginary part of the LO phonon self energy for the sample described in the previous section. Since details of the band structure have strong effects on the kinematic constraints of the intersubband transitions, the complicated GaAs quantum well band structure was calculated within a \( \vec{k} \cdot \vec{p} \) model and incorporated into the model. Screening of the free hole-LO phonon interaction was included in the LO phonon self energy through a calculation of the dominant contributions by free electrons and holes to the two-dimensional (2D) dielectric function. To the best knowledge of the author, this is the first calculation of contributions of intervalence band transitions to the LO phonon lifetime in GaAs quantum wells that takes energy and wavevector dependent ("dynamic") screening effects and a realistic band structure into account.

The theoretical results reported in Chapters 2 and 4 of this thesis reproduce the salient feature of the TRROD experimental data, lending support to the approach taken. It is expected that the formalism can also be successfully used for modelling LO phonon lifetimes in other III-V semiconductor systems as well as for modelling hole-screening effects and results of infrared absorption experiments in p-doped quantum wells.

1.5 Organization of the Thesis

The remainder of this thesis is organized as follows. In Chapter 2 the formalism for calculating free hole contributions to the LO phonon lifetime using the LO phonon self energy incorporating both many-body effects and a realistic model for the valence band structure are presented. The first part of the chapter contains
a review of the phonon self energy for parabolic bands and of $\vec{k} \cdot \vec{p}$ calculations of the band structure as well as a discussion of the free hole-LO phonon interactions in GaAs quantum wells. The second part of the chapter introduces the extension of the self energy formalism to nonparabolic bands that was developed by the author. Results of that model are presented for the quantum well parameters used in the experiment. Chapter 3 describes the experimental details of the TRROD experiment that was carried out by the author in order to measure the effective lifetime of LO phonons in GaAs quantum wells due to reabsorption by free holes. In that chapter results of measurements dealing with the sample characteristics are presented. In Chp. 4 the experimental results of the measurements of the effects of the cold electron-hole plasma on the LO phonon lifetime are discussed in detail. These results that demonstrate direct evidence for the reabsorption of LO phonons by free carriers are compared with the calculations from Chp. 2. In Chp. 5 the thesis is concluded with a summary of the results obtained and an outlook on possible extensions to the studies presented in this thesis is given.
In this chapter a theoretical model for calculating the absorption rate of LO phonons by free holes in GaAs quantum wells is described. The model, developed by the author, is based on a calculation of the contributions of intervalence band transitions to the LO phonon lifetime using the self energy formalism, taking a realistic band structure and screening effects explicitly into account. The theoretical results presented can be directly compared to the experimentally obtained results that are reported in Chapter 4.

This chapter is organized as follows: in Section 2.1 the rate equations that govern the interactions between the hot carriers and the LO phonons are presented and the effective lifetime of a LO phonon is introduced. In Section 2.2 a brief introduction to the relation between the self energy and the lifetime of an LO phonon is given. The free hole contributions to the LO phonon self energy for parabolic bands are reviewed in Section 2.3. To extend this formalism to nonparabolic bands, it is necessary to understand the valence band structure and the hole-LO phonon interaction in quantum wells. Section 2.4 therefore contains a review of the quantum well valence band structure and presents the results of calculations for the band structure of a the GaAs quantum well used in the TRROD experiments. Section 2.5 describes the model for the treatment on the hole-LO phonon interaction. Based on the results of Sections 2.4 and 2.5, the free hole contributions to the LO phonon self energy for nonparabolic bands are derived in Section 2.6. Special consideration is hereby given to the correct inclusion of screening effects through
the 2D dielectric function of the cold electron-hole plasma. As a conclusion to the development of the formalism, the numerical results for the LO phonon lifetime for the experiment described in Chapters 3 and 4 are presented and discussed.

2.1 Calculations of LO Phonon Lifetime

In the following, the author introduces the effective LO phonon lifetime $\tau_{\text{eff}}$ as a meaningful parameter to characterize free carrier-LO phonon absorption processes [6]. The energy loss rate $\frac{dE}{dt}$ of free carriers due to interaction with LO phonons of wavevector $q$ can be expressed as

$$\frac{dE}{dt} = \sum_{q} h\omega_{LO} (-R_{\text{em}}(n_{q} + 1) + R_{\text{abs}} n_{q}). \quad (2.1)$$

Here $R_{\text{em}}$ and $R_{\text{abs}}$ denote the LO phonon emission and absorption rates respectively and $h\omega_{LO}$ represents the LO phonon energy. The occupation number $n_{q}$ of LO phonons with wavevector $q$ is given by

$$\frac{dn_{q}}{dt} = R_{\text{em}}(n_{q} + 1) - R_{\text{abs}} n_{q} - \frac{n_{q}}{\tau_{\text{anh}}}. \quad (2.2)$$

Here $\tau_{\text{anh}}$ represents the decay time due to anharmonic relaxation into acoustic phonons. In these equations the equilibrium occupation number of LO phonons is assumed to be equal to zero. This approximation is made, since all experiments presented in this thesis were performed at $T = 5$ K, where the LO phonon equilibrium population is negligible. It is useful to rewrite the second rate equation in order to clearly separate LO phonon emission and absorption processes:

$$\frac{dn_{q}}{dt} = R_{\text{em}}(n_{q} + 1) - \frac{n_{q}}{\tau_{\text{eff}}}. \quad (2.3)$$

The effective lifetime $\tau_{\text{eff}}$ now incorporates all LO phonon absorption processes, i.e. anharmonic decay and reabsorption by the free carriers. Before proceeding, it is important to note that $\tau_{\text{eff}}$ corresponds to the population lifetime of a given LO phonon mode, which should be distinguished from the dephasing time of that specific vibrational excitation. The latter has been studied in bulk III-V semiconductors using coherent anti-Stokes Raman scattering [20].

The remainder of this chapter describes the calculation of contributions to $\tau_{\text{eff}}$ that are due to intervalence band transitions of free holes.
2.2 Self Energy and LO Phonon Lifetime

The concept of self energy plays an important role in many-body solid state physics, since it contains information on the energy renormalization and lifetime of elementary excitations. A strict derivation of an expression for the self energy is beyond the scope of this thesis and can be found for instance in [21]. Nevertheless, the following paragraph attempts to summarize the physical significance of the self energy.

In many-body physics, Green's functions, also called propagators, describe the dynamics of (quasi-)particles. In a quantum well, the unscreened LO phonon Green's function $G_{KK'}(\vec{q}_b, i\omega_n)$ describes the propagation of an unscreened ("bare") LO phonon with wavevector $\vec{q}_b$ and energy $\hbar \omega_n$ from state $K$ to state $K'$. In the presence of electrons and holes this propagation can of course be influenced by the other particles present. To take into account these many-body effects on the propagator one therefore really has to calculate the screened LO phonon Green's function $G^\text{sc}_{KK'}(\vec{q}_b, i\omega_n)$ that takes all interactions of the electrons and holes with the LO phonon into account. Interactions with single particle free carrier excitations as well as interactions with collective excitations ("plasmons") have to be taken into account. The screened Green's function is given by the Dyson equation [22]

$$G^\text{sc}_{KK'}(\vec{q}_b, i\omega_n) = G_{KK'}(\vec{q}_b, i\omega_n) - \sum_{LL'} G_{KL}(\vec{q}_b, i\omega_n) \Sigma_{LL'}(\vec{q}_b, i\omega_n) G^\text{sc}_{LK'}(\vec{q}_b, i\omega_n).$$ (2.4)

All information about the many-body effects is contained in the complex self energy $\Sigma_{LL'}(\vec{q}_b, i\omega_n)$. The real part of $\Sigma_{LL'}(\vec{q}_b, i\omega_n)$ is related to the energy renormalization of the LO phonon due to the interaction with all other particles. The imaginary part of $\Sigma_{LL'}(\vec{q}_b, i\omega_n)$ is related to the lifetime of the LO phonon by

$$\frac{1}{\tau(\vec{q}_b, \omega)} = \frac{2}{\hbar} Im[\Sigma_{LO}(\vec{q}_b, \omega)].$$ (2.5)

Eq. 2.4 for the self energy can be solved iteratively by diagramatic techniques. The treatment of the self energy in this thesis is done within the Random Phase Approximation (RPA)[21] and is restricted to contributions due to intersubband transitions of free holes.\(^1\)

\(^1\)The lifetime in Eq. 2.5 therefore corresponds to the population lifetime of the LO phonons.
2.3 Review of the Parabolic Two Band Model

To begin the consideration of the LO phonon self energy and to offer some insight the author starts by reviewing the results for the free hole contributions to the LO phonon self energy within a simple parabolic band model. Later, results for the more realistic non-parabolic band model are derived.

The free hole contributions to the LO phonon self energy $\Sigma_{LO}$ for a parabolic two subband (labeled 0 and 1) quantum well model can be directly inferred from the expression for bulk semiconductors [21]:

$$\Sigma_{LO}(q_\parallel, \omega) = \frac{|M_{01}(q_\parallel)|^2(P_{01}(q_\parallel, \omega) + P_{10}(q_\parallel, \omega))}{\epsilon_{0101}(q_\parallel, \omega)}$$  \hspace{1cm} (2.6)

Here, $M_{01}$ is the matrix element for 0-1 intersubband transitions of free holes that are driven by LO phonons. The bare 2D polarizability function for free carriers $P_{01}$ is given by [22, 23]

$$P_{01}(\vec{q}_\parallel, \omega) = -2 \int \frac{d\vec{k}_\parallel}{(2\pi)^2} \frac{f_1(\vec{k}_\parallel + \vec{q}_\parallel) - f_0(\vec{k}_\parallel)}{\hbar \omega - E_1(\vec{k}_\parallel + \vec{q}_\parallel) + E_0(\vec{k}_\parallel)}$$  \hspace{1cm} (2.7)

and $P_{10}$ can be obtained trivially by switching the indices 0 and 1. Here $f_0$ and $f_1$ stand for Fermi-Dirac occupation factors and $E_0$ and $E_1$ represent hole energies in band 0 and 1 respectively. In the case of quantum wells, the 2D dielectric function $\epsilon_{ijlm}(q_\parallel, \omega)$ is a matrix with indices $\{ij\}$ and $\{lm\}$ that designate pairs of levels involved in $i\rightarrow j$ and $l\rightarrow m$ inter- and intrasubband transitions. The contribution to the 2D dielectric function $\epsilon_{0101}(q_\parallel, \omega)$ from 0-1 intersubband transitions of free holes is given by

$$\epsilon_{0101}(q_\parallel, \omega) = 1 - V_{0101}(q_\parallel)(P_{01}(q_\parallel, \omega) + P_{10}(q_\parallel, \omega)),$$  \hspace{1cm} (2.8)

with the Coulomb matrix element (in cgs units)

$$V_{0101}(q_\parallel) = \frac{2\pi \varepsilon^2}{\varepsilon_\infty q_\parallel} \int dz \int dz' \psi_0^*(z)\psi_1(z)e^{-q_\parallel|z-z'|}\psi_0^*(z')\psi_1(z')$$  \hspace{1cm} (2.9)

The $\psi(z)$ are the envelopes of the hole wavefunctions as a function of the growth direction. Within the two subband model Eq. 2.5 can thus be written as

$$\frac{1}{\tau(q_\parallel, \omega)} = \frac{-2 \ |M_{01}(q_\parallel)|^2Im[P_{01}(q_\parallel, \omega) + P_{10}(q_\parallel, \omega)]}{\hbar \epsilon_{0101}(q_\parallel, \omega)^2}$$  \hspace{1cm} (2.10)
The interpretation of the terms in Eq. 2.10 is relatively straightforward. It can be shown that in the case of $\varepsilon_{0101}(q_{\parallel},\omega) = 1$, i.e. without any screening, Eq. 2.10 reduces to the familiar single particle Fermi's golden rule result for the LO phonon-free carrier scattering rate:

$$\frac{1}{\tau(q_{\parallel},\omega)} \propto |M_{01}(q_{\parallel})|^2 f_0[1 - f_1] \delta(E_0 + \hbar\omega - E_1)$$

(2.11)

This shows that the 2D polarizability functions are equivalent to the Fermi-Dirac occupation factors times the density of states. The effects of screening by free holes are included in Eq. 2.10 through the square of the magnitude of the 2D dielectric function $\varepsilon_{0101}(q_{\parallel},\omega)$. Since this function is of great importance in the calculations presented, its physical significance is discussed here in more detail.

Dielectric functions play a central role in calculating the response of linear systems to external perturbations. In general a dielectric function $\varepsilon(q,\omega)$ relates the Fourier components of an external potential $V_{\text{ext}}$ to the internal potential $V_{\text{int}}$ within a system [21] i.e.,

$$V_{\text{int}} = V_{\text{ext}}/\varepsilon(q,\omega).$$

(2.12)

The dielectric function is of great importance in the calculation of excitation spectra of coupled free carrier-LO phonon systems in semiconductors. If $\varepsilon(q,\omega) = 0$, an infinitesimally small external perturbation causes a large local response. Under this condition the system collectively oscillates. Examples for these collective oscillations in the case of an electron gas are plasmon excitations, corresponding to collective oscillations of the carriers. The imaginary part of the dielectric function is non-zero in regions of phase space where (uncorrelated) single particle transitions can take place.

For quantum wells, the collective excitations can be determined by finding the roots of the determinant of the dielectric matrix. The knowledge of the dielectric matrix for the coupled 2D free carrier-LO phonon system makes it possible in principle to calculate the Raman spectra of the system, since the Raman spectra under non-resonant conditions are related to the imaginary part of the determinant of the inverse of the dielectric matrix. However, the calculation of Raman spectra of an electron-hole plasma coupled to LO phonons is complicated for two reasons.
First, the dielectric matrix is difficult to set up and invert due to the number of possible inter- and intrasubband transitions and their coupling to the LO phonons. Second, in the case of resonant Raman scattering the coupling of the probe light to the excitations within the sample can lead to strong effects on the spectra. These are the reasons why this thesis focuses on a simple two subband model for the dielectric function to model screening effects on the LO phonon lifetime, but discusses the Raman spectra on a more qualitative level (see Chp. 4).

In the calculation of carrier and LO phonon relaxation processes, \( \epsilon(q, \omega) \) is also very important [24]. A hot carrier (or LO phonon), with energy \( h\omega \) and momentum \( q \), exerts an external potential on the surrounding system, described by \( \epsilon(q, \omega) \), that includes, for example, a density of cold carriers. The dielectric function describes the screening of the interaction of the hot carrier with the cold carriers and also determines if collective and/or single particle excitations can absorb the hot carrier (or LO phonon).

The main task in going from a single particle approximation to a many-body calculation for a quantum well is thus to evaluate the dielectric function \( \epsilon_{ijij}(q_h, \omega) \). If, as for most calculations in this thesis, only one set of intersubband transitions is being considered, \( \epsilon_{ijij}(q_h, \omega) \) reduces to a scalar. There are two different approaches to calculating the dielectric function. The simplest is to neglect the frequency dependence and to derive \( \epsilon_{ijij}(q_h, 0) \). This approximation is commonly called static screening. In general however, one has to perform a calculation that includes dynamic screening, i.e. retain the full frequency dependence in the dielectric function [24].

In the case of a single quantum well with parabolic bands, analytic expressions can be derived for the dielectric function [22]. For nonparabolic bands the situation is much more complicated. The dynamically screened formalism including nonparabolic bands is developed in Section 2.6 of this thesis. In order to understand the details of that formalism it is necessary to first review the valence band structure and the free hole-LO phonon interaction in GaAs quantum wells.
2.4 Valence Band Structure of GaAs Quantum Wells

The usual way of calculating the valence band structure of a GaAs quantum well (see e.g. Ref. [25]) is to match the bulk eigenstates of the well (A) and the barrier (B) materials (Fig. 2.1) at the interfaces so that they are continuous and current density is conserved. This is very similar to a “particle in a box” calculation [26], where the continuity of current density is expressed as a continuity in the first derivative of the wavefunction, since the particle mass is assumed to be the same in both materials.

Fig. 2.2 shows the band structure of bulk GaAs near the center of the Brillouin zone (Γ-point). The energy difference of the valence band transitions of interest for the calculations in this thesis are on the order of the LO phonon energy (36.2
Figure 2.2: Schematic diagram of the bulk GaAs band structure near the Γ-point.
meV), which is much less than the bandgap energy \( E_G = 1.519 \) eV or the split-off energy \( \Delta = 340 \) meV. The following discussion is thus restricted to the \( \Gamma_8 \) heavy and light hole bands.

A particle in a given band (label \( n \)) and material A or B is characterized by a dispersion relation \( E_n^{A,B}(\vec{k}_{A,B}) \) and a Bloch wavefunction

\[
\Psi_{n\vec{k}_{A,B}}^{A,B}(\vec{r}) = \Omega^{-1/2} u_{n\vec{k}_{A,B}}^{A,B} e^{i\vec{k}_{A,B} \cdot \vec{r}}.
\]

Here, \( \Omega \) denotes a normalization factor, \( n \) is a band index, \( u_{n\vec{k}_{A,B}}^{A,B} \) represents the lattice-periodic part of the Bloch function and \( \vec{k}_{A,B} \) is the wavevector of a carrier in material A or B, as determined from the bulk dispersion in the respective material.

The states of the \( \Gamma_8 \) hole valley originate from the p-orbitals of the underlying atoms. They can be characterized by the total angular momentum \( J \) and its projection \( m_J \) onto the z-axis. The basis states are thus the four wavefunctions that span the \((J,m_J)\) basis for \( J = 3/2 \)

\[
\begin{align*}
  u_{3/2}^+ &= |3/2,3/2\rangle = -\frac{1}{\sqrt{2}}|(X+iY)\uparrow\rangle, \\
  u_{1/2}^+ &= |3/2,1/2\rangle = \frac{1}{\sqrt{6}}|-(X+iY)\downarrow + 2Z\uparrow\rangle, \\
  u_{-1/2}^+ &= |3/2,-1/2\rangle = \frac{1}{\sqrt{6}}|(X-iY)\uparrow + 2Z\downarrow\rangle, \\
  u_{-3/2}^+ &= |3/2,-3/2\rangle = \frac{1}{\sqrt{2}}|(X-iY)\downarrow\rangle.
\end{align*}
\]

Here \( X, Y \) and \( Z \) stand for wavefunctions that transform like atomic p-orbitals and \( \uparrow \downarrow \) denote the direction of the spin. At \( k = 0 \) the \( \Gamma_8 \) level is four-fold degenerate. Away from \( k = 0 \) the level splits into two bands (heavy and light hole). Each of these is two-fold degenerate.

To derive the valence band structure for quantum wells it is useful to introduce the \( \vec{k} \cdot \vec{p} \) envelope function approximation. This approximation leads to valuable simplifications of the problem while still containing the essential physics. A detailed account of this formalism can be found e.g. in [27]. Recently Kash et al. [28] have shown that this way of calculating the GaAs quantum well valence band structure yields results that are in excellent agreement with data from conduction band to acceptor luminescence experiments.
The key to introducing the envelope function approximation is the fact that the two \( \vec{r} \) dependent terms in Eq. 2.13 vary on different length scales: the \( u_n \) have the periodicity of the underlying lattice structure, whereas the plane wave part oscillates with a spatial period that is on the order of the width of the quantum well. With the lattice constant of GaAs being 5.65 \( \text{\AA} \) and quantum wells usually having widths of 50 \( \text{\AA} \) or more, the length scales differ by at least one order of magnitude. The idea of the envelope function approximation is to deal only explicitly with the slowly varying plane wave part (envelope function) and have the rapidly varying component enter through \( a \ priori \) parameters. Instead of solving the original Schrödinger equation involving the periodic crystal potential, one can then solve an associated effective mass equation for the envelope functions [25].

For the \( \Gamma_8 \) states this approach leads to the so called Luttinger-Kohn Hamiltonian \( H \) [29]. The following equation gives their result for the Hamiltonian within the basis (\( \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2} \)) [25]:

\[
H = \begin{pmatrix}
P + Q & -S & R & 0 \\
-S^* & P - Q & 0 & R \\
R^* & 0 & P - Q & S \\
0 & R^* & S^* & P + Q \\
\end{pmatrix}
\] (2.18)

with

\[
P(\vec{k}) = \frac{\hbar^2}{2m} \gamma_1 (k_x^2 + k_y^2 + k_z^2), \]

\[
Q(\vec{k}) = \frac{\hbar^2}{2m} \gamma_2 (k_x^2 + k_y^2 - k_z^2), \]

\[
S(\vec{k}) = \frac{\hbar^2}{2m} 2\sqrt{3} \gamma_3 (k_x - ik_y) k_z, \]

\[
R(\vec{k}) = \frac{\hbar^2}{2m} \sqrt{3} [-\gamma_3 (k_x^2 - k_y^2) + 2i \gamma_3 k_x k_y].
\]

(2.19) \hspace{1cm} (2.20) \hspace{1cm} (2.21) \hspace{1cm} (2.22)

Here \( \gamma_1, \gamma_2 \) and \( \gamma_3 \) are the dimensionless Luttinger parameters that contain the physics of the lattice potential and are assumed to be known \( a \ priori \) in the envelope function approximation. The wavevector of the valence holes is denoted by \( \vec{k} \). Each quantum well valence band eigenstate is given by a four-component envelope function spinor \( \vec{F} \). The four components of this spinor are the projections of \( \vec{F} \) on the four orthogonal states that span the basis (\( \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2} \)). Due to time-reversal
symmetry of the Hamiltonian, all eigenstates are two-fold degenerate ("Kramers degeneracy"). This is similar to the situation for the $\Gamma_6$ conduction bands, which are two-fold degenerate with respect to the $+\frac{1}{2}$ or $-\frac{1}{2}$ electron spin. For holes however, as pointed out by Uenoyma and Sham [30], the role of the usual spin for the conduction band states is replaced by a parity quantum number $p = \pm 1$ of the spinor. This quantum number arises from the properties of the hole spinors under mirror reflection $z \rightarrow -z$. The parity of a given spinor is related to the parity of its spin components by

$$F_p(z) = \begin{pmatrix}
F_{\frac{3}{2},p}(z) \\
F_{\frac{1}{2},-p}(z) \\
F_{-\frac{3}{2},p}(z) \\
F_{-\frac{1}{2},-p}(z)
\end{pmatrix}. \quad (2.23)$$

This means that if the parity of $F$ is e.g. $+1$, then the $\frac{3}{2}$ and $\frac{1}{2}$ component also have parity $p = +1$, whereas the $-\frac{3}{2}$ and $-\frac{1}{2}$ components have parity $p = -1$. Due to the degeneracy mentioned above, each hole energy level $n, \vec{k}$ has one eigenstate with even (+1) and one with odd (-1) parity. It is straightforward to show that any of the two eigenfunctions can be obtained from the other by replacing the component $m_J$ with the component $-m_J$ of that same function. At any $k_\parallel$, each spinor envelope element is a superposition of all $k_\parallel = 0$ envelope wavefunctions with the same parity. This is in contrast to the conduction subbands, where the parity of a subband is determined by its level number $n = 0, 1, 2, ..., n$. The conduction subbands are degenerate with respect to spin, and the parity is determined by $p = (-1)^n$. The parity quantum number plays a central role in the analysis of inter- and intravalence band transitions, since the hole-LO phonon interaction Hamiltonian can be separated into parity-conserving and parity-altering components [30]. This leads to very useful parity selection rules for the hole/LO phonon interaction.

For the calculations of the free hole contributions to the LO phonon self energy, both the hole wavefunctions and the dispersion are required. In this thesis the axial approximation [31] to the Luttinger-Kohn Hamiltonian is used. Within this approximation, the constant energy surfaces are assumed to be cylindrically symmetric around the $k_z$-axis which means that band-warping in the $k_\parallel$ plane is neglected. Details of the calculations can be found in Appendix A.
Fig. 2.3 shows the result of the valence band structure calculation for a 200 Å GaAs/Al$_{20.70}$Ga$_{70}$As quantum well. The parameters for this calculation are given in the caption of Fig. 2.3. The bands are labelled by their heavy (hh) or light (lh) hole character at zone center. The result shows the well known strong mixing and nonparabolicity of the bands. To ensure reliability of the program written for the calculations, results of bandstructure calculations for a range of different quantum well parameters were successfully checked against published results [25]. Of specific interest for the subject of this thesis are bands hh$_0$ and hh$_3$ (corresponding to $v_0$ and $v_1$ in Fig. 1.3) since the energy spacing between these bands is close to the LO phonon energy and a strong contribution to LO phonon reabsorption by intersubband transitions can be expected. The hh$_0$ and hh$_3$ bands actually have opposite curvature at $k_{\|} = 0$, with hh$_3$ having an electron-like dispersion.

In Fig. 2.4 the individual $m_J$ components of the $p = +1$ hh$_0$ and hh$_3$ envelope functions are plotted for three different values of $k_{\|}$. This part of the program has also been successfully checked against published results for a 100 Å GaAs/Al$_{1.79}$Ga$_{2.41}$As quantum well [32]. At $k_{\|} = 0$ the only non-vanishing $m_J$ component is $\frac{3}{2}$ for hh$_0$ and $-\frac{3}{2}$ for hh$_3$. At $k_{\|} = 7.5 \cdot 10^5$ cm$^{-1}$ mixing with other bands is evident. For hh$_0$ there are obvious contributions from the hh$_1$, lh$_0$ and lh$_1$ bands, whereas the dominant admixture for the hh$_3$ wavefunction is due to the lh$_2$ band. At $k_{\|} = 1.5 \cdot 10^5$ cm$^{-1}$ the mixing is increasing. For hh$_5$, the lh$_0$ contribution is of comparable strength and there is also increased admixture of hh$_1$ and lh$_1$. For the hh$_3$ band, the effects of mixing are even stronger. It has essentially lost most of its hh$_3$ character. The dominant contributions are now from the lh$_1$ and hh$_4$ bands.

This strong dependence of the wavefunctions on $k_{\|}$ of course has direct consequences for the Coulomb matrix elements and the hole-LO phonon matrix elements between these states. This is discussed in Section 2.6.
Figure 2.3: In-plane valence band structure of 200 Å GaAs/Al$_{30}$Ga$_{70}$As quantum well with $V_b = 17.4$ meV, $\gamma_1^B = 5.66$, $\gamma_2^B = 1.49$, $\gamma_3^B = 2.35$, $\gamma_1^A = 6.80$, $\gamma_2^A = 1.9$, $\gamma_3^A = 2.9$. The energy plotted is the absolute value of the confinement energy of the holes with respect to the bulk GaAs valence band edge.
Figure 2.4: Valence band wavefunctions at different $k_y$. Solid curve: $m_J = \frac{3}{2}$; short dashed curve: $m_J = \frac{1}{2}$, dotted curve: $m_J = -\frac{1}{2}$; long dashed curve: $m_J = -\frac{3}{2}$. 

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2.5 Free Hole-LO Phonon Interaction in Quantum Wells

The free hole contributions to the LO phonon self energy depend on the hole-LO phonon matrix element $M_{01}$ (see Eq. 2.10). In quantum wells not only the electronic band structure is changed by the confinement effects but the phonons are also affected. This fact has to be taken into account for the calculation of $M_{01}$ and is discussed in the following as far as is relevant to the subject of this thesis. This matter is complicated by the fact that there still exists considerable controversy in the literature (see, e.g., the recent review by Ridley Ref. [33]) about different models for LO phonons in quantum wells. It seems that a satisfying description can only be achieved by using a microscopic “chain model” calculation. Unfortunately, these calculations are computationally very intensive. For the small wavevector optical modes of interest in the hot carrier relaxation process, it is often assumed that the microscopic details are not important and the quantum well and the barrier materials are replaced for simplicity by a dielectric and elastic continuum with dielectric constants $\epsilon_A$ and $\epsilon_B$ respectively (Fig. 2.5). Controversial in the ansatz of this model are the boundary conditions imposed at the interfaces. One class of dielectric continuum models assumes electromagnetic boundary conditions at the interfaces. This results in nodes of the electrostatic potential connected with the ion displacements at the interfaces. The other model assumes zero displacement of the two media at the interfaces. These two approaches lead to contradictory results, since a node in the electrostatic potential corresponds to an anti-node in the electric field and hence to a maximum in the ion displacement. Nevertheless, in terms of resulting optical phonon modes, both models lead to a qualitatively similar results in that they give bulk-like confined LO phonon modes and surface-like interface modes. The electrostatic potentials related to these modes can be classified in terms of parity under the transformation $z \rightarrow -z$. In general there exist odd and even parity confined modes and interface modes. The parity of these LO phonon modes plays a central role in the analysis of the relaxation processes through selection rules for the hole-LO phonon matrix element. These are dis-
cussed in the following section in terms of the dielectric continuum model with
electromagnetic boundary conditions. Without precluding the final outcome of
the boundary condition controversy, the author chooses this widely used model for
reasons of simplicity and the fact that symmetry related arguments also hold for
the corresponding modes from other models.

In contrast to the $\Gamma_6$ conduction band, where electrons and LO phonons only
interact via the Fröhlich interaction, holes and LO phonons in the $\Gamma_8$ valley can also
interact via the optical deformation potential. This interaction has been shown to
make strong contributions to the hole relaxation rate that can dominate the total
polar optical scattering at large wavevectors [34]. At the small $q_\parallel$ of interest in the
experiments presented in this thesis however, the Fröhlich interaction (including
screening effects) dominates due to its $1/q_\parallel$ dependence. At $q_\parallel = 5 \cdot 10^4$ cm$^{-1}$ the
strength of the optical deformation potential matrix element is estimated to be
four orders of magnitude smaller than the Fröhlich matrix element and is therefore
neglected.

The dielectric continuum model [35] assumes the quantum well material $\mathbb{A}$
and barrier materials $\mathbb{B}$ to be continuous dielectric media with dielectric constants
$\varepsilon_A$ and $\varepsilon_B$ as shown in Fig. 2.5. Maxwell's equations and electrodynamic boundary
conditions can then be used to calculate the possible LO phonon modes (labelled $\nu$) in this structure. The LO phonon modes resulting from this calculation can be
classified according to the $z$ dependence of the longitudinal part of their polariza-
tion $\mathcal{P}_\nu$ and their parity $p$:

- Symmetric interface modes ($p = +1$)

$$
\mathcal{P}_S(q_\parallel, z) \propto \begin{cases} 
  e^{q_\parallel(z+a)}, & z \leq -a \\
  \frac{\cosh(q_\parallel z/2)}{\cosh(q_\parallel a)}, & -a \leq z \leq a \\
  e^{-q_\parallel(z-a)}, & z \geq a 
\end{cases}
$$ (2.24)

- Antisymmetric interface modes ($p = -1$)

$$
\mathcal{P}_A(q_\parallel, z) \propto \begin{cases} 
  -e^{q_\parallel(z+a)}, & z \leq -a \\
  \frac{\sinh(q_\parallel z/2)}{\sinh(q_\parallel a)}, & -a \leq z \leq a \\
  e^{-q_\parallel(z-a)}, & z \geq a 
\end{cases}
$$ (2.25)
Figure 2.5: Dielectric continuum model for quantum well optical phonons.

- Confined LO phonon modes \((p = (-1)^{m-1})\)

\[
P_C(q_{||}, z) \propto \begin{cases} 
0, & z \leq -a \\
\sin\left(\frac{m \pi z}{2a}\right), & -a \leq z \leq a, m = 1, 2, ... \\
0, & z \geq a
\end{cases} \tag{2.26}
\]

- Barrier LO phonon mode

\[
P_H(q_{||}, z) \propto \begin{cases} 
\sin(q_z(z + a)), & z \leq -a \\
0, & -a \leq z \leq a \\
\sin(q_z(z - a)), & z \geq a
\end{cases} \tag{2.27}
\]

The square of the free carrier-LO phonon scattering matrix element \(M^{\nu\nu}_{ij}\) for a transition between two subbands \(i\) and \(j\) and for a particular LO phonon mode can be written as a product of a coupling function \(\alpha_\nu\) that depends on the strength of the interaction and a form factor \(f^{\nu\nu}_{ijij}\) that quantifies the overlap of initial and final states for the scattering process [35]:

\[
|M^{\nu\nu}_{ij}(k_{||}, q_{||})|^2 = \alpha_\nu \cdot f^{\nu\nu}_{ijij}(k_{||}, q_{||}) \tag{2.28}
\]

The form factor is given by

\[
f^{\nu\nu}_{ijij}(k_{||}, q_{||}) = \int dz dz' \psi_i(k_{||}, z) \psi_j(k_{||} + q_{||}, z) \eta^{\nu}_{\text{coupled}} \psi_i(k_{||} - q_{||}, z') \psi_j(k_{||}, z') \tag{2.29}
\]
This equation is written such that the $k_{||}$ dependence of the wavefunction for non-parabolic bands can be directly included. Expression 2.29 contains selection rules for free carrier-LO phonon scattering. If the parity of subbands $i$ and $j$ is different, the interaction function $\eta^p_{\text{coup}}$ has to have parity -1 to yield a non-vanishing form factor. In this case scattering only takes place by the antisymmetric interface modes and the odd parity confined LO modes. If the parity of the subbands $i$ and $j$ is the same, $\eta^p_{\text{coup}}$ has to have parity +1. Scattering can then only proceed via symmetric interface modes and even parity confined LO modes. The explicit form of the coupling functions $\alpha_p$ and $\eta^p_{\text{coup}}$ for the dielectric continuum modes can be found in Ref. [35].

The assumption made in interpreting the experimental results of this thesis is that the LO phonons probed by the TRROD experiment are generated by $c_2$: $c_1$ interconduction band transitions. This assumption is based on the kinematic constraints for intersubband transitions in the sample of interest. Conduction subbands have parity $(-1)^n$, where $n = 0, 1, 2, ...$ is the subband number. Intersubband transitions between neighbouring subbands, i.e. with $\Delta n = 1$, thus always involve a change of parity. If these transitions are LO phonon mediated, it follows that the LO phonon must have odd parity.

To estimate the contribution of different LO phonon modes for the experimental conditions dealt with in this thesis, the form factors for LO phonon emission via 2-1 interconduction band transitions have been calculated for an infinite quantum well model and are shown in Fig. 2.6. Due to the selection rules, only the scattering rates for antisymmetric interface modes and the odd confined modes are non-zero. Mori and Ando have shown that in the limit of $q_{||} \cdot a \rightarrow 0$ the coupling factors $\alpha_p$ for the confined LO phonon modes and the anti-symmetric interface modes equal the bulk Fröhlich interaction. The contributions of the modes to the scattering process are thus contained in the form factors. The result for $q_{||}a = 0.1$ (200 $\text{Å}$ well width and $q_{||} = 5 \cdot 10^4 \text{ cm}^{-1}$) clearly shows that the dominant contribution (about 70 percent) is due to the antisymmetric interface modes. For comparison the result for the bulk LO phonon form factor is also displayed in Fig. 2.6. The bulk LO phonon approximation assumes quantum confinement for the carriers but
Figure 2.6: Form factors for electron-LO phonon scattering in parabolic band approximation. Solid curve: antisymmetric interface modes, dot-dashed curve: odd parity confined modes, dashed curve: bulk modes.
not for the LO phonons and leads to the following result for the Fröhlich matrix element [36, 37]:

\[ |M_{ij}|^2 = 2\pi e^2\hbar\omega_{LO}(\frac{1}{\varepsilon_{\infty}} - \frac{1}{\varepsilon_0}) \frac{1}{2a_{\parallel}} \]

\[ \int \int dz_1dz_2\psi_i^\ast(z_1, k_\parallel + q_\parallel)\psi_j(z_2, k_\parallel - q_\parallel)e^{-q_\parallel|z_2-z_1|} \]

In the case of the valence bands, the four-component spinors have to be inserted for the \( \psi \). It is interesting to note that the form factor for the free carrier-LO phonon interaction in this case is the same as for the Coulomb matrix elements described in Section 2.6.

As shown in Ref. [35], the sum of the form factors for the dielectric continuum modes exactly equals the bulk form factor due to the complete orthonormality of the LO phonon modes. Again, assuming equal \( \alpha_\psi \), this means that the net free carrier-LO phonon scattering rate of all quantum well LO phonon modes is equal to the bulk phonon rate.

For modelling the results of the TRROD experiment, reabsorption of odd parity LO phonon modes has to be calculated. This means that the only reabsorption processes that can take place are between valence subband states with different parity. The approach taken by the author is to use a bulk LO phonon model for the calculation, but to take into account the parity constraints on the valence band levels in the calculation of the Fröhlich matrix element. This approach is chosen because of the uncertainties involved with selecting a particular quantum well LO phonon model and because of the fact that the spectral resolution in the TRROD experiment is not sufficient to resolve the contributions of confined and interface LO phonon modes.

### 2.6 LO Phonon Self Energy for Nonparabolic Bands

In the following the author gives an ad hoc derivation of the LO phonon self energy for nonparabolic bands including dynamic screening effects. A more rigorous derivation of the same result has recently been carried out by Hawrylak and Young [38].

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The starting point for the derivation is Eq. 2.6 for the LO phonon self energy function for a quantum well with two parabolic conduction bands. This equation can be extended to nonparabolic bands by noting the following points:

1. Each of the indices 0 and 1 now stand for two quantum numbers, the subband index and the parity. For parabolic conduction bands it is sufficient to have the subband index only, since it also determines the parity of the state. In general, the self energy is now a matrix with subband and parity indices, even for the two subband model. This matrix reduces to a scalar, if the parity-conserving and parity-altering contributions are written down separately. This can be done since parity-altering and conserving transitions do not couple.

2. The wavefunctions $\psi_i$ in the Coulomb matrix elements Eq. 2.9 now depend on the magnitude of $k_\parallel$.

3. The actual valence band dispersion has to be included in the expression for the 2D polarizabilities $P_{ij}$.

Taking considerations 1-3 into account, the LO phonon self energy can be written as

$$\Sigma_{LO}(q_\parallel, \omega) = \int \frac{d^2k_\parallel}{(2\pi)^2} |M_{0\parallel}^{\omega}(k_\parallel, q_\parallel)|^2 \left[ \frac{P_{01}(k_\parallel, q_\parallel, \omega) + P_{10}(k_\parallel, q_\parallel, \omega)}{\epsilon_{0101}^\parallel(q_\parallel, \omega)} \right]$$

(2.31)

with $P_{01}$ denoting the integrand of Eq. 2.7. Due to the nonparabolicity of the valence bands the equation takes into account that the hole-LO phonon matrix element and the 2D polarizability functions depend on $k_\parallel$.

The nonparabolicity of the bands also affects the screening through a modified dielectric function. In calculating $\epsilon_{0101}^\parallel(q_\parallel, \omega)$ all intervalence band transitions that contribute to the screening are summed up by carrying out the integration over $k_\parallel$. Now the Coulomb matrix elements also have to be integrated over, since the wavefunctions are $k_\parallel$-dependent. Furthermore, it can be shown [38] that the dielectric function also separates into parity-altering and parity-conserving contributions. This has the consequence that intersubband transitions that are driven by odd
(even) parity LO phonons are only screened by the odd (even) parity component of the dielectric function. The dielectric function can thus be written as:

\[
e^0_{0101}(\vec{q}_\|, \omega) = \left\{ 1 - \frac{2\pi e^2}{\varepsilon_\infty q_\|} \int \frac{d\vec{k}_\|}{(2\pi)^2} f^p_{0101}(q_\|, k_\|)[P_{01}(q_\|, \vec{k}_\|, \omega) + P^0_{10}(q_\|, \vec{k}_\|, \omega)] \right\} (2.32)
\]

\[
e^2_{0101}(\vec{q}_\|, \omega) = \left\{ 1 - \frac{2\pi e^2}{\varepsilon_\infty q_\|} \int \frac{d\vec{k}_\|}{(2\pi)^2} f^p_{0101}(q_\|, k_\|)[P_{01}(q_\|, \vec{k}_\|, \omega) + P^0_{10}(q_\|, \vec{k}_\|, \omega)] \right\} (2.32)
\]

The following equation gives the form factor for the Coulomb matrix element and hole-LO phonon matrix elements in terms of the hole spinors. The products of the spinors are calculated according to the rules for scalar products between vectors.

\[
f^{p}_{ijij}(k_\|, q_\|) = \int \int d\xi \, d\xi' \begin{pmatrix} F_{\frac{1}{2}}(k_\| + q_\|, z) \\
F_{\frac{1}{2}}(k_\| + q_\|, z) \\
F_{-\frac{1}{2}}(k_\| + q_\|, z) \\
F_{-\frac{1}{2}}(k_\| + q_\|, z) \end{pmatrix}_{i,j} \cdot \begin{pmatrix} F_{\frac{1}{2}}(k_\|, z) \\
F_{\frac{1}{2}}(k_\|, z) \\
F_{-\frac{1}{2}}(k_\|, z) \\
F_{-\frac{1}{2}}(k_\|, z) \end{pmatrix}_{i,j'} e^{-q_\|z-z'}.
\]

(2.33)

Fig. 2.7 shows the form factors for parity-altering and parity-conserving hh0-hh3 transitions. The LO phonon wavevector \(q_\|\) was set to zero in the wavefunctions to simplify the calculations. For comparison the form factor calculated from an analytical expression for a quantum well with infinite barrier potential and the finite barrier height parabolic band approximations are also shown. At \(k_\| = 0\), the hh0 and hh3 wavefunctions are identical to \(n = 0\) and \(n = 3\) parabolic conduction band wavefunctions (see Fig. 2.4). Thus a transition between these two levels can be driven only by an odd parity LO phonon. This is the reason for the parity-conserving form factor being identical to zero at \(k_\| = 0\). At \(k_\| = 0\) the parity-altering form factor is very close to the infinite well value, as expected. The small difference in magnitude is due to finite barrier height effects. With increasing \(k_\|\), both form factors show strong variations. The parity conserving form factor increases due to admixture of envelope function components with opposite parity to the hole wavefunctions whereas the parity-altering form factor decreases for the same reason. Very strong changes occur for both form factors.
Figure 2.7: Form factors $f_{ij|ij}(k_h, q_h)$ for hh$_0$-hh$_3$ Coulomb matrix element in 200 Å well. Dotted curve: finite quantum well, parity-altering transition; dashed curve: finite quantum well, parity-conserving transition; dot-dashed curve: finite quantum well, parabolic band approximation; solid curve: infinite quantum well, parabolic band approximation.
near $k_n = 1.3 \cdot 10^6 \text{ cm}^{-1}$. This is a direct reflection of the band structure, since both levels have anti-crossings and therefore strong mixing with other states near this wavevector, as is evident from the valence band structure diagram in Fig. 2.3. This strong dependence of the form factors on $k_{\parallel}$ compared to the parabolic band model clearly indicates the need for incorporating the full valence band structure into the calculations of the LO phonon self energy.

Since the form factors for the hole-LO phonon matrix element and the Coulomb matrix element are the same, the integrals in Eq. 2.31 and Eq. 2.32 are both of the form

$$I = \int \frac{d\vec{k}_n}{(2\pi)^2} f_{101}^{p}(q_{\parallel}, k_{\parallel}) [P_{01}(\vec{q}_{\parallel}, \vec{k}_{\parallel}, \omega) + P_{10}(\vec{q}_{\parallel}, \vec{k}_{\parallel}, \omega)]$$

The following strategy is used to evaluate this integral: first, the imaginary part of the integral is calculated and then, by using Kramers-Kronig relations, the real part can be obtained. This technique was successfully used by Young and Kelly [24] to model hot carrier relaxation in bulk GaAs. In the following, a zero temperature approximation is used, which simplifies the calculations considerably, since it reduces the Fermi-Dirac functions to step functions. This approach is justified at the hole densities of interest and since the experimental results that are compared with the calculations were obtained at $T \approx 5\text{K}$.

The imaginary part of the integral in Eq. 2.34 at $T = 0$ can be written as

$$Im(I) = \frac{\pi}{(2\pi)^2} \int_{0}^{2\pi} \int_{0}^{k_{F}} d\theta dk_{\parallel} \cdot k_{\parallel} \cdot f_{ijij}^{p}(k_{\parallel}, q_{\parallel}) \delta[E_i(\vec{k}_{\parallel} + \vec{q}_{\parallel}) - E_i(\vec{k}_{\parallel}) - \hbar\omega]$$

where $\theta$ denotes the angle between $\vec{k}_{\parallel}$ and $\vec{q}_{\parallel}$ and $k_{F}$ is the magnitude of the Fermi wavevector. For the valence band dispersion $E_i$, a polynomial fit to the $\vec{k} \cdot \vec{p}$ results is used and the integral is evaluated numerically. Details of this procedure can be found in Appendix B. The results obtained have been checked to agree with analytical results in the parabolic band limit.

Contributions of the conduction band to the screening can also be incorporated into the calculations. Since there are no electron single particle excitations around the LO phonon energy for the quantum well of interest, only the real part of the electron dielectric function due to interconduction subband transitions needs to be
considered. This contribution is calculated from the analytical expressions given in the literature and incorporated using the diagonal approximation [22].

2.7 Results

Fig. 2.8 shows the dependence of the free hole-LO phonon scattering rate on the free hole density \( n \) (expressed in terms of the Fermi wavevector \( k_F = \sqrt{2\pi n} \)) for three different approximations to Eq. 2.5:

1. Fermi's Golden Rule result, i.e. no screening.
2. Screening by \( hh_0-hh_3 \) transitions of free holes.
3. Screening by \( hh_0-hh_3 \) transitions of free holes and \( c_0-c_1 \) transitions of free electrons.

The common feature of all three cases is the onset of scattering at \( k_F = 7 \cdot 10^5 \text{ cm}^{-1} \). At this density, \( hh_0-hh_3 \) scattering for holes at the Fermi edge in the \( hh_0 \) subband becomes kinematically allowed. There also is an obvious second step in the scattering rate at \( \sim 1.3 \cdot 10^6 \text{ cm}^{-1} \). This step is due to the double minimum structure in the \( hh_3 \) band and would not occur within a parabolic band approximation. This structure leads to two scattering thresholds, one on the low wavevector side of the minimum and one on the high wavevector side.

The effects of screening are small at low density. The effects of hole screening alone are relatively small even at larger densities, but the electrons have a strong anti-screening influence due to their relatively small effective mass and the fact that the intersubband electron plasmon resonance is below the LO phonon frequency. This anti-screening effectively increases the Fröhlich coupling strength between the renormalized LO phonon and the single particle excitations.
Figure 2.8: Results for LO phonon scattering rates ($q// = 5 \times 10^4 \text{ cm}^{-1}$; triangles: no screening; circles: screening by electrons and holes; squares: screening by holes only.)

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Chapter 3

Experimental Technique and Apparatus

This chapter describes the details of the time-resolved Raman scattering/optical doping technique that the author developed to investigate free carrier-LO phonon interactions in GaAs quantum wells. First, the general requirements for the experiment are discussed. Then the apparatus used to obtain the results presented in this thesis is described in detail. In order to make meaningful comparisons between the results of the TRROD experiments and the theoretical results derived in Chapter 2, the sample parameters are very important. The general characteristics of the sample used are described at the end of this chapter. A more specific discussion of these parameters with respect to the interpretation of the results can be found in Chapter 4.

3.1 Introduction

As described in Chapter 1, the TRROD technique is a powerful tool for investigating free carrier-LO phonon interactions. In this technique, a pre-pulse (from laser 1) optically dopes a semiconductor sample with a cold electron-hole plasma of variable density. After a fixed delay time, hot carriers are created in the sample by a second laser pulse ("pump pulse", from laser 2). Subsequently Raman spectra are taken with a third laser pulse ("probe pulse", also from laser 2) at different delay times between the pump and the probe pulse. In this way the effect of the
cold plasma on the dynamics of the elementary excitations in the sample can be measured in a controlled manner. A variation of this technique is *time-integrated* Raman scattering with optical doping. Hereby only the probe pulse impinges on the sample and simultaneously excites and probes excitations in the sample. This is equivalent to performing the TRROD experiment with no time delay between pump and probe pulse.

The information extracted from the TRROD experiment is contained in the temporal evolution of the Raman spectra. The main parameters important for the apparatus used are therefore time resolution, spectral resolution and the range of injected cold plasma densities. The anharmonic decay of LO phonons into acoustic phonons in GaAs quantum wells takes place on a timescale of 5-8 ps [39, 10]. Thus, the time resolution of the experiment should ideally be much shorter than that. In a time-resolved Raman scattering experiment the time resolution is determined by the temporal width of the laser pulses used for the hot carrier injection and Raman probe. But since the spectral bandwidth of a transform limited laser pulse is inversely proportional to the duration of the pulse, there is a lower limit on the pulse duration that can be used in TRROD experiments. There are two reasons for this limitation: first, the rejection of the fundamental laser line by the spectrometer is not sufficient when using spectrally broad pulses. Second, the characteristic spectral features (from plasmons, LO phonons) have energies in the range of 10-100 meV and it is desirable to be able to resolve cold plasma induced energy shifts of these lines that amount to a few meV.

As a reasonable compromise between spectral and temporal resolution, a laser pulse duration of 1 ps was chosen for laser 2, corresponding to a FWHM bandwidth of 1.3 meV (assuming a sech² pulse). This pulse duration is achievable with mode-locked dye lasers with a saturable absorber jet. These lasers also have the advantage of a large wavelength tuning range that allows for flexibility in choosing an electronic resonance for resonant enhancement of the Raman signal [40].

The pulse duration of laser 1 does not influence the time resolution of the experiment. The only consideration is that its pulse duration should be shorter than the pre-pump delay. It is desirable for this laser to have enough power to
inject a wide range of carrier densities into the sample, since the cold plasma density is the main variable in the TRROD experiment.

3.2 Apparatus

This section deals with the details of the apparatus for the TRROD experiment. The experiment was set up by the author in the Quantum Physics Lab at the Institute for Microstructural Sciences of NRC (National Research Council of Canada). Parts of the apparatus had previously been used for time-resolved Raman scattering experiments. However, due to a move to a new lab location and the acquisition of a new Nd:YAG laser, the author had to completely set up, align and characterize the experiment. In addition, the author modified and expanded the experiment in order to perform the TRROD experiments.

Fig. 3.1 shows a detailed diagram of the experimental setup for the TRROD experiments. Each of the components will now be described in turn.

The pump laser for laser 1 and laser 2 is an actively modelocked Nd:YAG laser (Coherent Inc., model Antares). This laser emits 100 ps pulses at 1064 nm with a repetition rate of 76 Mhz and an average power of 30 W. The fundamental wavelength is frequency doubled in a LBO (Lithium Triborate) crystal, resulting in a train of 75 ps pulses with an average power of up to 5 W at 532 nm. This beam is then split and each of the resulting beams synchronously pumps one dye laser (Coherent Inc., model 702). Previous to the work of the author only one dye laser was pumped by the Nd:YAG laser in the Quantum Physics Lab. The simultaneous operation of two pulsed dye lasers significantly enhances the capabilities for pump-probe experiments. Laser 1 uses Styri 9 dye (tuning range 780-840 nm) in order to be tunable around the fundamental gap of GaAs (820 nm) for optical doping. Since short pulse duration is not critical for this laser, it is operated without a pulse shortening saturable absorber jet. It has a pulse duration of 5 ps FWHM as determined from autocorrelation measurements and assuming a sech² pulse shape. The average power of laser 1 is as high as 250 mW (~3.3 nJ energy per pulse). Laser 2 uses Rhodamin 640 dye (tuning range
Figure 3.1: Experimental setup for TRROD experiments.
590-640 nm) and a pulse shortening saturable absorber jet with DQTCI as the active substance. With this combination, 1 ps FWHM pulses (determined by autocorrelation measurements) at a wavelength around 635 nm can be achieved. This laser, which has an average power as high as 100 mW (~1.3 nJ/pulse), is used for hot carrier injection and Raman probing. An autocorrelator was set up permanently during the experiments so that the pulse durations could be checked at any time. The output powers of laser 1 and 2 were also monitored continuously by using beamsplitters and photodiodes to detect fluctuations of the output power during the experiments.

The output of laser 2 is sent through a pair of Pellin-Broca prisms to spectrally filter the beam. It was checked that these prisms have no measurable influence on the pulse duration. Each laser beam is steered through a combination of a \( \lambda/2 \)-plate mounted on a rotation stage and a polarizing cube to control the power (not shown in Fig. 3.1). The beam from laser 2 is then sent through an optical delay line to generate pump and probe pulses with variable delay. The first two elements of this delay line are a \( \lambda/2 \)-plate and a cube polarizer \( P_1 \) that splits the incoming beam into pump and probe beams with orthogonal polarizations. By changing the orientation of the \( \lambda/2 \)-plate, which is mounted on a rotation stage, the relative power between the two beams can be continuously adjusted. One of the two beams then passes through a combination of mirrors and a retroreflector mounted on a translation stage that enables a change in the optical path length and hence time delay. The position of the translation stage can be manually adjusted by means of a micrometer control. The other beam traverses a fixed optical path. Both beams are then recombined in polarizing cube \( P_2 \) and aligned to be collinear.

The most important point in calibrating the delay line is to align both optical paths to equal length \( l_0 \). This corresponds to zero time delay (\( \Delta t = 0 \)) between both pulses. Once this is established, the amount of delay is simply given by the path difference between the two beams as determined from the micrometer setting \( l \) of the translation stage divided by the speed of light \( c \)

\[
\Delta t = \frac{2(l - l_0)}{c}. \tag{3.1}
\]

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There are three steps in establishing $\Delta t = 0$: first, the optical path lengths are adjusted to be as equal as possible using a ruler. Then a fast photodiode is set up to adjust the optical paths to within 50 ps of $\Delta t = 0$. The final stage is to look for scattered light interference fringes from the reflection of the two overlapping beams. These interference fringes are only visible when both pulses are spatially and temporally overlapped. This last stage of alignment could in principle also be done by overlapping both pulses in a non-linear crystal and maximizing the second-harmonic signal generated.

After exiting the polarizer $P_2$, the collinear beams are focussed onto the sample with lens $L_1$ (focal length 100 mm). The sample is held in a Janis Supertran liquid Helium flow-through cryostat. The sample is mounted on a copper cold finger using heat sink compound. The temperature of the sample ($T = 5$ K) is monitored and controlled by a Lakeshore Cryogenics temperature controller.

The common spot of both beams on the sample surface was measured to have a diameter of $(100\pm10)\ \mu m$. The spot size measurement was carried out by measuring a magnified image of the spot and using the image of a pinhole with known diameter for calibration. The beam from laser 1 is brought onto the sample along a separate path of variable length so that the time delay between laser 1 and laser 2 can be adjusted. This delay is set to 50 ps between the pre-pump pulse of laser 1 and the hot carrier injection pulse of laser 2. The pulse jitter of the dye lasers is $\sim 3$ ps, which is much less than the delay time between pre-pump and hot carrier injection. The jitter of laser 1 does not affect the experiment since pump and probe pulses are derived from the same laser pulse. The spot of laser 1 has a diameter of $(350\pm10)\ \mu m$ on the sample in order to ensure homogeneity of the cold electron-hole plasma over the spot size of laser 2. The spatial overlap of the spots from the two lasers is monitored by imaging them into a video camera and observing the positions on a video monitor.

The scattered light from the sample is collected with lens $L_2$ ($f$-number = 2.6). The angle $\phi_1$ between the normal to the sample surface and the incident light beam and the angle $\phi_2$ between the normal and the scattered light beam in combination with the wavelength $\lambda_L$ of laser 2 determine the wavevector transfer.
$q_\parallel$ of excitations in the sample [41] (see Fig. 3.2):

$$q_\parallel = \frac{2\pi}{\lambda_L} (\sin \phi_1 - \sin \phi_2) \quad (3.2)$$

This equation only strictly holds for non-resonant Raman scattering. The implications of the relaxation of the $q_\parallel$ selection rule under resonant conditions are discussed in detail in Chapter 4.

For the experimental setup described here, $q_\parallel$ as determined from Eq. 3.2 is equal to $(5\pm2)\cdot10^4 \text{ cm}^{-1}$. The spread in $q_\parallel$ is calculated from the the finite diameter of the incoming laser beam and the acceptance angle $\Delta \phi$ of the collection optics.

The collected light is steered through an analyzing polarizer to reject the scattered light from the pump beam and to select collective or single particle excitations. Both single particle and collective excitations are observable in Raman scattering from semiconductor quantum well samples. It has been shown [42] that a judicious choice of the relative polarizations of incoming and scattered laser beams can be used to distinguish these two types of excitations in the spectra.

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The single particle excitations show up in the depolarized $z(x'y')\hat{z}$ spectra whereas the collective excitations appear in the polarized $z(x'x')\hat{z}$ spectra. $z$ and $\hat{z}$ are the propagating directions of the incident and scattered light, which are close to the (001) and (00\overline{1}) crystal axes of the sample. $x'$ and $y'$ are (110) and (1\overline{1}0) in-plane directions of the polarization vectors of the incident and scattered light.

A Spex Triplespectrometer is used to spectrally disperse the scattered light which is detected by a liquid Nitrogen cooled two-dimensional CCD array with 384 x 576 pixels (Thomson CSF model TH 7882CDA) that is interfaced to a computer. Multichannel detection greatly reduces the time required to acquire the Raman spectra. A range of wavelengths of $\pm 8$ nm around the center wavelength can be detected simultaneously with the spectrometer operating in second order. The combined spectral resolution of the spectrometer and CCD array in this configuration is 0.14 nm FWHM with the entrance slit set to 100 $\mu$m. The initial alignment of the detector and the spectrometer with respect to maximum throughput is achieved by lining up a HeNe laser beam from the detector side through the spectrometer so that the HeNe spot impinges on the sample at the same position as lasers 1 and 2. Once this is achieved the performance can be optimized by maximizing the photoluminescence and Raman signal from the sample.

3.3 Sample Parameters and Characterization

The sample used in the TRROD experiments is a piece (dimension $\sim 7\times7$ mm$^2$) of a GaAs/Al$_{30}$Ga$_{70}$As multiple quantum well wafer that was grown at the Institute of Microstructural Sciences of the National Research Council of Canada. The sample consists of 50 wells of 200 Å width and separating barriers of 130 Å width. Details of the nominal layer structure as given by the growth parameters are given in Table 3.1.

Fig. 3.3 shows the band structure for one of the quantum wells in the growth direction at $T = 5$ K. The height of the barriers was deduced from photoluminescence (PL) measurements. These PL measurements were performed by using the cryostat and spectrometer described in the previous section and laser 2 as an exci-
Table 3.1: Nominal layer structure of multiple quantum well sample from bottom to top, as grown on semi-insulating GaAs substrate.

<table>
<thead>
<tr>
<th>layer #</th>
<th>composition</th>
<th>thickness (Å)</th>
<th>repeat</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>GaAs substrate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>GaAs</td>
<td>700</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>AlAs</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>3</td>
<td>GaAs</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>30</td>
<td>AlAs</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>GaAs</td>
<td>1300</td>
<td></td>
</tr>
<tr>
<td>32</td>
<td>Al₃₀Ga₇₀As</td>
<td>130</td>
<td>50</td>
</tr>
<tr>
<td>33</td>
<td>GaAs</td>
<td>200</td>
<td>50</td>
</tr>
<tr>
<td>102</td>
<td>Al₃₀Ga₇₀As</td>
<td>130</td>
<td></td>
</tr>
<tr>
<td>103</td>
<td>GaAs</td>
<td>170</td>
<td></td>
</tr>
</tbody>
</table>

Figure 3.3: Band structure in growth direction.
In order to characterize the sample, band edge PL measurements were performed by the author. Fig. 3.4 contains a spectrum of the quantum well band edge PL as excited with a cw Ti:Sapphire laser tuned to 1545 meV (802.5 nm). This spectrum shows a strong peak at 1535 meV originating from the recombination of the $c_0$-$hh_0$ quantum well exciton. The high energy shoulder on this line is due to luminescence from the 170 Å GaAs cap layer. The low energy line at 1527 meV is most likely due to an acceptor bound exciton and the line at 1545 meV is due to scattered laser light.

Fig. 3.5 shows a resonance profile for the Stokes intensity of the Raman signal.
Figure 3.5: LO phonon Stokes resonance profile. Solid dots are data points; the dashed curve is a guide to the eye.

from the LO phonon as measured from polarized Raman spectra. These results were obtained by taking time-integrated Stokes spectra as a function of photon energy with no cold plasma present. The photon energy of laser 2 was hereby tuned around the split-off (so) band resonance. The identification of the individual peaks is complicated by the fact that there are a number of possible electronic transitions in this energy range. First, there are transitions between split-off hole subbands and conduction subbands. Second, there is the possibility of hh /lh to conduction band transitions in the AlGaAs barrier material. The resonance at ~ 1975 meV is due to this AlGaAs transition, since it is associated with the emergence of a luminescence line. This assignment is supported by the fact that the AlAs-like LO phonon signal from the AlGaAs barrier material shows strong resonant
enhancement at this energy. The line at ~ 1920 meV is most likely due to a s0 to c0 transition, since the c0-c1 intersubband plasmon is also resonantly enhanced for this photon energy. This resonance was chosen for most of the TRROD experiments described in this thesis, since at this energy free carriers are only excited in the well and not in the barrier material. Furthermore, the LO phonon and c0-c1 intersubband plasmon lines are enhanced and a study of the coupling between the two excitations is made easier.

For the interpretation of the TRROD results, the energy spacings between the lowest and the first excited conduction subband (c0-c1) and between the first and second excited level (c1-c2) are important parameters. These spacings were deduced from Raman pump-probe experiments using the set-up in Fig. 3.1 with a z(x'y')z geometry for the probe pulse and a z(x'x')z geometry for the pump. The spectra obtained in this manner have the pump-only background subtracted from them. A typical result is shown in Fig. 3.6. The conduction subband splittings are found to be 27±0.5 meV for the c0-c1 spacing and 41±0.5 meV for the c1-c2 spacing. These values are in agreement with a calculation of the conduction subband structure using a one-dimensional, finite barrier height, “particle in a box” model [26] and agree within errors with the 200 Å well width and the 34 % Al content of the barriers.
Figure 3.6: Solid curve: Stokes spectra of $c_0$-$c_1$ (at 27 meV) and $c_1$-$c_2$ (at 41 meV) single particle intersubband transitions. Dashed curve: fit to the single particle lines using two Lorentzians. The structure around 36 meV is a remnant of the LO phonon line in the pump-only spectrum after subtraction. The line at 48 meV is due to the AlAs like LO phonon in the AlGaAs barrier material.
Chapter 4
Experimental Results and Discussion

In this chapter the experimental results of the TRROD experiments are presented. The first part of the chapter discusses in detail the effects of the cold electron-hole plasma on the Raman spectra observed in the experiment. The second part describes the results of the time-resolved measurements of the LO phonon dynamics in the presence of the cold plasma. These results are compared with the calculations described in Chapter 2. The last section of this Chapter presents results on the dependence of the hot LO phonon occupation as determined from time-integrated Raman scattering experiments. These results represent corroborative evidence for the observed reduction of the LO phonon lifetime in the presence of a cold electron-hole plasma.

4.1 Effects of the Cold Electron-Hole Plasma on Excitation Spectra

Before dealing with the TRROD results that address the dynamics of the LO phonons in the presence of a cold electron-hole plasma it is helpful to discuss the effects of this plasma on the excitation spectra of the quantum well used in the TRROD experiments.

Fig. 4.1 shows a series of time-integrated Stokes and anti-Stokes spectra taken at different cold electron-hole plasma densities $n_{PL}$. For these spectra, laser 2 was

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Figure 4.1: Time-integrated Stokes and anti-Stokes Raman spectra \( z(x',x')z \) with a) no cold electron-hole plasma, b) an intermediate cold plasma density (\( \sim 1.5 \times 10^{11} \text{ cm}^{-2} \)) and c) the maximum density (\( \sim 3 \times 10^{11} \text{ cm}^{-2} \)) attained. The solid curves are fits to the individual lines. The dashed curves in c) indicate the individual contributions of the unshifted (iii) and shifted (ii) LO phonon-like modes. The carrier densities are estimated from laser and sample parameters and the calculations in this section.
operated without the saturable absorber jet at a pulse duration of 4 ps FWHM, in order to enhance spectral resolution. The cold plasma was injected by using a cw Ti:Sapphire laser (laser 1). With no cold plasma present (a), both Stokes and anti-Stokes spectra show a strong signal at the bare GaAs LO phonon energy near 36 meV. There is also a weak signal at 27 meV from the c₀-c₁ intersubband plasmon. The assignment of this feature is made on the basis of parabolic band energy level calculations for the conduction subband structure. Furthermore, the signal from the conduction subband excitations is enhanced since the Raman probe laser is tuned to a conduction-split off valence band resonance.

When the cold plasma is added (b,c), the spectra change dramatically. There are three features evident in the spectra and labeled in Fig. 4.1: i) The growth of the c₀- c₁ intersubband-like mode, which shifts from 27 meV to higher energies with increasing plasma density; ii) the phonon-like coupled plasmon-LO phonon mode (CPPM), also shifted to higher energies from the bare LO phonon energy; and iii) a peak at the bare LO phonon energy that is not influenced by the cold plasma, and which does not appear in the anti-Stokes spectra. The growth and shift of the intersubband plasmon signal (feature (i)) demonstrates that the density of cold carriers far exceeds that of the carriers injected by the visible pump pulses, hence all many-body effects can be attributed only to the cold carriers. Feature (ii), which is blueshifted (by 1.4 meV at nₚL = 3·10¹¹ cm⁻²) and broadened by the cold plasma, is due to allowed Raman scattering from CPPM with q‖ = 5·10⁴ cm⁻¹. The presence of such small wavevector nonequilibrium CPPM in the anti-Stokes spectra (which are directly sensitive to the population of the modes) implies that these hot excitations are generated via intersubband carrier relaxation processes. The c₂-c₁ interconduction subband transitions capable of generating the observed hot CPPM do occur, because the measured c₂-c₁ intersubband separation (41 meV, and significantly lifetime broadened) is nearly resonant with the LO phonon energy, and the 1.92 eV pump photons directly inject electrons into the c₂ subband. Intervalance subband transitions from hh₃ to hh₀ may also be contributing to the nonequilibrium CPPM population; however a direct measurement of these contributions is not possible. It should be noted that the q‖ selection rules are

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relaxed under resonance conditions. Sood et al. [44] have shown evidence of Raman scattering processes under resonance conditions in GaAs quantum wells that violate momentum (wavevector) conservation. These non-allowed processes can be explained by scattering of the LO phonons with impurities. Under resonant conditions one can then in general expect to observe contributions from a range of $q_\parallel$. This effect complicates the comparison of Raman scattering experiments with calculations, because of the uncertainty in the value of $q_\parallel$ for the excitations observed. It is important to emphasize that, as described above, the parameters of the TRID experiment presented in this thesis were chosen in such a way that allowed Raman scattering processes can occur. Even though it is not possible to estimate the additional contributions from non-allowed processes due to the lack of an appropriate theory, the observed strong interaction of feature ii) with the cold electron-plasma indicates that allowed Raman scattering processes with small wavevector ($q_\parallel = 5 \times 10^4$ cm$^{-1}$) are indeed the dominant contributions to features i) and ii).\footnote{It should also be noted that because the sample is a multiple quantum well, the experiment in fact probes a number of intersubband-like modes that share a $q_\parallel = 5 \times 10^4$ cm$^{-1}$. On the basis of calculations it is expected that the coupling between wells for this sample and the $q_\parallel$ probed is extremely weak and can be neglected [45].} However, the unshifted feature (iii) appearing at the bare LO phonon energy in the Stokes spectra could be due to non-allowed Raman scattering from LO phonon modes with large $q_\parallel$ that couple only weakly with either the hot carriers or the plasmons. A second possible explanation for this line is that it is due to allowed Raman scattering from the even parity confined LO phonon modes and symmetric interface LO phonon modes. These modes can not be generated by $c_2-c_1$ intersubband transitions and thus do not show up in the anti-Stokes spectra. Due to their even parity they are not able to interact with the $c_0-c_1$ plasmon and hence show no shift with increasing carrier density. The even parity modes are coupled with the $c_0-c_0$ intrasubband plasmon and the $c_0-c_2$ intersubband plasmon, but both of these contributions to the shift are expected to be weak due to the energy difference between these plasmons and the LO phonon energy. Furthermore, the $c_0-c_0$ plasmon energy is smaller than that of the LO phonon and the converse is true for the $c_0-c_2$ plasmon. The contributions of these
modes to the LO phonon shift are thus expected to compensate each other to some extent. A similar unshifted LO phonon feature has been observed in resonant Raman scattering from p-doped GaAs quantum wells by Dahl et al. [46] and was interpreted in terms of the interaction of carriers with different parity LO phonon modes [47]. The fact that allowed Raman processes are most likely enhanced near the allowed $q_n$ leads the author to favour the second explanation for the origin of feature iii). The experimental results described in this thesis would thus present evidence for a parity dependent mode shift in an optically doped quantum well.

In the following, the author discusses the energy shifts of features (i) and (ii). The shifts of these features provide interesting information on many-body interactions between the LO phonons and the free carriers and show the potential of TRROD experiments for investigating those effects that are not easily probed by any other technique. The LO phonon-like branch of the CPPM shifts to higher energies with increasing plasma density. This so called anti-screening [48] or descreening [49] effect is indicative of an increase in the Fröhlich coupling strength between the carriers and the LO phonons. As shown by Collet [50], anti-screening can have significant effects on the relaxation of electron-hole plasmas in bulk GaAs. In quantum wells the nature of the plasmon coupled to the LO phonons determines whether the anti-screening effect occurs. Intra- and intersubband plasmons associated with subband spacings that are less than the LO phonon energy anti-screen the LO phonons with appropriate parity, since the oscillations of the free carriers and the lattice are in phase. Intersubband plasmons associated with subband spacings larger than the LO phonon energy screen the LO phonons, i.e. cause a shift of the modes to lower energies. In the latter case, the lattice and the carriers oscillate $180^\circ$ out of phase. The results presented above give evidence of the anti-screening effect in an optically-doped quantum well, where the $c_0$-$c_1$ plasmon with energy less than the LO phonon energy dominates the shift of the LO-like mode. A quantitative explanation of the mode shifts could in principle be derived by calculating the carrier density dependent Raman spectra for the coupled LO phonon-free carrier system. As already discussed in Section 2.3, this is a complex task that is beyond the scope of this thesis. However, in order to get an indica-
Figure 4.2: Coupled plasmon-phonon modes and single particle excitations calculated for $q_\parallel=5\times10^4$ cm$^{-1}$.

...tion of the influence of the free electrons and holes on the shift of the CPPM the author estimated the mode shifts by solving for zeroes of the $T = 0$ 2D dielectric functions of the $c_0$-$c_1$-LO phonon and the $hh_0$-$hh_3$-LO phonon systems. A similar approach was used by Richards et al. [51] to model Raman spectra of n-doped GaAs quantum wells. Coupling effects between electrons and holes are neglected in these calculations. Fig. 4.2 shows the single particle excitation (SPE) and CPPM energies calculated from this model. The $c_0$-$c_1$ intersubband transition SPE and the $hh_0$-$hh_3$ SPE start at the $k_\parallel = 0$ intersubband spacings for $k_F = 0$. The hole SPE broadens to lower energies for increasing $k_F$. This is due to the fact that the energy difference between $hh_0$ and $hh_3$ initially decreases for increasing $k_\parallel$ (See
the band structure diagram in Fig. 2.3). Near $1.5 \times 10^6 \text{ cm}^{-1}$ the energy difference between the two bands starts to increase. This causes the small broadening of the hole SPE to higher energy at the largest $k_F$ plotted. In contrast to the hole SPE, the electron SPE broadens symmetrically with increasing $k_F$, which is due to the fact that the $c_0$ and $c_1$ bands are assumed to have the same in-plane dispersion. The electron SPE also is narrower than the hole SPE, because of the stronger curvature of the conduction bands. For the hole CPPM, the intersubband-like mode is Landau damped [52] by $hh_0$-$hh_3$ single particle excitations for $k_F < 1.1 \times 10^6 \text{ cm}^{-1}$. The LO phonon-like mode shifts to lower energies with increasing $k_F$, since the LO phonon energy is less than the $hh_0$-$hh_3$ splitting. The absolute shift of the hole CPPM is on the order of 1 meV, much less than that of the electron CPPM. This is due to the strong damping of the hole plasmon-like branch that reduces the coupling between the modes. The Landau damping would be even stronger if finite temperature effects and band warping were included in the calculations. In this case, even less coupling of the two modes is expected. A similar reduction in the coupling of a CPPM due to strong damping of the hole plasmon was observed by Kam and Young in p-doped bulk GaAs [13]. The two branches of the $c_0$-$c_1$ CPPM exhibit a very different density dependence compared to the holes. They show a strong shift to higher energies ($\sim 3 \text{ meV at } k_F = 1.5 \times 10^6 \text{ cm}^{-1}$). As mentioned above, the direction of this shift is due to the fact that the $c_0$-$c_1$ spacing is smaller than the LO phonon energy and hence anti-screening of the LO phonon occurs. Since the plasmon-like branch is not Landau damped, there is strong coupling between the modes that leads to the large energy shift. The calculations thus indicate that the energy shift of the LO phonon-like CPPM is dominated by the electrons whereas the holes only contribute very little. It is important to note that at the LO phonon-like branch of the $c_0$-$c_1$ CPPM starts to overlap the $hh_0$-$hh_3$ SPE at $k_F \approx 6 \times 10^5 \text{ cm}^{-1}$. This indicates that broadening and a reduction in lifetime of this mode is expected. Section 4.2 explicitly deals with this topic. In Fig. 4.3 the experimentally measured modeshifts for the $c_0$-$c_1$ CPPM are compared with the calculations. The experimental carrier densities were calibrated by simultaneously fitting the observed shifts of both modes to the calculated shift. The results for
Figure 4.3: Experimentally observed Stokes shift of LO phonon-like mode (squares) and $c_0$-$c_1$ plasmon-like mode (circles). The solid lines are results of calculations as described in the text.
the carrier densities are in reasonable agreement with an estimate based on laser fluence, spot size and absorption in the sample. The simultaneous fit of the theory to both modes is very restrictive, since the carrier densities for both branches were not independently scaled. The good agreement of data and calculations seems to indicate that the shift of the $c_0-c_1$ CPPM is indeed not significantly influenced by the $hh_0-hh_3$ transitions and is mainly due to contributions from the electrons.

4.2 Density Dependence of LO Phonon Dynamics

The LO phonon-like part of the $c_0-c_1$ CPPM not only shifts with increasing plasma density, but also broadens, which is evident especially in the anti-Stokes spectra (Fig. 4.4). This broadening is indicative of Landau damping of the LO phonon-like mode by $hh_0-hh_3$ single particle transitions and agrees qualitatively with the overlap of this mode with the $hh_0-hh_3$ SPE, starting at $k_F \approx 6 \times 10^5$ cm$^{-1}$, in Fig. 4.2. This damping is related to a reduced lifetime of the LO phonon-like branch of the CPPM. However, the $c_0-c_1$ plasmon-like branch of the CPPM exhibits an almost density-independent width in both Stokes and anti-Stokes spectra and shows a negligible change in intensity in the anti-Stokes spectra. This is indicative of the fact that the LO phonon-like modes play the major role in the carrier relaxation processes at the observed $q_\parallel$.

Unfortunately, the extraction of the LO phonon lifetime from the measured line width is complicated by convolution effects with the spectrometer resolution and the spectral width of the probe laser, especially when sources with pulse duration of the order of the LO phonon lifetime are being used. Even in cw Raman scattering experiments, the line width often only leads to a lower limit on the lifetime of a specific excitation due to inhomogeneous broadening effects [53]. It is thus preferable to perform a direct measurement of the LO phonon lifetime using a time-resolved Raman scattering technique.

The anti-Stokes intensity of the LO phonon-like feature is directly proportional to the LO phonon occupation $n_{q_\parallel}$. Anti-Stokes spectra taken as a function of time
Figure 4.1: Experimentally determined width of anti-Stokes LO phonon-like mode.

are thus a direct probe of LO phonon dynamics. This statement is strictly true only under non-resonant conditions. However, for the time-resolved measurements presented in this thesis, resonance effects are of secondary importance. Under the usual conditions for time-resolved Raman scattering experiments the anti-Stokes cross-section does not change over the typical lifetime of an LO phonon. A change in the cold plasma density could in principle change the resonant enhancement but the time scale for recombination of the cold carriers (about 1 ns) is much longer than the lifetime of the LO phonon (a few ps). Since the cold plasma density thus can be assumed to be constant over the LO phonon lifetime, the anti-Stokes intensity is directly proportional to the LO phonon occupation number at all time delays of interest and the LO phonon lifetime can be extracted even though the absolute value of the occupation number is only known within considerable systematic error. Fig. 4.5 shows the temporal behaviour of the anti-Stokes CPPM signal at different cold plasma densities. The data in this figure were obtained from fits to anti-Stokes spectra that were corrected for the background of probe-only
Figure 4.5: The time-resolved anti-Stokes intensity of the LO phonon-like branch of the CPPM at different cold plasma densities. The cold plasma densities are 0 (squares), $1.5 \times 10^{11}$ cm$^{-2}$ (circles) and $3 \times 10^{11}$ cm$^{-2}$. The corresponding decay times are $4.5 \pm 0.5$, $3.0 \pm 0.5$ and $1.9 \pm 0.5$ ps respectively. For reasons of clarity, the rising edge of the signal is shown for only one cold plasma density. The line centered around $t = 0$ indicates the autocorrelation trace of the probe laser pulse.

generated excitations. For these measurements the average powers for pump and probe pulses (laser 2) were 20 mW and 5 mW respectively, corresponding to an injected hot plasma density of $\sim 1 \times 10^{10}$ cm$^{-2}$. At each cold plasma density the anti-Stokes spectrum measured by the Raman probe pulse has an essentially time invariant shape, but its intensity (plotted in Fig. 4.5) changes dramatically with time delay. In the absence of a cold plasma, the lifetime of the hot LO phonons was found to be $4.5 \pm 0.5$ ps, in good agreement with results obtained by Ryan and Tatham [9] on a GaAs/Al$_{35}$Ga$_{65}$As sample with 147 Å wide wells. The lifetime...
decreases to 3.0 ± 0.5 ps in the presence of a cold plasma of density 1.5·10^{11} \text{ cm}^{-2}
and even further to 1.9 ± 0.5 ps for a density of 3·10^{11} \text{ cm}^{-2}. Since the density
of the hot plasma is fixed for all time delays associated with a particular cold
plasma density, there are no processes which could make the Raman-scattering
mechanism itself change with time delay. These data therefore provide direct,
unambiguous evidence that the population lifetime of the nonequilibrium CPPM
decreases substantially in the presence of a cold electron-hole plasma and provide
proof from the lattice perspective for efficient reabsorption of LO phonons by free
carriers.

The experimental results can be directly compared to the results from the
many-body calculations for the LO phonon self energy described in Chapter 2.
Fig. 4.6 shows a plot of the experimental results in comparison to the theoretical
results obtained using the screened LO phonon self energy (Eq. 2.5, including the
screening effects of both electrons and holes). The screening was included by using
the diagonal approximation [22] for the electron-hole dielectric function. Since the
c_0-c_1 transitions only contribute to the real part of the dielectric function at the
LO phonon energy, the LO phonon scattering rate can be expressed as

\begin{equation}
\frac{1}{\tau} \sim \frac{1}{(\text{Re}[\epsilon_{001}])^2} \frac{|M_{01}(q_\|)|^2 \text{Im}[P_{01}(q_\|, \omega) + P_{10}(q_\|, \omega)]}{|\epsilon_{001}^{\text{hole}}(q_\|, \omega)|^2}
\end{equation}

where \(\epsilon_{el}\) and \(\epsilon_{hole}\) stand for the contributions to the dielectric function from
electrons and holes respectively. The theoretical results are obtained by adding the
scattering rate due to intervalence band transitions \(\Gamma_{IVB}\) and the bare \((n_{PL} = 0)\)
scattering rate \(\Gamma_{n_{PL}=0}\) that reflects the decay of the LO phonon modes into acous-
tic phonons. \(\Gamma_{n_{PL}=0}\) is obtained from the experimentally obtained value for the
lifetime with no cold plasma present. The effective lifetime \(\tau_{eff}\) plotted in Fig. 4.6
is then calculated from

\begin{equation}
\tau_{eff} = (\Gamma_{n_{PL}=0} + \Gamma_{IVB})^{-1}
\end{equation}

The model calculations suggest that the holes should have a larger influence on
reducing the hot phonon lifetime than the experiments indicate. This discrepancy
between the experimental and model results for the hole-induced LO phonon
absorption rate is approximately an order of magnitude. Two approximations used in
Figure 4.6: Experimental results for the LO phonon lifetime obtained from the time-resolved Raman scattering experiment (circles) and theoretical results (at \( q_0 = 5 \times 10^4 \text{ cm}^{-1} \)) from calculations including both electron and hole screening (squares).

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developing the model are partially responsible for this over estimate of the phonon absorption rate; i) the use of fully-degenerate carrier distributions ($T = 0$ approximation), and ii) the neglect of lifetime broadening effects on the excited valence band levels. The consequences of these approximations can be appreciated by considering plots of the real and imaginary parts of $\epsilon_{\text{hole}}^{\text{el}}$ and the real part of $\epsilon_{0101}^{\text{el}}$ shown in Fig. 4.7. In particular, the imaginary part of the hole contribution to the intervalence band dielectric function plays a key role in determining the LO phonon absorption rate by cold holes, even in the unscreened limit. This can be seen from Eq. 2.10, in which the numerator is proportional to the imaginary part of $\epsilon_{0101}^{\text{hole}}$, regardless of what approximation is made for the screening function in the denominator. At non-zero temperatures the sharp discontinuities in $\epsilon_{0101}^{\text{hole}}$ would be eliminated due to the smearing of the hole distribution around the Fermi energy. A non-zero width (lifetime broadening) of the final states in the intervalence band transition would also tend to smear out $\epsilon_{0101}^{\text{hole}}$ (eg. the delta function in Eq. 2.11 would be replaced by a normalized function of energy with a width inversely proportional to the lifetime of the excited valence band states). Rough estimates of the hole temperature within 5 ps of the excitation pulse would be in the 50 K to 100 K range. The lifetime of the excited valence band states, that are coupled to the ground valence band via both acoustic and optical phonon scattering processes, is likely in the 150 fs to 300 fs range. More realistically then, the imaginary part of $\epsilon_{0101}^{\text{hole}}$ will be broadened over a range of at least 5 meV to 10 meV. Since this is substantially greater than the range over which it takes on non-zero values in Fig. 4.7, the net effect will be to significantly reduce the corresponding absorption rate of LO phonons.

The other point to consider when quantitatively comparing the model calculations with the experimental results is the fact that no effort has been made to tune the nominal physical parameters of the quantum wells. This means that the LO phonon energy is essentially at the peak of the imaginary part of $\epsilon_{0101}^{\text{hole}}$. Any deviation of the actual well width from this nominal value would therefore tend to further reduce the calculated hole contribution to the LO phonon reabsorption rate.
Figure 4.7: Contributions to the dielectric function near the LO phonon energy (arrow) at $k_F = 9 \cdot 10^5$ cm$^{-1}$. Solid curve: Re($\epsilon_{0101}^{\text{kole}} - 1$); dot-dashed curve: Im($\epsilon_{0101}^{\text{kole}}$); dashed curve: Re($\epsilon_{0101}^{\text{el}} - 1$).

In summary then, the parameters and assumptions embodied within the model calculation are such that it provides an upper limit on the expected LO phonon absorption rate by "cold" holes due to intervalence band transitions. The fact that the calculated rate is approximately an order of magnitude higher than the experimentally observed rate is therefore not inconsistent.

There are two other mechanisms that could, in principle, lead to a carrier-induced reduction in the hot LO phonon population lifetime and that are not included in the calculations presented here. The first involves a redistribution of the hot LO phonons in wavevector space, or phase space diffusion. Until recently, this process had been ignored in the context of reduced cooling rates; however, a calculation of hot phonon diffusion in momentum space has now been reported [54]. The second process involves a carrier-enhancement of the rate at which hot LO phonons decay into acoustic modes. Again, this process had been ignored.
until recently [55]. It is unlikely that either of these mechanisms would have a large effect on the experimental results presented above. The latter mechanism, if it were strong enough to explain our results, would be inconsistent with the large body of literature on the reduced coupling of energy from the carriers to the acoustic phonon bath at elevated densities. In addition, both of these mechanisms are higher order in the carrier-LO phonon interaction than the direct hot phonon reabsorption process.

As mentioned in the introductory chapter, LO phonon reabsorption by free holes had been proposed by Kash et al. to explain reduced LO phonon lifetimes in p-doped and highly excited bulk GaAs samples. The results presented in this thesis are qualitatively consistent with his results. The advantage of the TRROD technique used here lies in the fact that LO phonon reabsorption effects can be unambiguously isolated.

4.3 Stokes/Anti-Stokes Ratios

With the exception of Ref. [55], most hot carrier relaxation models include a constant, density-independent coupling strength between the hot LO phonons and the acoustic phonon bath. The plasma therefore transfers energy to the bath via a large number of hot LO phonons, with the instantaneous power transferred via each mode being proportional, at low lattice temperatures, to the corresponding hot LO phonon occupation number. Therefore, a reduced plasma cooling rate manifests itself as a reduction in the number of nonequilibrium LO phonons integrated over all modes. Fig. 4.8 shows experimental evidence that at elevated carrier densities there indeed is a reduction in the nonequilibrium occupation number of a specific LO phonon mode, and hence a reduction in the energy transfer rate from the plasma to the acoustic phonon bath through this particular LO phonon mode. The time-integrated nonequilibrium LO phonon occupation number $n_{q\parallel}$ with $q_{\parallel} = 5 \times 10^4 \text{ cm}^{-1}$ is plotted in Fig. 4.8 as a function of carrier density. The $n_{q\parallel}$ were extracted from fits (to feature (ii) in spectra like those in Fig. 4.1) to numerous Stokes and anti-Stokes spectra. The determination of an absolute value for the
LO phonon occupation number in a resonant Raman experiment is complicated by the fact that the results have to be corrected for resonance cross-sections $\sigma_S$ and $\sigma_{AS}$ of the Stokes and anti-Stokes signals. If the cross-sections are known, an absolute value for the occupation number $n_{\|}$ can be determined from the ratio of Stokes and anti-Stokes intensities. In practice it is necessary to perform a detailed study of the energy dependence of the Raman cross section ("resonance profiles"), in order to determine $n_{\|}$. To extract the occupation numbers from the time-integrated Raman spectra presented in this thesis, a technique commonly used in the literature (see e.g. [56, 57]) was used. To correct for resonance effects, the relationship

$$\sigma_{AS}(\omega_{in} - \omega_{AS}) = \sigma_S(\omega_{in})$$  \hspace{1cm} (4.3)

was used together with measured resonance profiles to correct the results for resonance effects. Here $\omega_{in}$ stands for the Raman probe frequency. One should note that Eq. 4.3 only strictly holds under non-resonant conditions and has been shown empirically to be valid under resonant conditions [57].

In the absence of a detailed theory to support the technique for converting Stokes and anti-Stokes data into occupation numbers under nonequilibrium conditions, a degree of uncertainty in the $n_{\|}$ values in Fig. 4.8 is noted. However, based on the time-resolved results in Fig. 4.5, one would expect that for a fixed generation rate of nonequilibrium LO phonons with $q_{\|} = 5 \cdot 10^4 \text{ cm}^{-1}$, their absolute numbers should decrease in relation to their reduced lifetime at elevated cold plasma densities. The data in Fig. 4.8, despite possible complications due to nonequilibrium effects on the resonant enhancement of the Raman signals, provide corroborating evidence for the observed reduction in LO phonon lifetime. Many-body calculations of the self energy of an electron in subband $c_2$ that can contribute to the generation of nonequilibrium LO phonon modes at the observed wavevector show that a decrease of the LO phonon population with increasing cold plasma density is not only due to a reduction in the effective LO phonon lifetime, but also to enhanced scattering into LO phonon modes other than those observed [38].
Figure 4.8: LO phonon occupation number as obtained from the measurement of Stokes/anti-Stokes ratios.
Chapter 5

Conclusions

The observation of a reduced hot carrier cooling rate at high carrier densities in polar semiconductors has attracted a large amount of attention in the last two decades. The interpretation of this effect is based on the assumption of carrier re-heating by small wavevector LO phonons. Despite the large number of experiments that demonstrate the effect by measuring the properties of the carrier system, direct evidence from the lattice perspective had been missing until now. This is mainly due to the fact that in previous experiments it was very difficult to isolate hot carrier from hot LO phonon effects. The results presented in this thesis give direct evidence for LO phonon reabsorption by free carriers. These results were obtained by introducing a novel experimental technique that allows one to unambiguously measure LO phonon reabsorption effects from the lattice perspective and by developing a new theoretical model for calculating LO phonon reabsorption by free holes.

The experimental investigation of LO phonon reabsorption processes necessitated the careful choice of a particular sample geometry and the development of an appropriate measurement technique. The sample chosen is a GaAs quantum well with one pair of subbands in each valence and conduction band that have an energy spacing that is close to the LO phonon energy. The TRROD technique used to isolate LO phonon reabsorption processes in this sample is based on the well established time-resolved Raman scattering technique for investigating the lattice dynamics in quantum wells. The pre-injection of a fixed density, cold electron-hole
plasma into the sample allows for an investigation of the hot LO phonon dynamics in the presence of a continuously variable but controlled density of cold electrons and holes that does not contribute to LO phonon generation processes but can absorb hot LO phonons. In the experiments presented here reabsorption is only possible by intervalence subband transitions of free holes. This choice makes it possible to investigate the role of holes in the reabsorption of LO phonons. The results of the TRROD experiments clearly show a strong reduction in the LO phonon lifetime with increasing cold plasma density. With no cold plasma present the LO phonon lifetime was measured to be 4.5 ps. At a cold plasma density of $1.5 \times 10^{11}$ the lifetime was reduced to 3 ps and at a cold plasma density of $3 \times 10^{11}$ it was measured to be 1.9 ps. These experimental results clearly demonstrate efficient reabsorption of hot LO phonons by the cold carriers from the lattice perspective. In addition to the measurement of the LO phonon dynamics in the TRROD experiment the author also investigated the influence of the cold electron-hole plasma on time-integrated Raman spectra. The determination of the cold plasma density dependence of the LO phonon occupation number from time-integrated Raman Stokes and anti-Stokes spectra supports the results of the TRROD experiments. These data show a strong reduction of the LO phonon occupation number as expected in the case of a fixed LO phonon generation rate and a reduction in LO phonon lifetime. In addition, the Raman spectra show a renormalization of the coupled LO phonon-c$_0$-c$_1$ intersubband plasmon mode to higher energies with increasing cold plasma density. This shift of 1.4 meV at $n_{pL} = 3 \times 10^{11}$ cm$^{-2}$ clearly demonstrates anti-screening of the LO phonons by the c$_0$-c$_1$ plasmon. The comparison of these results with calculations of the shift of the CPPM indicate that the blue-shift of the LO phonon-like CPPM is mainly due to interaction with the c$_0$-c$_1$ CPPM. The same calculations suggest that the interaction of the LO phonon-like CPPM is much weaker due to strong Landau damping of the hole plasmon. The experimental results presented in this thesis shows that the TRROD technique is a powerful tool for investigating free carrier-LO phonon interactions from the lattice perspective.

A model was also developed to quantitatively describe the hot phonon reab-
sorption process in quantum wells. The author calculated the cold plasma contributions to the LO phonon lifetime through the imaginary part of the LO phonon self energy within a two subband model. The LO phonon-cold plasma interaction is intrinsically many-body in nature and requires an adequate treatment of screening effects. Since the reabsorption of the LO phonons was restricted to intervalence band transitions, a realistic model for the complicated valence band structure of the quantum well had to be included. To the best knowledge of the author this is the first such calculation of free hole contributions to the LO phonon lifetime that explicitly includes screening and valence band dispersion effects. The importance of nonparabolicity effects is shown in the results of the calculations by the strong deviations in the kinematic constraints and the Coulomb matrix elements from the results of a parabolic band model.

The results of the calculations give a theoretical LO phonon absorption rate approximately an order of magnitude larger than the observed in the experiments. This discrepancy does not, however, invalidate the interpretation of the experimental observation as being due to hot phonon reabsorption by cold holes. The assumption of a fully degenerate hole distribution, the neglect of lifetime broadening of the excited valence band states, and the use of the nominal quantum well parameters in the model calculation all tend to increase the theoretical absorption rate over what it would be if more realistic (but complicating) conditions were taken into account.

In concluding the author wishes to make a few additional remarks on the relevance of the work presented in this thesis and possible extensions. The direct observation of the importance of intervalence band absorption processes lends further support to the suggestion by Kash et al., that the reduced phonon lifetime in p-doped and highly excited bulk GaAs samples is due to intervalence band re-absorption processes. This demonstrates the important influence of the valence bands on the dynamics of small wavevector LO phonons and suggests that any model dealing with these processes should include the contributions of the valence bands. The theoretical framework developed in this thesis could prove useful in the modelling of Heterojunction Bipolar Transistors (HBT’s). The dynamics of hot
electrons in the p-doped base region of these devices is crucial for the switching speed in these devices [61]. The two subband model for the hole dielectric function derived in this thesis could also be used in the interpretation of Raman [62] and infrared absorption experiments in p-doped GaAs quantum wells.
Appendix A

Computation of Valence Band Dispersion and Wavefunctions

The purpose of this appendix is to give detail on the calculation of the valence band structure and hole wavefunctions within the axial approximation that is not readily available in the literature. The Luttinger-Kohn Hamiltonian Eq. 2.18 can be simplified by utilizing the axial approximation [31]. The Hamiltonian can be transformed by a unitary transformation $H' = U H U^{-1}$ into block diagonal form $H'$.

$$H' = \begin{pmatrix} P + Q & \hat{R} & 0 & 0 \\ \hat{R}^* & P - Q & 0 & 0 \\ 0 & 0 & P + Q & \hat{R}^* \\ 0 & 0 & \hat{R} & P - Q \end{pmatrix} = \begin{pmatrix} H'^{uu} & 0 \\ 0 & \hat{H}' \end{pmatrix} \quad (A.1)$$

where

$$\hat{R} = \frac{\hbar^2}{2m} \sqrt{3(\bar{\gamma} k_y^2 - 2i\gamma_3 k_y k_z)} \quad (A.2)$$

and

$$\bar{\gamma} = \frac{\gamma_2 + \gamma_3}{2}. \quad (A.3)$$

The simplest result for the transformation matrix $U$ and the inverse $U^{-1}$ can be obtained if one sets $k_x = 0$. This is possible since in the axial approximation $k_x$ and $k_y$ are interchangeable. The transformation matrices for this case are given by [58]

$$U = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 0 & -1 \\ 0 & -1 & 1 & 0 \\ 1 & 0 & 0 & 1 \\ 0 & 1 & 1 & 0 \end{pmatrix} \quad (A.4)$$

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The eigenfunctions for the upper and lower Hamiltonians $H^u$ and $H^l$ are given by [25]

\[ F^u_{hh} = \frac{1}{N} \begin{pmatrix} U_1 \\ \tilde{R}^* \end{pmatrix} e^{i\tilde{\xi}r} \]  \hspace{1cm} (A.6)

\[ F^u_{lh} = \frac{1}{N} \begin{pmatrix} -\tilde{R} \\ U_2 \end{pmatrix} e^{i\tilde{\xi}r} \]  \hspace{1cm} (A.7)

\[ F^l_{hh} = \frac{1}{N} \begin{pmatrix} U_1 \\ \tilde{R} \end{pmatrix} e^{i\tilde{\xi}r} \]  \hspace{1cm} (A.8)

\[ F^l_{lh} = \frac{1}{N} \begin{pmatrix} -\tilde{R}^* \\ U_2 \end{pmatrix} e^{i\tilde{\xi}r} \]  \hspace{1cm} (A.9)

where

\[ U_1(\tilde{k}) = Q(\tilde{k}) - P(\tilde{k}) - E + V_b \]  \hspace{1cm} (A.10)

\[ U_2(\tilde{k}) = Q(\tilde{k}) + P(\tilde{k}) + E - V_b \]  \hspace{1cm} (A.11)

and $N$ and $V_b$ denote a normalization factor and the barrier potential respectively.

It must be noted that, due to the unitary transformation, the eigenstates of $H^l$ are not parity eigenstates as is the case for the eigenstates of $H$. But once the eigenstates for $H^u$ and $H^l$ have been obtained, the states in the original basis can be reconstructed using the unitary transformation. It can be shown that the eigenfunctions of $H^l$ can be obtained by a reflection $z \rightarrow -z$ of the eigenfunctions of $H^u$. These symmetry relations greatly simplify the calculation of the valence band structure.

To solve for the valence band structure and the related eigenfunctions of the quantum well, the wavefunctions and the current density have to be matched at the interfaces. In the case of $E < V_b$ there are exponentially decaying solutions to the Schrödinger equation outside of the quantum well (Material B) and confined states inside the well. In the case of $E > V_b$, there exist continuum states that also extend across the barrier region. These states are not considered in the following, since the focus here is on the confined states. Analogous to a finite quantum well calculation [59], the boundary conditions at the interfaces can be written in form
of a homogeneous system of linear equations

\[ \mathbf{A} \cdot \mathbf{b} = 0 \]  \hfill (A.12)

Here the solution vector \( \mathbf{b} \) contains the coefficients for the wavefunctions in the different regions. Inside the quantum well, the solution is a superposition of one forward and one backward travelling wave with two components each, requiring a total of four free coefficients. As already mentioned, there are exponentially decaying solutions outside of the quantum well and therefore only two free coefficients are required on each side. This leads to \( \mathbf{A} \) being a 8*8 matrix.

The boundary condition matrix \( \mathbf{A} \) for \( H^a \) is given in Fig. A.1. The indices l/h and b/w in \( \mathbf{A} \) stand for light/heavy hole and barrier/well respectively. All k’s in the matrix are \( k_z \) as determined from the bulk dispersion relations.

The current density operators used in this matrix are derived for the block diagonal form of the Hamiltonian following Ref. [25].

\[ J_1^h = \frac{\hbar}{m} (\gamma_1 - 2\gamma_2)k_z U_1 - \sqrt{3} \gamma_3 i k_q \hat{R}^* \]  \hfill (A.13)

\[ J_2^h = \frac{\hbar}{m} (\gamma_1 + 2\gamma_2)k_z \hat{R}^* + \sqrt{3} \gamma_3 i k_q U_1 \]  \hfill (A.14)

\[ J_1^l = -\frac{\hbar}{m} (\gamma_1 - 2\gamma_2)k_z \hat{R} - \sqrt{3} \gamma_3 i k_q U_2 \]  \hfill (A.15)

\[ J_2^l = \frac{\hbar}{m} (\gamma_1 + 2\gamma_2)k_z U_2 - \sqrt{3} \gamma_3 i k_q \hat{R} \]  \hfill (A.16)

The linear system A.12 only has a solution if the determinant \( \det(\mathbf{A}) = 0 \), i.e. when \( \mathbf{A} \) becomes singular. Since not only the eigenvalues are required for the calculation of the self energy, the author uses the technique of Singular Value Decomposition [60], which gives both eigen values and eigen functions. The upper and lower block Hamiltonians \( H^a \) and \( H^l \) have degenerate eigenvalues for a quantum well potential that is symmetric with respect to \( z = 0 \). It is therefore sufficient to solve for just one of these 2*2 Hamiltonians to obtain the energy eigenvalue for a given \( k_q \). The eigen functions of \( H^a \) and \( H^l \) are related through \( F_{H^a}(z) = F_{H^l}(-z) \). Using this relationship and the unitary transformation, the wavefunctions in terms of parity eigenstates can be constructed.
The strategy used to solve the quantum well eigenvalue problem is the following: first, a value for $k_\parallel$ is chosen. Then the matrix $A$ is evaluated for a range of energies $E$ and zeroes in the smallest of the singular values are searched for. Once a zero is found, Eq. A.12 has been fulfilled and an energy eigenvalue is found. The Singular Value Decomposition automatically determines the coefficient vector $\vec{b}$ for the eigen functions. A flowchart of the structure of the band structure program is given in Fig. A.1.
\[ A = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix} \]

Figure A.1: Matrix $A$
Input of quantum well parameters:
well width, barrier height, Luttinger
parameters for barrier and well.

Input of loop parameters: Range and number
of steps for k// and energy E.

Choose value for k//
Set E=0.
calculate k_z from dispersion relation
Set up matrix A
Do Singular Value Decomposition of A
Increment energy
No
Is A singular?
Yes
Save eigenvalue and parameters
for wave function.
Yes
E<E_{max}?
No
k//<k_{max}
No
End

Figure A.2: Flowchart of valence band structure program
Appendix B

Computation of Self Energy and Dielectric Function Integrand

This appendix summarizes the details of the numerical evaluation of the self energy and dielectric function integral Eq. 2.35.

Since the valence band dispersion is symmetric with respect to $k_n = 0$ in the axial approximation, it suffices to retain only even powers in the fit to the $\vec{k} \cdot \vec{p}$ band structure. It was found that an eighth order fit leads to very good agreement with the calculated band structure.

$$E'(\vec{k}_n) = a^1 + b^1 \vec{k}_{n1}^2 + c^1 \vec{k}_{n1}^4 + d^1 \vec{k}_{n1}^6 + e^1 \vec{k}_{n1}^8$$  \hspace{1cm} (B.1)

Fig. B.1 shows both, the fit and the original $\vec{k} \cdot \vec{p}$ calculation for the hh$_0$ and hh$_3$ bands.

The next step in evaluating Eq. 2.35 is to rewrite the delta function using the identity

$$\delta(f(x)) = \sum_i \frac{1}{\left| \frac{df(x)}{dx} \right|_{x=x_i}} \cdot \delta(x-x_i)$$ \hspace{1cm} (B.2)

in order to get rid of the $\theta$ integration. Here, $x_i$ denotes a root of the function $f(x)$.

In the above case this leads to

$$\text{Im}(P_{ij}) = \frac{\pi}{(2\pi)^2} \cdot \int_0^{k_F} d\vec{k}_n \cdot \left[ \frac{1}{\left| g_\| (1-(\frac{k_n^2-q_n^2-k_{n1}^2}{2k_n q_n})^{1/2}(2\nu + i\omega k_{n1}^2 + 6d^2 k_{n1}^4 + 2\delta ck_{n1}^6))^{1/2} \right|} \cdot \delta'(E_{\|}+\nu)-\delta'(E_{\|})-\hbar \omega \right]$$  \hspace{1cm} (B.3)
The quantum well valence subbands can be classified into two types. The first type are bands with a single minimum in the dispersion at $k_n = 0$. The second type are bands with two minima at finite $k_n$. This is evident for instance in Fig. 2.3, where hh$_0$, hh$_1$ and lh$_1$ belong to the single minimum class and lh$_0$, hh$_2$ and hh$_3$ belong to the double minimum class. The lowest energy subband is always of the single minimum type. For evaluation of the polarizability function, it is assumed that only the lowest (hh$_0$) subband is populated with carriers. Depending on which intersubband transition one wishes to calculate, one thus has to evaluate a single minimum to a single minimum transition or a single minimum to a double minimum transition. In the following, it is described how both of these cases can be treated and how the formalism is coded for numerical evaluation. To evaluate the integral Eq. 2.35 all kinematically allowed transitions have found and their respective contributions to the integrand has to be calculated.

For a transition from band $i$ to band $j$ a transition is energetically allowed
under the following condition (See Fig. B.2 and Fig. B.1a):

\[ E_i(k_i^1) + h\omega > E_j(k_\parallel = 0) \]  

(B.1)

If this is fulfilled for a given initial parallel wavevector \( k_\parallel^1 \) and excitation energy \( h\omega \) the wavevector \( k_\parallel^* \) is determined from energy conservation

\[ E_i(k_i^1) + h\omega - E_f(k_\parallel^*) = 0 \]  

(B.5)

This equation is solved by inserting the dispersion for \( E_i \) and \( E_j \) and using a bisection algorithm to search for solutions. Once \( k_\parallel^* \) is obtained, one has to check for momentum conservation. If

\[ |k_\parallel^* - k_\parallel| \leq |q_\parallel| \leq |k_\parallel^* + k_\parallel|, \]  

(B.6)

the transition is kinematically allowed and the integrand in Eq. B.3 contributes to the integral. Otherwise the transition is not allowed and the integrand equals zero.

Similar to the single minimum-single minimum case, the integrand can be calculated from Eq. B.3 if energy and momentum conservation are fulfilled but is zero otherwise. As can be seen from Fig. B.3 and B.4b there are the following special cases:

1. 

\[ E_i(k_i^1) + h\omega > E_j(k_\parallel = 0) \]  

(B.7)

This is equivalent to the single minimum case in the previous section.

2. 

\[ E_j(k_{\parallel, min}) < E_i(k_i^1) + h\omega < E_j(k_\parallel = 0) \]  

(B.8)

In this case, Eq. B.5 (energy conservation) has two solutions, \( k_{\parallel,1}^* \) and \( k_{\parallel,2}^* \). Depending on the magnitude of \( k_\parallel \) and \( q_\parallel \) there are the following possibilities

\[ |k_{\parallel,2}^* - k_\parallel| < |q_\parallel| < |k_{\parallel,1}^* - k_\parallel| \]  

(B.9)

Contributions to the integral arise from the \( k_{\parallel,2}^* \) shell only and therefore the integrand has to be evaluated at \( k_{\parallel,2}^* \).
Figure B.2: Intervalence subband scattering between single-minimum bands.

\[ |k^*_{||,2} + q_{||}| < |q_{||}| < |k^*_{||,1} + k_{||}| \]  \hspace{1cm} (B.10)

or

\[ |k^*_{||,1} - q_{||}| < |q_{||}| < |k^*_{||,2} - k_{||}| \]  \hspace{1cm} (B.11)

Contributions to the integral arise from the \( k^*_{||,1} \) shell only and therefore the integrand has to be evaluated at \( k^*_{||,1} \).

\[ |k_{||} - k^*_{||,2}| < |q_{||}| < |k^*_{||,1} - k_{||}| \]  \hspace{1cm} (B.12)

and

\[ |q_{||}| < |k^*_{||,2} + k_{||}| \]  \hspace{1cm} (B.13)

Contributions to the integral arise from both the \( k^*_{||,1} \) shell and the \( k^*_{||,2} \) shell. The integrand has to be evaluated at \( k^*_{||,1} \) and \( k^*_{||,2} \).
Figure B.3: Intervalance subband scattering between single-minimum band double-minimum band.
Figure B.4: Constraints for single minimum and double minimum scattering.
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