Interplay between coherent and incoherent phonons in optically excited biased quantum wells

T Papenkort¹, T Kuhn¹ and V M Axt²

¹ Institut für Festkörpertheorie, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany
² Institut für Theoretische Physik III, Universität Bayreuth, 95440 Bayreuth, Germany
E-mail: t.papenkort@uni-muenster.de

Abstract. We present numerical simulations of the generation of coherent LO phonons in an electrically biased quantum well. An ultrashort laser pulse is used to simultaneously excite two exciton levels, which leads to an oscillating dipole moment that couples to the polar lattice and drives the phonons. The generation of coherent phonons becomes resonantly enhanced if the splitting of the two exciton levels is tuned to the LO phonon energy. In this case relaxation by emission of incoherent phonons becomes also possible. Our calculation therefore takes into account incoherent phonons on a quantum kinetic level as well. The intersubband relaxation by excitation of incoherent phonons competes with the generation of coherent phonons and is dominant in terms of the energy transferred.

1. Introduction

The generation of coherent phonons by optical excitation with short laser pulses has been extensively studied both in bulk semiconductors [1–3] and in semiconductor quantum wells [4–6]. For different materials several different phonon generation mechanisms have been discussed [5]. They have in common that the ultrashort laser pulse either almost instantaneously creates a new carrier distribution which the lattice ions have to adjust to (displacive mechanism) or that it causes a short and intensive force on the ions (impulsive mechanism). In both cases the electronic subsystem changes on a very short time scale and after that it does not contribute to the generation process any more.

In this paper a resonant generation mechanism will be discussed. It is well known that ultrashort pulses may simultaneously excite different electronic levels of a quantum well. This leads to a quantum beat and, if the well is biased by an external electric field, to an oscillating dipole moment emitting THz radiation [7, 8]. Obviously this oscillating dipole moment also couples to LO phonons and, if the frequency of the oscillation coincides with the LO phonon energy, phonons are generated (resonant mechanism). In experiments the coupled state of quantum beat and LO phonons has been observed and it was shown that by tuning the subband splitting to the LO phonon energy both coherent LO phonon generation and THz emission become resonantly enhanced [6, 9, 10].

We present numerical simulations of the phonon generation in a biased quantum well modeled according to the experiment of Ref. [6]. Our calculations are based on a microscopic model with three subbands coupled to the LO phonon branch by Fröhlich interaction. The Coulomb interaction is taken into account on the mean-field level. Thus, excitonic effects in the carrier...
generation process are fully included. We make use of density matrix theory to derive our
equations of motion and cut off the resulting hierarchy by factorizing doubly phonon assisted
terms. Details of the derivation of the equations of motion may be found in Ref. [11]. The cut-off
of the hierarchy at this level means that both coherent and incoherent phonons are included. It
turns out that even a computationally much simpler model taking into account only coherent
phonons is able to reproduce the resonantly enhanced phonon generation. However, because
the subband splitting is equal to the LO phonon frequency in the resonant case, relaxation by
emission of incoherent phonons is very important, as will be shown below. In fact we expect
that this process might be at least partially responsible for the fact that the quantum beat did
not show up in the measurements of Ref. [6].

2. Model system
The system under consideration is a GaAs/AlAs quantum well with a width of 15.3 nm modeled
by a square potential. The potential has a depth of 1.58 eV which is distributed 60/40 to
conduction and valence band. The two optical transitions used for the phonon generation
process are the transitions from the uppermost two heavy-hole subbands to the second lowest
electron subband. We therefore reduce our model to these three subbands, leaving out the lowest
electron subband.

Figure 1(a) shows the subband structure for an external electric field of 110 kV/cm. At
this field strength, the splitting of the two hole subbands is close to the LO phonon energy.
The lowest electron subband, shown dashed in the plot, is not included in our calculation. In
Fig. 1(b) the calculated absorption spectrum, i.e. the imaginary part of the complex electric
susceptibility $\chi$, is shown. We observe two exciton peaks and the corresponding subbands. The
dashed line shows the energy distribution of the laser pulse with a full width at half maximum
of 100 fs and a central energy of 1.571 eV. The pulse shape overlaps with both excitons, so that
a coherent superposition of these states is excited.

3. Displacive and resonant coherent phonon generation
The oscillating carrier distribution couples to the lattice via Fröhlich interaction and thereby
generates coherent phonons. Figure 2 shows the lattice displacement $u(z,t)$ after an excitation
with the pump pulse centered at $t = 0$ for two different bias fields. As we assume a system
that is homogeneous perpendicular to the growth direction $z$, the displacement field $u$ points in
$z$-direction and it only depends on $z$. It is largely confined to the quantum well, which extends
from $z = 0$ nm to 15.3 nm.
Let us first take a look at the coherent phonons generated in an off-resonant case (Fig. 2(a)). The electric field of 30 kV/cm gives rise to a subband splitting of 15 meV, which is far from the LO phonon energy of 36.3 meV. The lattice displacement first goes up and then oscillates with the LO phonon frequency. This case is an example of displacive phonon generation: The sudden change in the carrier distribution results in a new equilibrium position for the ions, which start to oscillate around their new equilibrium. As is typical for the displacive case, the oscillation is cosine-like, i.e. the extrema lie at integer or half-integer multiples of the LO phonon period.

Figure 2 shows the resonant case, where the electric field of 110 kV/cm yields a subband splitting of 35.5 meV. The laser pulse is the same as above. Here, too, the phonon oscillation has a shifted equilibrium position, but in addition it gains amplitude with every period. The oscillation of the carrier distribution resonantly drives the phonon oscillation and therefore the amplitude increases approximately linearly in time. Already after only 0.5 ps the oscillation amplitude is much higher than for non-resonant excitation (note that the vertical axis is scaled differently in Figs. 2 (a) and (b)).

4. Energy density of coherent and incoherent phonons

We will now discuss the energy transferred into the phonon subsystem. This allows us to quantify the efficiency of the coherent phonon generation for different excitation conditions. After the optical excitation the Hamiltonian of our system can be divided into three different parts: $H = H_{el} + H_{ph} + H_{Fr}$, i.e., one part for the electronic subsystem, one for the phonon subsystem and the Fröhlich interaction Hamiltonian. The energy of the phonons is simply $\langle H_{ph} \rangle = \sum_{\mathbf{q}} \hbar \omega_{LO} \langle \hat{b}_{\mathbf{q}}^{\dagger} \hat{b}_{\mathbf{q}} \rangle$, where $\omega_{LO}$ is the LO phonon frequency and $\mathbf{q}$ is the phonon wave vector. The phonon number can be divided into the number of coherent and the number of incoherent phonons by $\langle \hat{b}_{\mathbf{q}}^{\dagger} \hat{b}_{\mathbf{q}} \rangle = \langle \hat{b}_{\mathbf{q}}^{\dagger} \rangle \langle \hat{b}_{\mathbf{q}} \rangle + \delta \langle \hat{b}_{\mathbf{q}}^{\dagger} \hat{b}_{\mathbf{q}} \rangle$ [2]. Only coherent phonons produce a non-vanishing mean lattice displacement $u(z, t)$, the incoherent phonons only add to the fluctuations $\Delta u$. The interaction energy $\langle H_{Fr} \rangle$ can be divided along the same lines and will be included into the phonon energy.

Figure 3 shows the energy transferred into coherent and incoherent phonons versus the electric field. For the coherent phonons this energy oscillates in time, so the maximum in the first 0.5 ps has been plotted. The coherent phonons peak where the subband splitting crosses the LO phonon energy. This is a clear indication for the resonant generation process and it agrees qualitatively with the experiments shown in Ref. [6].

For the incoherent phonons the energy at 0.5 ps is shown. As this energy is much larger, the energy of the coherent phonons has been multiplied by a factor of 500 relative to the incoherent phonon energy. Generation of incoherent phonons sets in when the subband splitting approaches the LO phonon energy, i.e. as soon as intersubband relaxation becomes possible. Even before
resonance is reached the energy transferred into incoherent phonons surpasses the energy of the coherent phonons.

5. Conclusion
We have shown numerical simulations of the generation of LO phonons in a biased quantum well excited by an ultrashort laser pulse. Two exciton levels are simultaneously excited and the resulting oscillating polarization drives coherent phonons. If the electrical bias is used to tune the exciton splitting to the LO phonon energy, coherent phonon generation becomes resonantly enhanced. Incoherent phonons have been considered on a quantum kinetic level as well, and it was shown that although intersubband relaxation by emission of incoherent phonons dominates the energy transfer to the lattice, resonant generation of coherent phonons still takes place.

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