Electron–LO-phonon quantum kinetics in semiconductor quantum wells

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Quantum correlations between electrons and phonons are demonstrated in two-dimensional semiconductors. The resulting phonon oscillation exhibits characteristic differences with respect to the three-dimensional parent system. We show how these correlations can unambiguously be distinguished from nonequilibrium coherent phonons. Furthermore, coherent control allows separation of the phonon oscillation from another oscillation observed here, specific to two-dimensional systems, which is due to the interference of two electron-electron scattering channels associated with different quantum-well subbands. [S0163-1829(98)01632-4]

According to quantum mechanics, any system possesses a complete set of orthonormal eigenfunctions φ_n and can always be prepared in an initial state described by the wave function $\Phi=\Sigma_n a_n\varphi_n$, where the amplitudes a_n are complex quantities. Therefore, it is possible, in principle, to adjust the amplitudes and phases of the a_n in such a way that the temporal behavior of any observable can be (almost) arbitrarily determined. The current interest in coherent control[1,2] stems from the fact that this scenario can actually be implemented, at least in the case of systems with a limited number of degrees of freedom, by use of coherent laser excitation. Macroscopic systems, such as a semiconductor, however, have a dense spectrum of energy levels and scattering processes among elementary excitations are often thought of as inherently irreversible processes. Yet even for these complex systems, in the early time of the quantum kinetics regime[3–9] relaxation processes are nothing but interference phenomena. Therefore, these processes are not just intrinsic properties of the solid, but a result of the specific way an experiment is conducted. It is only for long times that the interferences become so complicated and destructive that the dynamics looks irreversible. Only in that limit is the concept of effective scattering times meaningful.

In this paper we investigate how the effects of electron–LO-phonon quantum kinetics in a (quasi)two-dimensional (2D) semiconductor quantum well system (QWS) compare to those in three dimensions.[3,9] QWS's are of interest because their dynamics is more complex than that of the bulk parent material with, in addition, effects of electron-electron scattering involving different subbands. This complexity is first demonstrated in two-pulse experiments. We then show that, by preparing the QWS in different initial states through coherent control, it is possible to distinguish among the different interaction processes, e.g., between coherent phonon effects[10–13] and electron–LO-phonon quantum kinetics. In this way, we obtain more precise information on the dynamics of 2D electronic systems.

We have investigated two $Al_{0.3}Ga_{0.7}As/GaAs$ multiple QWS's, with 20 periods of 11.6 nm-wide wells and 15.0 nm-thick barriers for one (1), and 60 periods of 7.9 nm-wide wells for the other (2). The samples have been grown by molecular-beam epitaxy, the GaAs substrates were etched off, and they were glued onto sapphire disks and antireflection coated on the front side. During the experiments they were kept at $T=77$ K. The linear absorption spectrum of sample 1 is shown in the top inset of Fig. 1. One can distinguish clearly the heavy-hole (hh) and light-hole (lh) exciton peaks at the edge of the first subband (peaks labeled 1 and 2). The half width at half maximum of the hh-exciton line is 1.5 meV. Notice that the peak labeled 3 is due to a transition from the first lh subband to the second electronic subband. Such so-called forbidden transitions occur in many high-quality samples and indicate broken inversion symmetry in the growth direction (e.g., due to asymmetric heterointerfaces), a point that is relevant for the discussion on coherent phonons below. The transition from the second hh subband to the second electronic subband is labeled 4.

First, we have performed two-pulse four-wave mixing (FWM) measurements where the samples are excited by two sech^2-shaped, Fourier transform-limited optical pulses of 15 fs in duration and wave vectors $\mathbf{q}_1$ and $\mathbf{q}_2$, respectively, separated by the time delay $t_{21}=t_2-t_1$. We detect the total diffracted signal, time-integrated (TI) FWM, in direction $2\mathbf{q}_2-\mathbf{q}_1$ as t_{21} is varied. Figure 1 shows TI-FWM traces for increasing carrier density $n_{eh}=1.8\times10^{10}\rightarrow6.7\times10^{11}$ cm^{-2}. The dynamics of the signal, $S_{\text{TI-FWM}}$, is complex with one oscillation with a period $T_{\text{sub}}\approx45-50$ fs, a second much slower one with a period $T_{\text{hh-lh}}\approx500$ fs, and a third oscillation associated with the electron–LO-phonon quantum kinetics correlation around the LO-phonon period $T_{\text{LO}}=2\pi/\omega_{\text{LO}}=115$ fs in GaAs. The period of this oscillation is independent of n_{eh}. The slow oscillation is the well-known hh-lh quantum beat, recently also observed for continuum states.[14] It is consistent with the 8 meV hh-lh splitting seen in the absorption spectrum. The fast oscillation corresponds well to the 92 meV separation between the first and second subband as seen in the absorption spectrum. In sample 2, the subband separation is 130 meV, and this oscillation is correspondingly faster (not shown). Note, however, that the FWM spectrum, bottom inset of Fig. 1, shows no satellite (at a better than 1:1000 signal-to-noise ratio). This implies that this oscillation is neither a quantum beat nor a polarization interference.[15] Since in our conditions the dominant nonlinearity is due to excitation-induced dephasing,[16,17] it can be understood as arising from an interference between two different scattering channels: scattering of excitons with electrons in the first subband and scattering of excitons with electrons in the second subband.

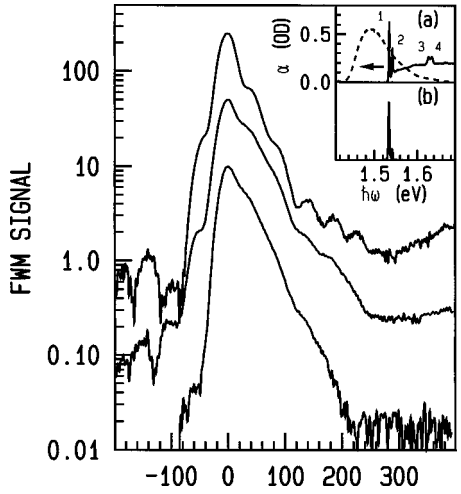

FIG. 1. Two-pulse four-wave-mixing signal of a GaAs quantum-well sample held at $T=77$ K and excited by optical pulses of 15 fs in duration. The carrier density n_{eh} increases from top to bottom: 1.8×10^{10}, 1.5×10^{11}, and 6.7×10^{11} cm^{-2}. Notice that the decay dynamics contains three different oscillations. The inset (a) shows the linear absorption (α) spectrum in units of optical densities (OD) and the laser spectrum, inset (b) the FWM spectrum ($n_{eh}=1.8\times10^{10}$ cm^{-2}) at zero time delay.

To investigate the electron–LO-phonon quantum kinetics in quasi-two-dimensions one would like to avoid interference with other scattering processes, i.e., eliminate the other two oscillations. This is impossible through the design of the QWS. In that respect the 11.6 nm QW width is, in fact, already an optimized choice. For a thicker QW the subband separation decreases, $T_{\rm sub}$ increases and comes yet closer to $T_{\rm LO}$; for a thinner QW it is the hh-lh splitting that increases, then $T_{\rm hh\text{-}lh}$ decreases and approaches $T_{\rm LO}$. We demonstrate now how coherent control can perform the desired operation. For this we replace the pulse propagating in the direction $\mathbf{q}_1$ by two phase-locked pulses 1 and $1'$.[9] These are derived from an apparatus described in Ref. 18, and are separated by the time delay $t_{11'}=t_1-t_{1'}$, which is controlled with an accuracy better than ±50 as. This is the key issue in our work, because $t_{11'}$ can be adjusted with respect to any optical transition frequency Ω for constructive interference, i.e., $\Omega t_{11'}=n2\pi$, or destructive interference $\Omega t_{11'}=(n2+1)\pi$; here n is an integer. For example, in a system, such as our QWS, whose dynamics involves three frequencies Ω_m, $m=1,2,3$, it is possible to adjust $t_{11'}$ to have destructive interference with respect to Ω_3 (second subband) but not for Ω_1 (first subband) and Ω_2 (first subband+one LO-phonon energy). In this case, we excite the phonon oscillation but suppress the subband oscillation. In the laboratory the two phase-locked pulses along $\mathbf{q}_1$ interact with the pulse along $\mathbf{q}_2$, arriving in the sample at $t=t_2=t_{1'}+t_{21'}$. All three pulses have approximately the same intensity and are linearly polarized in the plane of incidence. We still detect the diffracted signal along $2\mathbf{q}_2-\mathbf{q}_1$. A set of TI-FWM traces vs $t_{21'}$ for ten values of $t_{11'}=-19.00$ fs$\rightarrow-21.53$ fs is depicted in Fig. 2. Around $t_{11'}=-20$ fs all three oscillations are modulated, whereas around $t_{11'}=-53$ fs we exclusively modulate the phonon oscillation and the hh-lh beats (not shown). This is because the polarizations of the first and the second subband are again in phase after one beat period and

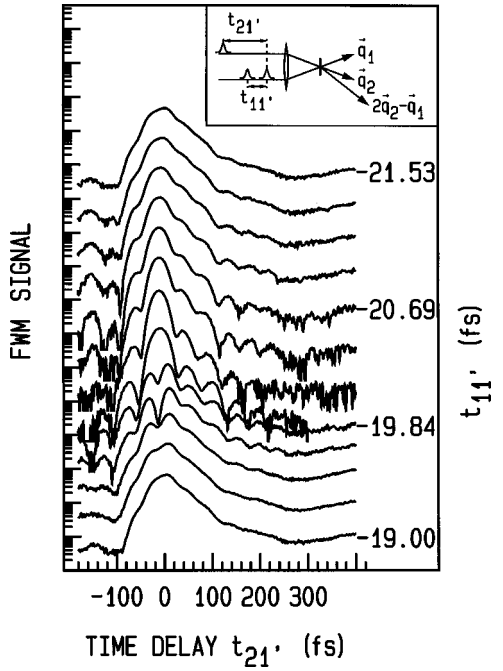

FIG. 2. Coherent control experiment under the same conditions as Fig. 1. The time delay $t_{11'}$ between the phase-locked pulses is parameter. It allows us to control the initial state wave function. The total carrier density (incoherent sum) is $n_{eh}=2.5\times10^{10}$ cm^{-2}. The curves are displaced vertically for clarity. The inset is a sketch of the experiment.

thus cannot be modulated separately. At more elevated carrier densities (Fig. 3) the 45–50 fs oscillation loses relative weight, while the phonon oscillation remains visible. In order to analyze the data more quantitatively, we fit the profile

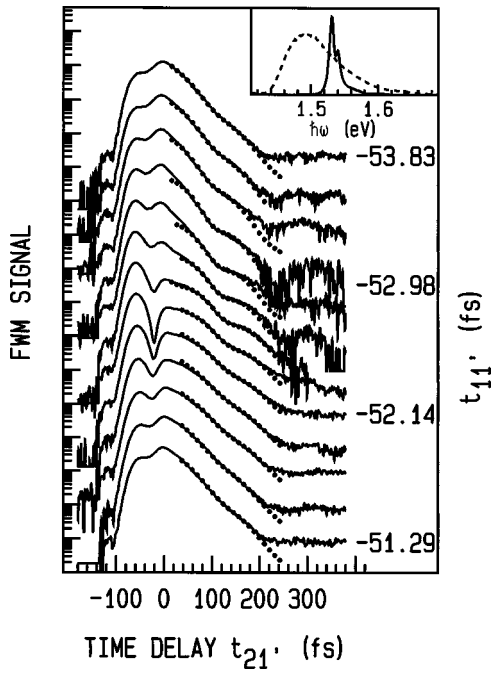

FIG. 3. Same as Fig. 2, however, for a 37 times higher excitation density. The dots are the result of a simple fit for the phonon oscillations. The inset shows the two-pulse FWM spectrum at zero time delay and the laser spectrum.

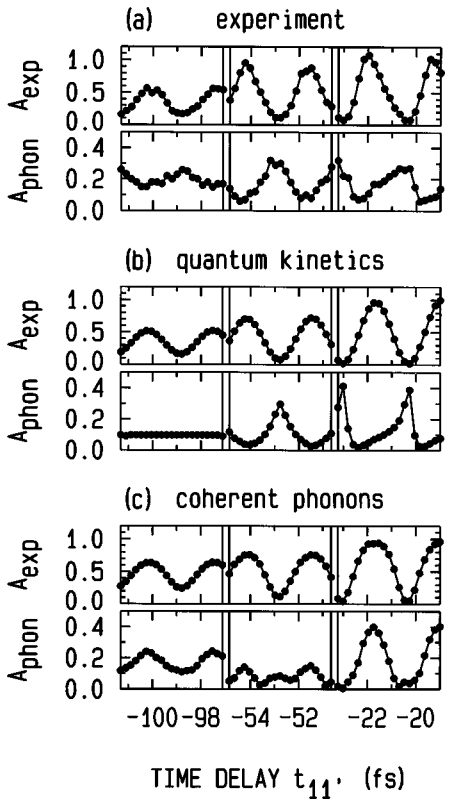

FIG. 4. The signal strength A_{exp} and the visibility of the phonon oscillations A_{phon} versus $t_{11'}$. Experiment (a): these parameters are extracted from fits to the data as shown in Fig. 3. Notice that A_{exp} and A_{phon} oscillate *out of phase* in the *quantum kinetic scenario* (b) in agreement with experiment, while they would oscillate *in phase* for the *coherent phonon scenario* (c).

of the TI-FWM traces to the formula $S_{\text{TI-FWM}} \propto A_{\mathrm{exp}}\{1 + A_{\mathrm{phon}}\cos(\omega_{\mathrm{osc}}[t_{21'} - t_0])\}\exp(-\gamma_{\mathrm{eff}} t_{21'})$, which we previously used for the bulk GaAs case.[9] This allows us to characterize the strength of the signal A_{exp}, the visibility of the phonon oscillations A_{phon}, the oscillation frequency ω_{osc}, and phase (through t_0). We obtain an oscillation period $2\pi/\omega_{\mathrm{osc}} = 115 \pm 20$ fs. The phonon visibility and the signal strength vs $t_{11'}$ are shown in Fig. 4(a) for three 4.2 fs time delay slots around $t_{11'} = -99$ fs, -53 fs, and -21 fs. The overall behavior is similar to that of bulk GaAs.[9] The phase of the phonon oscillation has an average value around $t_0 = 60$ fs while $t_0 = 25$ fs was found for the bulk GaAs.[9] This difference corresponds to a $\pi/2$ phase shift, which can already be guessed from Fig. 1. Numerical solutions of quantum kinetic theory in Ref. 19 also show a similar phase shift. The phonon visibility is comparable to that of the bulk. The theory, however, predicts in QW an oscillation amplitude lower approximately by a factor of 4 (Ref. 19) and, although there is a significant uncertainty on $2\pi/\omega_{\mathrm{osc}}$, the experimental value is not in agreement with the theoretical result of 80 fs. It should be noted, however, that the theory[19] assumes a very low carrier density, bulk phonons, and accounts for only one subband, whose valence-band structure is approximated by a single effective mass.

It is interesting to note in Fig. 4(a) that the strength of the TI-FWM signal, A_{exp}, and that of the phonon oscillations, A_{phon}, are *out of phase*. This signature allows us to assign the origin of the phonon oscillations to quantum correlations between electrons and phonons. These originate from a pure quantum kinetics process seen even if the phonons are in thermal equilibrium.[3,9] Nonequilibrium coherent phonons also can modulate electronic degrees of freedom,[10–13] even in the absence of correlations between electrons and phonons, but they would produce *in-phase* oscillations of A_{exp} and A_{phon}. This is explained in the following. Coherent phonons are observed in an optical experiment if a lattice displacement $x(t)$, generated by impulsive excitation, couples back to an electronic excitation. It is crucial to note that a lattice displacement cannot be generated efficiently by a laser field at $\hbar\omega \approx E_{\mathrm{gap}}$ in first-order processes. These are too far off-resonant, i.e., 1.5 eV $= \hbar\omega \gg \hbar\omega_{\mathrm{LO}} = 36$ meV. The lattice displacement can be generated by second-order processes, which require broken inversion symmetry. In QWS the inversion symmetry may be broken not only due to the symmetry of the parent material or surface electric fields but also due to asymmetric heterointerfaces, which can be interpreted as a built-in electric field. The photogenerated electron-hole pairs separate in this electric field, which they screen.[11] In that case the LO-phonons experience a steplike change of the electric field, $E_{\mathrm{LO}}(t)$, which drives the phonon displacement away from the equilibrium value $x = 0$ and lets it oscillate around a new stationary value, i.e., $E_{\mathrm{LO}}(t) \sim f(t)$, where f is the electron-hole pair density. The lattice displacement thus obeys the equation

$$\ddot{x} + \omega_{\mathrm{LO}}^2 x = \mathcal{C} f, \tag{1}$$

with a constant $\mathcal{C}$. This coherent oscillation of the lattice can couple back to the electronic excitations by modulating the electronic eigenfrequency, which becomes time dependent: $\Omega(t) = \Omega_0 + g x(t)$; here Ω_0 is the undisturbed eigenfrequency and g the electron-phonon coupling constant. Then the interband polarization, p and the pair density follow Bloch-equations of the form

$$\dot{p} + i\Omega(t)p + \gamma_2 p = i\hbar^{-1} dE, \quad \dot{f} = -2\,\mathrm{Im}(dEp^*), \tag{2}$$

where γ_2 is the constant dephasing rate, d is the dipole matrix element, and E is the sum of the laser fields. Pauli blocking has been neglected, which is the most favorable case for the coherent phonon scenario. Equation (2) can in fact easily be derived from the Hamiltonian discussed in Ref. 9 by assuming that phonons and electrons are uncorrelated. Note, however, that the conclusions of our discussion do not depend on the particular way x couples back to p. After impulsive excitation, coherent phonons induce an oscillation of $\Omega(t)$ at the phonon frequency with some amplitude. In the low density ($\chi^{(3)}$) limit, $S_{\text{TI-FWM}}$ shows no oscillations as the time delay varies. However, the FWM signal would exhibit oscillations in real time and, thus phonon satellites in the FWM spectrum, which are not observed experimentally here. For higher excitation density (beyond the $\chi^{(3)}$ limit), coherent phonon oscillations appear in the delay time domain also. In that case, as $t_{11'}$ varies, the excitation density is modulated by constructive or destructive interference and drives *in phase* the signal strength A_{exp} and the phonon visibility A_{phon}. As shown in Fig. 4(c), this qualitative discussion is confirmed by numerical calculations of $S_{\text{TI-FWM}} \sim \int_{-\infty}^{+\infty} |p_{2_{q_2 - q_1}}(t)|^2\, dt$, with parameters that correspond to

the experiment, $\gamma_2=(80\ \mathrm{fs})^{-1}$, $g=0.3\omega_{\mathrm{LO}}$, and each pulse corresponding to $g\mathcal{C}\Theta^2=10^{-4}/\mathrm{fs}^3$, where Θ is the pulse area. This behavior is contrasted by the results obtained with the quantum kinetics model of Ref. 9. This model (i) lumps together the conduction-band and valence-band states to form a two-level system, and (ii) retains in the electron-phonon interaction only the scattering terms in and out of the upper level. The results are shown in Fig. 4(b), they give an *out of phase* oscillation of A_{exp} and A_{phon}, both for low and high excitation in excellent agreement with the experiments.

In conclusion, we have demonstrated that it is possible by coherent control to isolate one type of scattering in the interaction of a subsystem with different reservoirs. Using this technique, electron–LO-phonon quantum correlations in a two-dimensional semiconductor are investigated. These correlations are unambiguously distinguished from scattering with coherent nonequilibrium phonons by the phase of the phonon oscillation amplitude relative to the FWM signal strength. This analysis is also applicable to other systems. The quantum correlations observed by us exhibit several specific features that are significantly different from those seen in the three-dimensional parent system.

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