Coherent Control of Electron-LO-Phonon Scattering in Bulk GaAs

M. U. Wehner, M. H. Ulm, D. S. Chemla,* and M. Wegener
Institut für Angewandte Physik, Universität Karlsruhe, Kaiserstraße 12, D-76128 Karlsruhe, Germany
(Received 7 October 1997)

Several tens of femtoseconds after optical excitation, scattering processes in semiconductors are not completed, thus, relaxation has not yet become irreversible. This opens the fascinating possibility of actually reversing or enhancing incompleted scattering events. Here, we use attosecond time scale coherent control to suppress or amplify the non-Markovian phonon oscillations in the model semiconductor GaAs. [S0031-9007(98)05370-8]

PACS numbers: 78.47.+p, 42.50.Md, 42.65.Re, 63.20.Kr

The evolution of the wave function of a crystal, prepared in a known state at earlier times, is completely determined by its state at a time t via its time-dependent Schrödinger equation. For complicated many-body problems, it is necessary to restrict the treatment to only certain degrees of freedom, for example, the electrons in a conduction and a valence band of a semiconductor. In this approach, we have to consider that this subsystem interacts with other degrees of freedom, e.g., the vibrations of the nuclei, on which we have only limited information and control and which are considered as a thermal bath. Then the equation of motion of the subsystem becomes nonlocal in time, i.e., the evolution at time t not only depends on the state of the subsystem at that time, but also on its history. In this time regime, the dynamics must be described in the framework of the so-called quantum kinetics theory [1], because a number of approximations on which the usual Boltzmann description of relaxation is based break down. This provides us with the unique opportunity to explore the physics of electronic excitations in condensed matter for conditions thus far inaccessible. For example, since scattering processes among elementary excitations are not instantaneous in time, for short times they have not yet become irreversible and a fascinating question arises: Is it possible to *reverse or enhance*, after it has already started, an interaction process that would become irreversible if the subsystem was left to itself?

In order to introduce the main concepts, it is useful to consider the analogy with the electron-photon interaction. When a short optical pulse excites a semiconductor at $t = 0$, a coherent oscillation of the interband transition amplitude is triggered [2]. As long as the quantum mechanical phase of the associated polarization has not been destroyed, the "absorption" process is not completed. Thus, an interference with a second time-delayed and phase-locked pulse can reverse (destructive interference) or enhance (constructive interference) the uncompleted absorption process. This technique, called coherent control of the electron (or exciton) density, has indeed been used [3]. A variety of related coherent control experiments has also been performed on semiconductors [4], metal surfaces [5], molecular systems [6], and atomic systems [7]. In the quantum kinetics regime, one does not only expect a coherent oscillation of the transition amplitude, but also a coherent oscillation of the scattering amplitude containing the thermal bath degrees of freedom. Its decay determines the memory time of the subsystem τ_m. For times $t < \tau_m$, it should be possible to adjust the phase of the two components of the scattering amplitude, generated by the two phase-locked pulses, such that they interfere destructively (constructively). This would reverse (enhance) a scattering process that has already started after the first pulse. In this Letter, we demonstrate experimentally for the first time that such a coherent control of scattering processes is feasible.

We consider the model system of electron-LO-phonon scattering in the polar semiconductor GaAs [8,9]. The subsystem, electrons in the conduction and valence band, couples to the LO-phonon bath. This situation presents the advantage of having a bath with a single characteristic frequency ω_{LO}. For this case, it has been shown that the decay of the polarization is non-Markovian, which leads to oscillations with a renormalized LO-phonon frequency [8]. For other scattering mechanisms, such as electron-electron scattering, the situation is more complicated [10,11]. In our experiment (Fig. 1), two phase-locked pulses 1 and $1'$, derived from an apparatus described in Ref. [12], are separated by the time delay $t_{11'} = t_1 - t_{1'}$, which is kept constant within less than 100 attoseconds, and propagate collinearly in the direction $\vec{q}_1$. They interact with another pulse propagating along $\vec{q}_2$ with time delay $t_{21'} = t_2 - t_{1'}$ in a GaAs sample (described in Ref. [8]). This produces two nonequivalent diffracted four-wave mixing (FWM) signals with wave vectors $2\vec{q}_2 - \vec{q}_1$ and $2\vec{q}_1 - \vec{q}_2$. Here, we focus our experiments on the emission in the direction $2\vec{q}_2 - \vec{q}_1$, for which, in the $\chi^{(3)}$ regime, the phase-locked pulses enter linearly. All beams are linearly polarized in the plane of incidence, and have approximately the same intensity. The intensity of the diffracted signal observed in the GaAs sample at $T = 77$ K with 15 fs pulses, the central frequency of which is tuned to the band gap, is depicted in Fig. 2. The excitation conditions in the frequency domain are shown in the inset. The excitation densities are comparable to two-beam FWM data published previously [8].

 0031-9007/98/80(9)/1992(4)$15.00

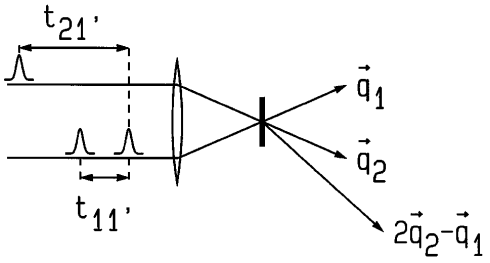

FIG. 1. Schematic of the coherent control experiment. The pulses 1 and 1′ are phase locked. The diffracted beam with wave vector $2\vec{q}_2 - \vec{q}_1$ is detected.

As seen in Fig. 2, when the delay between the phase-locked pulses is varied, we observe several remarkable features: (i) Around certain values, such as $t_{11'} = -43.69$ or -41.16 fs, the non-Markovian phonon oscillations seen in the time delay domain $t_{21'}$ [8] can almost disappear. (ii) Pronounced oscillations occur at low absolute signal levels, see $t_{11'} = -42.43$ fs trace, with a more pronounced fast initial decay. (iii) A more usual time profile with a decay superimposed by oscillations can be seen, for example, around $t_{11'} = -43.06$ and -41.79 fs. We have, of course, systematically investigated the full parameter space of time delays and excitation densities.

To analyze these data, we fit the experimental FWM signal by the expression $S_{\mathrm{FWM}} \propto A_{\mathrm{exp}}\{1 + A_{\mathrm{phon}} \cos[\omega_{\mathrm{osc}}(t_{21'} - t_0)]\} \exp(-\gamma_{\mathrm{eff}} t_{21'})$. It characterizes directly the visibility of the phonon oscillations A_{phon}, and was used previously to analyze our two-beam FWM experiments [8] with the result $A_{\mathrm{phon}} = 0.15$ to 0.2. We fix $\omega_{\mathrm{osc}} = 2\pi/95\ \mathrm{fs}^{-1}$ consistent with Ref. [8]. Here, again, the fits are very good, as can be seen from the dots in Fig. 2. This procedure provides us with the $t_{11'}$ dependence of the fit parameters presented in Fig. 3 around $t_{11'} \approx 40$ fs. The exponential amplitude A_{exp} oscillates almost harmonically while the other parameters A_{phon}, t_0, and γ_{eff} have a more complicated behavior. Let us note that the variation of t_0 is seen directly in Fig. 2 (see dashed straight lines). Figure 4 summarizes the $t_{11'}$ variation of these parameters for various time delays at the same excitation density. For $t_{11'} < 15$ fs, the laser pulses still overlap (not shown); for $t_{11'} \approx 20$ fs, we see a modulation of A_{phon}, which essentially disappears for $t_{11'} \approx 80$ fs, while the modulation of A_{exp} has decreased approximately by a factor of 2. Finally, we note that this overall behavior is essentially density independent in the range from 8×10^{14} cm^{-3} to 3.6×10^{16} cm^{-3} (incoherent sums).

The experimental results demonstrate clearly that we can coherently control the phonon oscillation visibility. We show now that this observation indeed corresponds to the coherent control of the scattering process. In our discussion, we neglect the rapidly decaying contribution around $t_{21'} = 0$, which is strongly influenced by mechanisms other than phonon scattering, essentially the Coulomb coupling between exciton and continuum states [13,14]. We base our discussion on a phenomenological and very simple

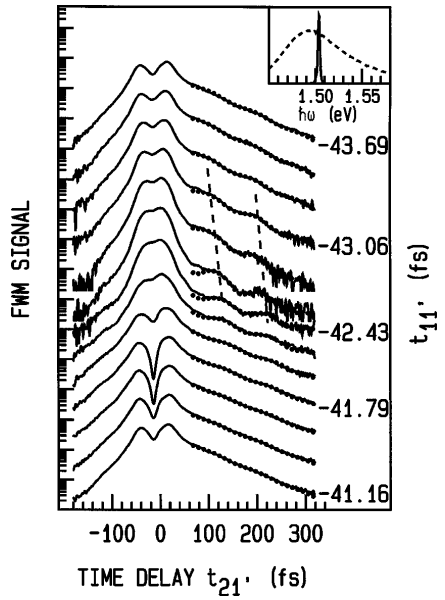

FIG. 2. Four-wave mixing signal as a function of the time delay $t_{21'}$ for various fixed time delays $t_{11'}$ in steps of 0.21 fs. The curves are vertically displaced for clarity. GaAs, $T = 77$ K, the excited carrier density (incoherent sum) is 3.6×10^{15} cm^{-3}. The dots are fits from which the parameters shown in Fig. 3 have been extracted. The inset shows the laser spectrum (dashed line) together with the two-beam FWM spectrum at zero time delay.

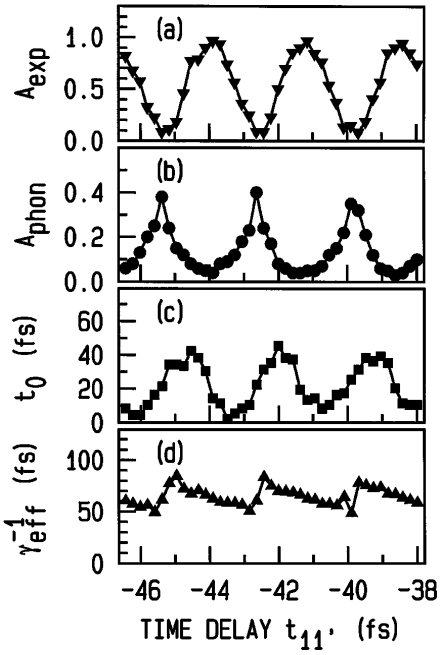

FIG. 3. The amplitude of the exponential A_{exp} (a), the visibility of the phonon oscillations A_{phon} (b), the phase shift t_0 (c), and the effective damping γ_{eff} (d) versus $t_{11'}$. These parameters are extracted from fits to the data as shown in Fig. 2. The excited carrier density is 3.6×10^{15} cm^{-3} (incoherent sum), GaAs, $T = 77$ K.

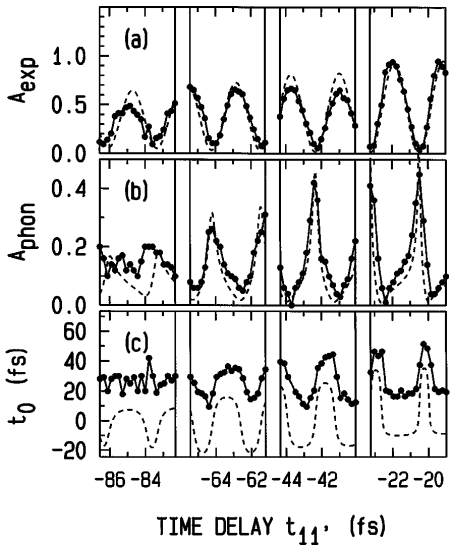

FIG. 4. As Fig. 3, however, for different time delay regimes. Notice the decay of the modulation in A_{phon}. The dashed lines are the result of the model discussed in the text.

model Hamiltonian that allows us to follow the arguments quite easily, but conveys the same physics as more thorough theoretical treatments [8,15]. It is justified by its consistency with the microscopic theories and by the agreement with experiment.

$$H = \hbar\Omega c_1^\dagger c_1 - dE(c_1^\dagger c_0 + c_0^\dagger c_1) + \hbar\omega_{\text{LO}} a^\dagger a + \hbar g(a^\dagger c_1^\dagger c_1 + a c_1^\dagger c_1). \quad (1)$$

The first line describes a fermionic two-level system with a ground state 0 at zero energy and an excited state 1 at energy $\hbar\Omega$, coupled to the light field E via the dipole matrix element d. This constitutes the optical Bloch equations. The terms of the second line describe the Fröhlich coupling to a single LO-phonon mode with energy $\hbar\omega_{\text{LO}}$ [16], with coupling constant g. The zero point energy of the phonon mode has been subtracted. Only scattering from the upper electronic level back into the upper level via emission or absorption of phonons is included.

These processes obviously violate energy conservation for the individual scattering event. Thus, they would not contribute in Fermi's "golden rule" or the Boltzmann equation; they are, however, especially important in the quantum kinetics regime. It is straightforward to compute the Heisenberg equations of motion of the observables. We use the rotating wave approximation, factorize the four-point functions into products of two-point functions within the second Born approximation, and assume thermal equilibrium of the phonons at $T = 0$. For a complete treatment, we have numerically solved the resulting equations of motion for optical pulses of finite duration. This will be discussed below. For an intuitive understanding, however, it is instructive to first discuss contributions linear in the electric field in the short pulse limit, where we can obtain simple analytical results. This is meaningful because the phase-locked pulses enter linearly into the diffracted signal in direction $2\vec{q}_2 - \vec{q}_1$ (in the $\chi^{(3)}$ regime). For the transition amplitude $p = \langle c_0^\dagger c_1\rangle = \tilde{p}\exp(-i\Omega t)$ and the scattering amplitude $s = i\langle a c_0^\dagger c_1\rangle = \tilde{s}\exp(-i\Omega t)$, we have $\partial\tilde{p}/\partial t + g\tilde{s} = i\hbar^{-1}d\tilde{E}$, and $\partial\tilde{s}/\partial t + i\omega_{\text{LO}}\tilde{s} = g\tilde{p}$, with $E = \tilde{E}\exp(-i\Omega t)$. Note that the scattering amplitude $\tilde{s}$ is driven by the polarization $\tilde{p}$ and, thus, it needs time to build up—a characteristic feature of quantum kinetics. Formal integration of $\tilde{s}(t)$ introduced into the equation of $\tilde{p}(t)$ gives

$$\frac{\partial}{\partial t}\tilde{p}(t) + g^2\int_{-\infty}^{t} e^{-i\omega_{\text{LO}}(t-t')}\tilde{p}(t')\,dt' = i\hbar^{-1}d\tilde{E}. \quad (2)$$

The solution for a δ function shaped right-hand side is the Green's function $G = \tilde{G}\exp(-i\Omega t)$:

$$G(t) = \frac{\Theta(t)}{1+2x}\,e^{-i\Omega t}[(1+x)e^{+i\omega_{\text{LO}}t} + xe^{-i(1+x)\omega_{\text{LO}}t}], \quad (3)$$

where $\Theta(t)$ is the Heaviside step function and $(1 + 2x) = \sqrt{1 + 4g^2/\omega_{\text{LO}}^2}$. Equation (3) shows that the polarization exhibits beats with the renormalized phonon frequency $(1 + 2x)\omega_{\text{LO}}$. For two phase-locked pulses, the solution is a linear superposition of two contributions delayed by $t_{11'}$. We can either adjust $t_{11'}$ for destructive interference with respect to the frequency $(\Omega - x\omega_{\text{LO}})$ or to its sideband $[\Omega + (1 + x)\omega_{\text{LO}}]$; destructive interference for the latter suppresses the phonon oscillations. Close to destructive interference for $(\Omega - x\omega_{\text{LO}})$, the amplitudes of the two contributions become comparable and the beat's visibility increases while the overall signal is small. The linear response for two phase-locked pulses can easily be put in the same form used for fitting the experimental data. The corresponding curves are shown as dashed lines in Fig. 4. Here, phenomenological damping has been added in the form $G \to G\exp(-\gamma t)$. Parameters are $x = 0.04$, $\hbar\omega_{\text{LO}} = 36$ meV, $\hbar\Omega = 1515$ meV, and $\gamma = \frac{1}{140}$ fs^{-1}. The agreement with the experiment is amazing. Note, e.g., in Fig. 4, the change of the $t_{11'}$ dependence of A_{phon} from an asymmetric profile at $t_{11'} \approx 20$ fs to a more symmetric one at $t_{11'} \approx 40$ fs and, finally, to a reversed asymmetry for $t_{11'} \approx 60$ fs. A related scenario is seen for the $t_{11'}$ dependence of t_0. Notice, also, that A_{exp} and A_{phon} are always out of phase, which is consistent with the above reasoning and with the experiment.

Turning now to the full numerical solution of the equations of motion for $\langle c_0^\dagger c_1\rangle$, $\langle c_1^\dagger c_1\rangle$, and the phonon assisted density matrix $\langle a c_i^\dagger c_k\rangle$, the time integrated FWM signal is extracted by a spatial Fourier expansion [17]. Again, the phenomenological damping rate γ for $\langle c_0^\dagger c_1\rangle$ and $\langle a c_i^\dagger c_k\rangle$ is added. The numerical results for 15 fs sech^2 pulses with a pulse area of $10^{-4}\pi$ for each pulse are shown in Fig. 5 together with the linear response as discussed above. The experimental findings are well

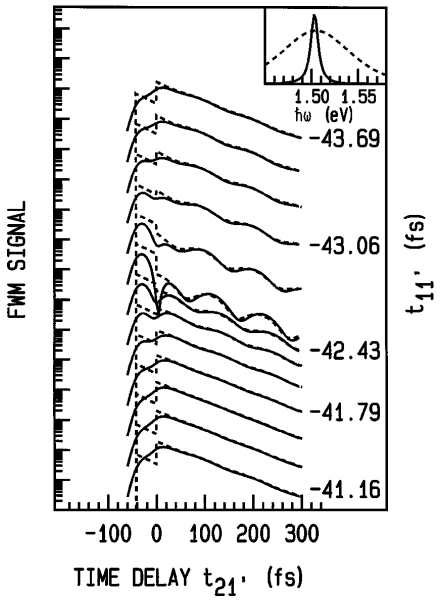

FIG. 5. Four-wave mixing signal (solid lines) as computed from the model Hamiltonian for parameters corresponding to the experiment. The dashed lines are the linear response. This result can be compared directly with the experiment (Fig. 2).

reproduced (compare Figs. 2 and 5), which confirms our above reasoning.

Thus, in summary, we have demonstrated for the first time, in the model case of electron-LO-phonon scattering in GaAs, that an interaction process can be manipulated even after it has already started.

We gratefully acknowledge support by the Krupp-Stiftung and by the DFG-Schwerpunktprogramm *Quantenkohärenz in Halbleitern.* The stay of D. S. C. has become possible due to funding by the Humboldt-foundation and the DFG-Graduiertenkolleg *Kollektive Phänomene im Festkörper.* We thank W. Stolz for the high quality sample and acknowledge stimulating discussions with H. Haug and L. Bányai.

*Permanent address: Material Sciences Division of Lawrence Berkeley Laboratory and Physics Department of the University of California, Berkeley, CA 94720.

[1] H. Haug and J.-P. Jauho, in *Quantum Kinetics in Transport and Optics of Semiconductors,* Springer Series in Solid-State Sciences Vol. 123 (Springer-Verlag, Berlin, 1996), and references therein.

[2] K. Leo, M. Wegener, J. Shah, D. S. Chemla, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. Lett. **65**, 1340 (1990); H. Wang, K. B. Ferrio, D. G. Steel, Y. Z. Hu, R. Binder, and S. W. Koch, Phys. Rev. Lett. **71**, 1261 (1993); M. Joschko, M. Woerner, T. Elsaesser, E. Binder, T. Kuhn, R. Hey, H. Kostial, and K. Ploog, Phys. Rev. Lett. **78**, 737 (1997).

[3] A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. **75**, 2598 (1995).

[4] A. Haché, Y. Kostoulas, R. Atanasov, J. L. P. Hughes, J. E. Sipe, and H. M. van Driel, Phys. Rev. Lett. **78**, 306 (1997); M. Gurioli, F. Bogani, S. Ceccherini, and M. Colocci, Phys. Rev. Lett. **78**, 3205 (1997).

[5] S. Ogawa, H. Nagano, H. Petek, and A. P. Heberle, Phys. Rev. Lett. **78**, 1339 (1997).

[6] M. S. Pshenichnikov, W. P. de Boeij, and D. A. Wiersma, Phys. Rev. Lett. **76**, 4701 (1996).

[7] V. Blanchet, C. Nicole, M.-A. Bouchene, and B. Girard, Phys. Rev. Lett. **78**, 2716 (1997).

[8] L. Bányai, D. B. Tran Thoai, E. Reitsamer, H. Haug, D. Steinbach, M. U. Wehner, M. Wegener, T. Marschner, and W. Stolz, Phys. Rev. Lett. **75**, 2188 (1995).

[9] C. Fürst, A. Leitenstorfer, A. Laubereau, and R. Zimmermann, Phys. Rev. Lett. **78**, 3733 (1997).

[10] S. Bar-Ad, P. Kner, M. V. Marquezini, D. S. Chemla, and K. El Sayed, Phys. Rev. Lett. **77**, 3177 (1996).

[11] F. X. Camescasse, A. Alexandrou, D. Hulin, L. Bányai, D. B. Tran Thoai, and H. Haug, Phys. Rev. Lett. **77**, 5429 (1996).

[12] M. U. Wehner, M. H. Ulm, and M. Wegener, Opt. Lett. **22**, 1455 (1997).

[13] M. U. Wehner, D. Steinbach, and M. Wegener, Phys. Rev. B **54**, R5211 (1996).

[14] S. T. Cundiff, M. Koch, W. H. Knox, and J. Shah, Phys. Rev. Lett. **77**, 1107 (1996).

[15] D. B. Tran Thoai and H. Haug, Phys. Rev. B **47**, 3574 (1993).

[16] C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1987), 2nd ed.

[17] M. Lindberg, R. Binder, and S. W. Koch, Phys. Rev. A **45**, 1865 (1992).