Band-edge quantum kinetics for coherent ultrashort-pulse spectroscopy in polar semiconductors

D. B. Tran Thoai* and H. Haug
Institut für Theoretische Physik, J.W. Goethe Universität Frankfurt, Robert-Mayer-Strasse 8, D-6000 Frankfurt, Germany
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A recently developed quantum kinetic description for the electrons in a two-band polar semiconductor coupled to LO phonons and excited by a femtosecond laser pulse is extended to include excitonic effects. Our numerical treatment yields a description of the relaxation of the electron-hole pairs which is valid on a time scale where the semiclassical Boltzmann description is no longer applicable. Oscillations with the period of the LO lattice oscillation are superimposed on the resulting polarization decay under excitation close to the band edge. The polarization oscillations are quantum beats between the direct interband transitions and their LO-phonon sidebands. The non-Markovian quantum kinetics is shown to yield, in contrast to the corresponding nonretarded kinetics for weak, nonresonant, stationary excitation, an Urbach absorption tail which is approximately exponential over several orders of absorption as universally observed in polar semiconductors.

I. INTRODUCTION

Both in transport theory[1-5] for semiconductor microstructures with their high electric fields and in theory of semiconductor ultrashort-pulse spectroscopy,[6-8] one needs a kinetic theory beyond the traditional Boltzmann kinetics with energy conservation for each of its successive, completed collisions. Quantum kinetic equations of rather similar structure have been derived with reduced density matrices[1,3,6] and with nonequilibrium Green's functions.[2,4,5] Common to all these quantum kinetic equations is that the energy conservation no longer holds strictly, but that the system has on short times scales a memory of its earlier states. In optics, Zimmermann[6] presented early investigations of retarded reduced density-matrix equations for electron-phonon interaction, while our studies of the Urbach tail absorption led one of the authors to introduce phenomenologically similar retardations for the interband polarization[8] in connection with the theory of the nonresonant optical Stark effect. In three preceding publications,[9-11] the authors *et al.* gave a derivation and a numerical analysis of the quantum kinetic equations for a two-band model with a coherent laser pulse and interactions of the carriers with LO phonons using nonequilibrium Green's-function theory. Similar equations have been derived by Kuznetsov.[7] The memory kernels of these equations are determined by retarded and advanced Green's functions. It is important that these Green's functions are also determined by the considered scattering processes. Only a consistent treatment of these functions results in numerically stable kinetic equations (i.e., the time-dependent electron and hole densities never exceed the interval between 0 and 1). Particularly, one has to take into account that the damping of the retarded and advanced functions are determined by the strength of the phonon scattering. This important fact has not been recognized, e.g., in the treatment of Zimmermann.[6] The obtained numerical solutions are very similar to solutions of Hartmann and Schäfer[12] obtained by direct but rather involved numerical evaluations of the two-point particle-propagator equations, showing that generalized Kadanoff-Baym ansatz[2] employed by us to relate two-point propagators to their equal-time limits is good for small and intermediate polaron coupling constants, as expected. They also found that the quantum kinetic theory yields an exponential Urbach absorption tail in the low-intensity limit.

II. INTERBAND QUANTUM KINETIC EQUATIONS WITH EXCITONIC EFFECTS

Because excitonic effects influence to a large extent the band-edge optical spectra of semiconductors,[13] we will extend our quantum kinetic equations[9,10] by taking the Coulomb interaction in the Hartree-Fock approximations into account. A similar combination of quasiclassical kinetic equations for LO-phonon scattering with Coulomb Hartree-Fock terms has been given by Kuhn and Rossi.[14] Naturally, the Hartree-Fock terms give only a simple mean-field approximation of the carrier interactions but omit any Coulomb scattering. We leave the considerably more involved numerical treatment of the quantum kinetics of Coulomb scattering[5] to subsequent investigations.

In the following, we use the notations for the densities of electrons and holes (e,h) in the conduction and valence bands (c,v):

$$f_{kcc}(t)=f_{ke}(t) \quad \text{and} \quad f_{kvv}(t)=1-f_{kh}(t) , \tag{1}$$

and for the interband polarization,

$$\begin{aligned} &f_{kcv}(t)=\langle a_{kv}^{\dagger}(t)a_{kc}(t)\rangle=p_k(t) , \\ &P_k(t)=p_k(t)e^{i\omega t} . \end{aligned} \tag{2}$$

In the rotating-wave approximation, the quantum kinetic equation for the polarization is

$$\left[\frac{\partial}{\partial t}+i\delta_k+i\Sigma_k(t)\right]P_k(t)=i\left[\frac{dE_0(t)}{2}+\sum_q V_q P_{k-q}(t)\right][1-f_{ke}(t)-f_{kh}(t)]-e^{i\omega t}\frac{\partial}{\partial t}p_k(t)\Big|_{\text{phon}}, \tag{3}$$

with the detuning $\delta_k=e_{ke}+e_{kh}-\omega$, where ω is the carrier frequency of the coherent laser pulse with an amplitude $E_0(t)$. The term $\sum V_q P_{k-q}$ describes the action of the attractive e-h Coulomb potential $V_q=4\pi e^2/q^2V$ on the pair function P formulated in k space. The corresponding e-h pair Hartree-Fock self-energy is given by

$$\Sigma_k(t)=-\sum_{q,i} V_q f_{k-q,i}(t) \quad \text{with } i=e,h \ . \tag{4}$$

For simplicity, the vector notations for the momenta are not given explicitly.

The corresponding quantum kinetic equation for the densities is

$$\frac{\partial}{\partial t}f_{ki}(t)=\text{Im}\left[\left[dE_0(t)+2\sum_q V_q P^*_{k-q}(t)\right]P_k(t)\right]-\frac{\partial}{\partial t}f_{ki}(t)\Big|_{\text{phon}}-\frac{\partial}{\partial t}f_{ki}(t)\Big|_{\text{pol}} . \tag{5}$$

The scattering terms in Eqs. (3) and (5) are given by

$$-\frac{\partial}{\partial t}p_k(t)\Big|_{\text{phon}}=\sum_{q,\pm} g_q^2\int_{t_0}^{t}dt'(p_k(t')\{e^{[i(\pm\omega_0-e_{k-q,e}-e_{kh})-\gamma_e-\gamma_h](t-t')}[(n_q+\tfrac{1}{2}\mp\tfrac{1}{2})\pm f_{k-q,e}(t')]$$
$$+e^{[i(\pm\omega_0-e_{ke}-e_{k-q,h})-\gamma_e-\gamma_h](t-t')}[(n_q+\tfrac{1}{2}\mp\tfrac{1}{2})\pm f_{k-q,h}(t')]\}-(k\rightleftarrows k-q)) \tag{6}$$

and

$$-\frac{\partial}{\partial t}f_{ki}(t)\Big|_{\text{phon}}=2\sum_q g_q^2\int_{t_0}^{t}dt'\,e^{-2\gamma_i(t-t')}(\cos[(e_{ki}-e_{k-q,i}+\omega_0)(t-t')]$$
$$\times\{n_q[f_{k-q,i}(t')-f_{ki}(t')]+f_{k-q,i}(t')[1-f_{ki}(t')]\}-(k\rightleftarrows k-q)) \ , \tag{7}$$

$$-\frac{\partial}{\partial t}f_{ki}(t)\Big|_{\text{pol}}=-2\sum_{q,\pm} g_q^2\int_{t_0}^{t}dt'e^{-2\gamma_i(t-t')}\text{Re}\{\pm e^{i(e_{ki}-e_{k-q,i}\pm\omega_0)(t-t')}P^*_k(t')P_{k-q}(t')\} \ . \tag{8}$$

The interaction matrix element of the Fröhlich coupling between the electrons and the longitudinal optical (LO) phonons with frequency ω_0 can be written as

$$g_q^2=\frac{\omega_0 V_q}{2\hbar}\left[\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right] . \tag{9}$$

ϵ_0 and ϵ_∞ are the dielectric constants in the static and high-frequency limit, respectively. It is convenient to express the interaction matrix element in terms of the dimensionless polaron constant α:[13]

$$g_q^2=\alpha\frac{4\pi\hbar(\hbar\omega_0)^{3/2}}{(2m_r)^{1/2}q^2V}$$

with

$$\alpha=\frac{e^2}{\hbar}\left[\frac{m_r}{2\hbar\omega_0}\right]^{1/2}\left[\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right] , \tag{10}$$

where m_r is the reduced e-h mass. The kinetics of the LO phonons should in principle be investigated simultaneously with that of the e-h pairs. Here, we take the phonons simply as a thermal bath with $n_q=(e^{\beta\hbar\omega_0}-1)^{-1}$. It has been pointed out in Ref. 9 that the imaginary part of the retarded e-h self-energies which determine γ_e and γ_h has to be calculated in a self-consistent way with the considered scattering process. We achieve a simple but qualitatively correct self-consistency by choosing

$$\gamma_i\simeq\alpha\hbar\omega_0 \ . \tag{11}$$

As shown in Ref. 9, the above formulation contains phonon-assisted sidebands, as can be seen, e.g., by an iterative solution of the polarization equation. The nondiagonal retarded and advanced Green's function $G_{cv}^{r,a}$ which also contribute to the sidebands have been neglected in the above quantum kinetic equations. Their contributions will enhance even further the phonon quantum beats derived below.

For comparison, we will also give the same collision terms in the nonretarded completed-collision approximation, where we take only the resonant terms (with $\Gamma=\gamma_e+\gamma_h$)

$$-\frac{\partial}{\partial t}p_k(t)\bigg|_{\text{phon}} = -\sum_{q,\pm} g_q^2 p_k(t)\left\{\frac{\Gamma + i(\pm\omega_0 + e_{ke} - e_{k-q,e})}{(\pm\omega_0 + e_{ke} - e_{k-q,e})^2 + \Gamma^2}\{(n_q + \tfrac{1}{2} \mp \tfrac{1}{2}) \pm f_{k-q,e}(t')\}\right.$$
$$\left. + \frac{\Gamma + i(\pm\omega_0 - e_{k-q,h} + e_{kh})}{(\pm\omega_0 - e_{k-q,h} + e_{kh})^2 + \Gamma^2}\{(n_q + \tfrac{1}{2} \mp \tfrac{1}{2}) \pm f_{k-q,h}(t')\}\right\}, \tag{12}$$

$$-\frac{\partial}{\partial t}f_{ki}(t)\bigg|_{\text{phon}} = +\sum_q g_q^2\left\{\frac{4\gamma_i}{(e_{ki} - e_{k-q,i} + \omega_0)^2 + 4\gamma_i^2}\{n_q[f_{k-q,i}(t) - f_{ki}(t)] + f_{k-q,i}(t)[1 - f_{ki}(t)]\} - (k \rightleftarrows k-q)\right\}, \tag{13}$$

and

$$-\frac{\partial}{\partial t}f_{ki}(t)\bigg|_{\text{pol}} \simeq 0\ . \tag{14}$$

In order to check the conjecture of Ref. 8 that a consistent description of the Urbach tail needs a quantum kinetic description, we will also specialize our theory to the case of a stationary, weak laser field with $E(t) = (E_0/2)e^{-i\omega t}$, for which we can neglect all population effects, i.e. $f_{ki} = 0$. For the constant polarization P_k we get from the quantum kinetic equations (3) and (6) by evaluating the time integrals

$$i\delta_k P_k = i\left[\frac{dE_0}{2} + \sum_q \mathbf{V}_q P_{k-q}\right]$$
$$-\sum_{q,\pm} g_q^2\left\{P_k(n_q + \tfrac{1}{2} \mp \tfrac{1}{2})\left[\frac{\Gamma + i(\omega \pm \omega_0 - e_{k-q,e} - e_{kh})}{\Gamma^2 + (\omega \pm \omega_0 - e_{k-q,e} - e_{kh})^2} + \frac{\Gamma + i(\omega \pm \omega_0 - e_{k-q,h} - e_{ke})}{\Gamma^2 + (\omega \pm \omega_0 - e_{k-q,h} - e_{ke})^2}\right] - (k \rightleftarrows k-q)\right\}. \tag{15}$$

Starting from the equation in the completed collision approximation, we find in this case

$$i\delta_k P_k = i\left[\frac{dE_0}{2} + \sum_q V_q P_{k-q}\right] - \sum_{q,\pm} g_q^2 P_k(n_q + \tfrac{1}{2} \mp \tfrac{1}{2})\left\{\frac{\Gamma + i(\pm\omega_0 + e_{ke} - e_{k-q,e})}{\Gamma^2 + (\pm\omega_0 - e_{k-q,e} + e_{ke})^2} + \frac{\Gamma + i(\pm\omega_0 - e_{k-q,h} + e_{kh})}{\Gamma^2 + (\pm\omega_0 - e_{k-q,h} + e_{kh})^2}\right\}. \tag{16}$$

From the linear polarization we get the absorption spectrum $\alpha(\omega) \simeq \mathrm{Im}\sum_k P_k / E_0$.

III. NUMERICAL RESULTS AND DISCUSSION

A. LO-phonon quantum beats

Quantum beats are the clearest manifestation of optical coherence in the induced polarization, and can, e.g., be measured in self-defracting four-wave mixing experiments.[15] Quantum beats arise when there are transitions from one initial state to two different final states. Quantum beats in exciton systems have, e.g., been observed between the light- and heavy-hole excitons in quantum wells,[16] between free and bound exciton states,[17] between exciton states split in a magnetic field,[18] and between excitons from the lower and upper polariton branch.[19]

Here we propose that a very basic process, namely the coupling of the electrons and holes to the LO phonons, also gives rise to quantum beats. Actually, polarization oscillations with the period of a longitudinal-optical-phonon mode have been observed in transient reflectivity measurements.[20] The excitation of these modes in the experiment was not predominantly due to the intrinsic stimulated Raman-like process which is considered here, but due to a nonlinear process caused by the electron-hole charge separation in the field of surface charges. The quantum beats due to LO phonons in our model can be seen as the beating between direct and LO-phonon-assisted transitions. The same coupling naturally gives rise also to the relaxation of the excited pairs and to the polarization decay, so that this partially coherent process is a typical example for quantum kinetics. Calculations for free carriers coupled to LO phonons[6,10] proved that the polarization of these band-to-band transitions showed, under realistic conditions, practically no oscillatory behavior. We will demonstrate that the situation changes if excitonic effects which increase the pair coherence are taken into account.

We will present numerical results for bulk GaAs and for the slightly more polar InP. The following material parameters are used. GaAs: $m_e = 0.067m_0$, $m_h = 0.46m_0$, $\epsilon_0 = 13.1$, $\epsilon_\infty = 11.1$, $\hbar\omega_0 = 36$ meV, $\alpha = 0.064$; InP: $m_e = 0.08m_0$, $m_h = 0.6m_0$, $\epsilon_0 = 12.6$, $\epsilon_\infty = 9.6$, $\hbar\omega_0 = 42$ meV, $\alpha = 0.12$.

For the pulses, we assume a Gaussian amplitude variation $E_0(t) = E_0 e^{-t^2/\tau^2}$ with $\tau = 50$ fs so that the spectral width of the excited energy range is larger than the LO-phonon energy. The field strength will be given in terms of the Rabi frequency $\hbar\omega_R = dE_0$. The temperature of the phonons has been assumed to be $T = 300$ K.

The numerical calculations are greatly simplified by the fact[10] that the memory kernels which consist of exponential functions factorize so that the coupled set of re-

tarded differential equations for f_{ke}, f_{kh}, and P_k can be transformed into a larger set of ordinary first-order differential equations which are local in time.

We will present first results for resonantly excited GaAs with $\hbar\Delta\omega=\hbar\omega-E_g>0$. Figures 1(a)–1(c) show the resulting densities for the electrons and holes and the absolute value of the polarization $P_k(t)$, respectively, as functions of energy and time. The Rabi frequency is chosen to be $\hbar\omega_R=13.20$ meV, and a relatively large excess energy $\Delta\hbar\omega=60$ meV is assumed. In the electron density, the structures due to the emission and also to the absorption of one LO phonon are clearly visible. After about 600 fs, the densities approach thermal distributions. The absolute amount of polarization $|P(t)|$ shows a slight oscillation with the phonon period of 115 fs on top of the decay. For the same parameters, the total density $N=\sum_k f_{ki}$ and the incoherently summed polarization $P(t)=\sum_k|P_k(t)|$ (Ref. 14) are shown for various approximations in Fig. 2. The full and dashed lines give the results with memory and without (i.e., in the completed collision approximation). The polarization oscillations are naturally only present in the solution of the quantum kinetic equations with memory. Figure 2(a) also gives the results for the quantum kinetic free-carrier model by the short-dashed line. The excitonic effects increase the oscillator strength in the band-gap region so that they cause a higher density of excitations after the pulse.

Again for an excess energy of 60 meV, Fig. 3(a) gives the time and energy dependence of the absolute amount of the polarization $P_k(t)$ as Fig. 1(c), but compared to Fig. 1 the Rabi frequency is doubled to 26.4 meV. The quantum beats are now more pronounced than for the lower excitation of Fig. 1. In the incoherently summed polarization of Fig. 3(b), three oscillation periods can be seen.

For excitations deeper in the band the quantum beats

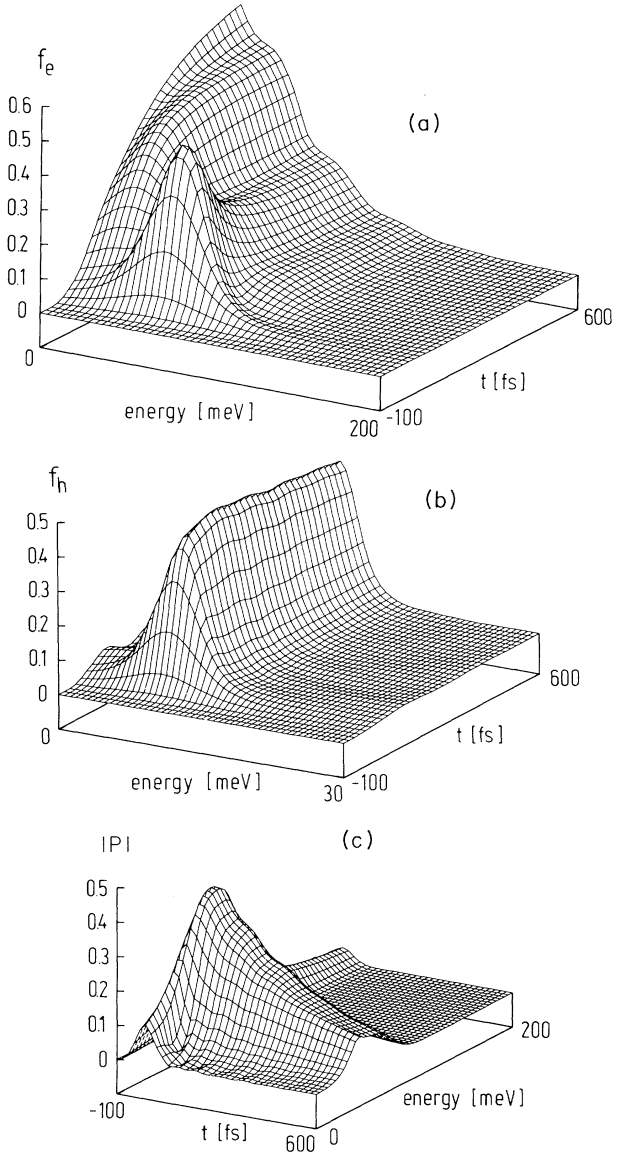

FIG. 1. Distributions (a) $f_{ke}(t)$ and (b) $f_{kh}(t)$ and absolute amount of the interband polarization (c) $|P_k(t)|$ vs energies e_{ke}, e_{kh}, and e_{ke}, respectively, and time for GaAs excited with a 50-fs pulse peaking at $t=0$ with 60-meV excess energy and a Rabi frequency of $dE_0=13.2$ meV.

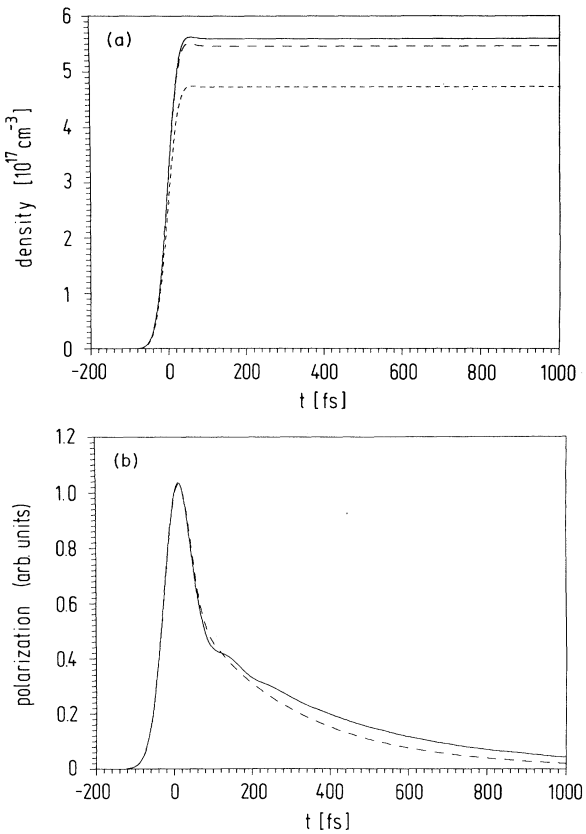

FIG. 2. (a) Total e-h density and incoherently summed polarization (b) $P(t)=\Sigma|P_k(t)|$ vs time for GaAs excited with a 50-fs pulse peaking at $t=0$ with 60-meV excess energy and a Rabi frequency of $dE_0=13.2$ meV. Full line: with memory; long-dashed line: without memory; short-dashed line: with memory but without excitonic effects.

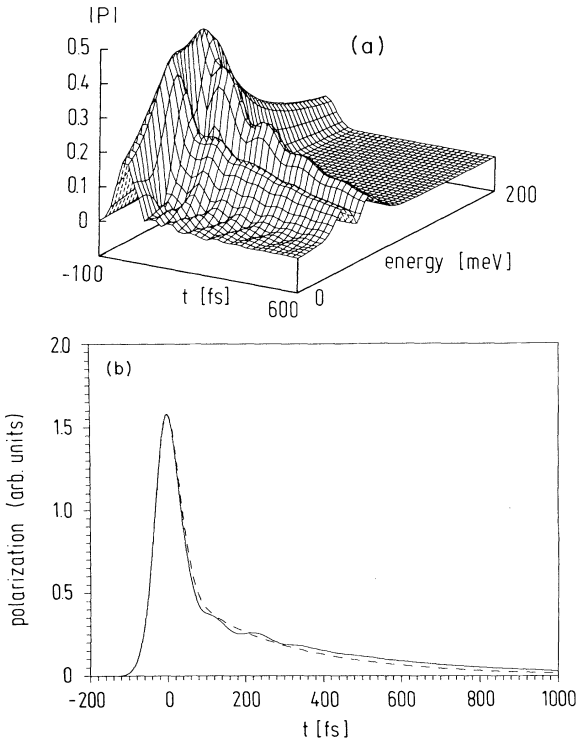

FIG. 3. Absolute amount of (a) the interband polarization $|P_k(t)|$ versus energy e_{ke} and time and (b) the incoherently summed polarization $P(t)=\Sigma|P_k(t)|$ vs time for GaAs excited with a 50-fs pulse peaking at $t=0$ with 60-meV excess energy and a Rabi frequency of $dE_0=26.4$ meV. Full line: with memory; long-dashed line: without memory.

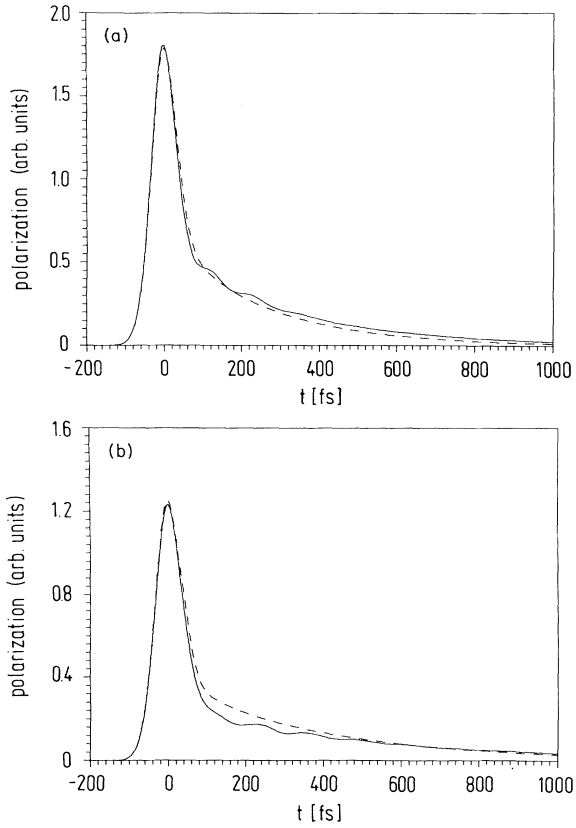

FIG. 4. Incoherently summed polarization $P(t)=\Sigma|P_k(t)|$ vs time for GaAs excited with a 50-fs pulse peaking at $t=0$ with an excess energy of (a) 100 and (b) 20 meV, and a Rabi frequency of $dE_0=26.4$ meV. Full line: with memory; long-dashed line: without memory.

become less pronounced, but their amplitudes increase for a still smaller excess energy. Figure 4(b) shows, for the same Rabi frequency as in Fig. 3, the incoherently summed polarization for the 20-meV excess energy. One can identify five oscillation periods.

Figure 5, as Fig. 1, shows the densities and polarization for a nonresonant excitation with $\hbar\Delta=-10$ meV and a Rabi frequency of 6.6 meV. For the electron distribution, one sees in Fig. 5(a) how, after a few hundred femtoseconds, a phonon-absorption process builds a population roughly one phonon energy above the band edge in a region which was not excited directly by the pulse. The polarization oscillations [Fig. 5(c)] are again visible, but for the considered conditions not very pronounced. Figure 6, as Fig. 2, again shows the corresponding total density and polarization in the various approximations. For the nonresonant excitation the differences between quantum kinetics and Markovian kinetics are more pronounced. In this exciton regime, the theory with Coulomb interaction (full curve) and without (short-dashed curve) naturally differ very strongly.

Next we consider the slightly more polar material InP for the same pulse excitations. Figure 7, obtained for a Rabi frequency of 26.4 meV, shows for three different excess energies of 60 and 20 meV the resulting oscillations in the incoherently summed polarization. One now obtains many deeply modulated oscillations, particularly for close to band-gap excitation [Fig. 7(b)]. For even more polar materials, the polarization decay becomes, with $\gamma=\alpha\omega_0$ increasingly fast so that the oscillations can only be observed for materials with α considerably smaller than 1.

For resonant excitation, the proposed LO-phonon quantum beats are expected to be smeared out by Coulomb scattering between the excited carriers. However, for nonresonant excitation conditions (i.e., with only virtual excitation of electron-hole pairs), one knows, e.g., from the optical Stark effect, that the influence of Coulomb scattering is negligible. Therefore, we expect that the proposed LO-phonon quantum beats can best be observed with nonresonant excitation.

B. Urbach tail absorption

Finally we will show that the description of the linear nonresonant absorption for stationary excitation needs the quantum kinetic equations with memory. Naturally, under stationary conditions, the time integrals can be evaluated explicitly as shown in Eq. (16). But the quantum kinetic equations and their Markovian approximation yield different frequency dependencies [compare Eqs.

(15) and (16)]. The corresponding absorption spectra are shown in Figs. 8(a) and 8(b) for GaAs with linear and logarithmic scales, respectively. The plot with a logarithmic scale (b) shows clearly how quantum kinetics yields an approximately exponential absorption tail over many orders of magnitude, while the Markovian kinetic model has a Lorentzian line shape. Following the arguments developed in Ref. 8, one reaches an exponential line shape starting from a Lorentzian one only if the linewidth $\gamma=\gamma(\omega)$ decreases rapidly with increasing detuning $\omega_0-\omega$ below the exciton resonance at ω_0. This decrease of $\gamma(\omega)$ away from the exciton resonance is due to the simple fact that with increasing detuning $\omega_0-\omega$ it is increasingly difficult to scatter by LO-phonon absorption into real final states with frequencies $\geq\omega_0$. A frequency-dependent damping $\gamma(\omega)$ of the polarization $P(\omega)$ in the time representation of the polarization equation gives rise to a convolution integral $\int dt'\gamma(t-t')P(t')$, i.e., to memory effects.[8] It has been seen above that such memory effects are the trademark of quantum kinetics. It is known (as discussed, e.g., in Ref. 8) that in more polar materials vertex corrections of the *e-h* self-energies have to be taken into account in order to get a quantitative description of the universally observed Urbach absorption tail. But irrespective of such refinements of the theory, the conjecture of Ref. 8 that the exponential nature of the absorption tail is a manifestation of the retardations and thus of quantum kinetics is proven here clearly. It should be mentioned that Hartmann and Schäfer[12] reached the same conclusion from their studies of the quantum kinetic equation of the two-time particle propagators.

It is known that at low temperatures LO-phonon sidebands grow out of the Urbach tail of linear absorption. This fact supports our proposal that the calculated LO-phonon quantum beats should be observable for nonresonant sub-band-gap excitation in time-resolved four-wave mixing experiments.

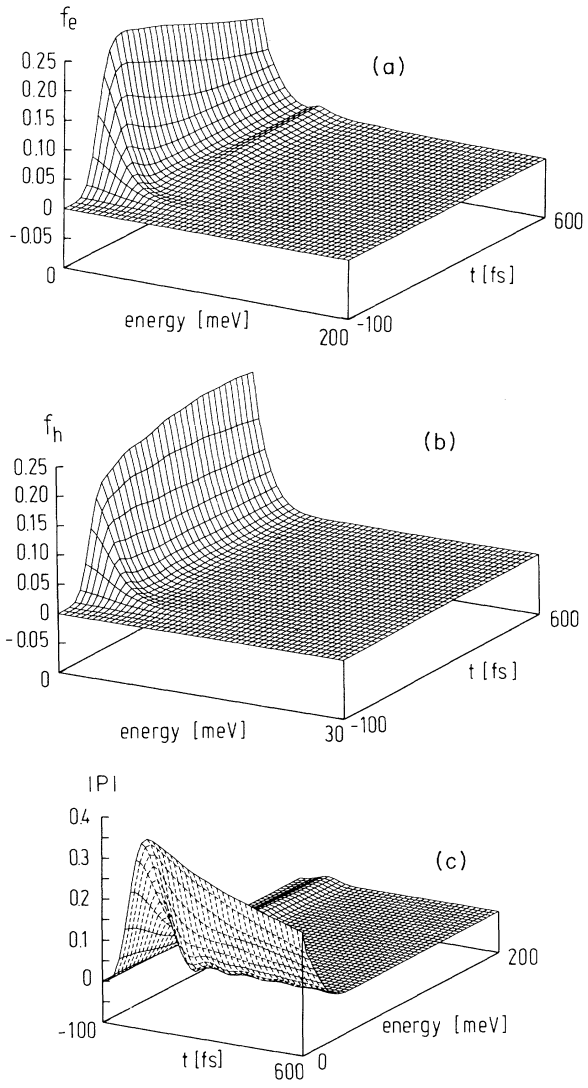

FIG. 5. Distributions (a) $f_{ke}(t)$ and (b) $f_{kh}(t)$, and absolute amount of the interband polarization (c) $|P_k(t)|$ vs energies e_{ke}, e_{kh}, and e_{ke}, respectively, and time for GaAs excited with a 50-fs pulse peaking at $t=0$ for nonresonant excitation 10 meV below the gap and a Rabi frequency of $dE_0=6.6$ meV.

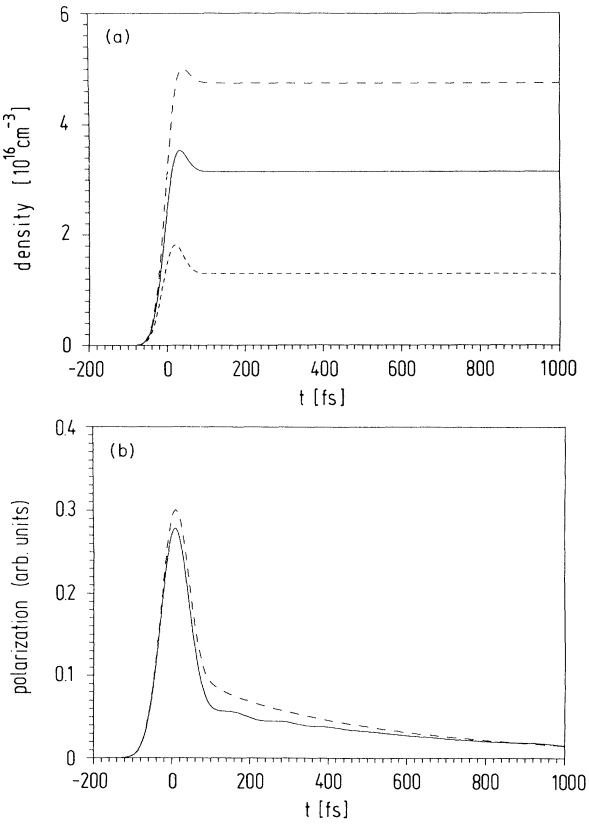

FIG. 6. (a) Total *e-h* density and (b) incoherently summed polarization $P(t)=\Sigma|\mathbf{P}_k(t)|$ vs time for GaAs excited with a 50-fs pulse peaking at $t=0$ for nonresonant excitation 10 meV below the gap and a Rabi frequency of $dE_0=6.6$ meV. Full line: with memory; long-dashed line: without memory; short-dashed line: with memory but without excitonic effects.

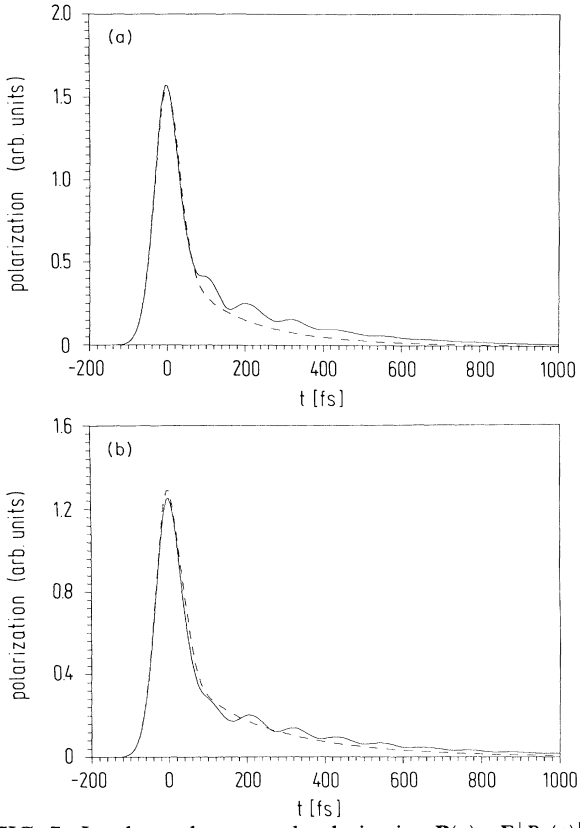

FIG. 7. Incoherently summed polarization $\mathbf{P}(t)=\Sigma|P_k(t)|$ vs time for InP excited with a 50-fs pulse peaking at $t=0$ with an excess energy of (a) 60 meV and (b) 20 meV, and a Rabi frequency of $dE_0=26.4$ meV. Full line: with memory; long-dashed line: without memory.

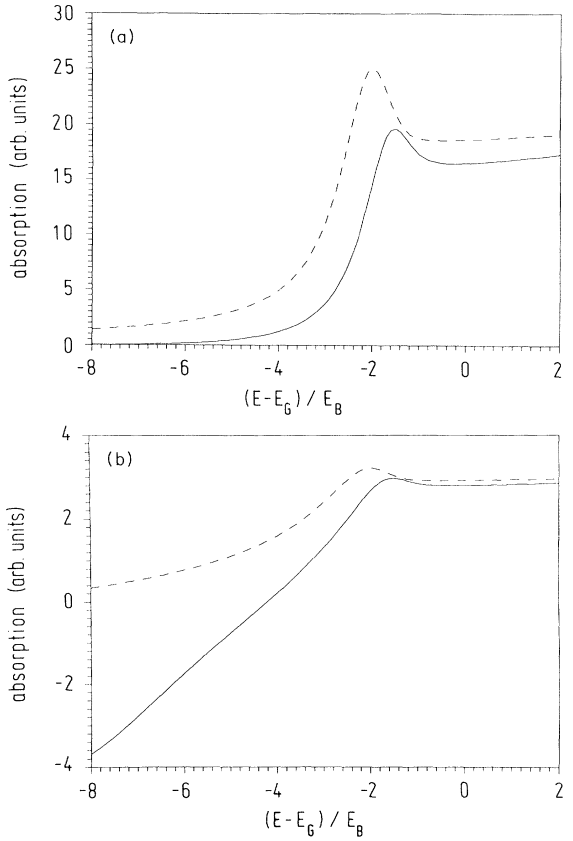

FIG. 8. Linear band-tail absorption spectrum of GaAs at room temperature. Full line: with memory; long-dashed line: without memory. (a) On a linear scale, (b) on a logarithmic scale.

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*Permanent address: Institute of Physics, National Center of Scientific Research, Mac Dinh Chi 1, Ho Chi Minh City, Vietnam.

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