Acoustic mode with time-dependent phase velocity in photoexcited semiconductors

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Using the Boltzmann equation, we study the collective modes of a photoexcited electron-hole plasma in a direct-gap, small-electron-mass semiconductor in which the electron distribution is evolving with time. In this situation, we find that there exists an acoustic mode of the electron-hole plasma with a *time-dependent* phase velocity. The phase velocity is approximately given by $\omega_{p,h}/q_{\mathrm{sc},e}(t)$, where $\omega_{p,h}$ is the hole plasma frequency and $q_{\mathrm{sc},e}(t)$ is the screening wave vector of the electron distribution $f_e(\mathbf{v},t)$. As the electron distribution cools, the $q_{\mathrm{sc},e}(t)$ increases and hence the phase velocity of the mode decreases with time.

There has been considerable interest in studying the ultrafast dynamics of carriers in semiconductors by photoexciting electron-hole plasmas and probing these plasmas on the picosecond and femtosecond time scales.[1] Measurement of these dynamics yields information on the carrier-phonon and carrier-carrier interactions, which are important in determining the characteristics of semiconductor devices. Recently, some effort has been directed at studying the transport properties of photoexcited systems in the femtosecond time scale,[2,3] where interesting transient effects due to the evolution of the carrier distribution functions can be seen. This activity has motivated us to study theoretically the linear response, and in particular the acoustic collective mode, of electron-hole plasmas in photoexcited semiconductors (with holes that are much more massive than the electrons) which are *not* in steady state. We note that this work is different from previous theoretical efforts[4] and experimental observations[5] of acoustic modes in photoexcited plasmas, where the carriers were assumed to be in a quasiequilibrium steady state. We find that in the non-steady-state photoexcited system, *the acoustic mode has a phase velocity that changes as a function of time.*

In an acoustic mode, as the hole density deviates from its uniform value, the lighter and more mobile electrons quickly respond to cancel out (or screen) the potentials created by the holes. However, due to thermal or quantum fluctuations, this screening is imperfect, and there is a residual, screened Coulomb interaction between the holes which causes the holes to oscillate. The dispersion relation for this mode, in a collisionless electron-hole plasma, over the full range of q is approximately[6]

$$\omega^2(q)=\omega_{p,h}^2\frac{q^2}{q^2+q_{\mathrm{sc},e}^2}\ , \tag{1}$$

where $\omega_{p,h}=(4\pi e^2 n_0/\epsilon_0 m)^{1/2}$ is the hole plasma frequency and $q_{\mathrm{sc},e}$ is the electron screening wave vector [which is equal to $(4\pi e^2 n_0/\epsilon_0 k_B T_e)^{1/2}$ for a Maxwellian distribution]. For small wave vectors ($q \ll q_{\mathrm{sc},e}$), $\omega(q)$ is approximately linear in q, with a phase velocity of $v_\phi=\omega(q)/q=\omega_{p,h}/q_{\mathrm{sc},e}$. The $q_{\mathrm{sc},e}$ is determined by the electron distribution. Therefore, if the electron distribution evolves with time, $q_{\mathrm{sc},e}$ and hence v_ϕ also change with time.

In this paper, we use the Boltzmann equation

$$\frac{\partial f_\alpha}{\partial t}+\mathbf{v}\cdot\frac{\partial f_\alpha}{\partial \mathbf{x}}+\frac{\mathbf{F}_\alpha}{m_\alpha}\cdot\frac{\partial f_\alpha}{\partial_{\mathbf{v}}}=I_\alpha[f_\alpha] \tag{2}$$

(f_α is the distribution function of component α, and I_α is the collision term) to study the collective modes of a bulk non-steady-state photoexcited system comprised of light electrons in the conduction band and holes in the valence band. We solve exactly the electron and hole distribution functions for the photoexcited system, within a simple collision model. Then, to study the collective modes, we add a small perturbation to these "unperturbed" distribution functions, and to linear order, we calculate the subsequent time-evolution of the electron and hole densities.

COLLISION MODEL

The main energy-loss mechanism for the carriers in III-V semiconductors such as GaAs is the emission of longitudinal-optic phonons because both the scattering rates and the frequencies of these phonons are large (e.g., in GaAs the scattering time is $\sim 2\times 10^{-13}$ s,[7] and phonon frequency is 36 meV). Therefore, we model the scattering by assuming that the carriers only emit dispersionless longitudinal-optic phonons of energy E_{op}. In this scattering model, which is similar to models used by others,[8] the

electrons cascade down from one level to another, each level being E_{op} removed in energy from the next (see Fig. 1). We ignore absorption of phonons (i.e., we assume that the lattice is at $T=0$), which is an adequate approximation for temperatures that are small compared to the Debye temperature ($\sim$400 K for GaAs).[9] We also assume that the holes are collisionless and are initially at rest. The rationale for this assumption is that for a typical case, the initial energy of the holes is small (e.g., in GaAs, when electrons are photoexcited to 0.3 eV, the initial hole energy is $\sim$40 meV) and therefore after one optic-phonon emission, which occurs quickly[10] ($\sim$100 fs), the holes possess very little energy and cannot scatter further because they are below the optic-phonon emission threshold.

To keep the calculation simple, we assume that the conduction and valence bands are parabolic. We also ignore intervalley scattering, and therefore our results are only valid for photoexcitation energies below the satellite valleys (e.g., $\lesssim$0.3 eV in GaAs). We do not explicitly treat carrier-carrier scattering, which becomes important[3] for densities greater than 10^{17} cm^3. However, we will argue that electron-electron scattering will not cause the acoustic plasma mode to disappear.

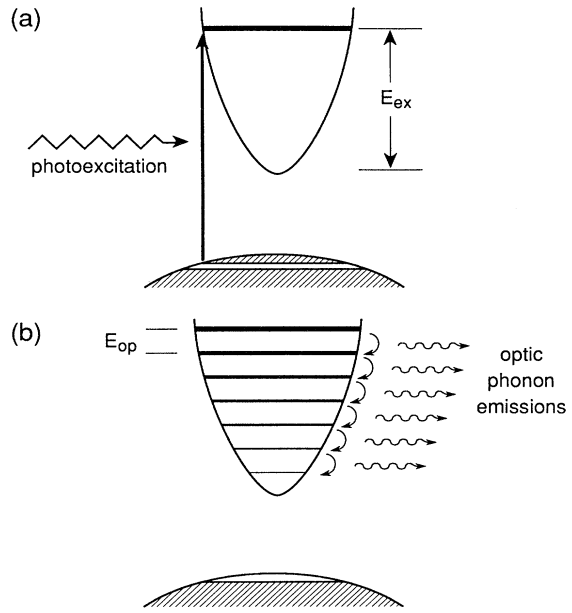

FIG. 1. Photoexcited carriers in a direct-band semiconductor, with $m_h \gg m_e$, (a) immediately after photoexcitation, and (b) scattering with the lattice, according to our model. (a) shows that, at the time of photoexcitation, the electrons have much more kinetic energy than the holes because the electrons are much lighter than the holes. (b) schematically shows our scattering model, in which the electrons emit optic phonons and lose energy in the quanta of the energy of the optic phonons, E_{op}. In the absence of any perturbations, the electrons are confined to levels with energies of $E_{ex}-jE_{op}$. The electrons cascade down the energy levels until they reach the bottommost level. The holes are assumed to have zero kinetic energy (since they have much less kinetic energy than the electrons) and are assumed to be collisionless because they are below the optic-phonon emission threshold.

UNPERTURBED DISTRIBUTION FUNCTIONS

Within our collision model, the distribution functions for the photoexcited system in the absence of perturbations, $f_{\alpha,0}(\mathbf{v},t)$, are as follows.[11] The holes are assumed to be at the valence-band edge at $t=0$, and hence the distribution function of the unperturbed holes is simply $f_{h,0}(\mathbf{v},t)=n_0\delta(\mathbf{v})$. Immediately after photoexcitation, all the electrons are concentrated on an energy shell E_{ex} above the conduction-band edge. Since the optic phonons in our model are assumed to remove energy in quanta of E_{op}, if the system is not perturbed by other forces the photoexcited electrons are only found in discrete levels indexed by $j=0,1,2,\ldots,N$ corresponding to the number of optic phonons emitted by the electron. Therefore, the electron distribution function for the unperturbed system is

$$f_{e,0}(\mathbf{v},t)=\sum_{j=0}^{N} P_j(t)\frac{n_0\delta(v-v_j)}{4\pi v_j^2}\ , \tag{3}$$

where $v_j=[2(E_{ex}-jE_{op})/m_e]^{1/2}$ is the electron velocity in the jth level. The coefficients $P_j(t)$ give the fraction of the photoexcited electrons populating level j.

The $P_j(t)$'s are determined by electron scattering. The rate of change of the population, $dP_j(t)/dt$, is equal to the rate at which particles are scattered in from the level above minus the rate at which particles are scattered out to the level below. Since the optic-phonon emission rate is fairly constant over a large energy range,[12] we assume that the scattering rate γ is the same for all levels. With this collision model, the populations are

$$P_j(t)=\begin{cases}\dfrac{(\gamma t)^j}{j!}e^{-\gamma t} & \text{if } j\neq N;\\ 1-\displaystyle\sum_{j=0}^{N-1}P_j(t) & \text{if } j=N.\end{cases} \tag{4}$$

THE PERTURBED SYSTEM

We study the behavior of the acoustic collective mode by perturbing the holes in the system and calculating the linear response of the system to that perturbation. Perturbations of the electron and/or hole distributions cause charge-density perturbations in the system, which result in electrostatic forces which in turn induce more perturbations of the electron and hole distributions. The time evolution of the perturbations is given by the Boltzmann equation, Eq. (2). We assume a small spatially sinusoidal initial perturbation in the distribution

$$f_\alpha(\mathbf{v},\mathbf{x},t)=f_{\alpha,0}(\mathbf{v},t)+f_{\alpha,1}(\mathbf{v},t)e^{i\mathbf{q}\cdot\mathbf{x}}\ , \quad (\alpha=e,h)\ . \tag{5}$$

Substituting Eq. (5) into Eq. (2), keeping only terms to

lowest order in the perturbation and using Poisson's equation to obtain $\mathbf{F}_\alpha$ gives

$$\frac{\partial f_{\alpha,1}}{\partial t}(\mathbf{v},t)+i\mathbf{q}\cdot\mathbf{v}f_{\alpha,1}(\mathbf{v},t) - \frac{4\pi i[n_{h,1}(t)-n_{e,1}(t)]ee_\alpha}{q^2 m_\alpha}\mathbf{q}\cdot\frac{\partial f_{\alpha,0}}{\partial \mathbf{v}}(\mathbf{v},t) = I_\alpha[f_{\alpha,1}](\mathbf{v},t)\ , \quad (6)$$

where e_α is equal to $-e$ for electrons and e for holes, and $n_{\alpha,1}=\int d\mathbf{v}\, f_{\alpha,1}(\mathbf{v})$ is the density perturbation of component α. We use Eq. (6) to solve for $f_{1,\alpha}(\mathbf{v},t)$. Then, integrating $f_{1,\alpha}(\mathbf{v},t)$ with respect to $\mathbf{v}$, we obtain a pair of coupled integral equations that describe the time evolution of the electron and hole density perturbations, $n_{e,1}(t)$ and $n_{h,1}(t)$ that result from an initial perturbation in the carrier distribution functions.

Since we are particularly interested in the acoustic mode, in which the holes oscillate and are screened by the rapidly moving electrons, we give the holes a small sudden impulse, which produces a perturbation of the hole distribution of

$$\delta f_{h,1}(\mathbf{v},t=0)e^{i\mathbf{q}\cdot\mathbf{x}}=\bar{n}_1 v_0\hat{\mathbf{q}}\cdot\frac{\partial\delta(\mathbf{v})}{\partial\mathbf{v}}e^{i\mathbf{q}\cdot\mathbf{x}}\ .$$

We then calculate the subsequent electron and hole density perturbations for $t>0$.

RESULTS

We numerically integrated the equations for $n_{e,1}(t)$ and $n_{h,1}(t)$ for experimentally accessible parameters. The hole and electron mass ratio was chosen to be $m_h/m_e=7$, and the wave vector was chosen to be $qv_0/\omega_{p,e}=1$. These parameters correspond to GaAs photoexcited to a density $n_0=3\times10^{18}$ cm^{-3} ($\omega_{p,e}\approx10^{14}$ s^{-1}) at an excitation energy of $E_{\rm ex}=0.3$ eV (i.e., just below the L valley in GaAs, with $v_0\approx10^8$ cm/s), and at $q=10^6$ cm^{-1}.

Using the above parameters, we performed the calculation of two cases. In the first case, the optic-phonon energy is taken to be $E_{\rm op}=36$ meV (as in GaAs), and hence $E_{\rm op}/E_{\rm ex}=0.12$, implying that an electron emits eight optic phonons in going from the initial to the lowest level. The scattering rate was assumed to be $\gamma=0.5\times10^{13}$ s^{-1}, so that $\gamma/\omega_{p,e}=0.05$. In the second case, we halved the optic-phonon energy ($E_{\rm op}/E_{\rm ex}=0.06$), hence doubling the number of optic-phonon emissions needed to get to the lowest level to 16, and we also doubled the phonon emission rate ($\gamma/\omega_{p,e}=0.1$). The second case roughly simulates the effect of the inclusion of electron-electron scattering (an important scattering mechanism at large densities),[3] since e-e scattering scatters electrons to states that interleave the levels connected by optic-phonon emissions but does not significantly change the energy-loss rate of the electrons. The results are shown in Fig. 2. In both cases, there is a well-defined mode, and the charge densities of the electrons and holes are almost completely out of phase, indicating that the mode is acoustic. *The oscillation frequency decreases notably as time increases, because the screening of the holes by the electrons increases when the electrons cool from optic-phonon emissions.* The fact that the results from the two cases do not differ significantly indicates that electron-

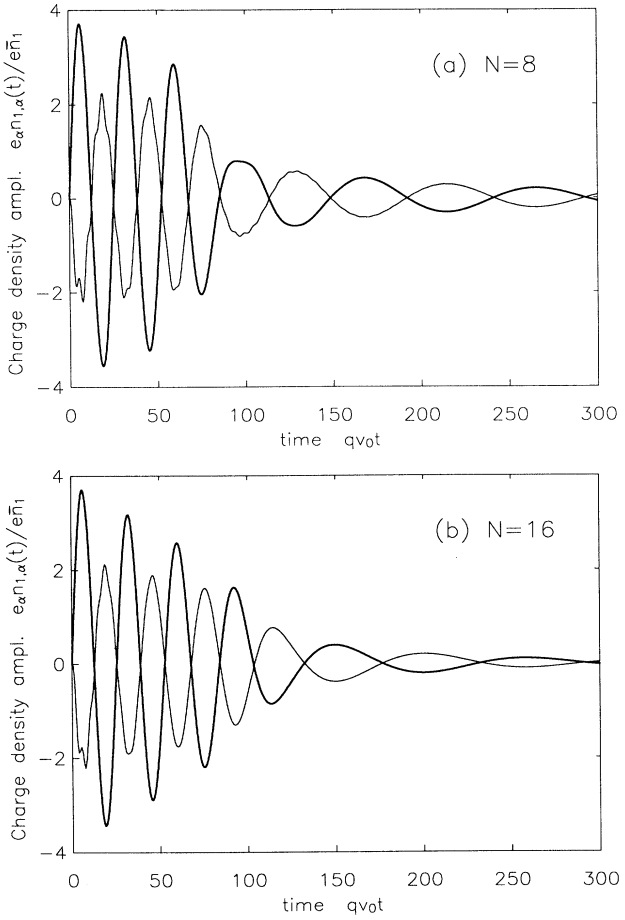

FIG. 2. The hole (thick line) and electron (thin line) charge-density amplitudes, as a function of time, for a photoexcited electron-hole plasma with the collision model shown in Fig. 1. The holes are perturbed at $t=0$ by $\delta f_h(\mathbf{v},\mathbf{x})=\bar{n}_1 v_0[\hat{\mathbf{q}}\cdot\partial\delta(\mathbf{v})/\partial\mathbf{v}]e^{i\mathbf{q}\cdot\mathbf{x}}$. (a) is for $m_h/m_e=7$, $qv_0/\omega_{p,e}=1$, $\gamma/\omega_{p,e}=0.05$, $E_{\rm op}/E_{\rm ex}=0.12$, and $N=8$ which corresponds to GaAs photoexcited to $n_0=3\times10^{18}$ cm^{-3}, with $E_{\rm ex}=0.3$ eV, and an optic-phonon energy of 36 meV and scattering rate of $\gamma=0.5\times10^{13}$ s^{-1}. The oscillation frequency decreases with time because the screening of the holes increases with time (due to the cooling of the electrons) which weakens the effective Coulomb interaction between the holes. In (b), we halved the optic-phonon energy and doubled the scattering rate ($N=16$, $\gamma/\omega_{p,e}=0.1$, and $E_{\rm op}/E_{\rm ex}=0.06$), which doubles the number of optic-phonon emissions while roughly keeping the energy-loss rate constant, in an attempt to simulate the effect of electron-electron scattering (which would scatter electrons to states interleaving the energy levels connected by optic-phonon emissions). A well-defined mode exists in (b), indicating that electron-electron scattering will not adversely affect this mode. The little ripples on the electron density in both cases are due to optic plasma oscillations.

electron scattering, which was roughly simulated in Fig. 2(b), will not cause the mode to disappear.

For a more quantitative discussion of this acoustic mode, we denote the ith time at which the hole density goes through zero as t_i ($i=0,1,2,\ldots$), and we define the angular frequency of the oscillation at time $(t_i+t_{i+1})/2$ to be $\pi(t_{i+1}-t_i)$. This is a rough measure of the "instantaneous" frequency of oscillation. These frequencies were taken from Fig. 2, and are shown as circles in Fig. 3.

We can understand the behavior of the oscillation frequencies shown in Fig. 3 by assuming that the instantaneous oscillation frequency, $\omega(q,t)$, is given by the form in Eq. (1), with $q_{\mathrm{sc},e}$ given by the screening wave vector for the electron distribution at time t. The $q_{\mathrm{sc},e}$ for the $f_{e,0}(\mathbf{v},t)$ given by Eq. (3), assuming that the electrons are collisionless, is[13]

$$q_{\mathrm{sc},e}(t)=\omega_{p,e}\left[\sum_{j=0}^{N}\frac{P_j(t)}{v_j^2}\right]^{1/2}. \tag{7}$$

Since the population of the lower levels (where the electron velocities are smaller) increases with time, $q_{\mathrm{sc},e}$ increases with time. The increased screening weakens the interaction between the holes, causing the acoustic-mode frequency to decrease with time.

Inserting Eq. (7) into Eq. (1), and using the expressions for the occupation fraction, $P_j(t)$, given by Eq. (4), we produce solid curves in Fig. 3. There is good agreement between the points obtained from the curves in Fig. 2, and the solid line, except that these points consistently lie below the solid line. This discrepancy can be qualitatively understood by noting that weak damping of a simple harmonic oscillator decreases the frequency of oscillation by an amount proportional to the square of the damping. Since the electrons scatter inelastically, they effectively act as a damping mechanism, and therefore the oscillation frequency from our calculation (which includes inelastic electron scattering) should be slightly lower than oscillation frequency given by Eq. (1) (which assumes that the electrons are collisionless). The discrepancy is slightly larger in the $N=16$ case than in the $N=8$ case, which is consistent with the damped simple harmonic-oscillator model, since the scattering rate (and hence the effective damping) for the $N=16$ case is twice that of the $N=8$ case. However, the quantitative dependence of the suppression of the oscillation frequency does not seem to be given by this model.

We also performed calculations for the same parameters as in the $N=8$ case, but with different wave vectors. Except for an overall change in the frequency of oscillations with q, the behavior of the oscillations is qualitatively the same, i.e., the period of the oscillations decreases down as the electrons cool. Equation (1), together with Eq. (7), is a good functional predictor of the oscillation frequency for all q tested. Therefore we conclude that, for small q, the acoustic mode in this model system

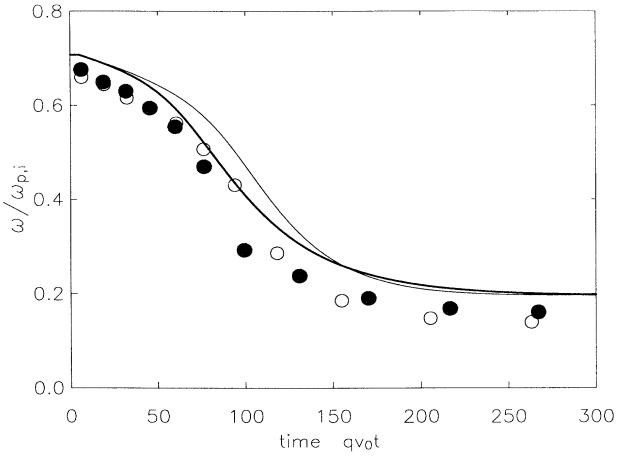

FIG. 3. The oscillation frequency vs time, of the modes shown in (a) Fig. 2(a) (solid circles) and (b) Fig. 2(b) (open circles). The periods of the oscillations were determined from the time taken for the amplitude of the hole density to pass from one zero to the next. The solid lines are the oscillation frequencies for (a) (bold line) and (b) (thin line) calculated using the collisionless plasma and static screening theory. This theory overestimates the oscillation frequencies because it does not take into account the electron scattering, which damps and slows down the oscillations.

(and presumably for real photoexcited systems) has a time-dependent phase velocity that is given by $v_\phi(t)=\omega_{p,h}/q_{\mathrm{sc},e}(t)$.

A possible way of experimentally observing this mode is through a femtosecond transient grating experiment.[14] In this type of experiment, a spatially periodic distribution of carriers is created by absorption from the interference pattern produced by two coherent light pulses. A third time-delayed pulse can then be used to probe the carrier density as a function of delay time.

To conclude, we used the Boltzmann equation within an optic-phonon emission model to show that a well-defined acoustic mode exists in a photoexcited intrinsic semiconductor that is still evolving in time. The phase velocity of the acoustic mode decreases with time, since it is inversely proportional to the electron screening wave vector, which increases as the electrons cool from optic-phonon emissions. We can understand the quantitative features of the change in the phase velocity with time by a naive theory which assumes that the electrons are collisionless and screen the holes statically.

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