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Electron-hole plasma-driven phonon renormalization in highly photoexcited GaAs

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Many-body renormalization of phonon frequencies, due to their interaction with a high-density electron-hole plasma, such as the one created by an intense laser pulse applied to GaAs, is calculated. Results for the change in phonon-dispersion curves are obtained, both for a distribution characterized by zero carrier temperature and for the electron-hole plasma equilibrium temperature. We use a many-valley model for the conduction and valence bands of GaAs, and the deformation potentials available from the literature for the carrier-acoustic-phonon interaction. We do not find complete softening of the phonons below the experimental electron-hole pair density ($<10^{22}$ cm^{-3}), although zone-boundary phonon softening is observed at $(1-5)\times 10^{22}$ cm^{-3}. From the increase in the mean squared amplitude of the atomic vibrations caused by the phonon softening, we obtain an effective lattice melting temperature which decreases with increasing carrier temperature. Thus, a nonthermal electronic mechanism could at best play only a partial role in semiconductor laser annealing experiments even on ultrafast subpicosecond time scales.

The effect of free carriers (electrons and holes) on the phonon properties in a semiconductor is substantially smaller than the effects of phonons on the electronic properties. This is due to the small ratio of the electron mass to the ionic mass. However, electronic renormalization of phonon properties increases with increasing carrier concentration, which can be varied by orders of magnitude in a semiconductor and one therefore has a control parameter to make these effects stronger. One way of controlling this concentration is by doping; there have been numerous studies, both theoretical and experimental,[1] of the changes in the lattice properties such as elastic constants and sound velocities in semiconductors caused by doping. The interpretation of the results is complicated by the difficulty in separating the effects of the free carriers from that of the dopant ions. Another way is by photoexciting electron-hole pairs. The larger the intensity of the radiation and the smaller the temporal resolution of the experiment, the more noticeable these effects of electronic renormalization of phonon properties become.

In fact, it was once conjectured that pulsed laser annealing of semiconductors would occur by an electronic mechanism called "electronic laser annealing," consisting in the photoexcitation of a large number of free carriers which screen the electron-phonon interaction, thereby weakening the interatomic forces so that the atoms could rearrange themselves in a time too short for the lattice to heat up.[2] In light of much experimental evidence it is now widely accepted that the actual annealing mechanism is the usual thermal one with the direct transfer of heat from the photoexcited plasma to the lattice, raising the lattice temperature to its melting point.[3] More recently, experiments involving ultrafast irradiation of bulk GaAs with 0.1-ps laser pulses associated with the observed loss of crystalline order within 0.2 ps of excitation[4] have revived interest in nonthermal mechanisms of laser annealing. The effect of a large concentration of electron-hole pairs on the phonons in silicon was earlier calculated by Biswas and Ambegaokar,[5] by directly diagonalizing the dynamical matrix to obtain the phonon frequencies with the effect of the electron-hole plasma being introduced in the model through a dielectric function that interpolated between the ground-state (insulator) dielectric function of the semiconductor and that of a free electron and hole gas. Their method, however, is not easily related to the usual treatment of the electron-phonon interaction in semiconductors using the deformation potential.

These considerations have motivated us to revisit the laser-annealing problem and to calculate the self-energy renormalization of the GaAs phonons due to their interaction with a high-density electron-hole plasma. We describe the electronic structure of GaAs by a multivalley model, taking into account contributions from a Γ valley, four L valleys, and three X valleys in the conduction band; and from a heavy-hole (hh) and a light-hole (lh) valley in the valence band. The phonon propagator for a given phonon mode is renormalized by the effect of the acoustic-phonon–electronic-carrier interaction through the deformation potential and is given by[6] the dressed propagator

$$D(q,\omega)=\frac{2\omega_q}{\omega^2-\omega_q^2-\dfrac{2\omega_q}{\varepsilon(q,\omega)}\sum_v |M_v(q)|^2\chi_v^2(q,\omega)}\ , \tag{1}$$

where

$$|M_v(q,\omega)|^2=\frac{D_v^2q^2}{2\rho\omega_q} \tag{2}$$

is the squared deformation potential matrix element for interaction with a single valley, ρ the mass density of the semiconductor, and ω_q the "bare" phonon frequency, which is the phonon frequency in the electronic ground state in the absence of any free electrons or holes. The sum in Eq. (2) is over all the valleys of GaAs ($v=\Gamma,L_1,\ldots,L_4,X_1,\ldots,X_3,\text{hh},\text{lh}$). For the conduction-band valleys the coupling to the strain tensor s_{ij} is given by

$$D_v=\Xi_{dv}\sum_{i=1}^{3}s_{ii}+\Xi_{uv}\sum_{i,j=1}^{3}\hat{k}_{iv}\hat{k}_{jv}s_{ij}\ , \tag{3}$$

where Ξ_{dv} and Ξ_{uv} are the two deformation potential coupling constants for the ellipsoidal valley v, and $\mathbf{k}_v$ are the vectors from the Γ point to the valley minimum. For the valence band valleys, the couplings are given by

$$D_{\text{hh,lh}}=a\sum_{i=1}^{3}s_{ii}\pm\left\{b^2[(s_{11}-s_{22})^2+(s_{11}-s_{33})^2+(s_{22}-s_{33})^2]/2+d^2(s_{12}^2+s_{31}^2+s_{23}^2)\right\}^{1/2}\ , \tag{4}$$

where a, b, and d are the deformation potential coupling constants for the valence band, and the plus (minus) sign applies to the heavy-hole (light-hole) valley. The strain associated with the phonon mode of direction $\hat{\mathbf{q}}$ and polarization $\hat{\mathbf{p}}$ is

$$s_{ij}=\tfrac{1}{2}(p_iq_j+p_jq_i)\ . \tag{5}$$

The materials parameters, in addition to the mass density and dielectric constant, are the valley minima energies, the effective density-of-states mass per valley, and the deformation potential constants. The values selected from the literature[7–11] that we use in the calculations are shown in Table I.

Free carrier effects are contained in the polarizabilities $\chi_v(q,\omega)$ and the dielectric function $\varepsilon(q,\omega)$. The renormalized phonon frequencies are given by the poles of Eq. (1), and since the phonon frequencies of interest are much smaller than the electronic energy scales (energy gap, effective Fermi energy, plasmon frequencies, etc.), we make the usual static approximation $\chi(q,\omega)\cong\chi(q,0)\equiv\chi(q)$ and $\varepsilon(q,\omega)\cong\varepsilon(q,0)\equiv\varepsilon(q)$, thus obtaining for the renormalized phonon frequencies

$$\omega^2(q)=\omega_q^2-\Omega^2(q) \tag{6}$$

with

$$\Omega^2(q)=\rho^{-1}\varepsilon(q)\sum_v D_v q^2\chi_v(q)\ . \tag{7}$$

The contribution of each valley to the polarizability $\chi_v(q)$ is calculated in the Lindhardt approximation, and depends only on m_v, the density-of-states mass per valley, and n_v, the concentration of carriers in the valley. The dielectric function is given by

$$\varepsilon(q)=1-\frac{4\pi e^2}{q^2}\sum_v\left\{\chi_v(q)+[\varepsilon_v(q)-1]\left[\frac{n-n_0}{n_0}\right]\right\} \tag{8}$$

and includes the effect of a q-dependent ground-state (i.e., without any free carriers) dielectric function $\varepsilon_0(q)$. The latter is usually approximated by a constant $\varepsilon_0=\varepsilon_0(0)$ in many-body calculations, but because we are interested in obtaining results across the Brillouin zone (BZ), we retain the q dependence and use an approximate form for $\varepsilon_0(q)$, obtained by fitting Srinivasan's results[12] for GaAs which are based on the Penn model for a spherical BZ (whose radius we choose to match the volume of the actual GaAs BZ).

At this point it is worthwhile mentioning that in the limit of small q the second term in Eq. (8) must dominate, and the self-energy correction becomes quartic in q:

$$\Omega^2(q)\cong\frac{q^4\sum_v D_v^2\chi_v(q)}{4\pi e^2\rho\sum_v\chi_v(q)}\ , \tag{9}$$

and, in the case of a doped semiconductor with only one valley occupied,

$$\Omega^2(q)\cong\frac{\Xi^2q^4}{4\pi e^2\rho}\ . \tag{10}$$

Corrections to the sound velocity can occur only at nonzero $q>q_s$, i.e., when the carrier concentration is low enough for the Thomas-Fermi screening wave vector (q_s) defined by

$$q_s^2=-4\pi e^2\chi(0) \tag{11}$$

to be smaller than q.

The evaluation of the free carrier response function $\chi_v(q)$ requires a knowledge of the actual (intravalley and intervalley) carrier distribution. The present method of calculation allows one to use as input an arbitrary distribution of the carriers within a valley and among different valleys as obtained from nonequilibrium calculations, and, therefore, the model can easily be extended to include a time-dependent calculation of the dynamics of photoexcited electron-hole plasma in a semiconductor. Here we assume that the carriers can be described by an equilibrium Fermi distribution with an effective carrier temperature T, and we obtain results in two extreme cases: $T=0$, with a common Fermi level for all the con-

TABLE I. Materials parameters used in the calculations presented in this paper.

Type	m/m_0	$E_{\min}$ (eV)[a]	Deformation potentials (eV)[c]	
Γ	0.063[a]	1.424	$\Xi_d=-17.5$	
L	0.22[a]	1.708	$\Xi_d=-22.0$	$\Xi_u=39.2$
X	0.41[a]	1.900	$\Xi_d=-1.6$	$\Xi_u=-6.3$
lh	1.47[b]	0	$a=-4.8$	
hh	0.43[b]	0	$b=-2.0$	
			$d=-5.4$	

[a]From Ref. 7, and for $T=300$ K; m is the density-of-states mass per valley.

[b]Since we are concerned with high hole concentrations, and to take into account the warped form of the valence bands, we use these high values of m_{hh} and m_{lh}, proposed by Reggiani (Ref. 8).

[c]See Ref. 9.

duction valleys and a different Fermi level for the holes; and $T=T_{eq}$, where T_{eq} is the temperature for an equilibrium electron-hole system, specified by electrons and holes having the same chemical potential. These are two simple extreme cases which catch the essence of the physics we are interested in.

In Fig. 1, we show the bare and renormalized phonon dispersion relations along the principal symmetry directions for $n=10^{22}$ cm^{-3} electron-hole pairs, assuming an equilibrium electron-hole distribution (i.e., $T=T_{eq}$). The results for the $T=0$ distribution are almost indistinguishable on this scale from the results shown. For this very-high-density electron-hole plasma the first mode to be driven to zero frequency is the (100) TA phonon at a density around 1.5×10^{22} pairs cm^{-3}. For $T=0$ (and presumably for any intermediate effective temperature $0\le T\le T_{eq}$) the instability also occurs at the BZ edge. This feature is consistent with the results obtained in Ref. 5 by an entirely different method.

It is reasonable to ask whether this electronic plasma-involved weak phonon renormalization contributes significantly to lattice melting. To quantify that contribution, we consider the real-space vibrations of the lattice atoms. Since little is known about the details of bulk melting anyway, we use the simple Lindemann criterion which relates the amplitude of the real-space vibrations to the melting temperature. By this criterion, the solid will melt when the average squared amplitude of the vibrations, given by

$$\langle u^2\rangle(T_{\mathrm{lat}})=(\mathrm{const})\times\sum_q \frac{n(T_{\mathrm{lat}},\omega_q)}{\omega_q}\ , \qquad (12)$$

reaches a certain specific value, which occurs at $T_{\mathrm{lat}}=T_M$, the melting temperature of the solid ($T_M=1513$ K for GaAs). Raising T_{lat} the lattice temperature is one way of increasing the amplitude of the vibrations; another way is lowering the phonon frequencies. We plot in Fig. 2 the calculated squared lattice vibrational amplitude $\langle u^2\rangle$ as a function of carrier concentration for several values of the lattice temperature including the effects of phonon softening by the plasma. The integration over the BZ in Eq. (12) is obtained by a weighted average of the integrals along the three principal symmetry directions. We find from these curves that, for example, at a free carrier concentration of $n=3\times10^{21}$ cm^{-3}, the restoring forces have become small enough for the vibration amplitudes at $T_{\mathrm{lat}}=0.9T_M$ to be as large as the ones corresponding to the melting of the semiconductor in the absence of the free carriers. This gives an effectively reduced melting temperature in the presence of the free carrier plasma. (Thus, for $n=3\times10^{21}$ cm^{-3}, the effective melting temperature is reduced by 10% due to the free carrier renormalization effect.) All the curves eventually diverge at the above-mentioned critical carrier concentration where the frequency of a particular phonon mode is renormalized down to zero, making the lat-

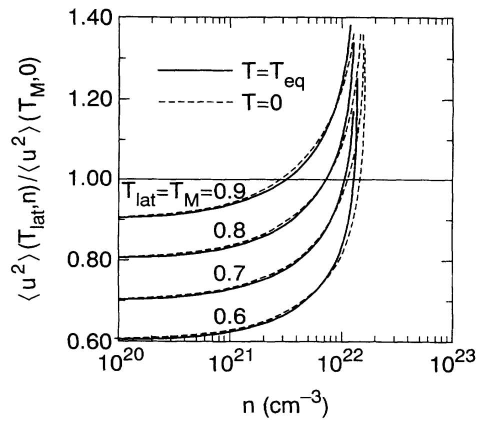

FIG. 2. Average squared amplitude of the atomic lattice vibrations as a function of electron-hole pair concentration for several values of the lattice temperature, normalized by the same amplitude, but calculated at zero carrier concentration, and, at $T=T_M=1513$ K, the melting temperature of GaAs.

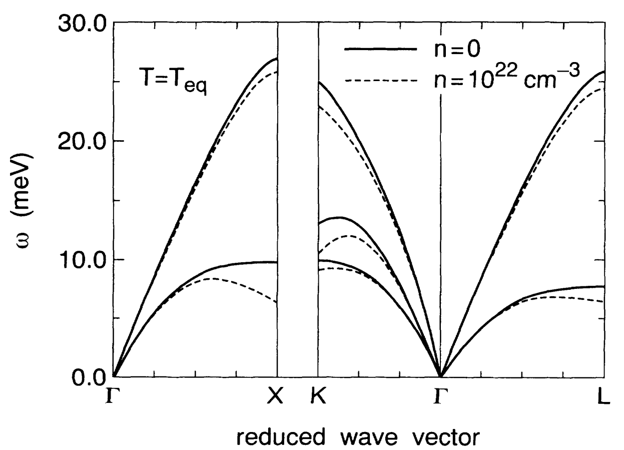

FIG. 1. Bare (solid line) and renormalized (dashed line) phonon dispersion relation along the [100], [110], and [111] directions. The renormalized curves were obtained using the equilibrium electron-hole distribution (the results with the $T=0$ distribution are almost indistinguishable from those in this scale).

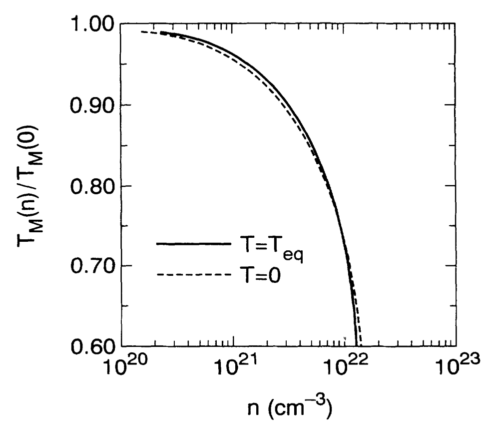

FIG. 3. Shows an "effective melting temperature," $T_M(n)$, obtained by applying the Lindemann criterion to the lattice at 300 K, but taking into account the softening of the phonons due to the presence of n electron-holes per cm^{-3}, and for the two cases of carrier distribution considered (as described in the text).

tice soft.

One can calculate the decrease in the melting temperature, as given by the Lindemann criterion, caused by the increase in the carrier density due to this phonon renormalization effect. One sees from the calculated results in Fig. 3 that the effects of large but reasonable carrier concentrations (of the order of 10^{21} cm^{-3}) are insignificant, and this "effective" melting temperature only becomes of the order of room temperature (300 K$=0.20T_M$ for GaAs) for electron-hole concentrations large enough for the occurrence of the BZ edge phonon instability anyway. Thus, while the free carrier plasma-induced phonon renormalization may play a small quantitative role in laser annealing, the main mechanism must still be the direct transfer of heat from the incident radiation field to the lattice without any interesting electronic mechanism. We do, however, expect the effect of the free carrier plasma on phonons to increase with a decrease in system dimensionalities and, therefore, it is possible that in lower dimensional semiconductor quantum wells (or quantum wires) one may see an enhanced role of the electron-hole plasma.

The results of this simple calculation are that nothing spectacular seems to happen in the phonon spectra of GaAs at the attainable concentrations of electron-hole pairs. It is clear that if a plasma mechanism is invoked[4] to explain the results of femtosecond pulsed laser experiments in GaAs, care must be taken to consider carrier distributions radically different from the quasiequilibrium, effective-temperature models considered here. In addition, the coupling of intervalley phonons to the electrons (Γ-L, Γ-X, X-X, and L-L) must be taken into account. We speculate, however, based on our results that it is unlikely that an electronic mechanism plays a significant role in the bulk laser annealing of GaAs under any (reasonable) circumstances.

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