Phonon Temperature Overshoot in GaAs Excited by Subpicosecond Laser Pulses

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Hot electrons and phonons excited in GaAs by subpicosecond laser pulses have been studied by inelastic light scattering for photoexcited electron densities varying between 10^{17} and 10^{19} cm^{-3}. Transient overshoot of longitudinal-optical (LO) phonon temperature above the electron temperature has been observed. This is explained by the fast production of zone-center LO phonons by hot electrons combined with slower reabsorption of the emitted phonons due to rapid cooling of Γ valley electrons by intervalley scattering.

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The relaxation of hot electrons in GaAs has been studied recently with subpicosecond dye-laser pulses using a variety of techniques.[1-6] It is known that the electron cooling rate decreases at electron densities higher than 10^{17} cm^{-3}.[7,8] This observation was explained as caused either by screening of the electron-phonon interaction[7] or by the generation of nonequilibrium optical phonons during the cooling of the hot electrons.[8,9] The latter explanation has sometimes been labeled as "hot-phonon effect." Recent studies[9-11] have suggested that the hot-phonon effect is more important for electron density in the range of 10^{18} cm^{-3}; however, direct experimental evidence is still lacking.

To study the effect of nonequilibrium phonons on the cooling of hot electrons, it is desirable to measure simultaneously both the phonon and electron distributions as functions of photoexcited electron-hole densities. One technique with this capability is inelastic scattering of light by photoexcited phonons and electrons. Previous light-scattering studies of hot phonons and electron-hole plasmas excited by picosecond (ps) laser pulses have only shown that the electrons and longitudinal-optical (LO) phonons attained the same temperature within a few ps after excitation.[12] In this Letter we have extended these measurements to the subpicosecond regime and to higher electron densities. We found that at densities above 10^{18} cm^{-3} the electrons and optical phonons are not in thermal equilibrium with each other within about 200 fs after excitation. In fact, the effective LO phonon temperature (definition given below) "overshoots" the electron temperature at high electron densities. From model calculations, we found that this overshoot of the LO phonon temperature in GaAs can be explained by a combination of two factors: fast cooling of electrons in the Γ conduction valley by intervalley scattering and efficient generation of zone-center LO phonons due to high electron density. The phonon temperature overshoot cannot be explained if LO phonon emission is the dominant relaxation mechanism of hot electrons in GaAs. Hence, our results strongly suggest that *intervalley scattering plays a dominant role in the subpicosecond cooling of hot electrons in GaAs*. The deformation potential for the Γ-to-*L* intervalley scattering in GaAs has been determined from our measurements to be 7×10^8 eV/cm. This value is consistent with recent measurements using other techniques.[1]

Our experiment was performed on a 0.3-μm-thick intrinsic GaAs layer grown by molecular-beam epitaxy (MBE) on a [100]-oriented GaAs substrate. The GaAs layer was sandwiched between two AlAs layers to confine the photoexcited carriers and to minimize surface recombination. The top AlAs layer was protected by an 80-Å GaAs cap layer. Hot electrons were excited in the GaAs layer by subpicosecond pulses from a colliding-pulse mode-locked ring dye laser operating at 2.0 eV and with an energy of 0.3 nJ per pulse.[13] To resolve details of the LO phonon Raman line shapes, the linewidth of the laser was reduced to 25 cm^{-1} by a one-plate birefringent filter inside the cavity. This also resulted in temporal broadening of the pulses whose lengths were measured with an autocorrelator. The autocorrelation traces are consistent with pulses with a sech^2 profile and full width at half maximum of 600 fs. The sample was maintained at room temperature during the experiment. Laser heating was found to be negligible.[14]

The light-scattering experiment was performed in a backscattering geometry with both the incident and scattered light polarized along the $[1\bar{1}0]$ direction. The photoexcited electron densities were both estimated by analysis of the line shape of this luminescence and calculated from the number of photons absorbed. The density was varied between 3×10^{17} and 10^{19} cm^{-3} by changing the size of the focal spot on the sample surface. A typical Raman spectrum, obtained after subtraction of the luminescence background, is shown in Fig. 1.

The Raman spectrum can be decomposed into relatively sharp structures, caused by scattering from the LO phonon, and a broader background due to scattering from single-particle excitations (SPE) of the photoexcited hot electrons.[15] As shown by Collins and Yu,[16] the coupling between the LO phonon and the plasmon should produce two peaks at the LO and transverse-

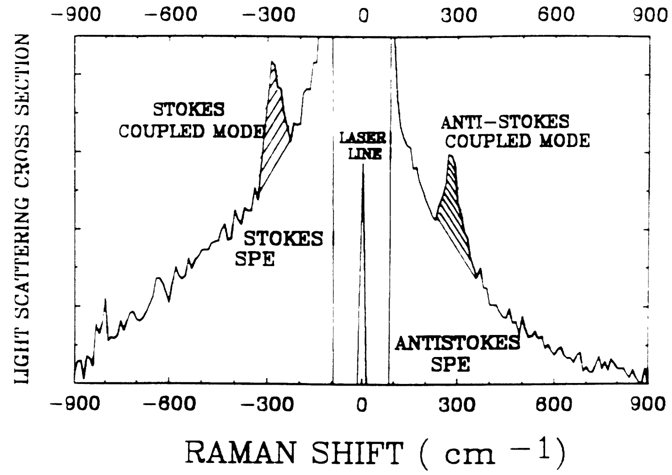

FIG. 1. A typical light-scattering spectrum of GaAs excited by intense subpicosecond laser pulses after subtraction of the luminescence background caused by recombination of hot-electron–hole pairs. The shaded peaks are due to scattering by the coupled plasmon-LO phonon modes. The broader peak on which the coupled modes are superimposed is caused by scattering by SPE of the photoexcited electrons. The density for this spectra is about 2×10^{18} cm^{-3}.

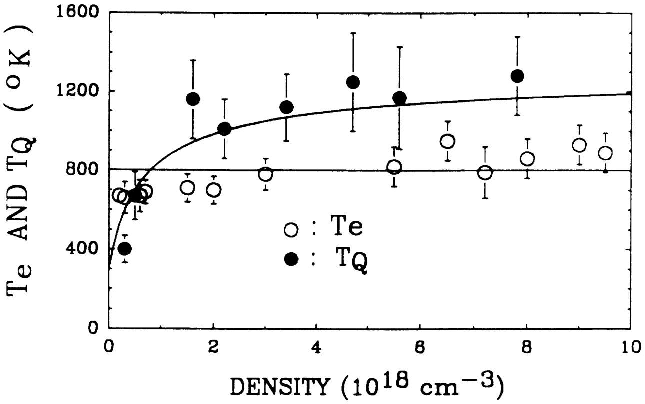

FIG. 2. The measured electron (T_e) and phonon temperatures (T_Q) plotted as functions of the photoexcited electron density. The solid lines are results of a fit obtained by our model calculation using $D_{\Gamma\text{-}L}=7\times10^8$ eV/cm.

optical (TO) phonon frequencies. These two peaks were not completely resolved because of our larger laser linewidth, so we observed instead a density-dependent shift in the phonon frequency. This shift in the phonon frequency is another way to estimate the electron density.

The phonon occupation numbers N_Q, corresponding to wave vector Q, were deduced from the ratios of anti-Stokes to Stokes Raman intensities by using the fact that[14,17] the anti-Stokes cross section is proportional to N_Q, whereas the Stokes cross section is proportional to N_Q+1. Care was taken to correct for the dispersion of the scattering efficiency by measuring the dispersion with a cw tunable dye laser and for the effect of the coupling between the plasmon and the LO phonon. The electron temperature T_e was obtained by comparing the Stokes and the anti-Stokes SPE scattering cross sections.[15] For convenience we define an "effective phonon temperature" T_Q by $N_Q=\{\exp(\hbar\omega/k_BT_Q)-1\}^{-1}$, where ω is the LO phonon frequency and k_B is Boltzmann's constant, because temperature is a more intuitive parameter in discussing energy exchange between phonons and electrons. This does not imply that we have any information on whether the phonons are in quasithermal equilibrium and have therefore a well-defined temperature. It should also be noted that our Raman measurement probes only LO phonons with a specific wave vector, $Q=7.5\times10^5$ cm^{-1}, determined by the photon wave vectors.

In Fig. 2 T_e and T_Q are plotted as functions of the electron density N_e. Since the same laser pulse was used to excite phonons and electrons and to perform light scattering from these electrons and phonons, it can be shown that T_e and T_Q determined in this way represent their temperatures about 200 fs after excitation by an infinitesimally short pulse.[18] As shown in Fig. 2 we find that T_e and T_Q behave differently as functions of N_e. While T_e is essentially constant (≈ 800 K) over densities from 10^{18} to 10^{19} cm^{-3}, T_Q increases rapidly when N_e is larger than 10^{18} cm^{-3}, "overshoots" T_e, and saturates at ≈ 1200 K for higher densities.

To our knowledge, this phonon temperature overshoot has not been predicted by Monte Carlo simulations. To understand our results, we have performed calculations of T_e and T_Q based on *models which either exclude or include intervalley scattering of electrons.* Models which exclude intervalley scattering can explain neither the overshoot in T_Q nor the relatively low T_e. Such models predict that, at the high densities and short-time scales of our experiment, the electrons saturate the relatively small number of LO phonon modes with wave vector Q so that $T_Q=T_e\approx 2500$ K. The model which successfully explains our result is shown schematically in Fig. 3(a). This model includes both intravalley LO phonon scattering (indicated by arrow 1) and intervalley scattering (indicated by arrows 2 and 3).

To keep the calculation simple, we have made a number of assumptions. Some of these assumptions are valid only within the subpicosecond time scale of interest to us. Hot electrons are assumed to be excited into a nonparabolic Γ valley from a parabolic valence band by an infinitesimally short pulse. The electrons thermalize instantaneously to a Fermi-Dirac distribution with initial electron temperature $T_e\approx 3000$ K determined by the photon energy and by the electron density through its chemical potential. This assumption is justified because it has been shown experimentally that, at the densities of interest to us, electrons reach thermal equilibrium in about 100 fs or less.[1,19-21] The hot electrons are assumed to cool by intravalley emission of LO phonons and by intervalley scattering into the X and L valleys only.

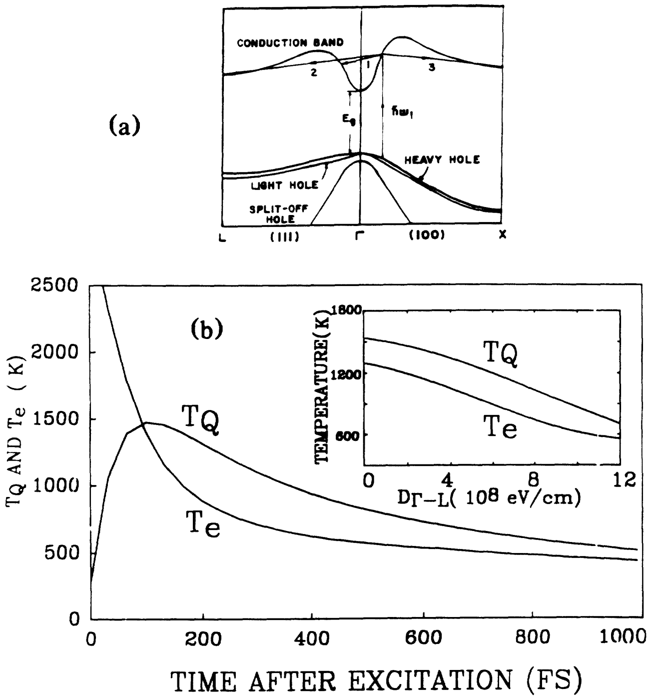

FIG. 3. (a) Schematic diagram of the band structure of GaAs used in the model calculation. Arrows 1 and 2 and 3 represent, respectively, intravalley scattering of electrons by LO phonons and intervalley scatterings of electrons by zone-edge phonons to the L and X conduction minima. (b) Calculated time dependence of the temperatures of Γ valley electrons (T_e) and of LO phonons with $Q = 7.5\times10^5$ cm^{-1} (T_Q) probed by Raman scattering assuming an initial electron density $N_e = 3\times10^{18}$ cm^{-3} and $D_{\Gamma\text{-}L} = 7\times10^8$ eV/cm. Inset: Calculated T_e and T_Q 200 fs after excitation plotted as a function of $D_{\Gamma\text{-}L}$ for $N_e = 3\times10^{18}$ cm^{-3}.

Other energy-loss mechanisms, such as emission of TO phonons and acoustic phonons by either electrons or holes and electron-hole scattering, are too slow to be significant.[22,23] The electron cooling rate caused by electron-hole scattering, as obtained by Osman and Ferry[24] using Monte Carlo simulations, is much smaller than that arising from intervalley scattering. Experimentally (see Fig. 2) we found that T_e *increased* with N_e rather than decreased, as would be expected if electron-hole scatterings were the dominant energy-loss mechanism. We found that intervalley scattering is the predominant mechanism for subpicosecond cooling of the hot electrons because they remove the high-energy electrons from the Γ valley in about 100 fs or less.[1,14] The return of electrons from the X and L valleys back to the Γ valley is negligible because such processes take more than 1 ps.[1] We have considered the effect of screening of the Fröhlich electron-LO phonon interaction in our calculation.[24,25] We have obtained results with both an unscreened Fröhlich interaction and with a Fröhlich interaction which includes Thomas-Fermi screening. We found that screening has almost no effect on T_e and only minor effect on T_Q at the time scale of our experiment. Finally, we have considered also the cooling of electrons in the X and L valleys by emission of LO phonons but found the effect to be insignificant.

Based on the above approximations we have calculated the time dependence of the electron and phonon temperatures by solving the following equations:

$$\frac{\partial N_q}{\partial t} = \frac{1}{q(q^2+k^2)}[-N_qA_q + (N_q+1)B_q]\,, \tag{1}$$

$$\frac{\partial N_\Gamma}{\partial t} = \int \left[\frac{\partial f}{\partial t}\right]_{\Gamma\text{-}L,X} \rho_\Gamma\, dE_\Gamma\,, \tag{2}$$

$$\frac{\partial (N_\Gamma\langle E_\Gamma\rangle)}{\partial t} = \int \left[\frac{\partial f}{\partial t}\right]_{\Gamma\text{-}L,X} \rho_\Gamma E_\Gamma\, dE_\Gamma - \hbar\omega \int \frac{\partial N_q}{\partial t}\rho_q\, dq\,. \tag{3}$$

In the above equations A_q and B_q are, respectively, the rates of absorption and emission of LO phonons by electrons in the Γ valley caused by intravalley scattering, k is the Thomas-Fermi screening wave vector, ρ_q is the density of states of LO phonons, ρ_Γ is the density of states of the Γ valley, N_Γ and $\langle E_\Gamma\rangle$ are, respectively, the number of Γ electrons and their average energy, and $[\partial f/\partial t]_{\Gamma\text{-}L,X}(E_\Gamma)$ is the rate of decrease of the Γ valley electron occupation number due to intervalley scattering evaluated at E_Γ. All the material parameters needed for the calculation are fairly well known except for the intervalley deformation potential $D_{\Gamma\text{-}L}$.[26,27] Thus we have treated $D_{\Gamma\text{-}L}$ as the only adjustable parameter.

Figure 3(b) shows the calculated time dependence of T_e and T_Q at $N_e = 3\times10^{18}$ cm^{-3} with $D_{\Gamma\text{-}L} = 7\times10^8$ eV/cm. It is clear that T_e decreases very rapidly in the first 100 fs while T_Q increases to $T_Q = T_e \approx 1500$ K. When T_e decreases from 3000 to 1500 K there is a loss of about 260 meV in $\langle E_\Gamma\rangle$. Within this short time an average electron in the Γ valley has emitted less than one LO phonon with energy of 35 meV. The contribution to the subpicosecond cooling of hot electrons in the Γ valley by intravalley emission of LO phonons is insignificant. The Fröhlich interaction is, however, responsible for the rapid rise in T_Q. For $N_e > 10^{18}$ cm^{-3}, the number of electrons capable of emitting LO phonons with wave vector Q is much greater than the density of phonon states at Q. *The most interesting result obtained by our model calculation is that T_Q overshoots T_e after 100 fs.* The reason for this is that the electrons have cooled so fast that after 100 fs there are fewer high-energy electrons to reabsorb the LO phonons emitted earlier. As shown in Fig. 3(b) it actually takes about 1 ps for the two systems to reach thermal equilibrium.

Using this model we have calculated T_e and T_Q at 200 fs after excitation as a function of N_e. The results are shown as the solid lines drawn through the experimental points in Fig. 2. In agreement with experiment, we found that T_e was relatively insensitive to N_e while T_Q

increased with N_e, overshot T_e around $N_e = 10^{18}$ cm^{-3}, and saturated at densities above 4×10^{18} cm^{-3}. To achieve the good fit to the experimental points, $D_{\Gamma\text{-}L}$ is adjusted to 7×10^8 eV/cm. The accuracy of the value of $D_{\Gamma\text{-}L}$ we determined depends on the value of $D_{\Gamma\text{-}X}$ since both Γ-to-X and Γ-to-L intervalley scatterings contribute to the phonon temperature overshoot. For the value of $D_{\Gamma\text{-}X} = 10\times10^8$ eV/cm used in our calculation, T_Q turns out to be higher than T_e even if Γ-to-L scattering is completely neglected. The effect of Γ-to-L scattering is to contribute additional cooling of the hot electrons and so to reduce both T_e and T_Q. This effect of $D_{\Gamma\text{-}L}$ is demonstrated by the theoretical curves shown in the inset of Fig. 3(b). From this curve we estimate an error of about 30% in our value of $D_{\Gamma\text{-}L}$ mainly resulting from the uncertainties in T_e and T_Q. Our value of $D_{\Gamma\text{-}L}$ is in good agreement with the value of 6.5×10^8 eV/cm determined by Shah *et al.*[1] but about 50% larger than the value deduced recently by Ulbrich, Kash, and Tsang.[27] Finally, we have measured the cooling rate of hot electrons in InP and InGaAs and found also good agreement with predictions.[28]

In conclusion, we have found that the phonon temperature became higher than the electron temperature when the electron densities were above 10^{18} cm^{-3}. Our results are in good quantitative agreement with results of a model calculation which includes intervalley scatterings.

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