Brief Reports

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Hot-phonon effects in bulk GaAs

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A theoretical model that accounts for both phonon disturbances and the electron-hole interaction is used to evaluate the role of hot-phonon effects in GaAs excited by a subpicosecond laser beam. Although the electron-hole interaction is not able to thermalize electrons and holes before significant phonon emission occurs it is mainly phonon heating that is found to be responsible for the experimentally observed reduction of the carrier cooling rates.

Experiments on both polar bulk materials and quantum-well structures have shown that the cooling rates of hot electron-hole pairs, excited by ultrashort laser pulses ($t_p < 1$ ps), are reduced as compared to simple theoretical estimates.[1] These estimates are based on the assumptions that (i) the photoexcited carriers thermalize instantaneously among each other, (ii) the carrier-phonon interaction is reduced by free-carrier screening, and (iii) the phonons remain in thermal equilibrium at any point in time. Within the last few years several attempts have been made to improve this simple theoretical picture. Free-carrier screening has been confirmed to reduce the cooling rates to some extent; however, not sufficiently to explain experimental results quantitatively.[1–4] On the other hand, inclusion of hot-phonon effects has lead to rather close agreement between theory and experiment for laser-excited GaAs.[3] The high LO-phonon emission rate of the hot electron-hole gas drives these modes out of equilibrium. Partial reabsorption of LO phonons at a later point in time reduces the effectiveness of the polar-optical coupling in the cooling process. In this stage of the time evolution, emission of TO phonons, which couple only to holes, also provides a significant contribution to the energy loss.[3] The relative simplicity of this model was based on the assumption that electrons and holes thermalize immediately after their creation. More recently it has been emphasized that this assumption is invalid in the initial stage of the cooling process and that it may lead to an overestimation of the importance of hot-phonon effects.[5,6] As a result of the band structure of GaAs, the electrons obtain the main portion of the photon energy in excess of the main energy gap. If the energy given to the hole is less than the optical-phonon energies, the cooling rates of the carriers will be significantly reduced. The large difference in the effective mass of electrons and holes will prevent rapid thermalization of the two carrier systems. This has been demonstrated by Asche and Sarbei, who estimated the energy transfer rate between electrons and holes.[5] More recently, ensemble Monte Carlo (EMC) studies have supported this idea.[6] For times less than one picosecond, the average energy of an electron-hole pair obtained from the EMC study, which did *not* include phonon heating, was higher than that obtained from the aforementioned analytical model that assumed thermalized electrons and holes, but *included* phonon heating.

This finding requires a reevaluation of hot-phonon effects. Clearly, the retarded thermalization of electrons and holes causes the holes to couple weakly to the lattice and phonon heating to be reduced. Here we address the question of whether these details in the carrier-carrier interaction render phonon heating irrelevant for explaining the slowing of the cooling process that has been observed experimentally. We report on the results of an analytical model for the cooling of laser-excited hot electron-hole pairs in GaAs. The model is an extension of a previously presented approach.[3] It includes coupling of the carriers to both optical- and acoustical-phonon modes, free-carrier screening in the long-wavelength limit according to Lindhart, heating of all optical-phonon modes, and electron-hole interaction via the self-consistently screened Coulomb interaction. Only electrons (e) and heavy holes (h) are considered in this study.

Electrons (holes) are assumed to be thermalized among each other at any point in time; however, the electron temperature (T) may differ from the hole temperature. (Therefore the present model will be referred to as the $2T$ model. The previous model will be called the $1T$ model.) This improvement is to account for the way electrons and holes share the excess photon energy. The initial thermalization processes of electrons (holes) from a narrow

Gaussian-like distribution function into a Fermi-Dirac distribution is ignored.[6,7] Except for time scales shorter than 0.1 ps, this can be considered reasonable because electron-electron (hole-hole) collisions (same mass of scatterers) will be associated with higher energy transfer rates than electron-hole collisions.

In the calculation of the energy transfer rate between electrons and holes, the electrons are allowed to be degenerate, but the holes are assumed to be nondegenerate. This is valid for the situation investigated here. Following Asche and Sarbei, but avoiding the assumption that $K_e/m_e \ll K_h/m_h$ (K_e, K_h, m_e, and m_h are the k vectors and effective masses of the electrons and holes, respectively), one can express this rate as a double integral which must be calculated numerically.[8] The long-wavelength limit of Lindhart's formula is used to implement screening.

The phonon Boltzmann equations are solved iteratively in time.[3] The nonelectronic decay of optical-phonon modes is included by a relaxation-time ansatz, using an experimental value for the LO-phonon lifetime of 7 ps.[9] For every phonon of type j, the phonon occupation number $N(q,j)$ is assumed to be isotropic in the wave vector **q**. The carrier-phonon interaction is screened statically, again using the long-wavelength limit of Lindhart's expression. The remaining material parameters are taken from Ref. 3.

The model outlined above allows the determination of the relative importance of the electron-hole interaction, phonon heating, and free-carrier screening. It has been used to perform a study of bulk GaAs excited by a 0.5-ps laser pulse that creates 1.35×10^{18} electron-hole pairs per $cm.^{-3}$ The excess photon energy over the gap is 150 meV and the lattice temperature is 10 K. Assuming parabolic bands with $m_e^* = 0.067m_0$ and $m_h = -0.45m_0$, the energies given to the electron and hole are 130.6 and 19.4 meV, respectively. The kinetic energy of the holes initially lies clearly below the LO-phonon energy of 36.2 meV. Without carrier-carrier interaction the holes would essentially not participate in the cooling process. Here it is assumed that before electrons (holes) can emit a significant number of LO phonons, the e-e (h-h) interaction thermalizes the electrons (holes) into Fermi-Dirac distributions at well-defined temperatures.

We made a crude comparison of the electron, hole, and e-h thermalization times. The latter was determined by turning off all carrier-phonon interactions and calculating the time required for electrons and holes to thermalize among each other. The electron (hole) thermalization time was estimated by creating 1.35×10^{18} electrons (holes) per cm^{-3}, exciting half the particles in a Fermi-Dirac distribution function of an average energy of 130.6 meV/particle and the other half into one with an average energy of 19.4 meV/particle. The resulting thermalization times were about 0.1, 0.5, and 2 ps for the electron, hole, and electron-hole system, respectively. These times are in good agreement with previous estimates and calculations.[5-7]

The main features of the time evolution of the carriers within the first 10 ps are shown in Figs. 1 and 2. Figure 1 displays the average kinetic energy per e-h pair as a function of time. Comparison of the results obtained from different versions of the model reveals the physics governing the cooling process. The assumption of instantaneous thermalization of the carriers made in the $1T$ model, which implies that the excess photon energy is equally shared among electrons and holes, strongly overestimates the cooling rate during the first picosecond. For times up to about 5 ps, the average energy per electron-hole pair as obtained from the $2T$ model is significantly higher. Carrier temperatures versus time are shown in Fig. 2. Up to about 2 ps, the electron temperature is considerably

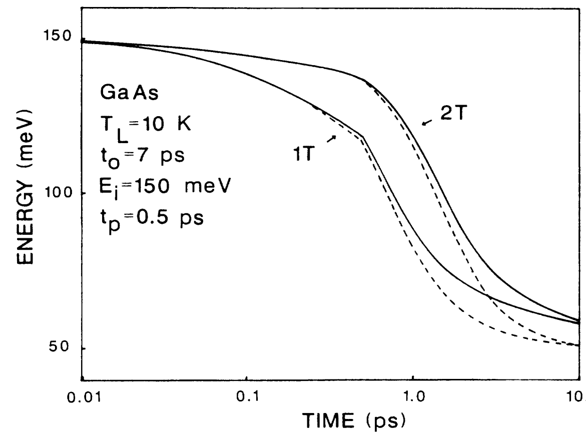

FIG. 1. Average kinetic energy per electron-hole pair as a function of time (solid lines, hot phonons included; dashed lines, hot phonons neglected). T_L, t_o, E_i, and t_p are the lattice temperature, optical phonon lifetime, the initial kinetic energy per e-h pair, and the pulse duration, respectively.

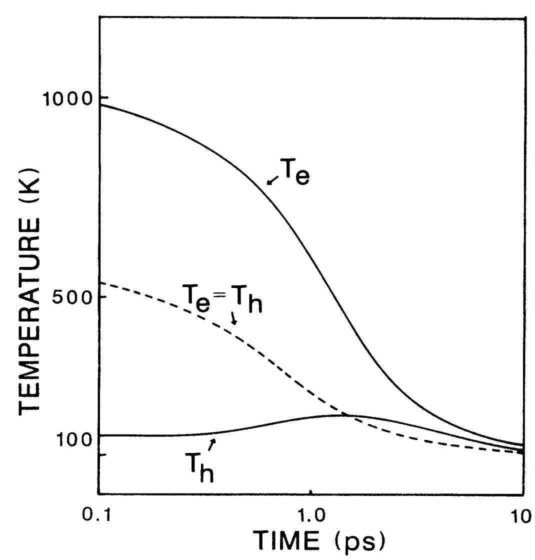

FIG. 2. The carrier temperatures as a function of time from the $2T$ model [solid lines for electrons (T_e) and holes (T_h)]. The dashed line is for the $1T$ model.

higher than the hole temperature. Even at 10 ps the electron temperature is about 20 K higher than the temperature of the holes. At this point in time, LO phonons are reabsorbed (mainly) by the electrons, preventing complete thermalization with the holes. After the first picosecond, phonon reabsorption becomes important. The cooling rate of the $1T$ model is found to drop below that predicted by the $2T$ model. In this time regime, phonon reabsorption is indeed overestimated by the $1T$ model. From about 10 ps on, however, the $1T$ model is as good as the $2T$ model. In this stage of the cooling process, the details of the carrier dynamics are well concealed. Hot-phonon effects are mainly responsible for the relatively high carrier temperatures observed in experiments that so far mainly investigated this time regime. Therefore the main conclusions drawn from the $1T$ model were correct for times larger than 10 ps.

The time evolution of the LO phonons as a function of phonon wave vector **q** is illustrated in Fig. 3 for $t=0.2$, 2, and 5 ps. Figure 4 shows the phonon population as a function of time for $q=2.4\times10^6$ cm^{-1}. Starting at about 0.05 ps, the phonon population rises significantly above its equilibrium value and reaches its maximum at about 2.3 ps, i.e., 1.8 ps after the end of the laser pulse. At about 15 ps, most of the LO phonons have either been reabsorbed or decayed into other phonon modes. The time evolution predicted from the $1T$ model occurs somewhat faster. The mode shown in Fig. 4 is dominated by LO-phonon emission of the electrons, which favorably emit LO phonons with q up to about 2.5×10^6 cm^{-1}, as compared to holes that favorably emit LO phonons with q around 6.5×10^6 cm.$^{-1}$ The occupation number of this particular mode is larger in the $2T$ model. Further details can be seen in Fig. 3. At $t=0.2$ ps a significant number of LO phonons has already been emitted, mostly by hot electrons, which is indicated by the peak at about $q=1.5\times10^6$ cm^{-6}. At 2.0 ps, the phonon population is fully developed, peaking at about 2×10^6 cm.$^{-1}$ A small shoulder, caused by the holes, can be seen at about $q=6\times10^6$ cm.$^{-1}$ This shoulder is more pronounced at 5 ps, where a good part of the LO-phonon population has already decayed. Comparison with the $1T$ mode shows that the latter overestimates the phonon rise and decay times as well as the importance of the holes in the cooling process. In other words, the $1T$ model overestimates the creation of LO phonons around $q=6\times10^6$ cm^{-1}, but underestimates LO-phonon population around $q=6\times10^6$ cm^{-1}. Overall, however, phonon heating is not significantly overestimated as compared to the $2T$ model. It should be noted that the $1T$ model predicts too much heating of the TO phonons. The deformation-potential coupling, although found to provide a significant contribution to the total energy loss at times $t>5$ ps, is less effective than originally thought because the holes remain rather cool (i.e., always below about 200 K). This is in good agreement with experimental finding.[10,11]

In summary, we have shown that, for times that have been accessible in most previous experiments (i.e., about 10 ps after the onset of the excitation pulse),[1] LO-phonon heating provides the dominant mechanism to explain the slow cooling of the carrier system. In the initial stage of the cooling process, the main energy loss must be attributed to the electrons which have considerably higher average kinetic energy than the holes, whereas the holes are too cool to emit a significant number of optical phonons. For approximately the first 0.5 ps this causes a dramatic reduction of the cooling rate and leads to a strong buildup of short-wavelength LO phonons (around $q=2\times10^6$ cm^{-1}). For up to about 5 ps, the average energy per *e-h* pair stays clearly above the prediction from a model that assumes instantaneous thermalization of electrons and holes.[3] About 0.5 ps after the end of the pulse, phonon heating effects become the dominant mechanism for reducing the cooling rates (particularly for electrons). Reabsorption of previously emitted LO phonons slows the cooling process as well as the thermalization of electrons and holes. This latter effect may explain the slow thermalization rate reported for carrier concentrations well above 10^{18} cm^{-3}.[12] LO-phonon heating, therefore, is confirmed as the dominant mechanism that slows the cooling of the carriers that has been observed typically 10

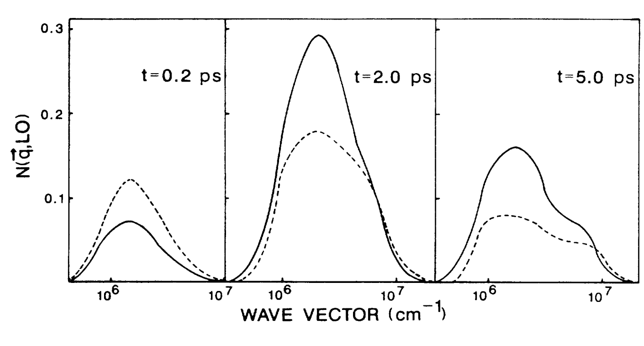

FIG. 3. The LO-phonon occupation number $N(q,\mathrm{LO})$ vs phonon wave vector **q** at $t=0.2$, 2.0, and 5.0 ps (solid lines for the $2T$ model, dashed lines for the $1T$ model). Note that a logarithmic scale is used for the q axis.

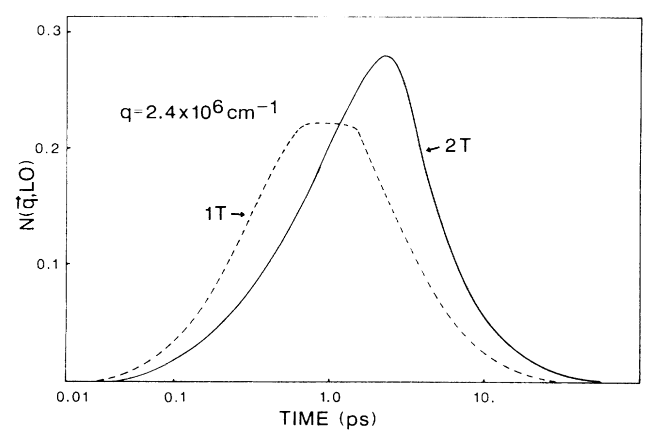

FIG. 4. Time evolution of the occupation number $N(q,\mathrm{LO})$ for the LO-phonon mode with $q=2.4\times10^6$ cm^{-1} (solid line, $2T$ model; dashed line, $1T$ model).

ps after the pulse. A significant buildup of LO phonons occurs around $q = 2\times 10^6$ cm^{-1}. Under the present conditions TO-phonon heating is found to be negligible. For times below 5 ps, the relatively slow thermalization of electrons and holes provides the dominant mechanism for reducing the energy transfer to the lattice. Finally it should be mentioned that, if phonon-heating is neglected in this model, our results agree well with those from the EMC studies.[6] The *e-h* energy exchange rate in the EMC study is somewhat lower than that obtained with the present approach. A more detailed presentation of results from this model and a comparison with the EMC model will be given elsewhere.

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