Direct observation of a reduced cooling rate of hot carriers in the presence of nonequilibrium LO phonons in GaAs:As

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We have directly observed the influence of nonequilibrium optical phonons on the cooling rate of hot carriers in GaAs:As. Carriers with density $>10^{18}$ cm^{-3} were generated by an 80-fs, 1.72-eV pulse and cooled *as well as* recombined on a 1-ps time scale leaving behind *only* nonequilibrium phonons. The cooling rate of low-density (10^{17} cm^{-3}) carriers generated by a second delayed pulse was studied through time-resolved luminescence. The rate changes with the intensity of the first pulse and delay of the second pulse in a manner consistent with reabsorption of the nonequilibrium phonons.

Hot-carrier energy relaxation in polar semiconductors such as GaAs has been investigated intensively for more than a decade using ultrafast spectroscopy techniques.[1] At low carrier densities the cooling rate is well described by the simple theory of LO-phonon emission as set out by Kogan.[2] However, for carrier densities well above 10^{17} cm^{-3} the energy-loss rate is strongly reduced. The reduction of the cooling rate of high-density carriers in polar and nonpolar semiconductors was originally suggested[3] to be due to the generation of a high density of nonequilibrium phonons ("hot phonons") in a small region of the Brillouin zone. However, in polar semiconductors screening[4] of the Fröhlich interaction between electrons and LO phonons has also been suggested as a mechanism to reduce the cooling rate. Detailed theoretical[5] and experimental[1,6] results have tended to favor the hot-phonon process as playing the key role although increasingly complex models have been used to try to reach this elusive conclusion. The difficulty is that for carrier densities greater than a few times 10^{17} cm^{-3} the carrier-lattice interaction has a many-body nature, with screening, Landau damping, the formation of coupled plasmon-phonon modes, etc., complicating the dynamics of energy transfer from the carrier to the phonon systems. The many-body processes are so complex that recent theoretical results[7] have tended to reach contradictory conclusions about the influence of, e.g., coupled modes on energy relaxation, and some authors have suggested that the formulation of the problem itself may be a problem.[8] Time-resolved Raman experiments certainly have detected nonequilibrium optical phonons in GaAs following carrier cooling and the lifetime has been determined to be 7 ps.[9] The influence of hot phonons on free-carrier absorption has also been seen[10] in InAs but to the best of our knowledge the influence of nonequilibrium phonons on carrier energy relaxation has never been *directly* observed.

Optical pump-probe techniques do not easily allow a separation of phonon and carrier effects on carrier cooling in intrinsic samples since both effects scale with the excitation level or carrier density. We have therefore used an extrinsic sample of GaAs with a carrier lifetime short compared to the phonon lifetime and a novel prepump-pump pulse technique whereby one can *independently* vary the carrier and nonequilibrium phonon densities to examine the effects of phonons alone on the carrier cooling rate. The cooling rate was determined from femtosecond, time-resolved luminescence spectroscopy.

The material used in our study was GaAs, which was grown at a low temperature (200 °C) by molecular-beam epitaxy on a GaAs substrate, and then annealed at 600 °C for 10 min.[11] Films of thickness between 1 and 5 μm were grown and gave essentially identical results in the experiments discussed below. The material, hereafter referred to as GaAs:As, is crystalline but nonstoichiometric with an approximately 1% excess of arsenic in the form of microclusters with density greater than 10^{17} cm^{-3}. The clusters behave[12] as buried Schottky barriers that can capture electrons and holes in approximately 1 ps; no significant carrier trapping is observed at longer times.[13,14] Samples were mounted in a cryostat and kept at 10 K. At this temperature the carrier lifetime is comparable to the energy relaxation time but short compared to the optical phonon lifetime of approximately (Ref. 15) 5 ps. We have taken advantage of these characteristics to study the cooling rate and temperature of hot carriers as a function of the nonequilibrium phonon population.

The principle of our method is illustrated in Fig. 1(a). A first (prepump) pulse with photon energy larger than the band gap of GaAs:As creates carriers with a density of about $n_{pp}=10^{18}$ cm^{-3}. Hot phonons of density $N_{\rm ph}$ are generated during the cooling process.[16] After a few picoseconds the carriers will have essentially disappeared but a substantial population of nonequilibrium phonons remains. The sample is then excited with a second (pump) pulse that injects a carrier density of approximately $n_p=10^{17}$ cm^{-3}, sufficiently low that many-body effects such as screening do not influence the carrier-phonon interaction. By changing the intensity of the prepump pulse or the separation (τ) between prepump and pump pulses, it is possible to produce a variable density of phonons that can interact with the carriers generated by the pump pulse. In this way, one can probe the

influence of hot phonons *alone* on carrier energy relaxation. The influence of the excess phonon population on carrier cooling is deduced from the characteristic temperature (T_c) extracted from the high-energy tail of the carrier luminescence.[14,17,18] Note that unlike the Raman experiments, this experiment is sensitive to optical phonons with a wide range of wave vectors.

Time- and energy-resolved luminescence spectra were obtained using femtosecond upconversion spectroscopy.[19] The optical pulses used to excite the sample and to upconvert the luminescence were obtained from an 82-MHz mode-locked Ti:sapphire laser operating at $\lambda=720$ nm (1.72 eV) with a pulse duration of 80 fs, and an average power of up to 2 W. The photon energy is chosen to be well below the threshold energy of Γ-L intervalley transfer.[17] Using dispersion-free parabolic mirrors to collect the luminescence and a 1-mm thick BBO crystal to upconvert the luminescence, we achieve a time resolution of less than 100 fs and an energy resolution of <30 meV for the whole energy range of interest here.

Figure 1(b) shows two different luminescence spectra at $\tau'=0.5$ ps after a pump pulse that is estimated to produce an initial density of $n_p=2\times10^{17}$ cm^{-3} (at the surface and the center of the laser spot; the absorption depth is about 0.5 μm). In the one case the pump pulse is delayed by $\tau=5$ ps relative to a prepump pulse that injects a carrier density of $n_{pp}=2\times10^{18}$ cm^{-3}; in the other case the prepump pulse is absent. The fitted T_c has a value of 680 K when the prepump is on and 510 K when absent, directly showing the influence of the nonequilibrium phonons induced by the prepump pulse. The nearly equal integrated intensities of both spectra confirm that the number of carriers remaining from the prepump pulse when the pump pulse arrives (about 1.3×10^{16} cm^{-3} based on a 1 ps lifetime) is negligible.

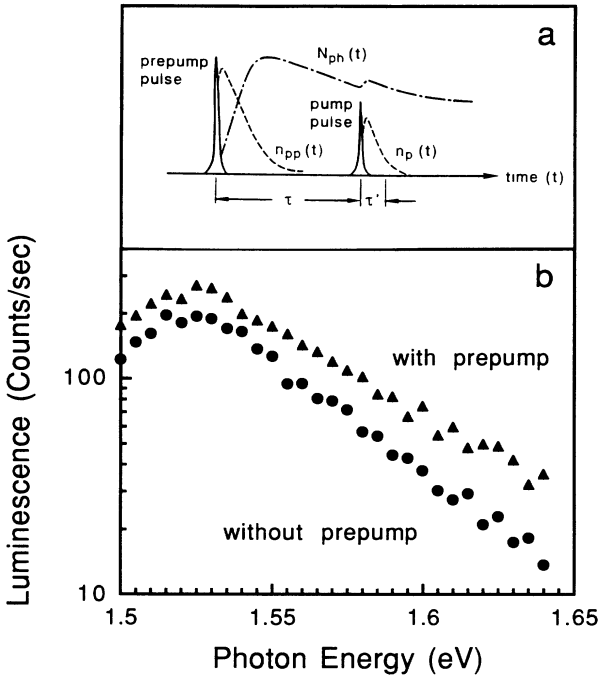

FIG. 1. (a) Schematic diagram of the two-pulse experiment with a prepump and pump pulse sequence. The time after the prepump pulse is denoted as τ, while time after the pump pulse is given as τ'. Following the prepump and pump pulses the time-dependent carrier densities are $n_{pp}(\tau)$ and $n_p(\tau')$, respectively, while the nonequilibrium phonon density following the prepump pulse is $N_{ph}(\tau)$. (b) Luminescence energy spectra obtained at $\tau'=0.5$ ps from the pump pulse; the upper data set (triangles) is obtained when the pump pulse is delayed from the prepump pulse by $\tau=5$ ps, while the lower set (circles) is obtained without a prepump pulse. The upper data set has plotted values that are 1.3 times higher than the measured values so as to allow the data sets to be separately displayed.

Figure 2 shows T_c valued at $\tau'=1$ ps as a function of time delay τ from the prepump pulse. Also shown is the value (360 K) of T_c obtained when no prepump is present. An exponential fit to the τ-dependent T_c data assumed to decay to the 360 K value yields a decay time of 3.9 ± 0.4 ps. This value correlates well with the phonon lifetime of 5 ps obtained from the Raman experiments.[15] The discrepancy is not considered significant in view of the small number of T_c data and the fact that our experiment neither monitors exactly the same phonons nor the identical physical process as the Raman experiment.

Figure 3 shows how T_c varies with τ' in the case where a prepump pulse arrives $\tau=5$ ps before the pump pulse; for comparison we show similar data when the prepump is absent. The values of T_c at $\tau'=200$ fs are nearly the same since these are near the initial temperature (approx. 1300 K), which is mainly governed by the difference between photon and band-gap energies. The separation of the T_c curves shortly thereafter can be understood if one considers how the nonequilibrium phonons are produced following the prepump pulse. At the onset of cooling, carriers are distributed over a large energy and momentum range and phonons are generated in a thick spherical shell in the Brillouin zone. The phonons are never far from equilibrium, and reabsorption of phonons is not significant in the first few hundred femtoseconds. As the

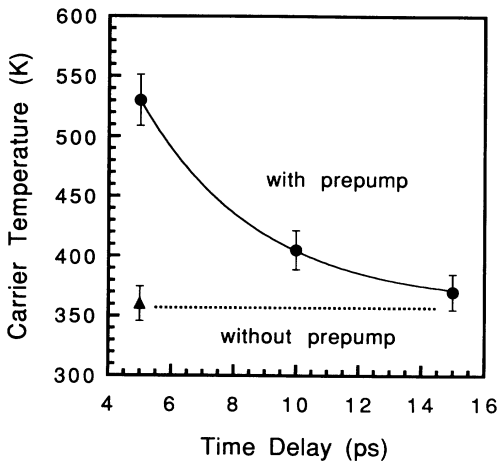

FIG. 2. Characteristic temperature T_c obtained at a delay of $\tau'=1$ ps after the pump pulse as a function of prepump-pump separation τ. The curve is based on an exponential fit with an e^{-1} decay time of 3.9 ps. The triangle and dotted line indicate the T_c obtained in the absence of the prepump.

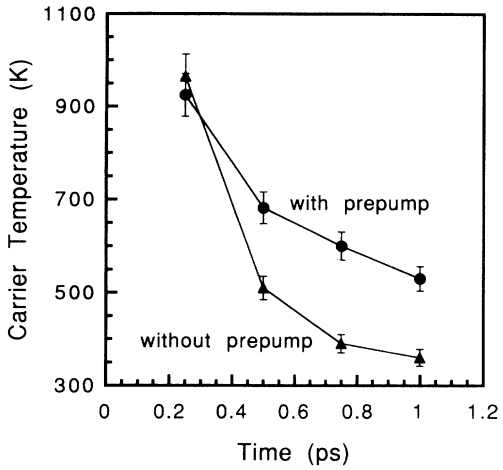

FIG. 3. Temporal evolution of $T_c(\tau')$ for carriers created by the pump pulse, which is delayed with respect to a prepump pulse by $\tau=5$ ps; the curves are guides to the eye.

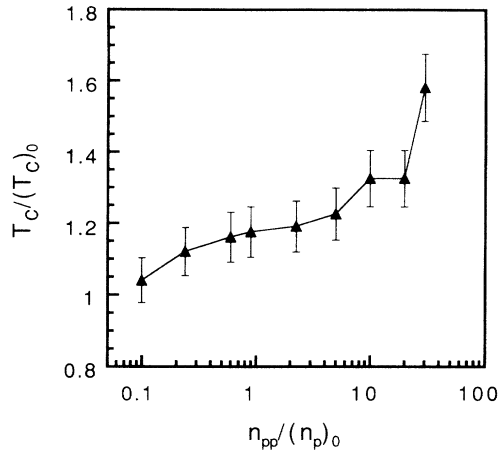

FIG. 4. Dependence of T_c at $\tau'=0.7$ ps on n_{pp} for $\tau=7$ ps; the curve is a guide to the eye. The T_c is normalized to a value of $(T_c)_0=400$ K measured without the prepump pulse. The values of n_{pp} are normalized to the pump density of $(n_p)_0=2\times10^{17}$ cm^{-3}.

electrons cool they become increasingly restricted to interacting with phonons in a thinner shell and these phonons are driven further from equilibrium. The carriers are strongly inhibited from reaching the lattice temperature until the nonequilibrium phonons decay. An additional reason for the subpicosecond onset of the reduced cooling rate is related to the buildup time of the nonequilibrium phonons. From Fig. 3 we observe that the risetime of N_{ph} is about 500 fs, consistent with previous estimates.[20]

Figure 4 shows how T_c depends on n_{pp} (through variation of the prepump pulse intensity) for $\tau'=0.7$ ps and $\tau=7$ ps. The temperature is observed to increase monotonically with n_{pp}. This indicates that carriers created by the pump pulse interact with an increasing population of nonequilibrium optical phonons with increasing n_{pp}, leading to an increased reabsorption rate. The behavior is consistent with previously reported reduced cooling rates with increasing carrier density in pump-probe experiments on GaAs.[6,20] To understand the details of the variation of T_c with n_{pp} (which reaches values near 5×10^{18} cm^{-3}) it would be necessary to consider how many-body carrier effects influence the rate of emission and the spectrum of phonons following the prepump pulse. For $n_{pp}<5\times10^{17}$ cm^{-3} electrons interact primarily with bare LO phonons through the Frohlich interaction but for higher carrier densities the interaction is with coupled LO-phonon-plasmon modes and TO phonons.[21] In all cases, however, n_p is sufficiently small that carriers induced by the pump pulse only interact with bare phonons and the main point of this work is to indicate directly how a nonequilibrium phonon population can influence the carrier cooling.

In conclusion, we have performed a two-pulse, time-resolved luminescence experiment on GaAs:As. The influence of hot phonons on reduced carrier cooling is separated from carrier-density-dependent processes. Our approach therefore offers an alternative technique for detailed studies of hot-carrier relaxation and carrier-phonon interactions. The data directly illustrate the important role of hot phonon effects on electron cooling. Experiments are now underway to compare the results here with those from normal GaAs.

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