Heating of cold electrons by a warm GaAs lattice: A novel probe of the carrier-phonon interaction

W. W. Rühle and H. -J. Polland
Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 12 March 1987)

Cooling of an electron-hole plasma after picosecond photoexcitation strongly decreases at carrier densities exceeding 1×10^{17} cm^{-3}. In contrast, the reverse process of heating a cool electron-hole plasma via phonon absorption from the warm lattice is independent of carrier density up to 4×10^{17} cm^{-3}, establishing the first experimental evidence for negligible screening of the Fröhlich interaction.

The relaxation processes of hot electrons and holes in compound semiconductors have been extensively studied by optical methods.[1-6] This interest is partly due to the search for a better understanding of the charge-carrier–lattice interaction, as, e.g., the Fröhlich coupling in polar semiconductors, and is partly stimulated by the quest for improved devices involving hot carriers. A strong reduction of the cooling process observed in time-resolved photoluminescence (PL) experiments in two- and three-dimensional systems at higher excitation densities remains to be explained. Nonequilibrium phonon population,[2] screening,[1,6] heating induced by radiative[7] or nonradiative[8] recombination, as well as effects due to a reduced dimensionality,[4] have been suggested as possible explanations.

In this paper we report a new conceptually simple approach to solve this fundamental problem. Time-resolved photoluminescence (PL) experiments are used to study the Fröhlich interaction with picosecond time resolution. The basic idea is to monitor the reverse process of carrier cooling, i.e., energy transfer from a warm lattice to cool carriers. Phonons are then mainly absorbed by carriers, thus contrary to plasma cooling an overpopulation of phonons cannot occur. A comparison of heating and cooling at identical plasma densities should reveal whether nonthermal phonons, screening, or any kind of recombination mechanism reduces the Fröhlich interaction at high densities. Finally, a complicating coupling of these effects as, e.g., enhanced phonon overpopulation due to screening,[9] is avoided.

Cool carriers are generated by using an excitation photon energy only slightly above the band gap of GaAs. The heating mechanism can simply be changed by the lattice temperature T_L: at 35 K deformation potential (DP) scattering dominates; at 55 K polar optical (PO) scattering dominates. In this temperature range the heating times are in the picosecond to nanosecond regime, i.e., are easily accessible to PL experiments with a streak camera.

We report measurements on undoped ($\leq10^{15}$ cm^{-3}) bulk GaAs. The liquid-phase epitaxial layers are imbedded in a (Ga,Al)As double heterostructure to avoid surface recombination and are only 0.2 to 0.3 μm thick in order to have homogeneous excitation with no disturbing diffusion of carriers. Samples are mounted on the cold finger of a temperature-regulated cryostat (±0.1 K). The heterostructures are excited by a tunable, synchronously pumped dye laser (80-MHz repetition rate, 4-ps pulse width). The wavelength of the dye laser is tuned to 819 nm, i.e., to a resonant excitation of the GaAs gap. Excitation density per pulse is changed between 1×10^{12} and 1×10^{14} photons per cm^2 by adjusting the laser focus or using neutral density filters. The minimum diameter of the focus was 15 μm, i.e., diffusion on the time scale of several 100 ps is negligible. Carrier concentration changes from 1×10^{16} to 4×10^{17} cm^{-3}. Bleaching reduces absorption at the highest densities.[10]

Figure 1 shows time-integrated spectra. The carriers assume a Maxwell-Boltzmann or Fermi-Dirac distribution since electron-electron scattering is fast. The carrier temperature T_C can be deduced from the exponential high-

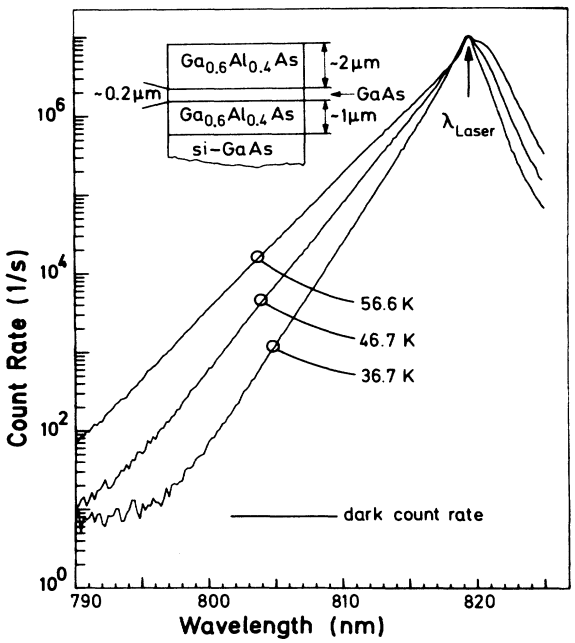

FIG. 1. Time-integrated PL spectra at different lattice temperatures. Excitation density is $n=1\times10^{16}$ cm^{-3}. The inset shows the sample structure.

energy tail.[2] Since recombination lifetime ($\geqq 4$ ns) is much longer than heating time, the time-averaged spectra yield the lattice temperature with high accuracy.

The temporal variation of the PL is monitored by a Hadland synchroscan streak camera with an optical multichannel analyzer as a readout system. The PL is wavelength dispersed by a $\frac{1}{4}$-m grating spectrometer limiting the time resolution to about 20 ps. The streaks on the high-energy tail of the PL are taken at 1-nm distance with 1-nm resolution. A prepulse of the laser on every streak assures a common time reference for all streaks. Examples of typical streaks are shown in the inset of Fig. 2. The initial offset is due to the fact that the decay time is comparable to the separation of subsequent laser pulses.

From the streaks and the time-averaged spectra we calculate the transient high-energy tails, as shown in Fig. 2. The time evolutions of the carrier temperatures T_C are plotted in Fig. 3(a) for low plasma densities of 1×10^{16} cm^{-3} at various lattice temperatures T_L. The solid and dashed lines represent theoretical fits as described in the following sections.

In the three-dimensional case the specific heat of the plasma is $3nk$ (n is the concentration of one carrier type).[11] Three processes lead to an increase in carrier temperature. First, the PO scattering can be written as[4,6]

$$\left[\frac{dT_C}{dt}\right]_{PO}=\frac{\hbar\omega_{LO}}{3k\tau_{av}}\left[\frac{e^{X_L-X_C}-1}{e^{X_L}-1}\right]F ,$$

where $\hbar\omega_{LO}$ is the energy of the optical phonon, $X_{L,C}=\hbar\omega_{LO}/kT_{L,C}$, and $F=0.98$.[12] The average scattering time τ_{av} per carrier[5,13] is determined by the effective masses and the dielectric constants of the material.[6] Second, the temperature increase due to DP scattering can be written as[14]

$$\left[\frac{dT_C}{dt}\right]_{DP}=C_1(T_L-T_C)T_C^{1/2} .$$

Third, piezoelectric (PE) scattering is given by[14]

$$\left[\frac{dT_C}{dt}\right]_{PE}=C_2(T_L-T_C)T_C^{-1/2} .$$

C_1 and C_2 are constants in K/ps which can be related to the effective masses, deformation potentials, and the PE modulus of GaAs.[14] Nonpolar optical scattering of holes can be neglected.[14] Fits to the experimental data clearly show the well-known dominance of PO scattering for higher T_L ($\geqq 45$ K). Assuming τ_{av} as an adjustable parameter, we obtain from the uppermost curves in Fig. 3(a) $\tau_{av}=(0.15\pm0.05$ ps), a value about 2 to 3 times larger than the theoretical one,[13] but, in agreement with other experimental data.[1]

For $T_L\leqq 30$ K PO scattering is negligible. The experi-

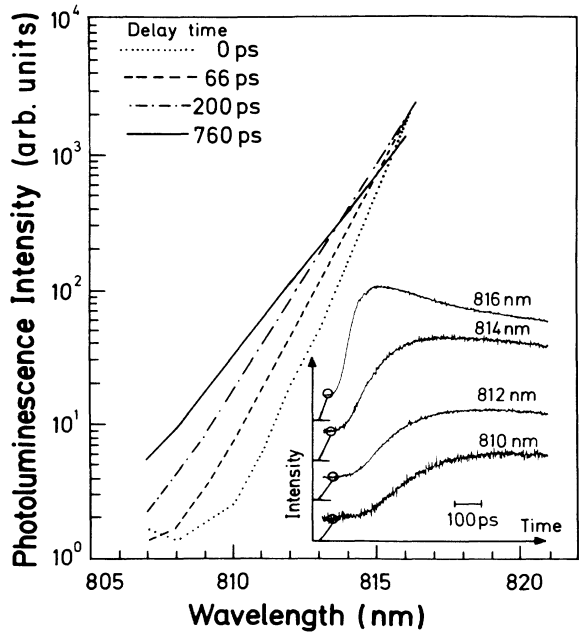

FIG. 2. Transient short-wavelength tails of PL spectra at $T_L=37.6$ K. The inset shows streaks at different wavelengths.

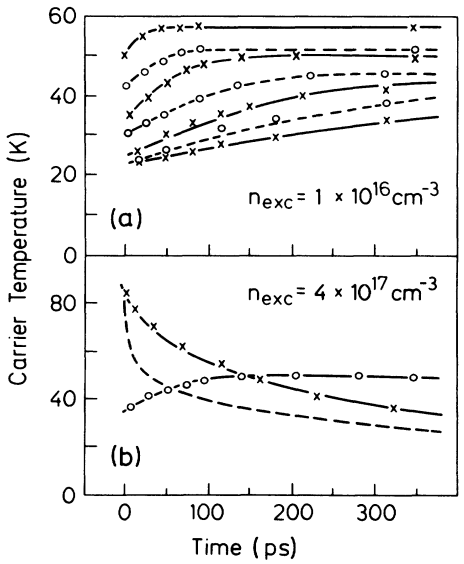

FIG. 3. (a) Heating of carriers at low plasma density (1×10^{16} cm^{-3}) and different lattice temperatures T_L (from top to bottom): 58.0, 52.5, 50, 46, 42.5, 40.5, 36.5 K. Solid and dashed lines are theoretical fits. The elevated temperatures at the beginning are caused by two effects. First, our zero time was chosen 50 ps after the very beginning of the laser pulse in order to be sure that the laser pulse (due to the limited time resolution) no longer influences the heating process. Second, due to the long lifetime there are hot carriers left from the preceding excitation pulse. (b) Heating (circles) and cooling (crosses) at a high plasma density of 4×10^{17} cm^{-3}. The heating curve (solid line fit to circles) uses an average relaxation time of 0.15 ps, whereas the fit to the cooling data requires $\tau_{av}=2$ ps. For comparison, a cooling behavior with a relaxation time $\tau_{av}=0.15$ ps is given by the dashed curve.

mental variation of T_C with time as given by the lowest curve in Fig. 3(a) requires that DP dominates PE scattering. This is expected using recently published values of the average effective hole DP, $E_{ac}=4.8$ eV.[15] Figure 3(a) shows all data between the two limiting cases of PO and DP scattering with excellent fits, using $\tau_{av}=0.15$ ps.

We now proceed to the case of high excitation. Figure 3(b) shows the heating curve at a plasma density of 4×10^{17} cm^{-3} (circles). Best theoretical fit of the experimental data is again obtained for the same τ_{av} as in case of low excitation. As in case of low excitation, excellent fits (not shown for clarity) are possible for various lattice temperatures between 30 and 57 K without any change of parameter. Therefore, PO and DP scattering are not affected by plasma density within our experimental accuracy.

In contrast, we observe a significant change with excitation density in the cooling behavior: In Fig. 3(b) we included cooling data measured with the excitation wavelength at 800 nm (i.e., the generated electrons are initially hot) and at a lattice temperature of 4.2 K. Care was taken to actually obtain the same plasma density as for the heating, i.e., bleaching effects were taken into account.[10] At 80 K, PO scattering is by far dominant. With our value of τ_{av} we would expect a fast cooling as shown by the dashed curve. The actual fit to the data (solid line), however, requires a τ_{av} as large as 2 ps, i.e., an apparent reduction of PO scattering by more than a factor of 10. This slower cooling has been already observed by several other authors.[2,4,6,16]

Our simultaneous measurements of carrier heating and cooling allow us to distinguish between the different mechanisms inducing the decrease of carrier cooling at high densities. Screening can be excluded since it would reduce both, absorption and emission of phonons, i.e., heating as well as cooling should be decelerated. Additional heating processes due to radiative or nonradiative recombination[8] are also negligible as clearly evidenced by the slow increase of carrier temperature at low lattice temperatures ($\leq$30 K). We thus attribute the reduction of carrier cooling to nonequilibrium nonthermal phonons, which are created only in a cooling experiment due to the high generation rate and long lifetime of optical phonons (i.e., the formation of a phonon bottleneck).

In conclusion, we have presented a new experiment, the observation of heating of a cool plasma by a hotter lattice, where a phonon overpopulation cannot occur. From this experiment we obtain PO and DP scattering rates which are independent on plasma density from 1×10^{16} cm^{-3} to at least 4×10^{17} cm^{-3}. We have also demonstrated that up to densities of 4×10^{17} cm^{-3} the following mechanisms cannot be responsible for a reduced plasma cooling in bulk GaAs: screening and heating via radiative or nonradiative recombination. Instead, the reduction of cooling appears to be caused by nonthermal phonons.

We gratefully acknowledge the sample preparation by E. Bauser and K. Löchner and the expert technical assistance by K. Rother and H. Klann. Thanks are due to H.-J. Queisser, J. Kuhl, and K. Leo for valuable discussions and for a critical reading of the manuscript.

[1]J. Shah, J. Phys. (Paris) Colloq. **42**, C7-445 (1981).

[2]K. Kash, J. Shah, D. Bloch, A. C. Gossard, and W. Wiegmann, Physica **134B&C**, 189 (1985).

[3]J. Shah, A. Pinczuk, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **54**, 2045 (1985).

[4]J. F. Ryan, R. A. Taylor, A. J. Tuberfield, A. Maciel, J. M. Worlock, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **53**, 1841 (1984).

[5]C. H. Yang, J. M. Carlson-Swindle, S. A. Lyon, and J. M. Worlock, Phys. Rev. Lett. **55**, 2359 (1985).

[6]H. J. Zarrabi and R. R. Alfano, Phys. Rev. B **32**, 3947 (1985).

[7]D. Bimberg and J. M. Mycielski, Phys. Rev. B **31**, 5490 (1985).

[8]W. W. Rühle, H.-J. Polland, and K. Leo, in Proceedings of the International Conference on the Physics of Semiconductors, edited by O. Engström (World Scientific, Göteborg, 1987), Vol. 1, p. 211.

[9]C. H. Yang and S. A. Lyon, Physica **134B&C**, 309 (1985).

[10]The effect of bleaching was determined experimentally by measuring excitation spectra, i.e., measuring the PL intensity at 805 nm (and $T_L=60$ K) and varying the excitation wavelength from 795 to 830 nm. These excitation spectra are a direct measure for the relative absorbed power. At the lower excitation levels absorption is almost constant for all wavelengths shorter than 820 nm. However, at the highest excitation level a decrease in absorption by a factor of 2.5 is observed for wavelengths between 819 to 820 nm with respect to shorter wavelength (795–800 nm) where bleaching cannot occur. The physical origin of bleaching is the limited density of states close to the renormalized band gap. Although the absolute values of carrier densities might inhibit a larger error, the densities for cooling and heating in Fig. 3(b) are very accurately equal. In particular, in this case the focus was not changed and the same excitation density was adjusted by neutral density filters.

[11]For simplicity, we use Maxwell-Boltzmann statistics. Comparative calculations with Fermi-Dirac statistics show no essential differences due to the long lifetime and the moderate excitation densities of 4×10^{17} cm^{-3}. The difference between the two statistics becomes important at higher excitation and will be discussed in a subsequent publication.

[12]F is a lengthy expression given in Ref. 6. However, for $T_L=25$ to 55 K, F changes monotonically from 0.99 to 0.97 only.

[13]For $1/\tau_{av}=1/\tau_e+1/\tau_h$ we expect $\tau_{av}=0.06$ ps.

[14]E. O. Göbel and O. Hildebrand, Phys. Status Solidi B **88**, 645 (1978).

[15]The average effective hole DP is defined by J. D. Wiley, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1975), Vol. 10, p. 91. With the newly compiled data on GaAs, in *Semiconductors*, Vol. 17 of *Landolt-Börnstein, New Series*, edited by O. Madelung (Springer-Verlag, Berlin, 1982), and the formulas given by Wiley, we obtain an effective DP $E_{ac}=4.8$ eV.

[16]J. F. Ryan, in *Two-Dimensional Systems: Physics and New Devices*, Vol. 67 of *Springer Series in Solid-State Sciences* (Springer-Verlag, Berlin, 1986), p. 315.