Direct Measurement of Hot-Electron Relaxation by Picosecond Spectroscopy

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The time dependence of the optical absorption and the gain of a hot electron-hole plasma in GaAs is measured by means of tunable picosecond light pulses. The electrons and holes ($\sim 10^{17}$ cm^{-3}) are photoexcited via two-photon absorption. Our measurements provide a detailed picture of the temperature evolution, showing that in 250 ps the plasma cools from over 100 K down to ~40 K and does not reach the lattice temperature. The energy relaxation process is shown to be polar LO-phonon emission of hot electrons and holes.

Information on the relaxation of electrons and holes in semiconductors has previously been obtained mainly from hot-electron charge-transport measurements[1] and also from steady-state optical spectroscopy.[2] The utility of directly studying energy relaxation in the time domain was first demonstrated by Ulbrich's photoluminescence measurements[3] of a low-density electron gas in GaAs on a nanosecond time scale. For very hot carriers, however, the relaxation is expected to occur on a time scale of 10^{-12} s. Picosecond laser spectroscopy provides a useful new tool for measuring these very rapid processes.[4] Shank *et al.*[5] concluded from picosecond reflectivity measurements on GaAs that an electron-hole (e-h) plasma having an initial temperature of about $k_B T = 0.6$ eV loses energy at a rate of approximately 0.4 eV/ps. In a very recent experiment[6] hot carriers were shown to relax to the lattice temperature (80 K) in about 4 ps. Our own preliminary picosecond absorption spectra of GaAs were tentatively interpreted[7] in terms of cooling and recombination of an e-h plasma. However, in all these cases the details of the temperature variation with time were not obtained.

In this Letter we report on picosecond measurements providing for the first time a comprehensive picture of the temperature evolution of a hot electron-hole (e-h) plasma in GaAs. Our experiments clearly demonstrate a distinct deceleration of the relaxation rate with time. We find that the electron-hole plasma does *not* reach the lattice temperature ($T_L = 7$ K) within the total observation time of about 250 ps. These observations are consistent with the polar LO-phonon relaxation mechanism.

Experimentally we use an excite-and-probe scheme[4] in which a first intense picosecond pulse ($t_p = 25$ ps) from a mode-locked Nd-YAlG (neodymium-yttrium-aluminum garnet) laser creates a hot e-h plasma via two-photon excitation. A second delayed, frequency-tunable pulse from a synchronously mode-locked optical parametric oscillator[8] measures the resulting changes of the optical transmission spectrum. Two-photon excitation provides uniform e-h density over the depth of the 8-μm-thick GaAs crystal (n-type, carrier concentration 10^{16} cm^{-3}, room-temperature mobility 3×10^3 cm^2/V s). Radial diffusion and recombination do not reduce the plasma density for our experimental geometry and time scale.

Figure 1 depicts the experimental data that we wish to discuss here. We plot the induced changes of the absorbance[9] A as a function of probe-pulse delay time, t_D, for an 8-μm-thick slab of GaAs. The data were measured at three different probe photon energies ($\hbar\omega$) below the exciton ground state, viz., 1.513, 1.510, and 1.507 eV.

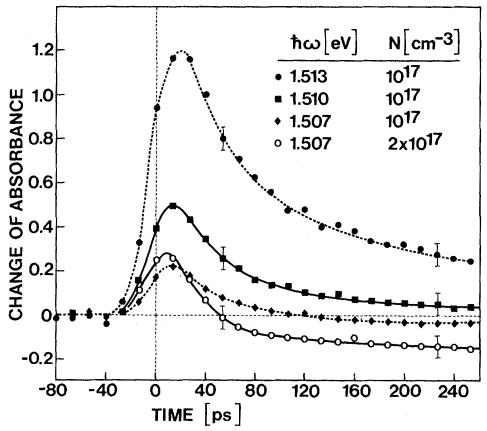

FIG. 1. Induced absorbance vs delay time. Time zero refers to the maximum of the pump pulse; $\hbar\omega$, photon energy of the probe pulses; N, density of e-h pairs determined from two-photon transmission measurements. Dotted curves, guide to the eye only; full curves, examples of the calculated temporal evolution of the absorbance. The calculated temperatures at the maxima of the curves are 160 and 120 K for 10^{17} cm^{-3} and 2×10^{17} cm^{-3}, respectively.

The e-h pair density N was determined to be 10^{17} cm^{-3} ($\pm 50\%$) from separate two-photon transmission experiments. For $\hbar\omega = 1.507$ eV we also show results for $N = 2\times10^{17}$ cm^{-3}. Each datum point in Fig. 1 represents an average of 100 individual measurements. The error bars correspond to twice the standard deviation. The energy of the excitation pulse must be kept constant within better than 10% in order to achieve an experimental accuracy of a few percent.

For all probe wavelengths we initially observe a rapid increase of the absorption approximately following the integrated pump pulse. At $t_D = 20$ ps the induced absorption begins to decrease. The decay pattern is distinctly nonexponential and characteristic of each probe wavelength. In particular, at 1.507 eV the induced absorption decays to zero and then becomes negative. Comparing the induced negative absorption with the background absorption in the absence of excitation we conclude that optical gain is observed. The final value of the optical gain and also the zero crossings of A depend very sensitively on the excitation level (e.g., Fig. 1 shows a fourfold increase of the gain upon doubling of N).

In the following analysis we show that these experimental observations represent direct evidence of the cooling of the e-h plasma, and we are able to determine the plasma temperature T as a function of time, and to identify the energy dissipation mechanism.

We now outline the physical concepts on which our analysis will be based. Two-photon pumping provides electrons and holes with large excess energies, viz., 0.716 and 0.095 eV. The light holes will be neglected because the density of states of the heavy-hole band is much larger. The carriers interact so strongly at high densities[10] that thermal equilibrium among the electrons and holes is established on a time scale much faster than the pump process. The temperature is determined by the energy W transfered to the plasma for each excited e-h pair. W is estimated[2] to be approximately 0.16 to 0.25 eV for densities ranging from 10^{17} cm^{-3} to 2×10^{17} cm^{-3}. W is substantially smaller than the total initial excess energy because during thermalization carrier-carrier collisions compete with efficient LO-phonon emission.[2] The measured changes of the absorption are related to the plasma temperature by calculating the optical absorption spectrum of a hot e-h plasma as a function of density, N, and temperature, T. The absorption coefficient is taken to be of the form[11,12]

$$\alpha(\omega) = C(\hbar\omega - E_g')^2 \int_0^1 (1-x)^{1/2} x^{1/2} (f_e - f_h) dx, \quad (1)$$

where C is a constant proportional to the transition matrix element. The renormalized energy gap E_g' takes into account exchange and correlation effects of the interacting Fermi system.[13] In Eq. (1) the Fermi distribution functions f_e and f_h are calculated for $\epsilon - \mu_e$ for electrons, and $-(\hbar\omega - E_g' - \epsilon - \mu_h)$ for holes. μ_e and μ_h are the chemical potentials, and $\epsilon = x(\hbar\omega - E_g')$ is the energy of the conduction electrons. Equation (1) is obtained by simply taking $\alpha(\omega)$ to be proportional to the number of optically coupled states and neglecting k selection rules. Although a rigorous justification is still lacking, Eq. (1) was shown to agree well with the experimental spectra of highly excited semiconductors.[14]

The calculated $\alpha(\omega)$ exhibits the following interesting features. For $\hbar\omega$ greater than the total chemical potential $\mu = E_g' + \mu_e + \mu_h$, $\alpha(\omega)$ is positive. For $E_g' < \hbar\omega < \mu$ there is optical gain, $\alpha(\omega) < 0$, and μ can be determined from the condition $\alpha(\omega) = 0$. At any given photon energy $\hbar\omega > E_g'$, $\alpha(\omega)$ decreases when T decreases. For the determination of C in Eq. (1) absorption and gain spectra were measured by varying the probe photon energy for different, fixed delay times between pump and probe. Excellent agreement with the theoretical spectra[15] was obtained for the very same value of C, viz. $C = 6\times10^7$ cm^{-1} eV^{-2}. Experimental spectra measured at longer delay times matched with calculated spectra of lower temperature, suggesting that the changes of the spectra with time are due to a decrease of the plasma temperature.

For a detailed analysis of the temperature variation Eq. (1) is solved for T with the measured values of A.[16] The results are shown in Fig. 2: We plot the values of T obtained from the four different data sets of Fig. 1. There is excellent consistency of the temperatures derived from measurements differing both in frequency and excitation level. The fact that the decay curves for completely different experimental conditions can be accurately described by the very same temperature function is strong evidence that the dynamics of absorption and gain are indeed determined by the cooling of the plasma.

Let us now consider how the observed cooling can be related to energy relaxation processes of the electrons and holes. The rate of change of

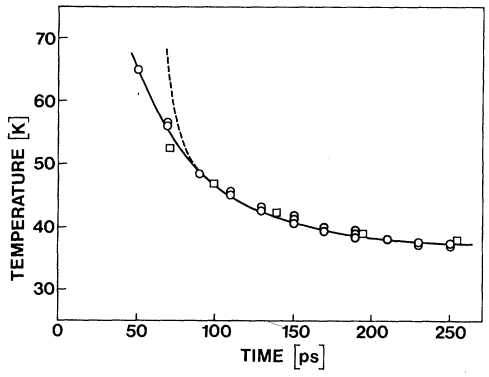

FIG. 2. Plasma temperatures vs time. Circles, temperatures evaluated from the measured absorbances using Eq. (1); squares, temperatures evaluated from the measured chemical potentials (see note in Ref. 16); full curve, temperature calculated from Eq. (6); dashed curve, solution of Eq. (6) with $H(t)=0$ (free cooling).

the average particle energy u can be written

$$du/dt = H(t) - L(t), \tag{2}$$

where $H(t)$ describes the increase in energy due to optical pumping, and $L(t)$ is the energy loss rate due to relaxation processes. For a Fermi gas du/dt can be expressed in terms of dT/dt by

$$\frac{du}{dt} = \tfrac{3}{2} k_B \left[\frac{\frac{5}{2} F_{3/2}(\eta)}{F_{1/2}(\eta)} - \frac{\frac{3}{2} F_{1/2}(\eta)}{F_{-1/2}(\eta)} \right] \frac{dT}{dt}, \tag{3}$$

where F_j are the Fermi integrals of order j, and $\eta = \mu_e / k_B T$ (for electrons). The bracket is very close to unity for our situation ($\eta < 2$), and will be neglected. $H(t)$ is given by the e-h pair generation rate, times the energy W, divided by the instantaneous density.[2] For two-photon pumping we have

$$H(t) = WI^2(t) \left[\int_{\infty}^{t} I^2(\tau) d\tau \right]^{-1}. \tag{4}$$

We show later that the energy dissipation is dominated by emission of polar LO phonons. The energy loss rate is then approximately given by[2,17]

$$L(t) = (\hbar\omega_{LO}/\tau_0) \exp(-\hbar\omega_{LO}/k_B T). \tag{5}$$

Here $\hbar\omega_{LO}$ is the energy of the LO phonons, and τ_0 is the time constant of the LO-phonon relaxation. Equation (5) differs from the exact loss rate (averaged for a Fermi distribution) by less than 10% for the temperature regime considered here. Combining Eqs. (2) to (5) we obtain a differential equation for $T(t)$:

$$\tfrac{3}{2} k_B \frac{dT}{dt} + \left(\frac{\hbar\omega_{LO}}{\tau_0} \right) \exp\left(\frac{-\hbar\omega_{LO}}{k_B T} \right) = H(t). \tag{6}$$

The solution of Eq. (6), together with Eq. (1), provides a complete theoretical description of the instantaneous $\alpha(\omega)$. For a comparison with the measurements, however, the finite time resolution must be taken into account by convoluting $\exp(-\alpha(\omega)l)$ with the shape of the probe pulse. Two examples of such calculations are given by the solid lines in Fig. 1. Both curves fit the experimental points very well over the entire time range. W was chosen to be 150 meV for the squares, and 300 meV for the open circles (note that for a larger N, a larger fraction of the excess energy is expected to be transferred to the e-h system[2]). The adjustment of W and the specific choice of the pulse shapes[18] affect the quality of the fit only near the maximum. The decaying portions are independent of both.

We established that the instantaneous $\alpha(\omega)$ and the convolution are identical for $t_D > 50$ ps. The data of Fig. 2 therefore represent the instantaneous temperature. For a comparison with these data the theoretical $T(t)$ [solution of Eq. (6)] is given by the solid line in Fig. 2. Equation (6) obviously gives an excellent account of the observed temperature variation. The time constant τ_0 was treated as an adjustable parameter, and from the fit of the data we obtain $\tau_0 = 0.12 \pm 0.05$ ps. This value compares favorably with the theoretical value[19] $\tau_0 = 0.11$ ps for the LO-phonon process in GaAs. From the good agreement between the solution of Eq. (6) and the measured temperatures we conclude that LO-phonon emission is indeed the dominant energy relaxation process in the temperature range investigated here.[20]

An important characteristic of the LO-phonon process is that the energy loss rate decreases exponentially with T. It explains the very slow decrease of T for long times and the fact that the lattice temperature is not reached. On the other hand, considering just the loss rate, T is expected to vary much faster than observed for $t_D < 100$ ps. The dashed curve in Fig. 2 is the solution of Eq. (6) with $H(t) = 0$, and $T = 65$ K as initial condition, i.e., it represents "free cooling" of the plasma with T determined by the relaxation process alone. The comparison with the complete solution (full curve) shows that for times much longer than the pulse duration T is still affected by the pump process.

In conclusion, the picosecond measurements

have revealed the time dependence of the optical gain and the absorption of a photoexcited e-h plasma. We were able to determine the plasma temperature as a function of time, and we have identified the observed cooling to be due to polar LO-phonon emission of electrons and holes in the energetic tail of the Fermi distribution.

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[9]$A = \ln(T_0/T)$, where T and T_0 are the transmission with and without excitation, respectively.

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[15]The experimental data were fitted allowing for a 5-meV shift to higher energies. A similar energy correction had to be used in previous work on GaAs, see, e.g., O. Hildebrand, B. O. Faltermeier, and M. H. Pilkuhn, Solid State Commun. 19, 841 (1976); O. Hildebrand, E. O. Göbel, K. M. Romanek, H. Weber, and G. Mahler, Phys. Rev. B 17, 4775 (1978).

[16]T was also obtained by a different method [without using Eq. (1)]: μ is determined from the experimental spectra using the fact that $\alpha(\omega) = 0$ for $\mu = \hbar\omega$. T is then evaluated with the help of the well-known expression for the chemical potential of a Fermi gas. The results of this independent analysis are given by the squares in Fig. 2.

[17]E. M. Conwell, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1967), Suppl. 9.

[18]For the calculations we used a rectangular shape (t_p = 20 ps) for the probe pulse, and a hyperbolic secant shape, viz., $I(t) = I_0/\cosh(2.63t/t_p)$ for the pump pulse (t_p = 25 ps). Streak camera (Ref. 5) studies established that these shapes represent good approximations of the experimental pulse shapes.

[19]$1/\tau_0 = (1/\tau_{0e} + 1/\tau_{0h})/2$, where $\tau_{0e} = 0.13$ ps is the contribution of the electrons (Refs. 2 and 17) and the hole contribution is given by $1/\tau_{0h} = (1/\tau_{0e})\,(m_h/m_e)^{1/2}/K$, with $K \sim 2$: See J. Wiley, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1975), Vol. 10.

[20]Contributions of acoustic phonons to the energy relaxation were also examined. The relaxation rate due to emission of acoustic phonons by holes is expected to be equal to the sum of the LO-phonon rate of electrons and holes at about 35 K. Including hole acoustic-phonon relaxation above 40 K gives temperatures differing by less than 2 K from those calculated for the LO process alone.