Influence of hot phonons on energy relaxation of high-density carriers in germanium

H. M. van Driel
Department of Physics and Erindale College, University of Toronto, Mississauga, Ontario, Canada L5L 1C6
(Received 12 July 1978)

Recent experiments on germanium have indicated anomalously long energy relaxation times for dense electron-hole plasmas produced by intense optical picosecond pulses. Since the carriers relax primarily through optical-phonon emission it is suggested here that the long equilibration time is due to a relaxation bottleneck produced by a buildup of the optical-phonon population on a picosecond time scale. A comparison of theory and experiment gives excellent agreement.

Over the past several years the investigation of ultrafast transient effects in semiconductor plasmas using picosecond pulse lasers has provided direct insight into many electronic processes.[1-5] Germanium, in particular, has been employed in many of these investigations since its band-gap energy is comparable to, but less than, the photon energy of the 1.06-μm glass:Nd laser. Since this laser is easily mode locked to produce intense pulses $\sim$ 5 psec in duration, it is possible through absorption of such pulses to achieve electron-hole plasmas of density greater than $10^{20}/cm^3$. Such a photoexcited plasma is initially at a temperature of at least 1800 K because of the difference in the photon energy 1.17 eV, and the minimum energy (band-gap energy), $\sim$0.7 eV, needed to create an electron-hole pair. Recently, Elci *et al.*[1] (henceforth referred to as ESSM) have reported results of extensive experimental and theoretical investigations on the generation and subsequent temporal evolution of hot plasmas in 5-μm-thick Ge. In one type of experiment the nonlinear transmission of a single-picosecond 1.06-μm pulse was investigated as a function of intensity. In a second type of experiment to study the temporal evolution of the plasma, the sample is first irradiated by an excitation pulse of sufficient intensity to cause enhanced transparency. It is then followed at various delay times by a probe pulse at the same wavelength but at considerably reduced intensity so as not to significantly alter the density of the plasma. Probe-pulse transmission data for various delay times and two different sample temperatures are shown in Fig. 1, reproduced from ESSM. It should be pointed out that these data are essentially in agreement with similar data obtained by Shank and Auston.[6] Very briefly, the probe-pulse transmission can be interpreted in the following way. After the absorption of the excitation pulse, the electrons (holes) are distributed in the conduction (valence) bands appropriate to the high distribution temperature, thereby leaving some optically coupled states available for absorption. Initially, then, the probe transmission is small. As the electron-hole plasma cools by phonon emission, the carriers fill the states needed for absorption and the probe transmission increases. The subsequent slow fall in the probe transmission is attributed to recombination of electron-hole pairs, which reduces the carrier density and once again frees the optically coupled states for absorption. Diffusion has also been suggested as a contributing factor in the fall (as well as the rise) of the probe-pulse transmission.[6,7] From Fig. 1 one is tempted to conclude that the carrier relaxation time is $\sim$40 psec for a sample temperature of 297 K and

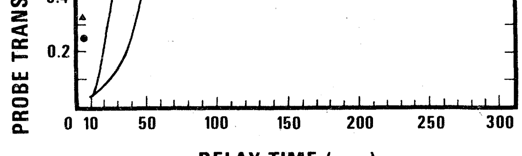

FIG. 1. Probe-pulse transmission data for two values of the sample temperature. Relative transmission units are given by the ratios of the excitation transmission to probe transmission for each delay, normalized such that the peak of the 77-K curve is unity. The solid lines indicate theoretical fits of ESSM.

~ 75 psec at 77 K. The observation of such long relaxation times has generated considerable interest since these times are considerably longer than the typical time of ~ 2 psec observed in other semiconductors.[8,9]

The detailed theoretical model of ESSM used to explain the probe-pulse transmission considers carrier relaxation via phonon emission with optical-phonon emission being the dominant process in accord with other experimental observations.[10] In order to simplify the calculations the complex conduction-band structure of germanium with its multiple-valley structure was replaced by ten equivalent valleys. This simplification is not expected to affect relaxation-time calculations significantly. An additional simplification which is also well justified[10,11] is to assume only intravalley phonon-assisted relaxation, since the intervalley scattering rate is small. A detailed calculation using first-order perturbation theory gives for the energy-loss rate of the plasma,

$$\frac{\partial E}{\partial t} = \Gamma_0(t) = -AQ_0^2 T_e(T_e - T_L)[m_h^3 \ln(1+e^{-h/kT_e}) + 5m_c^3 \ln(1+e^{-(E_g-\epsilon)/kT_e})] \ . \quad (1)$$

Here A is a universal constant, Q_0 is the optical phonon-electron coupling constant, T_e is the temperature of electrons (and holes), T_L is the lattice temperature, m_h and m_c are the hole and conduction-band edge effective masses, h and ϵ are the hole and electron Fermi levels, and E_g is the energy gap. This equation can be easily transformed to give the time dependence of the electron temperature. Because of the considerable energy dissipated through phonon-assisted relaxation, ESSM have taken into account the possible influence of a rise in the lattice temperature. This simply gives

$$\frac{dT_L}{dt} = \frac{\Gamma_0(t)}{C_L} \ , \quad (2)$$

where C_L is the specific heat of germanium. The temperature rise of the lattice can be expected to be ~ 20 K for plasma densities of $\sim 10^{20}/\text{cm}^3$. These equations, together with similar equations for the time variation of the electron and hole Fermi levels, can be used to obtain the temporal dependence of the electron and hole Fermi distributions and hence the time dependence of the absorption constant for the 1.06-μm probe pulse. Figure 1 shows their theoretical fits to the probe-pulse data for an initial plasma density of $\sim 10^{20}/\text{cm}^3$ with $T_e = 5000$ K at $T_L = 297$ K and $T_e = 3000$ K at $T_L = 77$ K.[12] In general the fit can be regarded as satisfactory. However, a number of features indicate that the model is incomplete. First of all, the fitted electron-phonon coupling constants are 3.7×10^8 eV/cm for $T_L = 297$ K and 1.2×10^8 eV/cm at $T_L = 77$ K. These values are well below $(6.2 \pm 1.3) \times 10^8$ eV/cm, which represents the mean and average deviation from the mean of eight values reported by different experimenters.[13] Since the cooling rate of the plasma is proportional to Q_0^2, the two fitted values lead to relaxation times which are approximately 3 and 25 times the values one obtains by using $Q_0 = 6.2 \times 10^8$ eV/cm. Second, a strongly temperature-dependent Q_0 has to be employed to account for the different relaxation times observed at the two temperatures, whereas it is known that the electron-phonon constant is essentially a temperature-independent material property by the way it is defined.[10] Lastly, it can be seen that the data show a steep rise in probe transmission with a gradual leveling off, whereas the theory predicts a delayed, steep rise.

In order to account for the above discrepancies, many additional mechanisms not associated with the electron-phonon interaction have been and are being considered.[7,9] Among these considered are diffusion, band-gap renormalization, and enhanced indirect absorption. At the moment it is difficult to assess what contribution these various processes make towards an explanation of the probe-pulse data. What we wish to propose here is a simple extension of the ESSM model based upon the small reservoir of optical phonons that the carriers interact with while relaxing. Such an extension removes the objections outlined above while providing a consistent explanation of all probe transmission data, including those which show an apparent density as well as a temperature dependence of the relaxation time.

The basis of the model is shown schematically in Fig. 2. The laser beam provides photons of energy $h\nu$. Of this amount, E_g is used to produce electron-hole pairs, while the remainder, $h\nu - E_g$, goes into kinetic energy of the carriers. This leads to the establishment of a Fermi distribution at a temperature of

$$T_e = 2040 \text{ K for } T_L = 297 \text{ K}$$

and

$$T_e = 1800 \text{ K for } T_L = 77 \text{ K} \ ,$$

the difference being attributable to the temperature dependence of the band gap. The carriers relax in a characteristic time of τ_e by emission of optical phonons. Since the electron transitions are intraband

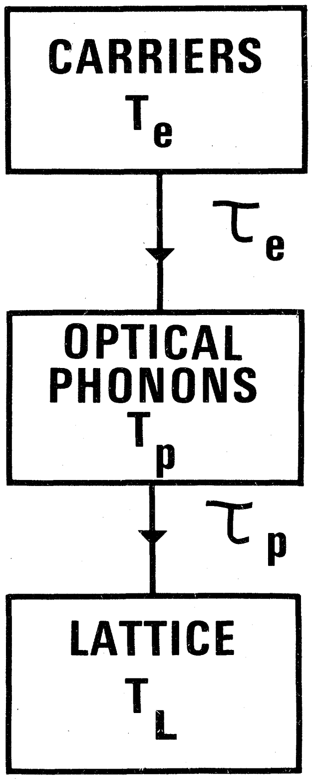

FIG. 2. Schematic description of the physical system considered in the text.

(and predominantly intravalley) they occur between states which are separated by much less than a reciprocal lattice vector, i.e., the phonons created are near the center of the Brillouin zone. The same is true for hole transitions which occur near the top of the valence band, located at the center of the Brillouin zone. Reasonable estimates based on the band structure of germanium and the initial carrier distribution indicate that only those optical phonons located within $\sim 10^{-2}$ of the volume of the Brillouin zone are involved in the hot carrier relaxation. Therefore, an optical-phonon population[14] in large excess of the equilibrium value is established near zone center. The optical phonons decay via the emission of two longitudinal acoustic phonons near the zone boundaries[15] with a characteristic time constant τ_p. This latter interaction can be considered to bring the optical phonons into equilibrium with the lattice. Quasiequilibrium distribution of all three systems is assumed to be established through collisons and multiple emission and absorption of optical phonons so that the electrons, optical phonons, and lattice have time-dependent temperatures T_e, T_p, and T_L, respectively.

In terms of this model, the relevant equations which take the place of Eqs. (1) and (2) are

$$\frac{\partial E}{\partial t} = \Gamma_0'(t) \;, \quad = -AQ_0^2 T_e (T_e - T_p)[m_h^3 \ln(1+e^{-h/kT_e}) + 5m_c^3 \ln(1+e^{-(E_g-\epsilon)/kT_e})] \;, \tag{3}$$

$$\frac{\partial T_p}{\partial t} = \frac{\Gamma_0'(t)}{C_p} - \frac{N(T_p)-N(T_L)}{\tau_p}\left[\frac{\partial T_p}{\partial N}\right] \;, \tag{4}$$

$$T_L(t) = T_L \;. \tag{5}$$

Here $N(T_p)$ is the Bose-Einstein factor appropriate to optical phonons (of energy 37 meV). It should be noted that the essential difference between Eqs. (1) and (3) is that the lattice temperature has been replaced by the optical-phonon temperature. This reflects the fact that it is the optical-phonon reservoir with which the carriers are attempting to reach equilibrium. In Eq. (4) for the time dependence of the phonon temperature, the first term tends to increase the phonon temperature reflecting carrier relaxation. The second term arises because of optical-phonon decay with

$$\frac{\partial N(T_p)}{\partial t} = -\frac{N(T_p)-N(T_L)}{\tau_p} \;. \tag{6}$$

This term tends to decrease the phonon temperature. The overall lattice temperature according to Eq. (5) is assumed to be constant. At this point we wish to note that as τ_p approaches zero then this set of equations reduces to Eqs. (1) and (2). However, according to Safran and Lax[15] $\tau_p \simeq 10$ psec at 77 K and $\simeq 5$ psec at 297 K. These relatively long lifetimes[16(a)] have the effect of maintaining a high phonon temperature as the plasma cools. This results in a relaxation bottleneck for the hot carriers. This can be seen from the time evolution of the electron and phonon temperatures. The numerical solution of the differential Eqs. (3), (4), and (5), along with similar equations for the electron and hole Fermi level, yields the temporal dependence of the electron and phonon temperatures shown in Fig. 3 for $T_L = 77$ K. In solving these equations, we assumed the same initial density as ESSM with a temperature-independent electron-phonon coupling constant of 6.2×10^8 eV/cm, the mean value obtained from other experiments. It can be seen that the electron temperature initially decreases quite rapidly from its initial value of 1800 K. During this time the optical-phonon temperature rises to a maximum value of ~ 1050 K. From then on the two temperatures cool at a relatively slow rate, the phonons because of their long lifetime and the electrons because their temperature, differs little from the phonon temperature. As a result it takes 75 psec for the carriers to come to equilibrium with the lattice. If we allow τ_p to approach zero then we find that the equilibration time

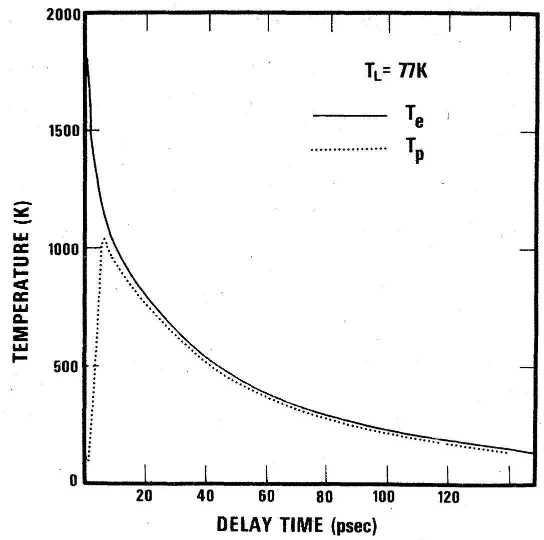

FIG. 3. Time dependence of the electron and optical-phonon temperatures.

of the carriers with the lattice is calculated to be ~5 psec, in agreement with observations in other semiconductors.[8] A similar calculation performed for 297 K with the same electron-phonon coupling constant but with the shorter phonon lifetime gives an equilibration time of ~40 psec. Once again, if τ_p approaches 0 then the equilibration time is ~5 psec. Further analysis based on our model leads to the probe-pulse transmission at the two lattice temperatures given in Fig. 4. The agreement between the theoretical model and experimental data dealing with the rise in the probe-pulse transmission is excellent in view of the simplicity of the extension of the ESSM model. It can be seen that the theoretical model yields the experimentally observed probe-pulse transmission rise times at the two lattice temperatures. In the case of Fig. 4 this is all we wish to demonstrate. No attempt was made, for example, to reproduce the absolute transmissivities because of uncertainties in the plasma density. The curves therefore represent normalized transmissivities. As was pointed out earlier as well, the fall in the probe-pulse transmission with time can be attributed to recombination, diffusion, etc., which are not within the realm of the model presented here.

To further test the model we have considered the data of ESSM on the variation of the probe-pulse transmission under various initial excitation conditions for $T_L = 77$ K, as shown in Fig. 5. Ratios labeling these curves are the relative excitation-pulse per probe-pulse energies. It is seen that the carrier relaxation time apparently decreases with decreasing initial excitation. Estimates of the initial carrier densities achieved by the excitation pulse are $2\times10^{20}/\text{cm}^3$, $4\times10^{19}/\text{cm}^3$, and $10^{19}/\text{cm}^3$ for highest to lowest excitation pulse energies, respectively. Theoretical fits to the data using our model with $T_e = 1800$ K initially are also shown in Fig. 5. Once again the fit to the data is seen to be very good. In terms of the model the decrease in relaxation time with decreasing density is due to a reduction in the amount of phonon heating, allowing the plasma to cool more quickly. Were it not for phonon heating effects, the relaxation time should be independent of carrier density, as has been pointed out by ESSM as well.

In summary, we have shown that a simple extension of the model of Elci *et al.*, taking into account optical-phonon heating but only a single temperature-independent electron-phonon interaction, is able to account for the major features of their data. It should be pointed out, however, that in view of the simple extension of the overall model the agreement of theory and experiment must be regarded as very good. In the spirit of the model, no at-

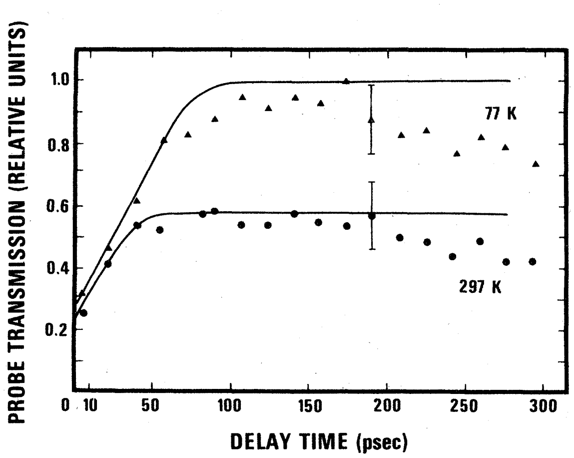

FIG. 4. Same data as in Fig. 1 with the solid lines indicating theoretical curves based on hot-phonon effects.

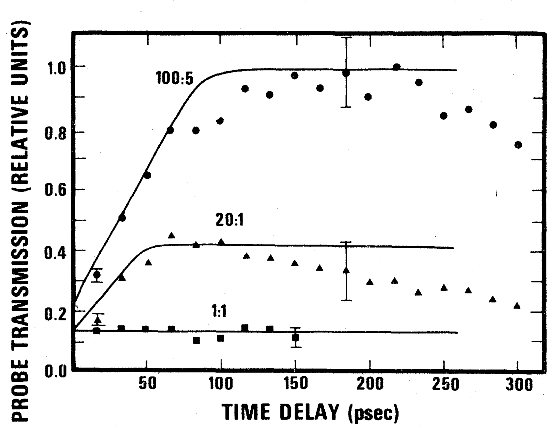

FIG. 5. Probe-pulse transmission data for $T_L = 77$ K. The ratios represent relative excitation per probe-pulse energies. The solid curves indicate theoretical fits based on the model discussed in the text.

tempt was made to perform the equivalent of a least-squares fit to determine the optimum parameters Q_0, τ_p, etc. Because of the scatter in the data one would not be able to determine these parameters to a better accuracy than already has been reported in the literature. Rather the emphasis has been to choose the values of these parameters from the literature and to use them to demonstrate the effects of phonon heating that we note here. It would be interesting to perform similar experiments on other semiconductors to see if the influence of hot phonons can be seen to alter the carrier relaxation rate. It is doubtful that the effects noted here would be nearly as pronounced, since the phonon lifetimes in other common semiconductors are much smaller than in Ge. It would also be interesting to test the model proposed here by investigation of the time dependence of the relative intensities of Stokes to anti-Stokes Raman lines associated with the optical phonons. This would directly yield the phonon temperature. Such experiments have already been performed, for example, in connection with high-intensity cw excitation in various other semiconductors to observe elevated phonon temperatures.[16(b)]

ACKNOWLEDGMENTS

We thank M. Gallant for checking some of the calculations and for helpful comments concerning the manuscript. Research supported by NRC of Canada and a Univ. of Toronto Connaught Grant.

---

[1]A. Elci, M. O. Scully, A. L. Smirl, and J. C. Matter, Phys. Rev. B 16, 191 (1977).

[2]D. H. Auston and C. V. Shank, Phys. Rev. Lett. 32, 1120 (1974).

[3]G. A. Jamison, A. V. Nurmikko, and H. J. Gerritsen, Appl. Phys. Lett. 29, 640 (1976).

[4]A. L. Smirl, J. C. Matter, A. Elci, and M. O. Scully, Opt. Commun. 16, 118 (1976).

[5]D. H. Auston, C. V. Shank, and P. LeFur, Phys. Rev. Lett. 35, 1022 (1975).

[6]C. V. Shank and D. H. Auston, Phys. Rev. Lett. 34, 479 (1975).

[7]A. Elci, A. L. Smirl, C. Y. Leung, and M. O. Scully, Solid-State Electron. 21, 151 (1978).

[8]D. H. Auston, S. McAfee, C. V. Shank, E. P. Ippen, and O. Teschke, Solid-State Electron. 21, 147 (1978).

[9]A. L. Smirl, J. R. Lindle, and S. C. Moss, Phys. Rev. B 18, 5489 (1978).

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

[11]C. Herring, Bell Syst. Tech. J. 34, 237 (1955).

[12]The initial temperatures of the plasma used by ESSM are much higher than one would expect on the basis of photoexcitation of the carriers. This has been attributed to various additional heating processes such as plasmonassisted recombination. Assuming these as initial temperatures, as opposed to the temperatures arrived at by photoexcitation alone, has a negligible effect on the probe transmission model of ESSM since the initial temperature drop is quite rapid.

[13]W. P. Latham, Jr., A. L. Smirl, and A. Elci, Solid-State Electron. 21, 159 (1978).

[14]No distinction is made between longitudinal and transverse optical phonons here, it being assumed that the electron-phonon coupling constant is the same for both. In addition, we will assume that in the region of the Brillouin zone occupied by hot phonons, the population is independent of wave vector.

[15]S. Safran and B. Lax, J. Phys. Chem. Solids 36, 753 (1975).

[16](a) Lifetimes in comparable semiconductors like GaAs, Si, and InSb are in the vicinity of 3 psec; J. C. V. Mattos and R. C. C. Leite, Solid State Commun. 12, 465 (1973); (b) J. C. V. Mattos and R. C. C. Leite, Solid State Commun. 12, 465 (1973).