RADIATIVE RECOMBINATION FROM PHOTOEXCITED HOT CARRIERS IN GaAs

Jagdeep Shah and R. C. C. Leite
Bell Telephone Laboratories, Holmdel, New Jersey 07733
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We report the first observation of the radiative recombination from photoexcited hot carriers in GaAs. Our results show that the hot carriers have an effective temperature which implies a Maxwellian distribution. This temperature increases with the excitation intensity. An empirical relationship between this temperature and the power transferred from the hot carriers to the lattice is in quantitative agreement with the theoretical predictions for carrier scattering by polar optical modes.

Benoit à la Guillaume and Lavallard (referred) to as BL)[1] have reported the observation that the emission band due to band-to-band recombination in InSb narrows and shifts to lower energies as the intensity of excitation is increased by a factor of 6. They have interpreted this observation in terms of the following model: At the lower excitation intensity the electrons are hot; when the excitation is increased, electron-hole scattering reduces this temperature thus giving rise to their observed results.

The experimental results we present for GaAs are in contrast to those of BL. We find that both electrons and holes are thermalized among themselves and have a temperature which increases with increasing excitation intensity. Our interpretation of these results is also in contrast to that of BL. We conclude that the hot-carrier system loses energy to the lattice via polar optical scattering instead of collisions with cold holes.

Epitaxial GaAs (impurity concentration ~10^{15} cm^{-3}) was used in this experiment. The samples were immersed in liquid helium (pumped to ~2°K) and were excited by a high-power (~1 W) argon laser focused into a spot ~100 μm in diameter. A double spectrometer (Spex Model No. 1400) was used to analyze the emitted radiation.

The emission spectra of the n- and p-type samples were measured at several excitation intensities. The spectra were highly intensity dependent. At low excitation intensities, both n and p samples showed a large number of sharp lines, details of which were different for n and p samples.[2] The spectra of n and p samples showed a similar qualitative behavior with increasing excitation intensity. The sharp lines (width ~0.1 meV) present at low intensities merged to form a rather broad (width ~5 meV) emission band (at ~1.512 eV) at higher intensities. The peak of this band shifted slightly to lower energies (a total of <1 meV at our highest intensity) as the excitation intensity was further increased. At very high intensities, the n- and p-type spectra were essentially the same.

The most striking feature of the spectra of both the n- and p-type samples at high excitation intensities is the fact that the emission extends to energies much above E_g. This high-energy tail is plotted in Fig. 1 as a function of the energy of the emitted photon for several excitation intensities. Note that the high-energy tail can be characterized by an effective temperature[3] T_c which increases with increasing excitation intensity.[4] Note also that T_c is much larger than the temperature T_B of the helium bath surrounding the sample even at the lowest excitation intensity

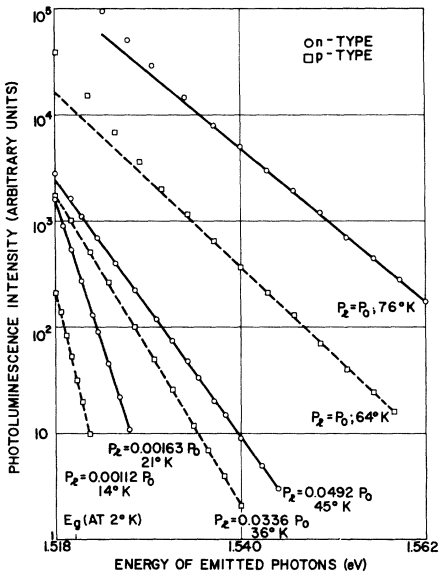

FIG. 1. The high-energy tails in the photoluminescence spectra of n-type and p-type epitaxial GaAs. The data for three different excitation intensities were chosen (from a set of eight) for this plot. The relation between the emission intensities of any two curves is arbitrary. The points are experimental points selected from a continuous recording. Straight lines drawn through the high-energy points define an effective temperature (T_c) for each curve. P_0 is the laser power absorbed by the sample at the maximum excitation intensity.

used in this experiment. This implies one of two things: (i) The lattice temperature $T_L > T_B$; or (ii) $T_L \simeq T_B$, but one or both types of carriers (electrons and holes) are "hot" and have Maxwellian distribution. However, we observe very strong donor-acceptor pair recombination from the sample at the same time as we see the high-energy tail. The donor-acceptor pair recombination in our samples becomes very weak for $T_B > 15°K$ and disappears for $T_B \gtrsim 35°K$. In order to make sure that the "active" region of the crystal had a uniform temperature, we measured the carrier diffusion length by a new technique, to be described elsewhere.[5] We found that under the conditions of our experiments the ambipolar diffusion length was less than 1 μm (~0.3 μm). Since the thermal conductivity of high-purity GaAs at the temperatures of our experiments is $\gtrsim 1$ W/deg cm,[6] the maximum temperature difference in the active region cannot be larger than 1°K. Therefore the observation of donor-acceptor pair emission implies[7] that T_L is not significantly larger than 15°K.[8] Hence the high-energy tail must be due to hot carriers thermalized among themselves.

The effective temperature T_c of the hot carriers is plotted in Fig. 2 as a function of the photon flux F for both n- and p-type samples. Notice that the relationship of the form $F \propto \exp(-T_0/T_c)$ seems to describe the data well except at lower intensities. Notice also that T_0 corresponds to 33 meV which is very close to the LO-phonon energy in GaAs (36 meV[9]).

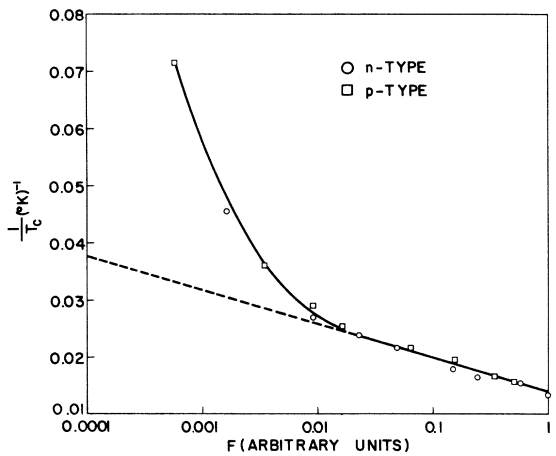

FIG. 2. T_c^{-1} (obtained by the method of Fig. 1) as a function of F, the incident photon flux ($F = P_l/A\hbar\omega_l$). The dashed line is an extrapolation of the straight line at the higher values of F. Arbitrary units of F are chosen so that $F = 1$ for the maximum excitation intensity. Note $F \propto \exp(-T_0/T_e)$, where $T_0 = 33$ meV.

We now present a model to explain these results. The exciting laser beam (of photon energy $\hbar\omega_l$) creates monoenergetic electrons ($E \simeq \hbar\omega_l - E_g$). These high-energy electrons lose energy mainly by two competing processes, namely emission of optical phonons and collisions with other carriers. The latter process raises the temperature of the carriers, which are thermalized among themselves due to carrier-carrier collisions. In GaAs the dominant mechanism for energy loss from the carrier system to the lattice is polar optical-mode scattering.[10] Therefore, the temperature of the carrier system is determined by the interplay between the power supplied to it by the high-energy, photoexcited electrons and the power it loses to the lattice by the emission of optical phonons.

In what follows, we show that our experimental results can be explained quantitatively by the above model.

The experimental fact that the carriers are thermalized among themselves is in agreement with theoretical expectations. It can be shown[11] that when polar optical scattering is the dominant energy-loss mechanism, the carriers are thermalized among themselves if their density exceeds a critical density n_c given by

$$n_c = |e| E_0 k\theta (T/\theta)^{1/2} (2\pi e^{*4})^{-1} \exp(-\theta/T), \quad (1)$$

where e is the carrier charge, k is the Boltzmann constant, $k\theta$ is the phonon energy, T is the carrier temperature, and $(e^*/e)^2 = \bar{\epsilon}^{-1}$, where $\bar{\epsilon}$ is a suitable dielectric constant. E_0 is an effective electric field given by $|e| E_0 = m^* e^2 k\theta \hbar^{-2} \times (\epsilon_\infty^{-1} - \epsilon^{-1})$, where m^* is the carrier effective mass, $\hbar$ Planck's constant, and ϵ_∞ and ϵ are the high-frequency and static dielectric constants, respectively. Taking numbers appropriate for GaAs[9] (i.e., $m_h^* = 7m_e^* = 0.5m_0$, $\epsilon_\infty = 10.62$, $\epsilon = 12.53$, $\bar{\epsilon} = 11.7$, and $\theta = 400°K$) and taking T as the highest temperature (76°K) obtained from Fig. 1, we obtain $n_c = 3 \times 10^{14}$ cm^{-3} for electrons and $n_c = 2 \times 10^{15}$ cm^{-3} for holes. We estimate the actual carrier density from photoconductivity measurements to be $(5 \pm 2) \times 10^{16}$ cm^{-3} at the highest excitation intensity. Therefore, the observed Maxwellian distribution is consistent with theoretical expectations. The numbers also seem to indicate that even the electron-hole collisions are more efficient than the carrier-lattice collisions. This implies that the electrons and holes are in equilibrium with themselves and with each

other, i.e., $T_e \simeq T_h \neq T_L$. As a consequence the values of T_c(Fig. 1) can be identified with the carrier temperatures ($T_e \simeq T_h$) at various values of P_l, the laser power absorbed by the sample.

We can now establish an empirical relationship between T_e and P_l. This relationship provides information on how $P(T_e)$, the power per electron transferred from the hot-electron system to the lattice, depends on T_e. This is because $P(T_e)$ must equal P, the average power transferred from the laser to each electron in the electron system, i.e., $P(T_e) = P \simeq P_l(\hbar\omega_l - E_g)f/dA\hbar\omega_l n$, where $A(\simeq 100\ \mu\text{m})$ is the laser beam area at the sample, $d(\simeq 0.3\ \mu\text{m})$ is the carrier ambipolar diffusion length, f is defined below, and the other symbols have the same meaning as before. f is the fraction of P_l given to the electron system by the photoexcited electrons and is given by $f = n/(n + n_c^*)$, where n_c^* is the density of electrons in the electron system at which the photoexcited high-energy electrons lose the same amount of power to the electron system as to the lattice by the emission of optical phonons. It can be shown[11] that for the monoenergetic electrons of energy $E \gg k\theta$,

$$n_c^* = \frac{8\pi}{e^{*4}} |e| E_0 k\theta \ln\left[2\left(\frac{E}{k\theta}\right)^{1/2}\right].$$

For GaAs, $n_c^* \simeq 7\times 10^{17}\ \text{cm}^{-3}$ when $E = 1$ eV. Thus $n \ll n_c^*$ in our experiments so that $P(T_e) \simeq P_l(\hbar\omega_l - E_g)/dA\hbar\omega_l n_c^*$. But $P_l \propto F$ and $F \propto \exp(-T_0/T_e)$ from Fig. 2. Therefore we obtain the empirical relationship $P(T_e) \propto \exp(-T_0/T_e)$ with $T_0 = 33$ meV (Fig. 2).

Theoretically, it can be shown[12] that for polar optical-mode scattering,

$$P(T_e) = \left(\frac{2k\theta}{\pi m_e^*}\right)^{1/2} |e| E_0 \exp\left(\frac{-\theta}{T_e}\right) \times \left[\left(\frac{\theta}{T_e}\right)^{1/2} K_0\left(\frac{\theta}{2T_e}\right)\exp\left(\frac{\theta}{2T_e}\right)\right]$$

if the phonon energy $k\theta \gg kT_L$. Here K_0 is the modified Bessel function of the order zero. In the temperature range of our experiments the quantity in the square bracket is nearly a constant. Hence Eq. (2) reduces to $P(T_e) = 8.7\times 10^{-8} \times \exp(-\theta/T_e)$ W for values of m_e^*, etc., appropriate to GaAs.[9]

Notice that the experimental result that $P(T_e)$ varies exponentially with T_e is in agreement with theoretical expectations. Moreover, $kT_0 = 33$ meV is in agreement with the LO-phonon energy of 36 meV in GaAs.[9] Finally, the magnitude of $P(T_e)$ should be compared with the average power given by the laser to each electron which is $P \simeq P_l(\hbar\omega_l - E_g)/dA\hbar\omega_l n_c^*$. At our highest excitation intensity $P \simeq (1 \pm 0.2)\times 10^{-10}$ W, which is in reasonable agreement with the theoretical value $P(T_e) \simeq 2.7\times 10^{-10}$ W at $T_e \simeq 76$°K. Thus our results are basically in agreement with the theory of energy loss by polar optical-mode scattering. The main deviation from the theory occurs at lower excitation intensities (see Fig. 2). But we note [Eq. (2)] that the rate of loss of energy via optical-phonon scattering decreases drastically with the reduction in electron temperature. Thus a possible explanation of the deviation (Fig. 2) may be that other energy-loss mechanisms become important at lower excitation intensities.

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[1]C. Benoit à la Guillaume and Ph. Lavallard, in Proceedings of the Seventh International Conference on the Physics of Semiconductors, Paris, France, 1964 (Academic Press, Inc., New York, 1964) Vol. 4, p. 53.

[2]Jagdeep Shah, R. C. C. Leite, and R. E. Nahory, to be published.

[3]From Sturge's absorption data [M. D. Sturge, Phys. Rev. 127, 768 (1962)], we estimate that the correction in T_c due to self-absorption will not exceed 0.3%.

[4]It was shown by A. Mooradian and H. Y. Fan [Phys. Rev. 148, 873 (1966)] that when electrons and holes are at the same temperature (T), the emission intensity as a function of photon energy ($\hbar\omega$) is given by $I(\hbar\omega) \propto \exp(-\hbar\omega/kT)$ for direct transitions. Therefore the observed straight lines of Fig. 1 imply an effective temperature T_c. This expression is strictly true only for nondegenerate distribution of carriers. However, it will be valid even for degenerate carrier distributions for the long energy tail that we are considering.

[5]R. C. C. Leite and Jagdeep Shah, to be published.

[6]M. G. Holland, Phys. Rev. 134, A471 (1964).

[7]The rate of escape of electrons from donors depends on the lattice temperature rather than the electron temperature.

[8]If the shift in the position of the emission peak is attributed to temperature rise, the shift (<1 meV) would correspond to a temperature rise of <15°K (from 2° to 76°K, the bandgap would be reduced by ~7 meV; M. D. Sturge, Ref. 3). This is consistent with the conclusion in the text. It is also possible that the shift is due to many-body effects. [N. G. Basov, O. V. Bogdankevich, V. A. Goncharov, B. M. Lavrushin and V. Yu. Sudzilov-

skii, Dokl. Acad. Nauk SSSR 168, 1283 (1966) [translation: Soviet Phys.-Doklady 11, 522 (1966)].
[9]O. Madelung, Physics of III-V Compounds (John Wiley & Sons, Inc., New York, 1964).
[10]H. Ehrenreich, Phys. Rev. 120, 1951 (1960); also E. Haga and H. Kimura, J. Phys. Soc. Japan 19, 658 (1964).
[11]R. Stratton, Proc. Roy. Soc. (London), Ser. A 246, 406 (1958).
[12]B. V. Paranjape, Rept. Brit. Elec. Allied Ind. Res. Assoc. No. L/T 285 (1953); see also E. M. Conwell, Solid State Phys. Suppl. 9, 1 (1967).

MAGNETISM IN Ni-Cu ALLOYS*

C. G. Robbins, Helmut Claus, and Paul A. Beck
Department of Metallurgy and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801
(Received 10 April 1969)

On the assumption that in Ni-Cu alloys the spin moment on a Ni atom depends on the local atomic environment, it was possible to find moment values for the various atomic configurations so as to give average moments in reasonable quantitative agreement with the values measured in the ferromagnetic composition range. The local environment is specified by the number of Ni nearest neighbors and the number of Ni second-nearest neighbors. This model allows also a consistent qualitative interpretation of the effect on the average moment of low-temperature annealing treatment and of plastic deformation.

The linear decrease in the average atomic moment with Cu content[1,2] of the ferromagnetic Ni-Cu solid solutions has long been interpreted in terms of filling a rigid d band by the extra electron of Cu,[3] with the filling completed at about 53 at.% Cu. However, magnetization measurements[4] showed the presence of permanent moments (and of superparamagnetism) at 70 and even at 80 at.% Cu. The temperature-independent susceptibility[5] and the electronic specific heat[6-8] of Cu-rich alloys increase with the Ni content, suggesting that d states lie at the Fermi surface. Photoelectron energy-spectrum and optical-reflectivity data for Cu-rich alloys[9] show that the Ni solute atoms give rise to a high density of virtual bound d states at and just below the Fermi-energy level. As estimated from the published electronic specific-heat value,[7] the contribution per Ni atom to the density of states at the Fermi surface for alloys with less than 30 at.% Ni is only slightly less than that for pure Ni. Mössbauer isomer-shift measurements with ^{61}Ni in Ni-Cu alloys led to the conclusion[10] that the electronic charge density at the Ni nucleus remains approximately the same over the entire composition range, in agreement with the proposition that the number of d holes per Ni atom stays approximately constant at all concentrations.[11] The interpretation of Cu NMR measurements with alloys containing as little as 1% Ni allows the conclusion that, even at this concentration, d holes are present at the Ni.[12] Thus, various types of evidence indicate that the decrease in ferromagnetic moment with increasing Cu concentration cannot be due to the filling of the d band. A different interpretation is, therefore, needed.

Jaccarino and Walker[13] suggested that the moment associated with an Fe atom in dilute solid solution in Nb-Mo alloys is determined by the number of nearest-neighbor Nb atoms. A similar relationship was found by those authors for the moment on Co atoms dissolved in Rh-Pd alloys. The magnetic moment on Fe in Fe-Al alloys, both disordered and ordered, can be described in terms of the nearest-neighbor atomic environment.[14] The coherent magnetic scattering of neutrons from ordered Fe_3Al showed[15] that Fe atoms, all eight nearest neighbors of which are Fe, have a moment of $2.14\mu_B$ at room temperature, or nearly the same as that of Fe in bcc iron, even though in ordered Fe_3Al all second-nearest neighbors are Al and the moment on the nearest-neighbor Fe atoms is only $1.46\mu_B$. When the number of nearest-neighbor Fe atoms decreases from five to three (or less) the moment at $T=0$ decreases from a value just slightly below the full $2.2\mu_B$ to zero.[14] The near disappearance of Langevin paramagnetism from VAu_4 when the alloy is disordered was also interpreted on the basis of nearest-neighbor atomic environment.[16] In ordered VAu_4 the V atoms, which have a full complement of 12 Au nearest neighbors, possess a permanent moment, and the alloy becomes ferromagnetic at about 45°K.[17] In the disordered alloy nearly all V atoms have at least one V nearest neighbor and, under such conditions, they have no moment.[16] Recent Ag NMR linewidth data taken as a function of the V content in $V_x(Au_{0.8}Ag_{0.2})_{1-x}$ alloy[18] and careful magnetic susceptibility versus temperature mea-