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Translated by J. G. Adashko

Spectrum and polarization of the luminescence emitted from GaAs in the energy range $E_g + \Delta$

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(Submitted October 7, 1975)

Zh. Eksp. Teor. Fiz. **70**, 1092-1099 (March 1976)

An investigation was made of the spectrum and polarization of the 1.48-1.94 eV photoluminescence emitted from *n*-type GaAs crystals excited with linearly and circularly polarized He-Ne laser radiation (1.48-1.94 eV). A detailed study was made of the luminescence band located near 1.86 eV, which was due to transitions between the conduction band and the split-off (by the spin-orbit interaction) valence band Γ_7 . The short lifetime of holes in the band Γ_7 , due to the relatively high probability of transitions between the valence subbands, resulted in a considerable deviation of the hole distribution function from the Boltzmann form (the holes did not become thermalized during their lifetime). For this reason the degree of circular polarization of the luminescence excited by circularly polarized laser radiation was close to unity. This result was evidence of almost complete optical orientation of nonequilibrium holes in the split-off band (the holes maintained the initial spin direction during their lifetime). The principal quantitative relationships obtained in the present investigation were explained satisfactorily by assuming the dominant role of electron-hole collisions in the energy relaxation of holes.

PACS numbers: 78.60.Dg, 71.30.Mw

In contrast to the optical absorption spectra, studies of the photoluminescence spectra of semiconductors have been limited mainly to the range of frequencies near the fundamental absorption edge. The "edge" luminescence in the range $\hbar\omega \leq E_g$ may be related to the radiative recombination of free or bound excitons, interband electron transitions, impurity-band transitions, etc. (see, for example, ^[1]).

An analysis of the luminescence line profile in the range $\hbar\omega > E_g$ made it possible to obtain information on the distribution function $f(\epsilon)$ of the energy of nonequilibrium carriers. In this way it has been possible to determine the effective temperature of nonequilibrium electrons, which—under certain conditions—may exceed the lattice temperature, ^[2] and also to detect the deviation of the function $f(\epsilon)$ from the Boltzmann distribution in the high-energy range. ^[3,4]

An investigation of the polarization of recombination radiation makes it possible to investigate, in particu-

lar, the optical orientation of free carriers in semiconductors. The phenomenon of optical orientation, i. e., the establishment of a preferential orientation of carrier spins on absorption of circularly polarized light, has been used successfully in studies of the kinetics of recombination and spin relaxation of electrons in semiconductors (see, for example, ^[5]).

The cited investigations were concerned with the luminescence due to electron transitions between two nearest bands (in the case of GaAs between the conduction band Γ_6 and the quadruply degenerate, at $k=0$, valence band Γ_8). An investigation of the Raman scattering in *n*-type GaAs crystals, carried out by Burstein *et al.*, ^[6] revealed a weak luminescence band in the energy range 1.86 eV, which complicated an analysis of the scattered-light spectra. It was shown in ^[6] and also in ^[7], where the preliminary results of the present study were reported, that this band was due to transitions between the conduction band Γ_6 and the va-

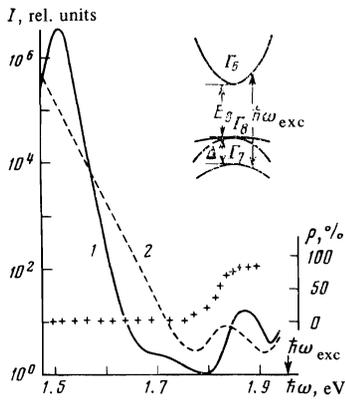


FIG. 1. Photoluminescence spectra of sample of GaAs ($n = 3.4 \times 10^{17} \text{ cm}^{-3}$) above the fundamental absorption edge: 1) 82°K; 2) 200°K. Here, “+” is the degree of circular polarization of the luminescence excited at 82°K by circularly polarized pump radiation. The top right-hand corner shows the energy band structure of GaAs in the vicinity of $k=0$.

lence band Γ_7 split-off by the spin-orbit interaction from the band Γ_8 (in GaAs we have $\Delta = \epsilon_{\Gamma_8} - \epsilon_{\Gamma_7} = 0.341 \text{ eV}$ at 20°K^[8]); the energy band structure of gallium arsenide is shown in Fig. 1.

The hole lifetime τ_0 in the split-off band should be short (it is clearly governed primarily by the relatively high probability of the $\Gamma_7 \rightarrow \Gamma_8$ transitions accompanied by optical phonon emission). This is supported, in particular, by the observation that the $\Gamma_8 - \Gamma_7$ (1.86 eV) photoluminescence band was several orders of magnitude weaker than the “edge” photoluminescence band $\Gamma_6 - \Gamma_8$ (~1.52 eV) for comparable excitation rates.^[7] The shortness of τ_0 may result in a strong deviation of the distribution function from the Boltzmann form (holes do not become thermalized during their lifetime) and in a high degree of optical orientation of holes (holes do not lose the preferred spin direction during their lifetime).

These qualitative hypotheses were confirmed by us in a detailed investigation of the spectrum and polarization of the $\Gamma_8 - \Gamma_7$ luminescence, which is reported below.

EXPERIMENTAL METHOD

The luminescence was investigated using n -type GaAs crystals with electron densities from 5×10^{15} to $7 \times 10^{17} \text{ cm}^{-3}$. These densities were deduced from the conductivity and Hall coefficient. Before the measurement the samples were polished and etched in a polishing solution ($\text{H}_2\text{SO}_4 : \text{H}_2\text{O}_2 : \text{H}_2\text{O} = 5 : 1 : 1$). An investigated sample was placed on a heat sink in a cryostat and its temperature could be varied from the room value to 82°K. The luminescence was excited by He-Ne laser radiation ($\hbar\omega_{\text{exc}} = 1.96 \text{ eV}$, 25 mW). The photocarrier density did not exceed $\sim 10^{15} \text{ cm}^{-3}$. This was estimated from the observation that the 1.86 eV luminescence was not observed for samples with $n < 10^{15} \text{ cm}^{-3}$. The luminescence spectra $I(\hbar\omega)$ were recorded with a DFS-24 spectrometer with a double grat-

ing monochromator. A cooled FEU-79 photomultiplier and a photon counting system were used. The spectral characteristic of the recording system (including that of the photomultiplier) was determined with a TRSh-2850 calibration lamp. In all measurements the spectral slit width did not exceed $5 \times 10^{-4} \text{ eV}$.

EXPERIMENTAL RESULTS

Curve 1 in Fig. 1 shows the luminescence spectrum $I(\hbar\omega)$ obtained in the energy range 1.48–1.94 eV for a sample with an electron density of $3.7 \times 10^{17} \text{ cm}^{-3}$ when the temperature of the heat sink was 82°K. In addition to the main luminescence band with a maximum at 1.52 eV and associated with transitions between the Γ_6 and Γ_8 energy bands, there was also a band with the maximum at 1.865 eV, due to transitions from the conduction band Γ_6 to the valence band Γ_7 . The intensity at the maximum of this band was approximately 10^5 times less than the intensity of the main band. The high-frequency tail of the main band was described satisfactorily by the exponential dependence with a characteristic temperature 100°K. This temperature was somewhat higher than the “dark” temperature of the crystal, 82°K, and the difference was clearly due to the local heating of the sample by the exciting radiation. Control experiments indicated that reduction of the exciting radiation power made the temperature found in this way approach the temperature of the heat sink (82°K) to which the sample was bonded. Therefore, in analyzing the experimental data we assumed that the true temperature of a crystal was that deduced from the edge luminescence tail. We included in Fig. 2 the spectrum obtained at 200°K. This spectrum exhibited clearly the thermal shift of the $\Gamma_6 - \Gamma_7$ luminescence band in the direction of lower energies.

When carriers were excited with circularly polarized light, the $\Gamma_6 - \Gamma_7$ luminescence was found to be largely circularly polarized as well, whereas the degree of polarization ρ of the $\Gamma_6 - \Gamma_8$ luminescence was close to zero (Fig. 1). Hence, we concluded that under the experimental conditions the spin orientation of the majority carriers (electrons) was negligible and the ob-

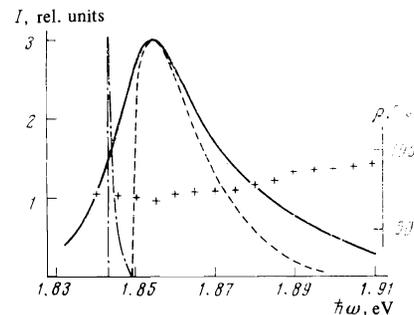


FIG. 2. Photoluminescence spectrum of a sample of GaAs ($n = 7.6 \times 10^{15} \text{ cm}^{-3}$) in the region of $E_g + \Delta$ recorded at $T = 100^\circ\text{K}$. The continuous curve represents the experimental results, the dashed curve is calculated on the basis of Eq. (1) (interband transitions), the chain curve is calculated on the basis of Eq. (2) (donor-band transitions), and “+” is the degree of circular polarization.

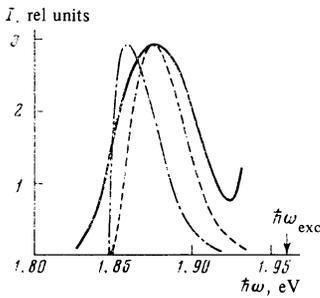


FIG. 3. Photoluminescence spectrum of a sample of GaAs ($n = 3.4 \times 10^{17} \text{ cm}^{-3}$) recorded in the $E_g + \Delta$ range at $T = 100^\circ \text{K}$. The continuous curve is experimental, the dashed curve is calculated on the basis of Eq. (3) (indirect transitions), and the chain curve is calculated on the basis of Eq. (1) (direct transitions).

served circularly polarized $\Gamma_6 \rightarrow \Gamma_7$ luminescence was due to spin-oriented holes in the Γ_7 band.

The degree of polarization across the latter luminescence band increased somewhat in the direction of higher frequencies (Fig. 2). The degree of polarization ρ measured at the luminescence band maximum increased with the carrier density. For samples with $n = 7.6 \times 10^{15} \text{ cm}^{-3}$, $4.4 \times 10^{16} \text{ cm}^{-3}$, and $3.4 \times 10^{17} \text{ cm}^{-3}$ the measured degree of polarization was 68, 72, and 83%, respectively.

The luminescence spectrum in the region of the $\Gamma_6 \rightarrow \Gamma_7$ direct interband transitions could be described by

$$I(\epsilon) = A e^{-\epsilon} f_e f_h, \quad (1)$$

where A includes the matrix element of the transition and functions dependent weakly on the energy; $\epsilon = \hbar\omega - (E_g + \Delta)$; Δ is the spin-orbit splitting; f_e and f_h are the distribution functions of electrons and holes corresponding to the energy of the emitted photons.

Figure 2 shows the experimental spectrum $I(\hbar\omega)$ (continuous curve) for a sample with $n = 7.6 \times 10^{15} \text{ cm}^{-3}$ recorded at 100°K . This figure includes also the spectrum calculated from Eq. (1) (dashed curve). In these calculations we used $E_g(100^\circ \text{K}) = 1.508 \text{ eV}^{[9]}$ and $\Delta = 0.341 \text{ eV}$,^[8] and we assumed an equilibrium energy distribution. It is clear from Fig. 2 that the positions of the maxima of the calculated and experimental curves agreed well but the experimental spectrum was much more extended, compared with the calculated one, in the direction of low and high frequencies. The origin of this spread will be discussed later.

An increase in the degree of doping shifted the $\Gamma_6 \rightarrow \Gamma_7$ band maximum in the direction of higher frequencies and the band became broader. The spectrum obtained for a sample with $n = 3.7 \times 10^{17} \text{ cm}^{-3}$ at 100°K is shown in Fig. 3.

DISCUSSION OF RESULTS

The distribution function of nonequilibrium holes $f_h(\epsilon)$ in the Γ_7 band can be determined by comparing the theoretical and experimental luminescence spectra.

However, we have to know which (interband, impurity-band, etc.) transitions are responsible for the experimental spectrum. The main criteria in the determination of the type of transition are the energy position of the luminescence band and its profile.

It is frequently possible to identify transitions on the basis of the first criterion. In the present case this is difficult because, on the one hand, the threshold energies of the $E_g + \Delta$ interband transitions and of the $E_g + \Delta - E_D$ donor-band transitions are similar (the donor ionization energy is $E_D \approx 6 \text{ meV}^{[1]}$) and, on the other, the spin-orbit splitting Δ is not known sufficiently accurately at 100°K . However, if we use the above values of $E_g(100^\circ \text{K})$ and $\Delta(20^\circ \text{K})$ (it is shown in^[8] that Δ depends much less on temperature than E_g), the maxima of the experimental spectrum are found to agree well with the interband spectrum calculated from Eq. (1).

We shall now consider the donor-band transitions. Eagles^[10] used the effective mass approximation in a calculation of the form of the luminescence spectrum for transitions from the ground state of an impurity to the valence band:

$$I(\epsilon) = B [x^2 / (1+x)^4] f_h(\epsilon_h), \quad (2)$$

where B is a quantity which depends weakly on the energy; $\epsilon = \hbar\omega - (E_g + \Delta - E_D)$; $x = m_s \epsilon / m_D E_D$; m_s and m_D are the effective masses of holes in the Γ_7 band and of electrons, respectively; $f_h(\epsilon_h)$ is the hole distribution function.

The spectrum calculated using this formula and $E_g = 1.508 \text{ eV}$, $\Delta = 0.341 \text{ eV}$, and $E_D = 6 \text{ meV}$, differs considerably from the experimental spectrum (Fig. 2). Firstly, the maximum of the calculated curve is shifted toward lower energies by about 10 meV (which is much greater than a reasonable discrepancy between theory and experiment bearing in mind some indeterminacy in Δ). Secondly, the half-width of the experimental spectrum ($\sim 30 \text{ meV}$) is considerably greater than the half-width of the calculated spectrum ($\sim 2 \text{ meV}$). The narrow theoretical curve is typical of transitions from shallow donor states.^[11] This is due to the fact that the matrix element of the transition decreases rapidly with increasing wave vector \mathbf{k} (and, consequently, with increasing kinetic energy of holes in a band).

All that was said above gives us grounds for assuming that the observed spectrum (at least its high-frequency part beginning from the maximum) is due to interband transitions. The discrepancy between the theory and experiment at low frequencies is clearly due to the contribution of transitions from impurity states and transitions between the energy band tails.

When the degree of doping is increased, the luminescence maximum shifts toward higher energies faster than one would expect on the assumption of direct interband transitions [Eq. (1)].¹⁾ It is shown in^[12] that in the case of sufficiently heavily doped InSb crystals ($n > 10^{17} \text{ cm}^{-3}$), the interband luminescence spectrum is dominated by indirect transitions which are accompanied by the scattering on impurities. The lumines-

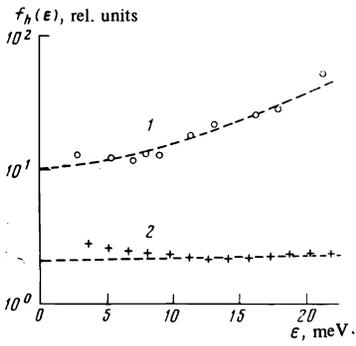


FIG. 4. Dependence of the hole distribution function in the Γ_7 band on the energy ($T = 400^\circ\text{K}$): 1) $n = 7.6 \times 10^{15} \text{ cm}^{-3}$, 2) $n = 6.7 \times 10^{17} \text{ cm}^{-3}$.

cence spectrum due to these transitions is

$$I(\hbar\omega) = C \int_0^{\hbar\omega - (E_g + \Delta)} |M(\epsilon_e, \hbar\omega)|^2 f_e(\epsilon_e) f_h(\epsilon_h) e^{-\epsilon_e/\hbar\omega} d\epsilon_e,$$

where $|M|$ is the matrix element of the transition and the electron and hole energies are related by $\epsilon_e + \epsilon_h = \hbar\omega - (E_g + \Delta)$.^[12] It is difficult to calculate the spectrum on the basis of Eq. (3) because the energy dependence of the matrix element is not known. In the first approximation, $|M|$ can be calculated assuming that $I(\hbar\omega)$ is constant and independent of energy. A comparison of the spectrum calculated on this assumption with that obtained experimentally for a sample with $n = 3.7 \times 10^{17} \text{ cm}^{-3}$ (Fig. 3) demonstrates that indirect transitions play the dominant role in the luminescence spectrum of GaAs even at these dopant concentrations.

A comparison of the experimental and calculated spectra $I(\hbar\omega)$ for an assumed (governed by the electron density) form of $f_h(\epsilon)$ was used to determine the hole distribution function $f_h(\epsilon)$ for the Γ_7 band. It should be stressed once again that $f_h(\epsilon)$ was calculated using the high-frequency part of the luminescence spectrum [the beginning of this spectrum was 15 meV higher than the energy corresponding to the $I(\hbar\omega)$], which was attributed—as pointed out earlier—to interband transitions. This made it possible to eliminate indeterminacy in the determination of the density-of-states function near the bottom of the conduction band of a sample with impurities.

Figure 4 shows the distribution functions for two samples with different electron densities. For the sample with $n = 7.6 \times 10^{15} \text{ cm}^{-3}$, the function $f_h(\epsilon)$ not only does not decrease but even rises with the energy. Similar curves were obtained also for several samples with $n = (1-4) \times 10^{16} \text{ cm}^{-3}$.

This type of distribution function can be explained by the fast recombination of holes due to the $\Gamma_7 \rightarrow \Gamma_8$ transitions between the valence subbands, so that the majority of the optically generated holes does not reach the top of the valence band. In the investigated case the equation for the hole distribution function $f_h(\epsilon)$ in the Γ_7 band is given by the following expression obtained on the assumption of quasielastic scattering (see, for example,^[13]):

$$\frac{f_h(\epsilon)}{\tau_e} = g^{-1}(\epsilon) \frac{d}{dt} \left[A(\epsilon) \left(1 + kT \frac{d}{d\epsilon} \right) f_h(\epsilon) \right] + g^{-1}(\epsilon_0) I_0 \delta(\epsilon - \epsilon_0), \quad (4)$$

where $g(\epsilon)$ is the density of the hole energy states ϵ ; I_0 is the rate of hole generation; ϵ_0 is the initial energy holes; $A(\epsilon)$ is a quantity characterizing the relaxation of the energy of holes in the Γ_7 band; $A(\epsilon)$ and the energy relaxation time τ_e are related, in the range $\epsilon \gg kT$, by the simple expression $\tau_e^{-1} = [g(\epsilon)\epsilon]^{-1} A(\epsilon)$; τ_0 is the lifetime of holes in the Γ_7 band (governed mainly by the $\Gamma_7 \rightarrow \Gamma_8$ transitions associated with optical phonon emission).

In the range $\epsilon \gg kT$ the solution of Eq. (4) can be represented in the form

$$f_h(\epsilon) = \frac{I_0}{A(\epsilon)} \exp \left\{ - \int_{\epsilon}^{\epsilon_0} \frac{d\epsilon'}{\epsilon'} \frac{\tau_e}{\tau_0} \right\} \quad \text{for } \epsilon < \epsilon_0, \\ f_h(\epsilon) = 0 \quad \text{for } \epsilon > \epsilon_0. \quad (5)$$

If τ_0 is independent of ϵ but $\tau_e \propto \epsilon^\nu$, where the exponent ν depends on the actual energy dissipation mechanism, we find that

$$f_h(\epsilon) \propto \frac{1}{\epsilon^{-\nu}} \exp \left\{ \frac{1}{\nu} \frac{\tau_e}{\tau_0} \right\}. \quad (6)$$

If the holes lose their energy in the Γ_7 band by collisions with free electrons,^[2] we can assume that $\tau_e^{-1} \propto n\epsilon^{-3/2}$ (n is the free-electron density). Consequently, in this case we have

$$f_h(\epsilon) \propto \exp(a n^{-1} \epsilon^{3/2}), \quad (7)$$

where a is a coefficient which is independent of energy. The numerical value of a was determined from the experimental value of $f_h(12 \text{ meV})$ for the sample with $n = 7.6 \times 10^{15} \text{ cm}^{-3}$. The dashed lines in Fig. 4 give the results of calculation based on Eq. (7). It is clear from Fig. 4 that the formula (7) is in qualitative agreement with the experimental results. When n is increased, the relaxation of the hole energy becomes faster and if the electron density is sufficiently high, the function $f_h(\epsilon)$ depends weakly on the hole energy (curve 2 in Fig. 4).

We shall now consider the results obtained in the determination of the degree of circular polarization of the luminescence. Studies of the optical orientation of free carriers in semiconductors have only established the orientation of electron spins in the conduction band^[5] and not of holes in the valence band. This is due to the rapid relaxation of the hole spin in the band Γ_8 which is degenerate and $k=0$. In this case the spin relaxation time of holes τ_s is comparable with the momentum relaxation time τ_p and much shorter than the lifetime τ_0 . Therefore, the value of $\tau_s(\tau_s + \tau_0)^{-1}$, governing the degree of spin orientation of holes in the Γ_8 band is negligible.^[14] The situation is different in the simple band Γ_7 . In this case the spin relaxation time τ_s is not linked rigidly to the value of τ_p and the latter can be exceeded considerably, whereas the lifetime τ_0 is—as mentioned above—governed by the relatively high probability of $\Gamma_7 \rightarrow \Gamma_8$ transitions and it may be considerably smaller than τ_s . Therefore, holes created in the Γ_7 band do not lose, during their life-

time τ_0 , their spin, and consequently, the $\Gamma_6 \rightarrow \Gamma_7$ luminescence is largely circularly polarized.³⁾ As in^[14], the degree of the circular polarization of the $\Gamma_6 \rightarrow \Gamma_7$ luminescence observed in the direction opposite to that of the exciting radiation is equal (in the absolute sense) to the degree of spin orientation of holes.

In accordance with the selection rules, ρ_0 is the degree of spin polarization of holes excited in the Γ_7 band by circularly polarized light of $\hbar\omega_{exc} = E_g + \Delta$ energy and at the moment of creation of holes the value of ρ_0 is 100%^[7,14] (for $\hbar\omega_{exc} = 1.96$ eV, $\rho_0 \approx 99\%$ ^[7]). This circumstance, and the smallness of τ_0/τ_s result in high values of the degree of circular polarization of the luminescence observed in our investigation. The value of ρ is highest at the moment of creation of holes ($\rho = \rho_0$) and may decrease in the course of their energy relaxation. This results in some dependence of ρ on the energy within a luminescence band and in its reduction with decreasing hole energy (Fig. 2). On the same assumptions as were used to obtain Eq. (5) for the hole distribution function $f_h(\epsilon)$, the expression for the degree of spin orientation of holes of energy ϵ can be found by analogy with^[14]:

$$\rho(\epsilon) = \rho_0 \exp \left\{ - \int_{\epsilon'}^{\epsilon} \frac{d\epsilon' \tau_s(\epsilon')}{\epsilon' \tau_s(\epsilon')} \right\}. \quad (8)$$

If holes lose their energy mainly by collisions with electrons in the conduction band, then $\tau_s \propto n^{-1}$ and the degree of circular polarization of the luminescence near the $\Gamma_6 \rightarrow \Gamma_7$ band maximum is generally higher for samples with greater electron densities, as found experimentally.

Thus, the principal experimental observations made by us in a study of the luminescence spectrum and polarization can be explained satisfactorily on the assumption that the lifetime of holes in the split-off band Γ_7 is short and that electron-hole collisions dominate the process of energy relaxation of holes.

The authors are grateful to E. L. Ivchenko for discussing our results and to I. L. Drichko for measuring the electrical parameters of the samples.

¹⁾If the photoluminescence spectrum had been solely due to direct transitions, the shift of the line position with increasing dopant concentration would (in this case) have been only due to the shift of the chemical potential level into the conduction band.

²⁾Estimates indicate that in our case the maximum energy of holes in the Γ_7 band is less than the optical phonon energy $\hbar\omega_{L0}$ so that we shall ignore the participation of optical phonons in the energy relaxation processes.

³⁾The lifetime τ_0 can be determined from the reduction in the degree of polarization in a transverse magnetic field^[5] (Hanle effect). However, in the case under consideration, the lifetime τ_0 is short and one would need strong magnetic fields for this purpose (estimated to be of the order of 100 kOe).

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Translated by A. Tybulewicz