Polarization of hot photoluminescence in semiconductors of the GaAs type

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(Submitted June 21, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **24**, 96–99 (20 July 1976)

We investigate the frequency and polarization dependences of the photoluminescence of hot electrons in semiconductors of the GaAs type. The anomalously large degree of circular polarization and the appearance of linear polarization in the recombination-radiation spectrum are interpreted with account taken of the onset of anisotropy in the momentum distribution when the electrons are photoexcited.

PACS numbers: 78.60.Dg

We report here the first investigation of the frequency and polarization dependences in the photoluminescence spectrum of hot electrons in semiconductors. The measurements were performed, in particular, at frequencies as close as possible to the frequency of the exciting light, i. e., under conditions of minimal energy relaxation of the photoexcited electrons. A linear polarization of the recombination radiation of the free carriers was observed upon excitation with linearly polarized light, as well as anomalously large degrees of circular polarization in the case of circular excitation. The objects of the investigations were p-GaAs single crystals and epitaxial layers of solid solutions $Al_xGa_{1-x}As$ and $GaAs_{1-x}P_x$ of p-type at a temperature 90 °K. The luminescence was excited by an He-Ne laser ($\hbar\omega_{ex}=1.96$ eV). The measurements were made with a DFS-24 spectrometer with a double grating monochromator and a photon-counting system.

Figure 1 shows, in semilog scale, the luminescent spectrum of an

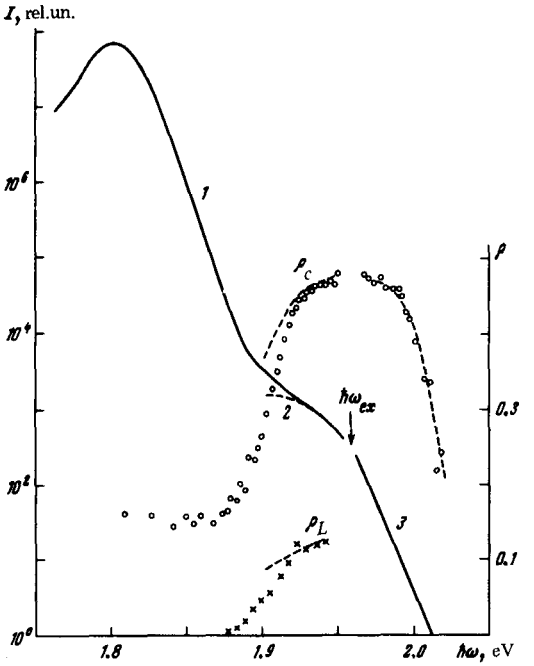

FIG. 1. Recombination radiation spectrum $I(\hbar\omega)$ for an $Al_{0.25}Ga_{0.75}As$ sample (solid curve) and frequency dependences of ρ_c (circles) and ρ_L (crosses) (the dashed curves are obtained after subtracting the contribution of the thermalized radiation).

$Al_{0.25}Ga_{0.75}As$ sample with $N_A = 10^{18}$ cm^{-3}. On the high-frequency side of the peak one can see clearly three regions: two exponential regions (1 and 3), $\exp(-\hbar\omega/T)$ with T close to the crystal temperature, separated by region 2 with a weaker frequency dependence. In this crystal, the initial energy ϵ_0 of the photoexcited is approximately 0.15 eV. Region 1 is due to the usual recombination luminescence of the thermalized electrons. The plateau 2 (distinguished from the thermalized luminescence on Fig. 1 by the dashed line) is due to recombination of the photoelectrons during the course of the thermalization.[1,2] Of particular interest is region 2. In addition to the relaxation of the nonequilibrium electrons to the bottom of the conduction band, the electrons diffuse from the source in energy space. It is this which makes the electron distribution function proportional to $\exp[(\epsilon - \epsilon_0)/T]$ at $\epsilon > \epsilon_0$,[1] and give rise to the exponentially decreasing section in the luminescence spectrum at $\hbar\omega > \hbar\omega_{ex}$.

Figure 1 shows also the spectral dependences of the degree of circular polarization of the luminescence, $\rho_c(\hbar\omega)$, following excitation by circularly polarized light, and also the degrees of linear polarization following excitation by linearly polarized light, $\rho_L(\hbar\omega)$. It is known that when the electrons are excited with circularly polarized light from the valence band, the electron spins are optically oriented and this manifests itself, in particular, in a circular polarization of the recombination luminescence.[3,4] The spectral dependence of $\rho_c(\hbar\omega)$ is distorted in this case by the contribution of the thermalized radiation from region 1. The dashed line in Fig. 1 shows the variation of ρ_c with this contribu-

tion subtracted. As seen from the figure, ρ_c decreases with increasing distance from $\hbar\omega_{ex}$ on both sides, i.e., the degree of orientation of the electrons decreases both in the case of relaxation to the bottom of the band and in the case of diffusion towards energies higher than $\hbar\omega_{ex}$. One would expect the decrease to be steeper in the latter case, as is indeed the case.

When electrons from the valence band Γ_8 are excited in A_3B_5 crystals by circularly polarized light into the conduction band Γ_6, the degree of circular polarization of the recombination radiation usually does not exceed 0.25.[3,4] Attention is called in this connection to the fact that the degree of circular polarization ρ_c^0 at the instant of production is much larger in this case and is close to 0.5. The appearance of linear polarization in the recombination-radiation spectrum of the free carriers ($\rho_L^0 \approx 0.15$) was quite unexpected. A probable explanation of these "polarization anomalies" was proposed by M. I. D'yakonov and V. I. Perel' and is based on the following. The value $\rho_c^0 = 0.25$ is obtained only as a result of averaging over all the directions of the quasi-momenta in the Brillouin zone.[3] Upon excitation by circularly polarized light, owing to the symmetry properties of the wave functions and owing to the connection between the angular momentum and the momentum, a preferred direction along the momentum of the produced holes is created in the valence band (and by the same token along the electron momentum, by virtue of the conservation of the momentum in absorption), with a symmetry axis that coincides with the direction of the light beam. The number of photo-holes in the investigated p-type samples at 90 °K is small in comparison with the number of equilibrium holes, and the introduced anisotropy (prior to its relaxation) is significant and is in fact the cause of the excessively large degree of circular polarization.

In the case of linearly polarized pumping, an anisotropy is likewise produced in the momentum distribution of the photo-holes, and consequently also of the hot electrons, with a symmetry axis parallel to the electric vector of the light wave (the heavy holes absorb light polarized only perpendicular to the direction of their momentum [5]). The selection rules for the radiative recombination

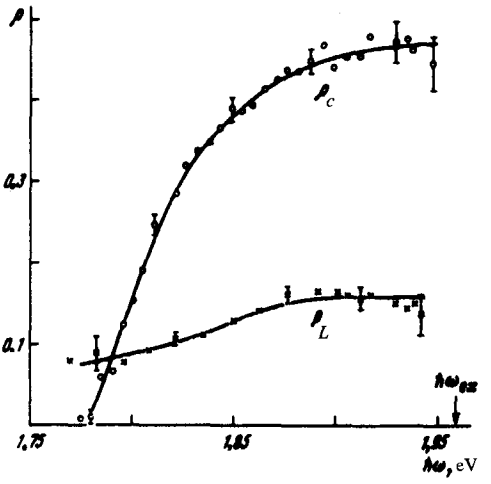

FIG. 2. Spectral dependences of ρ_c and ρ_L in a p-GaAs sample ($N_A = 4 \times 10^{17}$ cm^{-3}).

and the photoexcitation are identical, and therefore the anisotropy of the momentum distribution of the electrons leads also to linear polarization of the radiation.

Figure 2 shows plots of $\rho_c(\hbar\omega)$ and $\rho_L(\hbar\omega)$ for GaAs in a wider frequency range, corresponding to the relaxation of the hot electrons excited from the heavy-hole band. In this case ϵ_0 amounts to 0.38—0.42 eV when the excitation is by an He-Ne laser. The smearing of ϵ_0 is due to the corrugation of the equal-energy surfaces in the valence band. This probably causes also the initial gently sloping sections of the $\rho_c(\hbar\omega)$ and $\rho_L(\hbar\omega)$ curves. As seen from Fig. 2, ρ_c decreases rapidly to zero in an interval less than 0.1 eV, whereas ρ_L decreases only to approximately half its value. In light of the foregoing, the frequency dependence of ρ_L is due to momentum relaxation, and the decrease of ρ_c is governed also by the spin relaxation. In the case of spin relaxation of free carriers, owing to spin flips in the scattering processes, the ratio of the momentum-relaxation time τ_p to the spin-relaxation time τ_s contains the product of two small parameters, $(\epsilon/E_g)^2$ and $(\Delta/E_g)^2$, which are smaller in this case than 10^{-2} (E_g is the width of the band and Δ is the spin-orbit splitting). From the $\rho_c(\hbar\omega)$ and $\rho_L(\hbar\omega)$ curves, however, it follows directly that τ_s and τ_p are of the same order. This result is apparently due to the spin-relaxation precession mechanism, which is connected with the spin splitting of the conduction band.[6] If we use for this mechanism the estimate given in [7] for GaAs, namely $\tau_s^{-1} = 10^{18}\epsilon^3[\text{meV}]\cdot\tau_p$, then we actually get $\tau_s \approx \tau_p$ at $\tau_p = 10^{-13}$ sec (scattering by optical phonons or holes) and $\epsilon = 400$ meV.

The authors thank M. I. D'yakonov, E. L. Ivchenko, V. I. Perel', G. E. Pikus, and I. N. Yassievich for very useful discussions during different stages of this work.

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