Optical orientation of carriers in a nonequilibrium electron-hole plasma in CdS crystals

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Optical orientation of the magnetic moments of the holes in a dense electron-hole plasma has been observed in CdS crystals. The dependence of the degree of circular polarization of the recombination of CdS on the excitation level and on the wavelength of the exciting light is investigated.

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It is well known that the absorption of circularly polarized light by a semiconductor can lead to a preferred orientation of the intrinsic magnetic moments of the excited carriers.[1] By now, the optical orientation has been investigated for a large class of crystals of hexagonal symmetry.[2,3] In the case of cubic crystals (such as Si and III-V compounds), owing to the degeneracy of the valence band at the Γ point, rapid spin relaxation of the holes takes place, so that it is possible to orient only the electrons.[4] In hexagonal crystals (such as CdS, CdSe, and others), the anisotropic

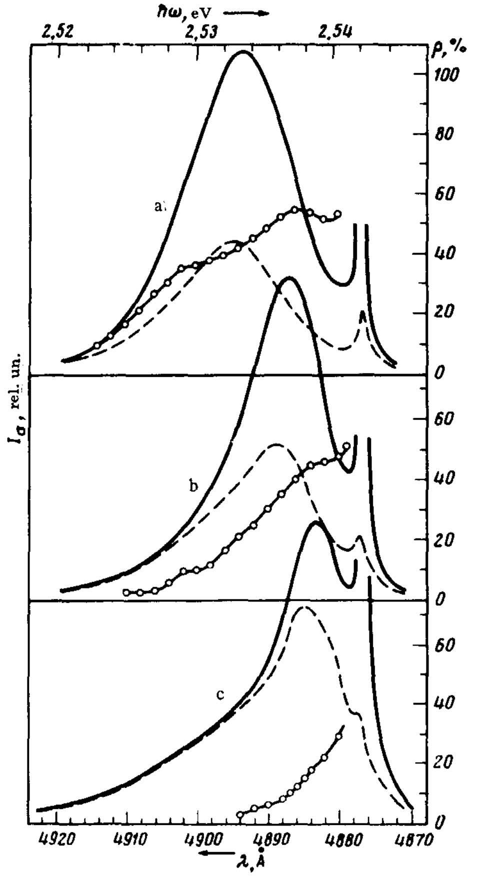

FIG. 1. Optical orientation spectra of electron hole plasma at an excitation power J: a—3×10^6 W/cm^2, b—5×10^5 W/cm^2, c—10^5 W/cm^2. Excitation with σ^+ polarized light. Solid lines—registration in σ^+ polarization, dashed—in σ^- polarization. Circles—degree of circular polarization $\rho=(I_+-I_-)/(I_++I_-)$ in %.

crystal field leads to a degeneracy at $\mathbf{K}=0$, splitting the upper valence band Γ_8 into subbands Γ_9 and Γ_7.[5] Consequently, it becomes possible to orient both types of carriers in these crystals, as well as to ensure a higher degree of spin orientation compared with cubic crystals.[6-8] The degree of orientation $P=(n_+-n_-)/(n_++n_-)$ of the magnetic moments of the carriers (where n_+ and n_- are the numbers of the carriers produced with an angular momentum directed parallel and antiparallel to the wave vector of the photon) depends only on the symmetries of the valence and conduction bands and amounts to $P=1$ in transitions from Γ_9 to Γ_7.

Optical orientations have heretofore been investigated at low nonequilibrium-carrier densities, when the interaction between particles could certainly be neglected. The purpose of the present study was to investigate the optical orientations of carriers in a nonequilibrium electron-hole plasma (EHP) of high density, when the distances between particles become comparable with the radius of the Bohr orbit of the exciton and the excitonic states vanish as a result of the screening of the Coulomb interaction. We are interested here in the extent to which the spin-relaxation times are altered under conditions of strong interparticle interactions in the EHP, and also the manifestation of the spin orientation of the carriers in the shape of the recombination radiation (RR) of the EHP.

We investigated CdS single crystals with donor density$N_D\sim3\times10^{16}$ cm^3 at helium temperatures. The circular polarizer (analyzer) was a combination made up of a polaroid and a quarter-

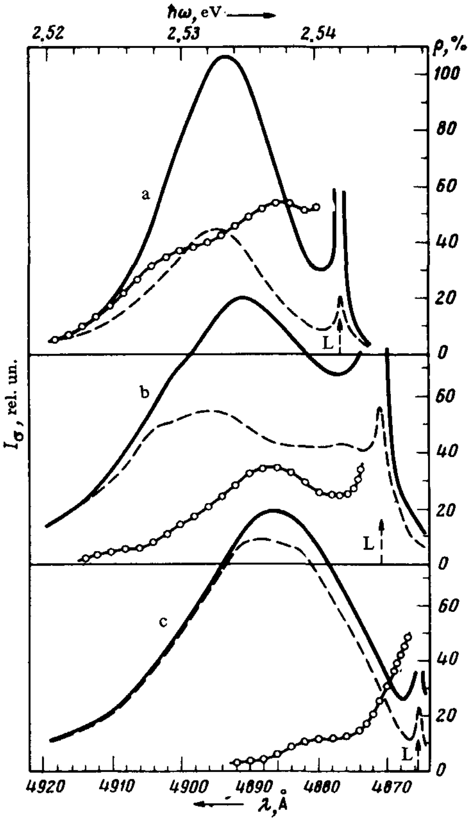

FIG. 2. RR spectra of EHP at different wavelengths of the exciting laser λ_L: a—$\lambda_L = 4877$ Å, b—$\lambda_L = 4872$ Å, c—$\lambda_L = 4866$ Å. Excitation with σ^+ polarized light $J = 3 \times 10^6$ W/cm². Solid lines—registration in σ^+ polarization, d—in σ^-polarization, circles—degree of circular polarization $\rho = (I_+ - I_-)/(I_+ + I_-)$ in %.

wave plate. The excitation was perpendicular to the surface of the crystal, and the incident-wave vector **K** was parallel to the hexagonal axis. A long-focus optical projection system was used to analyze the luminescence emerging from the sample in a direction close to normal ($\mathbf{K}_l \| C_6$). The excitation source was a pulsed tunable-frequency organic-dye-solution laser with monochromaticity $\Delta\lambda \leqslant 2$Å. The pulse repetition frequency and the duration of a single pulse were respectively 100 Hz and 10^{-8} sec. The recombination radiation was photoelectrically recorded in a strobing regime with a time resolution 2–3 nsec.

It was shown earlier that an EHP can be produced in CdS both by exciting carriers deeply in the band, or with the aid of light close in frequency to the exciton-impurity complex on a neutral donor, producing thereby carriers near the Fermi surfaces of the electron and hole bands in the EHP.[9] Figure 1 illustrates the development of the EHP emission spectrum and the behavior of the degree of circular polarization as a function of the exciting-light intensity. With increasing pump, the intensity and the width of the band increased. At pumps 3×10^6 W/cm² [Fig. 1(a)], the radiation corresponds to a degenerate electron-hole plasma of density $\sim 10^{18}$ cm^{-3} and an approximate binding energy 12 meV.[10] The degree of circular polarization of such a plasma is maximal on the "violet" edge of the spectrum and reaches 55%. With increasing concentration in the plasma, the degree of spin orientation of the carriers increases, apparently as a result of the decrease of their lifetime.

At helium temperatures, the "violet" end point of the emission spectrum of the degenerate electron-hole plasma corresponds to the energy of the photons emitted in recombination directly

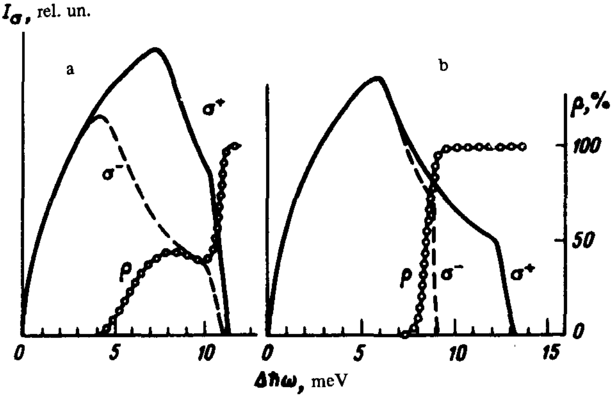

FIG. 3. Calculation of the degree of circular polarization of electron-hole plasma recombination radiation in the case: a) of oriented holes $n_h^-/n_h^+=0.5, n_e^+=n_e^-$. b) of oriented electrons $n_e^-/n_e^+=0.5, n_h^+=n_h^- \times T=0, n_n^+ + n_h^- = n_e^+ + n_e^- = 0.8\times 10^{18}$ cm^{-3}

from the Fermi surfaces of the electron and hole bands.[10] It was therefore of interest to determine the degree of optical orientation of the EHP as a function of the wavelength of the exciting light (Fig. 2). It is seen that the degree of circular polarization of the recombination radiation decreases rapidly with increasing frequency difference between the exciting laser and the "violet" end point of the EHP emission spectrum. This means that rapid ordering of the carrier spins takes place in the course of the thermalization, and that the times of the spin relaxation in this case are much shorter than in the EHP itself.

Because of the substantial difference between the Fermi energies of the carriers and because of the strong anisotropy of the effective masses of the holes, the electron-spin orientation manifests itself predominantly at the "violet" edge of the spectrum, whereas the hole orientation manifests itself within the entire EHP emission band (Fig. 3). Comparison with the experimental plots indicates that it is mainly the holes that preserve the spin orientation in the EHP. This conclusion is confirmed by measurements of the degree of circular polarization in a transverse magnetic field $H \perp C_6$. It turns out that up to fields $H=20$ kG there is no noticeable depolarization of the radiation (there is no Hanle effect). This is not surprising if it is recognized that in hexagonal CdS crystals the g-factor of the holes is anisotropic and is equal to zero at $H \perp C_6$.

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