Electron–hole plasma with completely spin-polarized carriers in ZnTe:Mn crystals

S. I. Gubarev, N. I. Vitrikhovskiĭ, A. V. Komarov, and V. B. Timofeev
Institute of Solid State Physics, Academy of Sciences of the USSR; Institute of Physics, Academy of Sciences of the Ukrainian SSR

(Submitted 24 December 1980)

Pis'ma Zh. Eksp. Teor. Fiz. **33**, 202–206 (20 February 1981)

A giant paramagnetic splitting of spin subbands is observed in ZnTe:Mn crystals in a magnetic field as the result of an exchange interaction of the carriers with the magnetic impurity. This giant splitting is greater than the electron and hole Fermi energies in the nonequilibrium electron–hole plasma. Under these conditions the electron and hole subsystems in the plasma are completely spin-polarized. The spin ordering of the carriers is accompanied by a decrease in the binding energy per particle pair in the plasma.

PACS numbers: 71.35. + z, 71.70.Gm

Study of the magnetic reflection spectra in the exciton-resonance region in ZnTe:Mn crystals has revealed an intensification of the paramagnetic susceptibility of the carriers by an exchange interaction with bound d electrons of the magnetic impurity. This intensification is seen clearly in the anomalously large Zeeman splittings of the spin states of the band electrons (or holes).[1,2] In magnetic-impurity crystals of this type it is possible to excite a degenerate electron–hole plasma with a high degree of spin ordering among the nonequilibrium carriers. With a rapid spin relaxation, we would expect the greatest spin ordering of the carriers in the plasma to occur under conditions such that the spin splittings of the electron and hole states are greater than the corresponding Fermi energies in the plasma. We have accordingly studied the recombination-radiation spectrum of the electron–hole plasma in ZnTe:Mn crystals in a magnetic field. Information on the carrier redistribution among spin states was drawn from the degree of circular polarization of the plasma emission.

In the experiments we studied ZnTe:Mn samples with various manganese concentrations, which we estimated from the amount of manganese in the material used to grow the crystals. In this letter we are reporting results for the sample with the highest concentration of this magnetic impurity, on the order of 2×10^{21} cm^{-3}. The samples were immersed in superfluid helium in a superconducting solenoid. Measurements were carried out in the Faraday configuration in magnetic fields up to 50 kOe. The electron–hole plasma was produced by bombarding the sample with a dye laser, which was itself pumped by a 100-kW pulsed N_2 laser. The excitation pulses were 5 ns long and had a repetition frequency of 25 Hz. The spectra were measured with a high-luminosity monochromator with a dispersion of 2 nm/mm. The intensity of the recombination radiation was measured with a fast-response photomultiplier and a strobe integrator. The circularly polarized light was analyzed with a quarter-wave plate and a polarizer.

In pure ZnTe crystals the equilibrium carrier density in the electron–hole plasma, corresponding to the minimum energy per particle pair, is $\sim 3\times10^{17}$ cm^{-3} (corresponding to the dimensionless parameter $r_s' \approx 2$).[3,4] In the absence of a magnetic field the shape of the spectrum and the energy position of the emission band of an electron–hole plasma with this equilibrium carrier density are both insensitive to the concentration of the magnetic impurity. In a magnetic field the emission spectrum of the electron–hole plasma in a heavily doped ZnTe:Mn sample splits into two circularly polarized bands: σ^+ and σ^- (Fig. 1). As the magnetic field is strengthened, the σ^+ component drops in energy and intensifies, while the σ^- component shifts toward a higher energy and becomes much less intense. The σ^- component weakens by more than an order of magnitude in a field ~ 7 kOe, and as H increases further we lose track of this component. At $H \geqslant 10$ kOe the emission from the electron–hole plasma becomes essentially 100% circularly polarized.

Figure 2 shows how the magnetic field affects the positions of the σ^+ and σ^- peaks in the emission band of the electron–hole plasma and the reflection band of the $1S$ excition, according to measurements for the same crystal, with a manganese concentration $\sim 2\times10^{21}$ cm^{-3}. These similar curves have a common origin in the exchange interaction of the carriers with the manganese impurity. As a result of this interaction, the electron and hole bands undergo a strong spin splitting in the magnetic field[2,5]:

$$\Delta E^e_{(\pm 1/2)} = J_e < S_M >_{H,T} S_e \qquad \Delta E^h_{(\pm 1/2,\ \pm 3/2)} = J_h < S_M >_{H,T} S_h \, .$$

Here J_e and J_h are the exchange-interaction constants, $\mathbf{S}_e$ and $\mathbf{S}_h$ are the effective electron and hole spins, and $\langle \mathbf{S}_M \rangle_{H,T}$ is the average magnetic moment at the Mn^{2+} ion; this magnetic moment determines the degree of spin polarization of the magnetic-impurity subsystem. The inset in Fig 1. shows the splitting of the spin subbands of the carriers in the electron–hole plasma in ZnTe:Mn for the case in which the splittings ΔE^e and ΔE^h are greater than the Fermi energies in the electron and hole bands. The 100% circular polarization observed experimentally over the entire width of the plas-

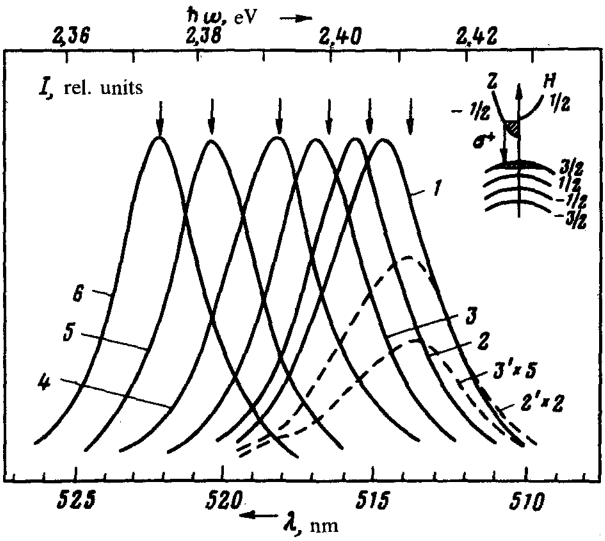

FIG. 1. Emission spectra of the electron–hole plasma in ZnTe:Mn at $T = 1.75$ K in various magnetic fields. 1) 0; 2, 2′) 3.5 kOe; 3, 3′) 7 kOe; 4) 14 kOe; 5) 25 kOe; 6) 47 kOe. In the σ^+ polarization (solid curves) the spectra correspond to the transitions $|-1/2\rangle_e \to |3/2\rangle_h$ and $|1/2\rangle_e \to 1/2\rangle_h$; in the σ^- polarization (dashed curves), the spectra correspond to the transitions $|-1/2\rangle_e \to |-1/2\rangle_h$ and $|1/2\rangle_e \to -3/2\rangle_h$. All the spectra are normalized to the maximum intensity in the σ^+ polarization. The arrows near the top show the position of the $1S$ exciton.

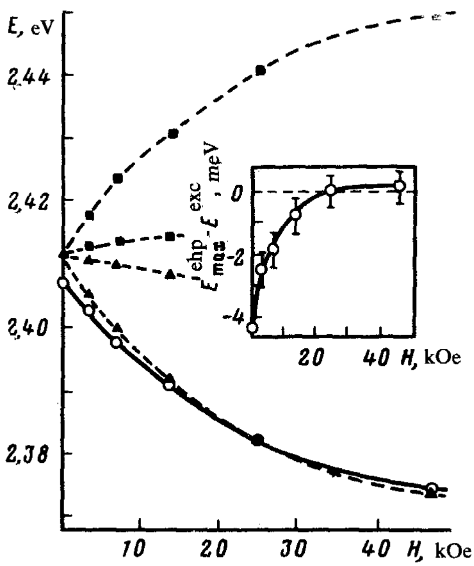

FIG. 2. Spin splitting of the $1S$ exciton in ZnTe:Mn (the filled squares represent σ^- polarization and the filled triangles denote σ^+ polarization) and the spectral position of the emission peak of the electron–hole plasma (open circles), both as functions of the magnetic field.

ma band implies a rapid carrier relaxation to the lowest-spin states.

The magnitude of the spin splitting in the valence band is greater than in the conduction band, since the holes interact more effectively with the magnetic impurity here ($J_e/J_h = 0.5$; Ref. 2). Furthermore, the Fermi energies in the hole subsystem are smaller than in the electron subsystem, in accordance with the known ratio of the effective masses of the state densities in the electron and hole bands.[3] In weak magnetic fields, therefore, the increase in the degree of circular polarization of the emission of the electron—hole plasma is caused primarily by a carrier redistribution among spin subbands in the hole subsystem. In magnetic fields $H \geqslant 40$ kOe the spin splittings exceed the Fermi energies in the electron subsystem as well as in the hole subsystem. (At $H = 47$ kOe, the width of the plasma band yields the estimates $E_F^e \approx 9$ meV, $E_F^h \approx 5.5$ meV, in comparison with the spin splittings of $\Delta E^e \approx 11$ meV and $\Delta E^h \approx 22$ meV.) At such splittings in our experiments the situation is such that the electrons and holes in the plasma are completely spin-polarized.

The spin ordering of the plasma carriers is accompanied by an interesting effect. Comparison of the spectral positions of the components in the plasma emission band and in the exciton reflection band reveals that an increase in the magnetic field is accompanied by a decrease in the difference $E_{\text{exc}} - E_{\text{ehp}}^{\text{max}}$ (see the inset in Fig. 2). This decrease amounts to 4.5 meV in a field of 47 kOe. The difference is independent of H when the spin splitting is large enough, such that there is complete spin ordering in the electron and hole bands. This behavior can be explained qualitatively on the basis of a decrease in the stability of the plasma with respect to the lowest exciton state due to the lifting of the spin degeneracy, which is equivalent to a decrease in the effective number of valleys.[6,7]

In crystals with a lower manganese concentration ($\sim 6 \times 10^{19}$ cm^{-3}) the circular polarization of the plasma emission remains less than complete, even in the strongest magnetic fields used (~ 50 kOe), and the degree of polarization varies in a nonmonotonic manner over the width of the plasma band, changing sign near the "violet" boundary. These results mean that the nonequilibrium carriers in this crystal are not completely spin-ordered and that the spin temperature is higher than the electron temperature.

The nonequilibrium carriers in the plasma could, in principle, heat the spin subsystem of the impurity. Such a heating was not seen in the results discussed here, however, since a change in the excitation level by more than an order of magnitude did not affect the spin splitting.

We thank S. M. Ryabchenko for extremely useful discussions.

[1]A. V. Komarov, S. M. Ryabchenko, O. V. Terletskiĭ, I. I. Zheru, and R. D. Ivanchuk, Zh. Eksp. Teor. Fiz. **72**, 608 (1977) [Sov. Phys.–JETP 46, 318 (1977)].
[2]A. V. Komarov, S. M. Ryabchenko, and N. I. Vitrikhovskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **27**, 441 (1978) [JETP Lett. **27**, 413 (1978)].
[3]G. Beni and T. M. Rice, Phys. Rev. **B18**, 768 (1978).
[4]R. Baltrameyunas and É. Kuokshtis, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 588 (1978) [JETP Lett. **28**, 542 (1978)].
[5]J. A. Gaj, J. Ginter, and R. R. Gałązka, Phys. Status Solidi **B89**, 655 (1978).

[6]V. S. Bagaev, T. I. Galkina, O. W. Gogolin, and L. V. Keldysh, Pis'ma Zh. Eksp. Teor. Fiz. **10**, 309 (1969) [JETP Lett. **10**, 195 (1969)].
[7]R. Zimmermann and M. Rösler, Solid State Commun. **25**, 651 (1978).

Translated by Dave Parsons
Edited by S. J. Amoretty