Kinetics of the exciton luminescence of the magnetically mixed semiconductor $Zn_{1-x}Mn_xSe$ in a magnetic field

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Experiments reveal that the intensity and shape of the exciton luminescence spectrum of the magnetically mixed cubic semiconductor $Zn_{0.99}Mn_{0.01}Se$ depend strongly on the direction of an external magnetic field. It is shown that this effect stems from a dependence of the luminescence kinetics on the magnetic-field induced anisotropy of the effective masses of free excitons in this crystal.

In magnetically mixed semiconductors, an external magnetic field $\mathbf{H}$ causes a giant spin splitting of the energy bands of free electrons and holes at low temperatures. The corresponding exciton states are formed by Coulomb-coupled carriers from certain spin subbands.[1] Such states, known in the literature as "spin excitons,"[1] have a binding energy E_{ex} in wide-gap magnetically mixed semiconductors with a cubic lattice which is essentially the same for all directions of $\mathbf{H}$ and for all possible spin subbands corresponding to certain projections of the electron spin, $n = \pm 1/2$, and of the effective spin of a hole, $p = \pm 3/2, \pm 1/2$, onto $\mathbf{H}$ (Refs. 2 and 3). It was noted in Ref. 2 that E_{ex} is independent of $\mathbf{H}/|\mathbf{H}|$, n, and p. The reason is that the effective inertial masses of the spin excitons do not depend on these factors in a cubic magnetically mixed semiconductor.[3] A significant anisotropy should arise from the effective mass M associated with the translational motion of a spin exciton under conditions of a giant spin splitting.[2] For the spin exciton with the lowest energy ($n = -1/2$, $p = +3/2$), for example, the mass $M_\parallel$, corresponding to the direction of $\mathbf{H}$, is much larger than the effective mass in the direction transverse with respect to the field, $M_\perp$. For a spin exciton with $p = \pm 1/2$ the situation is qualitatively the opposite: $M_\parallel < M_\perp$. These effects stem from a corresponding anisotropy of the effective masses in the hole spin subbands formed as a result of the giant spin splitting by the effective exchange field[4] $\mathbf{G}_h$, since the effective spin of a hole is now quantized along the direction of $\mathbf{G}_h \parallel \mathbf{H}$, rather than along the direction of the wave vector.

This difference between $M_\parallel$ and $M_\perp$ might be manifested in spatial-dispersion effects in polariton reflection spectra,[5] but the pronounced broadening of a spin exciton in a magnetically mixed semiconductor has prevented observation of such effects so far.

In this letter we are reporting the first observation of a manifestation of an effective-mass anisotropy of spin excitons in the shape and intensity of an exciton luminescence line. It is determined by the kinetics of the energy and spatial distributions of the spin excitons. It was shown in Ref. 6 that since the excitons generated by light near the surface of a crystal become redistributed deeper into the sample over their lifetime,

there is a decrease in the part of the exciton luminescence which comes from excitons which are relatively far from the surface of the crystal. As a result, the change in effective mass, which is related to the mobility of a free exciton in the direction of its density gradient (i.e., along the normal to the plane of the sample, $\mathbf{n}$), may lead to change in both the intensity and the shape of the spin-exciton-luminescence signal. Changes of this sort in M in magnetically mixed semiconductors are easily arranged by rotating $\mathbf{H}$ with respect to $\mathbf{n}$ (Ref. 3).

An experiment was carried out on a $Zn_{0.99}Mn_{0.01}$ Se crystal with a cubic lattice at $T = 2$ K. The sample was inside a superconducting solenoid with a transverse slot. The sample was excited by the beam from a He–Cd laser with $h\nu = 2.807$ eV (at a power $\leqslant 10$ mW). The exciton luminescence spectrum was recorded for two directions of the magnetic field: (1) $\mathbf{H}\|\mathbf{n}$ and (2) $\mathbf{H}\perp\mathbf{n}$. The directions of the wave vectors of the incident light, $\mathbf{k}$, and of the emitted light, $\mathbf{k}' \simeq -\mathbf{k}$, remained along the normal to the cleaved surface ($\mathbf{k}\|\mathbf{n}\|[110]$), while the polarizations of the exciting light and the detected light were the same. They corresponded to σ^+ and to a linear polarization $\mathbf{E}\perp\mathbf{H}$ ($\mathbf{E}$ is the polarization vector of the light wave) in the first and second cases, respectively. It was thus possible to excite and detect excitons for which the projection of the resultant spin of the electron and the hole onto the direction of $\mathbf{H}$, $n + p = 1$, corresponded to the lowest-energy spin-exciton component, $n = -1/2$, $p = 3/2$. In the former case, the diffusion of the excitons into the crystal was thus controlled by the effective mass $M_{\|}$, while in the latter case it was controlled by $M_{\perp}$.

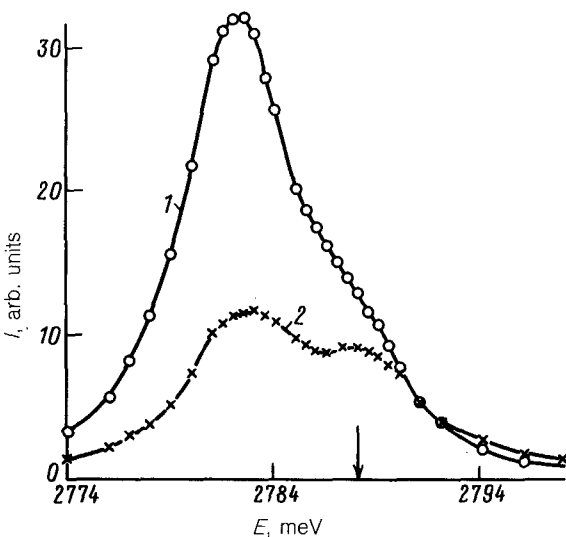Figure 1 shows emission spectra detected in a field of 26 kOe in different direc-

FIG. 1. Emission spectra in the exciton region, normalized to the amplitude of the luminescence at $H = 0$ in the corresponding polarization. 1—$\mathbf{H}\|\mathbf{n}$; 2—$\mathbf{H}\perp\mathbf{n}$. The strength of the magnetic field is $H = 26$ kOe; the temperature is $T = 2$ K. For convenience in comparison, curve 2 has been multiplied by a factor of 2, as discussed in the text proper. The arrow shows the energy of the σ^+ exciton (the lowest-lying one) found from the reflection spectra at the same field and at the same temperature.

tions with respect to **n**. The intensities shown here for the exciton luminescence have been scaled for convenience in comparison. Each intensity has been normalized to the amplitude of the emission in the corresponding polarization and configuration of the experiment for $H = 0$. The amplitude of the light for the configuration $H \perp \mathbf{k}$ (curve 2) was multiplied by another factor $r_\perp = 2$, which reflects the expected ratio of the intensities in a strong field to the intensity at $H = 0$ under the experimental conditions. The corresponding matrix elements are taken into account here, as is the fact that there is a complete transfer of the exciton population to the lowest-energy spin state. We see that, when the direction of **H** with respect to **n** is changed, the change in the intensity is quite different from the expected factor of 2. In addition, there is a substantial change in the shape of the band. In the case $H \| \mathbf{n}$, for example, the maximum intensity normalized to $H = 0$ exceeds the corresponding value for $H \perp \mathbf{n}$ by a factor of essentially 6, i.e., by a factor three times that expected.

To describe these results, we consider the contributions from the lower and upper polariton branches, $I_1(\epsilon)$, and $I_2(\epsilon)$, respectively, to the intensity σ^+, of the exciton luminescence corresponding to the lowest-lying spin exciton:[6]

$$I_i(\epsilon) = I_{1i}(\epsilon) + I_2(\epsilon) = \frac{r_i \Phi_1(\epsilon)}{L_i \alpha_1(\epsilon) + 1} + I_2(\epsilon), \tag{1}$$

where $i = \|, \perp$ specifies the configuration ($H \| \mathbf{n}$ or $H \perp \mathbf{n}$); $r_\| = 1$; $r_\perp = 2$; the factor $\Phi_1(\epsilon)$ is independent of the direction of the energy transfer; $\alpha_1(\epsilon)$ is the optical absorption coefficient; and L_i is the diffusion length for a spin exciton, determined along the direction of the density gradient of the spin excitons ($\|\mathbf{n}\|\mathbf{k}$). The low-energy part of the spectrum is dominated by $I_1(\epsilon)$, while both polariton branches contribute to the short-wavelength wing. The intensity of the main peak of the exciton luminescence line in this figure is thus given approximately by the first term in (1). We can therefore write the relative change in this intensity due to the change in the direction of **H** as follows:

$$\frac{I_\|(\epsilon_{m1})}{I_1(\epsilon_{m1})} \approx 2 \frac{1 + \xi (M_\|/M_\perp)^\gamma}{1 + \xi}, \tag{2}$$

where the parameter $\xi = L_\| \alpha_1(\epsilon_{m1})$ is determined at the energy of the exciton-luminescence peak, ϵ_{m1}, in the Faraday geometry ($H \| n$), while γ is found from $(M_\|/M_\perp)^{-\gamma}$. It takes on values in the interval 1–1.5, depending on the particular exciton scattering mechanism.[7] The ratio in (2) may be considerably greater than unity if ξ is not too small and if $M_\| > M_\perp$. To calculate M, we used equations from Ref. 3 and the band parameters of pure ZnSe: $\gamma_1 = 3.77$; $\gamma_2 = 1.24$; $\gamma_3 = 1.67$, and $m_e = 0.14 m_0$. The calculations yielded $M_\| = 1.69 m_0$ and $M_\perp = (0.32–0.34) m_0$. We have thus succeeded in finding a quantitative description of the experimental ratio in (2) (≈ 5.8) with $\gamma = 5/4$ and $\xi = 0.4$. Corresponding to this value of ξ with $\alpha \approx 10^4$ cm^{-1} are the values $L_\| \approx 0.4 \times 10^{-4}$ cm and $L_\perp \approx 3 \times 10^{-4}$ cm. These values agree in order of magnitude with the values found for L in Refs. 6 and 8 for crystals which exhibit a doublet structure in their exciton luminescence.

The appearance of a high-energy hump on the exciton luminescence line upon the switch to $H \perp \mathbf{n}$ is explained in this model as resulting from a relative increase in $I_2(\epsilon)$

against the background of a decrease in the $I_{11}(\epsilon)$ component in (1), because of a higher mobility of a spin exciton in the $n\perp H$ direction. This explanation may not be complete, since the exciton damping is large (comparable to or greater than the longitudinal-transverse splitting). Moreover, the reason for the two humps in the exciton luminescence might also be, according to Ref. 8, a resonant $\alpha_1(\epsilon)$ dependence at $L_i\alpha_1(\epsilon) > 1$ (a reabsorption accompanying an effective diffusion). However, the ambiguity in the interpretation of the reason for the dip in the exciton luminescence does not alter our conclusion that the observed effect is generally related to an anisotropy of the translational mass of the exciton, nor does it alter the estimates made on the basis of the analysis of the low-energy maximum.

The depth at which light is absorbed under our experimental conditions, in general, increases in a field $H\perp K$ after the high-energy spin-exciton component, which is active in σ light, splits off by an energy greater than the amount by which the energy of the exciting light exceeds the energy of the exciton at $H = 0$. This situation is reached at $H \geqslant 10$ kOe. In the absence of an exciton diffusion, this circumstance should have absolutely no effect on the intensity or shape of the luminescence, since the absorption coefficient for the light emitted from the crystal changes to the same extent that the absorption accompanying the excitation changes. When there is a diffusion, on the other hand, this circumstance could cause at most some lowering of the observed manifestation of the anisotropy of the translational effective masses, according to (1). The reason is that there is a decrease in the ratio of the diffusion length L to the absorption depth α^{-1}.

A change in $I(\epsilon)$ upon a reorientation of H similar to that described above was observed not only during nearly resonant exciton excitation but also during excitation with $h\nu = 3.397$ eV, i.e., at an energy well above the energy of a spin exciton. This observation agrees with the general thrust of the interpretation above. However, the effect was weaker than in the case of excitation with $h\nu = 2.807$ eV. The latter circumstance should be analyzed with allowance for possible spatial redistributions of excitons during their energy relaxation.

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