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Hole spin polarization in the exchange field of the dilute magnetic (Ga,Mn)As semiconductor studied by means of polarized hot-electron photoluminescence spectroscopy

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We have studied the hole spin polarization in the exchange field of ferromagnetic (Ga,Mn)As by means of polarized hot-electron photoluminescence (HPL). We found that the holes contribute to the HPL spectrum in two very different states. Most of the holes are strongly localized on Mn acceptors whose spin polarization under circularly polarized excitation is not sensitive to the exchange field arising in the ferromagnetic state below the Curie point. In contrast, the spin polarization of weakly localized or delocalized holes strongly decreases below the Curie temperature. This effect is explained by the hole spin orientation in the exchange field. The spin splitting of the weakly localized or delocalized holes in the mean exchange field is estimated $\Delta \simeq 6$ meV (equivalent to the built-in exchange field $\simeq 90$ T).

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Introduction. The dilute magnetic semiconductor (DMS) (Ga,Mn)As is of great interest since the discovery of a transition into a ferromagnetic (FM) phase at low temperatures.[1] The unique combination of semiconducting and magnetic properties in this material opens up new prospects for realizing semiconducting spintronic devices.[2,3] However, in spite of intense experimental and theoretical studies and even successive demonstration of prototype spintronic devices,[4] a profound understanding of physics of (Ga,Mn)As has not been achieved. Most of the proposed models assume that holes produced by the Mn dopants play a key role in mediating the interaction between spins ($S=5/2$) of Mn ions. However, the principally unresolved problem in the physics of (Ga,Mn)As concerns the state of the holes mediating the ferromagnetism. The Zener model, based on a Rudermann-Kittel-Kasuya-Yosida (RKKY)-type exchange interaction,[5–8] implies that the chemical potential of the holes lies in the valence band. Alternatively, it has been suggested that the holes in (Ga,Mn)As reside in the impurity band.[9–12] Strong evidence for the formation of an impurity band has been provided by optical[13–16] and transport[17,18] studies.

In this Rapid Communication, we present a study of the effect of ferromagnetic phase transition on the hole spin orientation in (Ga,Mn)As by means of hot-electron photoluminescence (HPL).[13,15] The HPL circular polarization under circularly polarized excitation provides detailed information on spin-relaxation mechanisms as well as on direction and value of exchange fields acting on the hole spins. Our results on the temperature dependence of the HPL polarization demonstrate that two types of holes contribute to the HPL spectrum of the ferromagnetic (Ga,Mn)As state. The HPL circular polarization related to the recombination of the delocalized holes (most probably in the impurity band) shows a strong decrease below the Curie temperature. In contrast, the HPL circular polarization related to the recombination of the Mn-acceptor-bound holes is almost insensitive to the transition into the ferromagnetic state. This difference in HPL polarization is explained by the different nature of the wave functions of delocalized holes and Mn-acceptor-bound holes. The exchange field acting on delocalized holes is found to significantly (about three times) exceed the exchange field of individual Mn ions in Mn-acceptor-bound holes.

Samples and experimental. The 500-nm-thick (Ga,Mn)As films for this study were grown at 250 °C by molecular-beam epitaxy on semi-insulating GaAs (001) substrates covered with 100 nm GaAs buffer layers. The studied paramagnetic (PM) and three FM samples have a Mn content of $x=0.01$ (PM), and $x=0.025, 0.043, 0.07$ (FM), respectively. In addition, a Mn-doped 1000-nm-thick GaAs film $x \simeq 10^{-5}$ grown at 540 °C was used as reference sample (R). Superconducting quantum interference device (SQUID) magnetometry was used to determine the magnetic properties of the (Ga,Mn)As DMS. The laser power densities focused on the sample were varied in the range from 100 to 200 W cm^{-2}. The photoluminescence (PL) spectra were dispersed by a Jobin-Yvon U-1000 monochromator equipped with a cooled GaAs photomultiplier. The HPL circular polarization under circularly polarized excitation was measured in the backscattering geometry using a photoelastic modulator in the wide temperature range of $T=4-200$ K (see Refs. 13 and 15 for details). The degree of circular polarization was defined by the common expression $\rho_c=(I^+-I^-)/(I^++I^-)$, where I^+ and I^- are the intensities polarized like the exciting light or opposite to it, respectively.

Results and discussion. Figure 1 shows intensity (solid lines) and circular polarization of the HPL obtained from the PM and FM GaMnAs DMS samples at $B=0$ under circularly polarized excitation. The HPL spectra obtained from (Ga,Mn)As DMS at low temperatures (see spectra 1 and 2 for $T=4$ K) are due to the recombination of hot electrons with holes bound to single Mn acceptors or to the Mn-acceptor impurity band (i.e., $1hh \rightarrow 1e \rightarrow A^0$ type transitions) as illustrated schematically in the inset of Fig. 1(a).[13,15] In the doped GaAs:Mn reference (R) sample the HPL spectrum spreads from the point of electron generation in the conduction (c) band (marked as $\hbar\omega_0$ on the high-energy side of the spectra in Fig. 1) to the bottom of the conduction band. In the DMS samples, the impurity-band formation leads to the blueshift of the HPL high-energy cutoff.

In our previous studies we have demonstrated that HPL in (Ga,Mn)As DMS is linearly/circularly polarized under linearly/circularly polarized excitation. A further careful in-

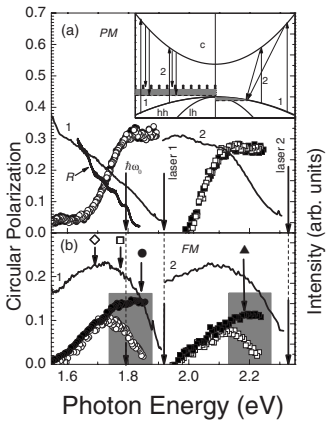

FIG. 1. Intensity (solid lines) and polarization (open and solid symbols) HPL spectra of samples (a) PM (Ga,Mn)As and doped GaAs:Mn *R* (b) FM (x=0.043) excited with a Kr^+ [1.916 eV, shown by arrow down (1)] and Nd YAG laser [2.33 eV, shown by arrow down (2)] at T=4 K (open symbols) and T=100 K (solid symbols). Inset of (a) is schematic of HPL spectroscopy for acceptor and impurity-band holes (left) and for valence-band holes (right) and for two excitation energies [arrows (1) and (2) pointing up].

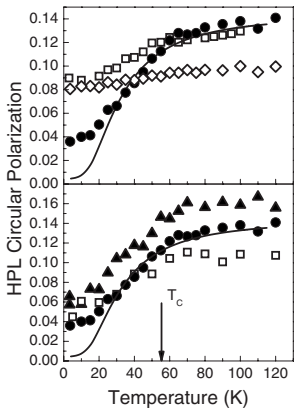

FIG. 2. Temperature dependence of the HPL circular polarization measured under circularly polarized excitation with laser/HPL detection photon energies: (a) 1.916/1.835 (solid circles), 1.916/1.773 (open circles), and 1.916/1.686 eV (open diamonds). (b) 1.833/1.748 (solid triangles), 1.916/1.835 (solid circles), and 2.33/2.182 eV (open squares) for bulk FM (x=0.043) DMS. The solid curve in (a) and (b) represents the fit obtained with Eq. (4).

spection of the HPL circular polarization spectra from PM and FM DMS samples reveals their very different temperature behavior. In PM DMS samples, the HPL circular polarization spectra are not sensitive to the sample temperature over a wide range, as shown for T=4 K and T=100 K in Fig. 1(a) (compare open and solid circles for excitation at 1.916 eV). Similarly, the effect of temperature on the HPL circular polarization is absent for excitation with a much higher laser energy of $\hbar\omega_{ex}$=2.33 eV [see open and solid squares in Fig. 1(a)]. In contrast, the HPL circular polarization of the FM DMS samples exhibits a strong dependence on temperature for both excitation energies (1.916 and 2.33 eV). Comparison of the spectra obtained from the (x=0.043) FM sample at T=4 K and T=100 K [see Fig. 1(b)] clearly demonstrates the strong increase in the circular polarization with temperature measured on the high-energy side of the HPL spectrum for both excitation energies [gray regions in Fig. 1(b)]. On the other hand, the polarization measured at energies below the HPL maximum only slightly increases with temperature. This means that two kinds of optical transitions contribute to the HPL spectrum. More intense and almost temperature-independent transitions contribute predominantly at energies below $\hbar\omega_0$, while less intense but strongly temperature sensitive transitions dominate on the high-energy tail shown by the gray area. To demonstrate more clearly the effect of temperature on the HPL polarization properties, the $\rho_c(T)$ has been measured [see solid circles and open squares in Fig. 2(a)] at two spectral points of the high-energy tail indicated in Fig. 1(b) by (●) and (□). Both dependences indicate an abrupt decrease in the HPL circular polarization at temperatures below $T_C \sim 60-65$ K which is close to the Curie temperature measured in this FM (x=0.043) sample by SQUID ($T_C \simeq 55$ K). The temperature dependence of the HPL circular polarization measured at an energy below $\hbar\omega_0$ [see e.g., spectral point indicated by (◇) in Fig. 1(b)] depicts much smaller decrease at temperatures below T_C [see open diamonds in Fig. 2(a)]. A similar behavior of the HPL circular polarization on temperature was observed for higher ($\hbar\omega_{exc}$=2.33 eV) [spectrum 2 in Fig. 1(b)] as well as for lower excitation energy ($\hbar\omega_{exc}$=1.833 eV). The temperature dependences of the HPL circular polarization measured in the high-energy tail at these excitation energies are presented in Fig. 2(b) by solid triangles (1.833 eV) and open squares (2.33 eV, measured in the spectral point marked by (▲) in Fig. 1(b).

We now discuss the effect of temperature on the HPL circular polarization in (Ga,Mn)As DMS. The absorption of circularly polarized light in GaAs-type semiconductors leads to the spin orientation of the excited electrons. The HPL can thus be circularly polarized[19] when oriented electrons recombine with equilibrium nonpolarized holes. The HPL circular polarization in GaAs significantly exceeds the maximum theoretical value (0.25) obtained for band-gap PL due to spin momentum (s-p) correlation, which occurs for electrons excited and recombining with finite k vector.[19] In the FM DMS samples, the circular polarization is lower than in the PM samples due to fast electron momentum relaxation, leading to partial distraction of the s-p correlation in the point of hot-electron generation.

The effect of temperature on the HPL circular polarization in FM DMSs can be understood by a model assuming the presence of a built-in exchange field B_{eff}, which in the ferromagnetic phase is proportional to the sample spontaneous magnetization $B_{eff} \propto \lambda M_s(T)$, where λ is the Weiss constant and $M_s(T)$ is the sample spontaneous magnetization, expected within the textbook Weiss mean-field theory.[20] Circularly polarized laser excitation creates spin-oriented electrons in the conduction band whose spins are perpendicular to the built-in field lying in the plane of the studied samples. Spin polarized electrons recombine with acceptor- or impurity-bound holes, whose polarization depends on the magnitude and direction of the built-in exchange field.

The destruction of the ferromagnetic order, i.e., built-in magnetic field ($\boldsymbol{B}_{eff} \rightarrow 0$) with temperature increase restores the HPL circular polarization. Qualitatively the effect of the

built-in field on HPL circular polarization manifests itself for hot electrons recombining with holes bound to Mn acceptors as well as with weakly localized or free holes. However, the effect of B_{eff} on the hole spin polarization strongly depends on the character of the hole wave function, which determines the selection rules for HPL. In the following, we will demonstrate that holes contribute to the HPL circular polarization of FM DMS samples in two different electronic states. We start with holes bound to Mn acceptors (Mn ion $3d^5$ shell plus valence-band hole) whose model has been developed in Ref. 21. The Mn-acceptor ground state is characterized by the total angular momentum $F=1$. The splitting of this state in the internal field B_{eff} is given by the following Hamiltonian:

$$H = \mu_B g_{F=1} F_x B_{eff}, \tag{1}$$

where F_x is the angular-momentum projector on the x axis, which coincides with the B_{eff} (easy axis) direction and perpendicular to the wave vectors of the laser and PL photons. For the sake of simplicity we assume that the excited states $(F=2,3,4)$ of A^0_{Mn} are empty and $\mu_B g_{F=1} B_{eff} \ll |E_{F=2} - E_{F=1}|$ as well as the final state of neutral Mn acceptors after recombination is polarized. The expression for the HPL circular polarization induced by external magnetic field in Faraday geometry (magnetic field parallel to the wave vector of the HPL photon) has been obtained in Ref. 21. The results of this calculation can be easily adopted to the present case by proper choice of the angle between built-in magnetic field and direction of the HPL detection. This angle equals $\pi/2$ in the present study. With these assumptions one can get the following expression for the polarization of the optical transition hot-electron-neutral Mn acceptor:

$$\mathcal{P}_c = 4S \frac{\left(3Q^2 + \frac{32}{15}\gamma_2^2 k_0^4\right)\cosh(\tilde{y}) + 2Q^2 - \frac{32}{15}\gamma_2^2 k_0^4}{\left(13Q^2 + \frac{32}{15}\gamma_2^2 k_0^4\right)\cosh(\tilde{y}) + 7Q^2 - \frac{32}{15}\gamma_2^2 k_0^4 + \frac{1}{4}\alpha\chi}, \tag{2}$$

where

$$\chi = \left(\frac{32}{105}\gamma_2^2 k_0^4 + \frac{48}{5}\gamma_2^2 k_0^2 (E_0 + \gamma_1 k_0^2)\right)\cosh(\tilde{y}) - \left(\frac{32}{105}\gamma_2^2 k_0^4 - \frac{32}{15}\gamma_2^2 k_0^2 (E_0 + \gamma_1 k_0^2)\right),$$

$$\tilde{y} = \frac{\mu_B g_{F=1} B_{eff}}{kT}, \quad Q = (E_0 + \gamma_1 k_0^2)^2 + 4\gamma_2^2 k_0^4.$$

The Luttinger parameters γ_1 and γ_2 determine the spectrum of heavy $E_{hh}=(\gamma_1-2\gamma_2)k^2$ and light $E_{lh}=(\gamma_1+2\gamma_2)k^2$ holes in GaAs, while $E_0=110$ meV is the Mn-acceptor binding energy. The wave vector of the recombining electrons k_0 is determined by the energy of the exciting photons and the carrier masses in valence (m_{hh}) and conduction band (m_e): $k_0^2 = 2\mu \frac{\hbar\omega_0 - E_g}{\hbar^2}$, where $\mu^{-1} = m_e^{-1} + m_{hh}^{-1}$, while $2S$ is the mean spin of electron in the point of creation with k_0.[21] The parameter α is determined by the character of the valence band supplying electrons into the conduction band ($\alpha=-1$ for $hh \rightarrow e$ excitation). The temperature dependence of $\mathcal{P}_c$ is described by the parameter $\tilde{y}$. The analysis of Eq. (2) reveals that we can expect only a small increase in the HPL circular polarization with temperature increase above the Curie temperature $T \rightarrow \infty$ (i.e., $y \rightarrow 0$) $\mathcal{P}_c(T=\infty)/\mathcal{P}_c(T=0)=1.05$. The weak effect of temperature on $\mathcal{P}_c$ is caused by the complex character of the hole wave function in Mn acceptors, whose ground state is formed by antiferromagnetically coupled $3d^5$ electrons $(S=5/2)$ and holes $(J=3/2)$. Our calculations have demonstrated that interaction of the hole with two or more Mn ions leads to the further decrease in the $\mathcal{P}_c(T=\infty)/\mathcal{P}_c(T=0)$ ratio. This means that Eq. (2) can be used also for the qualitative analysis of the $\mathcal{P}_c(T)$ in DMS samples, where the Mn-acceptor impurity-band formation is expected. This theoretical prediction explains well the observed weak temperature dependence of the $\mathcal{P}_c(T)$ on the low-energy side (at energies below the point labeled as $\hbar\omega_0$) of the HPL spectrum, but it fails to explain the temperature dependence on the high-energy side.

The strong effect of temperature on the HPL polarization observed on the high-energy side can be explained by the assumption that holes are not bound to individual Mn acceptors but localized on disorder potential fluctuations. Such states, for example, can be formed in the impurity band by excited states of Mn acceptors. The wave functions of holes localized on different potentials can overlap, thus forming the band of delocalized states. The states of this narrow band can be calculated in the model of zero radius potential.[22] In this model, the binding energy (E_l) is considered as a parameter, while the symmetry of the hole wave function is considered exactly. The effect of magnetic field on this state is given by the following Hamiltonian:

$$H_B = \mu_B g_1 (J_x B_{eff}), \tag{3}$$

where g_1 is the g factor of the hole, J_x is the x component of the angular-momentum projector of the hole total angular momentum $J=3/2$. We assume that the hole binding energy significantly exceeds the Zeeman energy $|E_l| \gg \mu_B g_1 B_{eff}$. With this assumption and using the hole wave functions of the Γ_8 valence subband, the HPL polarization is obtained:

$$\mathcal{P}_c = \frac{4}{5}S\left[\frac{16}{5}\gamma_2^2 k_0^4 \cosh\left(\frac{3}{2}\widetilde{y}_1\right) + \left(E_l^2 + \frac{8}{5}\gamma_2^2 k_0^4\right)\cosh\left(\frac{1}{2}\widetilde{y}_1\right)\right] / \left\{\left(\frac{3}{10}E_l^2 + \frac{81}{25}\gamma_2^2 k_0^4 + \frac{4}{5}\alpha\gamma_2^2 k_0^4\right)\cosh\left(\frac{3}{2}\widetilde{y}_1\right) + \left[E_l^2 + \frac{68}{25}\gamma_2^2 k_0^4 + \frac{16}{5}\alpha\gamma_2 k_0^2\left(E_l - \frac{\gamma_2 k_0^2}{7}\right)\right]\cosh\left(\frac{1}{2}\widetilde{y}_1\right)\right\} \tag{4}$$

where $\tilde{y}_1 = \frac{g_1 \mu_B B_{eff}(T)}{kT}$ and E_l is the binding energy of holes weakly bound to Mn acceptors. The Eq. (4) was obtained by assuming that holes occupy energy states in accordance with the Boltzmann statistics. This assumption is valid in the present case when the wave-function overlap of holes located on neighboring disorder potentials can be neglected. For GaAs valence-band parameters, $g_1=1$ and hole binding energy $E_l \sim 10$ meV the Eq. (4) predicts a significant increase in the HPL circular polarization above the Curie temperature, which is in good agreement with the experimental observation. The solid curve in Fig. 2(a) represents a fit of $\mathcal{P}_c(T)$ obtained for the excitation energy $\hbar\omega_{ex}=1.916$ eV ($k_0 \simeq 10^7$ cm^{-1}).[23] We believe that the deviation of the fit from the experimental data at low temperature is related to the

admixture of Mn-acceptor-related transitions, which is almost temperature independent [see Eq. (2)]. The best agreement of the data (also for 1.833 and 2.33 eV excitations) is obtained with an effective exchange field $B_{eff}(T) \simeq 90$ T at $T=0$ K. For the sake of simplicity we assume that the temperature dependence of the exchange field $B_{eff}(T)$ is given rather by the standard textbook Weiss mean-field theory than a more complex behavior.[24] At $T=0$ K this spontaneous exchange field induces a significant Zeeman splitting of the delocalized hole states $\Delta_{pd}=g_1\mu_B B_{eff}=\beta M_s(T=0)/g\mu_B$ $=6$ meV [where β is p-d exchange integral, $M_s(T=0)$ is the spontaneous magnetization of the sample, and $g=2$ is the electron g factor in the Mn d shell). This value of 6 meV is five times smaller than that estimated in the RKKY-like model of Ref. 6 (30 meV) for the Mn content of ($x=0.05$), which is close to the present case ($x=0.043$). Note that the spin splitting of the weakly bound or delocalized holes is about three times larger than that measured for Mn-acceptor-bound holes in doped samples.[25] This leads to the conclusion that weakly bound or delocalized holes interact with a larger number of Mn ions due to their collective nature. Therefore, we conjecture that the impurity-band delocalized holes are responsible for ferromagnetism in (Ga,Mn)As DMS despite their low density compared to the acceptor-bound holes. Coexistence of the holes in two states means that (Ga,Mn)As is microscopically not uniform. The presence of the noninteracting neutral Mn acceptors can be related to the interstitial Mn (Mn_I) or As antisites (As_{Ga}), both acting as double donors. Due to inhomogeneous distribution, these defects significantly reduce the local density of holes inside some regions where the exchange coupling between the remaining uncompensated neutral (Mn_{Ga}) acceptors is strongly suppressed. Such nanoislands consisting of substitutional Mn_{Ga} acceptors surrounded by interstitial Mn_I donors have been recently detected by Raman spectroscopy of FM DMS.[26]

Comparison of the HPL spectra from PM and FM samples clearly demonstrates that in both cases the high-energy tail extends about up to the laser line, but only the high-energy tail of FM samples reveals a strong temperature dependence of its circular polarization. This is clear evidence for the existence of delocalized states in the impurity band of FM (Ga,Mn)As DMS. Our analysis of the HPL circular polarization spectra of FM samples shows that the delocalized states cover a wide range of states [shown by gray regions in Fig. 1(a)], which extends from the isolated Mn-acceptor state (arrow labeled by $\hbar\omega_0$) almost up to the top of the valence band. In agreement with our previous studies[13,15] we assume that these weakly bound or delocalized states rather belong to the Mn-acceptor impurity band than to the valence band. Only weakly localized holes can contribute to the HPL for high-energy excitations due to the uncertainty of the wave vector k_l, which is inversely proportional to the hole localization length l. In contrast, for free valence-band holes we expect a decrease in their contribution to the HPL spectrum with an increase in the excitation energy; as such HPL would be caused by less probable indirect transitions [see inset of Fig. 1(a)]. However we cannot exclude the presence of the holes in the valence band while they were not detected in the HPL spectra so far. The contribution of these holes into HPL is expected near the band-gap PL, where it can be masked by PL from substrate.

Conclusions. In summary our study of the temperature dependence of the HPL circular polarization has revealed the existence of holes in two states in ferromagnetic (Ga,Mn)As DMS. Most of the valence-band holes are bound to Mn acceptors, while minority of the holes occupies a broad band of weakly bound or delocalized states. These states retain the polarization properties of the Mn acceptors and can thus be assigned to the Mn-acceptor impurity band. The exchange field acting on delocalized holes is found to be three times stronger than that acting on holes in the Mn-acceptor-bound state. This supports our assumption that impurity-band delocalized holes are responsible for the expansion of ferromagnetic order over the DMS layer.

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