

Picosecond dynamics of magnetic polarons governed by energy transfer to the Zeeman reservoir

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(Received 1 May 1997)

We suggest a mechanism of magnetic polaron formation in semimagnetic quantum wells in external magnetic fields, which is characterized by a transfer of energy from the carrier-magnetic-ions exchange reservoir to the Zeeman reservoir. This mechanism is related to the anisotropic exchange interaction due to the spin structure of the heavy-hole state. The polaron formation is determined by a precession of a total spin of magnetic ions inside the exciton volume in the sum of external and exchange field and happens inside the short time of about 10 ps. Experimentally, this mechanism manifests itself in photoluminescence spectra as an increase of the magnetic polaron shift at magnetic fields applied parallel to the quantum-well plane. [S0163-1829(97)00739-X]

I. INTRODUCTION

The electronic properties of semimagnetic (diluted magnetic) semiconductors such as $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ are determined by the strong exchange interaction of carrier spins with the magnetic moments of magnetic ions (Mn ions). This exchange interaction induces considerable changes of the energy spectra of free carriers at external magnetic fields and manifests itself in the giant Zeeman splitting of band states and in the giant Faraday rotation.¹ In the absence of external fields it leads to a magnetic polaron formation via the ferromagnetic alignment of magnetic-ion spins inside the volume of carrier localization.²

Compared to the nonmagnetic semiconductors, semimagnetic ones have an additional system of magnetic ions, which couples with the carrier and phonon systems.^{3,4} The dynamics of energy and spin relaxation of photocarriers in semimagnetic semiconductors is strongly influenced by carrier interaction with the system of magnetic ions, which in turn dissipates excess energy into the phonon system via spin-lattice relaxation. The effective interaction between the carriers and the Mn-ion system has been well documented in experiments on photoinduced magnetization, where a magnetic moment of optically oriented photocarriers is transferred into the system of Mn ions.⁵⁻⁷ Nowadays, interest in the mentioned effects is stimulated by the successful preparation of quantum-confined heterostructures based on semimagnetic semiconductors. Spin superlattices formed at external magnetic fields,⁸ two-dimensional magnetic polarons,

modification of the magnetic properties in quasi-two-dimensional semimagnetic-semiconductor layers,⁹⁻¹¹ and subpicosecond spin dynamics of the magnetic-ions system,¹² are only some examples of the new physical phenomena, which became accessible for investigation.

The dynamics of spin organization in the process of exciton magnetic polaron formation has been studied recently experimentally and theoretically in bulk materials and in quantum-well structures (see Refs. 10 and 13 and references therein). It is contributed by various relaxation processes: dephasing and spin relaxation of photocarriers, spin relaxation of magnetic ions, and exciton recombination, which limits a polaron lifetime. The hierarchy of characteristic relaxation times contributing to the magnetic polaron formation has been established recently.¹³⁻¹⁶ After photogeneration and localization of excitons, the polarization of the localized Mn spins inside an exciton volume emerges in the time scale of about a hundred picoseconds. The process of magnetic polaron formation involves both spin and energy relaxation. In the formation process, energy and spin momentum of the localized spins aligned by the exciton exchange field are dissipated with a relaxation rate of about 10^{10} s^{-1} . The spin-lattice relaxation rate of $10^4\text{-}10^7 \text{ s}^{-1}$ is too slow to determine the time scale of magnetic polaron formation. It has been suggested that the excess energy in the polaron formation is transferred to the spin-spin reservoir of interacting Mn spins and the conservation of spin momentum is broken by strong nonscalar spin-spin interactions.¹³ The polaron formation times are such that an equilibrium polaron

energy is not always achieved during the exciton lifetime. This dynamical effect on the magnetic polaron energy has been found to be strong at structures with low Mn concentrations.¹⁵

In this paper we suggest a faster mechanism for the magnetic polaron formation in semimagnetic quantum wells present at external magnetic fields, which has its origin in a transfer of the carrier–magnetic-ions exchange energy to the energy reservoir determined by interaction of localized Mn spins with the external magnetic field, known as the Zeeman reservoir.¹⁷ The dynamics of magnetic polaron formation due to this mechanism is determined by the precession of Mn spins polarized by external magnetic field in the sum of external and exchange field, with a typical time of about 10 ps which is one order of magnitude shorter than the polaron formation time at zero field. This mechanism allows that the polaron in magnetic fields applied in the quantum-well (QW) plane comes closer to the equilibrium situation during the exciton lifetime. In the following, we first present our model and then report the experimental data.

II. THEORY

In semimagnetic semiconductors, the exchange interaction of charge carriers and electrons from d shells of magnetic ions is described by the Heisenberg Hamiltonian

$$\hat{H}_{\text{ex}} = a \sum_i (\hat{\mathbf{J}}\hat{\mathbf{S}}_i) W_i = -\mu_B g_{\text{Mn}} \sum_i \frac{b_i}{J} (\hat{\mathbf{J}}\hat{\mathbf{S}}_i), \quad (1)$$

where a is a parameter of the exchange interaction, $\hat{\mathbf{J}}$ is the operator of a carrier spin (for holes, which give dominant contribution in the magnetic-polaron formation in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ crystals, $J = \frac{3}{2}$). g_{Mn} , $\hat{\mathbf{S}}_i$, and W_i are the g factor, the spin of i th magnetic ion, and the probability of finding the carrier at the location of this ion, respectively. $b_i = -aJW_i/\mu_B g_{\text{Mn}}$ is the value of the mean-exchange field at the i th ion location, produced by the carrier with the maximum average value of spin. μ_B is the Bohr magneton.

In addition, the magnetic-ion spins are affected by local fields created by adjacent magnetic ions, which are given by the Hamiltonian

$$H_{ii} = \sum_{i,j} C_{\alpha\beta} S_i^\alpha S_j^\beta. \quad (2)$$

At low temperatures, the interaction among ionic spins brings forth spin clustering, which results, for instance, in a decrease of the magnetic susceptibility.¹ The spin-spin interaction also provides the effective channel of the energy relaxation for the magnetic polarons to form.¹³

A magnetic polaron is contributed for by a large number of magnetic ions inside the orbit of the localized carrier. Therefore, it is convenient to consider the carrier spin which couples with the total spin of all these ions, that is practically classical.^{18–20} It is natural to define the total spin of magnetic ions as having its average given by the vector $\mathbf{I} = \sum_i \langle \mathbf{S}_i \rangle W_i$ that governs the exchange field affecting the carrier. In order to do this, one can use the adiabatic approximation. In this particular case, one can write down the spin part of the wave function of the whole system ‘‘carrier+magnetic ions’’ as a product of the carrier-spin wave function and the wave

function of the ion spins: $\Psi(\mathbf{J}, \mathbf{S}_1, \mathbf{S}_2, \dots) = \Phi_c(\mathbf{J}, \mathbf{I}) \Phi_{\text{Mn}}(\mathbf{S}_1, \mathbf{S}_2, \dots, \langle \mathbf{J} \rangle)$. Then, the exchange Hamiltonian expressed by Eq. (1) can be recast in the following equivalent form:

$$\begin{aligned} \hat{H}_{\text{ex}} = & a(\hat{\mathbf{I}}\hat{\mathbf{J}}) - \mu_B g_{\text{Mn}} \left[\sum_i b_i (\hat{\mathbf{S}}_i \langle \mathbf{J} \rangle) \right. \\ & \left. + \sum_i b_i [(\hat{\mathbf{S}}_i - \langle \mathbf{S}_i \rangle) \cdot (\hat{\mathbf{J}} - \langle \mathbf{J} \rangle)] - \sum_i b_i (\langle \mathbf{S}_i \rangle \langle \mathbf{J} \rangle) \right]. \end{aligned} \quad (3)$$

Here $\langle \mathbf{S}_i \rangle$ and $\langle \mathbf{J} \rangle$ are the mean values of the i th ion spin and of the carrier spin, respectively. They are calculated by averaging over the functions $\Phi_{\text{Mn}}(\mathbf{S}_1, \mathbf{S}_2, \dots, \langle \mathbf{J} \rangle)$ and $\Phi_c(\mathbf{J}, \mathbf{I})$. The first term of the Hamiltonian results in a rotation of the carrier spin in the mean-exchange-field of the magnetic ions. The second term causes a rotation of the i th ion spin in the mean field of the rapidly revolving carrier spin \mathbf{J} . The third term describes an interaction of fluctuations in the carrier-ion spin system, which lead to deviations from the adiabatic approximation. Specifically, it is responsible for the indirect exchange interaction among ions via flip-flop transitions involving the carrier spin. Finally, the last term just allows for the fact that the mean exchange energy in the previous terms of the Eq. (3) is counted twice: as the energy of the carrier spin in the ionic exchange field and as the energy of the ions in the exchange field of the carrier.

Within the adiabatic approximation, the dynamics of the spin system is traced by use of a self-consistent routine. Namely, the mean value of the carrier spin, $\langle \mathbf{J} \rangle$, is found as a function of the mean total spin of the magnetic ions $\langle \mathbf{I}(t) \rangle$. Then, $\langle \mathbf{J} \rangle$ is used to calculate exchange fields produced by the carrier at each ion, thus finding out differential equations for all $\langle \mathbf{S}_i \rangle$ as functions of time. Finally, an averaging over the ionic positions closes the loop, giving the rates of changing the components of $\langle \mathbf{I}(t) \rangle$.

The next step towards a more simplified, if not quite precise, description of the spin-system dynamics can be done by considering a precession of $\langle \mathbf{I}(t) \rangle$ as a whole in the mean exchange field

$$\mathbf{B}_{\text{ex}} = \frac{\langle \mathbf{J} \rangle}{J} \sum_i W_i b_i = \frac{\langle \mathbf{J} \rangle}{J} B_p$$

(which, in turn, depends on \mathbf{I}), instead of doing calculations for each $\langle \mathbf{S}_i \rangle$ with a consequent averaging. Here, $B_p = \sum_i W_i b_i$ is a parameter, which is equal to the mean exchange field that would affect the ions, if the carrier is completely polarized. The main shortcoming of this simplest approach is a coherent description of the dynamics of individual spins. Actually, spins precess independently with various frequencies, due to inhomogeneity of the exchange field. The effect of the field inhomogeneity can be accounted for by introducing a relaxation of the components of \mathbf{I} transversal to the mean field \mathbf{B}_{ex} , which should result from the dephasing of ionic spins. As the variance of the carrier field at different ions is on the order of the mean value of this field, the estimate of the corresponding relaxation time is close to the period of precession of \mathbf{I} .

In bulk crystals with the cubic symmetry, such as $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, the mean spin of the carrier just traces the direction of \mathbf{I} . Therefore, \mathbf{I} is collinear to the exchange field, and does not precess. If, however, the symmetry is reduced (for instance, in wurtzite-type crystals, in strained cubic crystals, or in low-dimensional structures), the directions of \mathbf{I} and $\langle \mathbf{J} \rangle$ may differ significantly. The difference is much more important for holes, than for electrons, due to the strong spin-orbit interaction in the valence band. In particular, in quantum wells, the direction of the heavy-hole spin is governed by confinement and strain effects.^{16,19–21} This property of the hole spin in QW structures plays the dominant role in the fast energy relaxation mechanism considered in this paper.²²

A. Instructive limiting case: Extreme anisotropy

In order to give a qualitative insight into the model suggested, we consider first the demonstrative extreme case of the magnetic polaron (MP) formed by a strongly confined heavy hole. Assuming the splitting between the light-hole and heavy-hole states (Δ_{lh-hh}) to be much more than the hole exchange energy, we put the in-plane components of the heavy-hole spin equal to zero: $J_x = J_y = 0$. The exchange interaction, given above by the Heisenberg Hamiltonian [see Eq. (1)], now reduces to the Ising form $-\mu_{BG_{Mn}}(B_p/J)(I_z J_z)$ (in this case $\mathbf{B}_{ex} = \pm B_p \mathbf{z}$, where \mathbf{z} is a unit vector along the normal to the QW plane). The exchange field \mathbf{B}_{ex} is “switched on” by the creation of a localized hole, which can be realized experimentally by a direct photoexcitation of localized excitons or by localization of excitons excited non-resonantly. It is obvious that the exchange field is “switched off” by an exciton recombination.

Let us consider an ensemble of magnetic ions aligned in an external field \mathbf{B} and having an average spin \mathbf{I} . After the creation of a localized hole with the orbit covering the spin ensemble, the average spin \mathbf{I} is exposed to the effective field, which is the sum of exchange and external fields: $\mathbf{B}_\Sigma = \mathbf{B} + \mathbf{B}_{ex}$. If \mathbf{B}_{ex} and \mathbf{B} are not parallel to each other, the spin \mathbf{I} starts to precess around \mathbf{B}_Σ (see inset in Fig. 1). In the following, we consider for simplicity the case $\mathbf{B} \perp \mathbf{B}_{ex}$, i.e., \mathbf{B} is applied parallel to the quantum-well plane. In this case, the field \mathbf{B}_Σ is directed at the angle φ to the normal to the QW plane, with φ defined by the relations $\cos \varphi = B_p / \sqrt{B_p^2 + B^2}$ and $\sin \varphi = B / \sqrt{B_p^2 + B^2}$. As a result of the precession, the projection of \mathbf{I} on \mathbf{B} decreases, and its projection on \mathbf{B}_{ex} increases. Respectively, the Zeeman energy of magnetic-ion spins $E_z = -\mu_{BG_{Mn}} \mathbf{B} \mathbf{I}$ increases, and the exchange energy $E_{ex} = -\mu_{BG_{Mn}} \mathbf{B}_{ex} \mathbf{I}$ decreases. The decrease of the exchange energy corresponds to the magnetic polaron formation with an energy $E_{MP} = -E_{ex}$. This polaron formation process is provided by the energy transfer from the exchange to the Zeeman reservoir, with the total energy of the system “hole+magnetic ions” conserved.

Keeping in mind that the MP energy is determined by the projection of the average spin \mathbf{I} on the direction of the exchange field, we distinguish three stages of the magnetic polaron formation at external magnetic fields applied perpendicular to the hole exchange field.

(i) After the exchange field \mathbf{B}_{ex} is switched on by photo-generation of an exciton at the time moment $t=0$, the

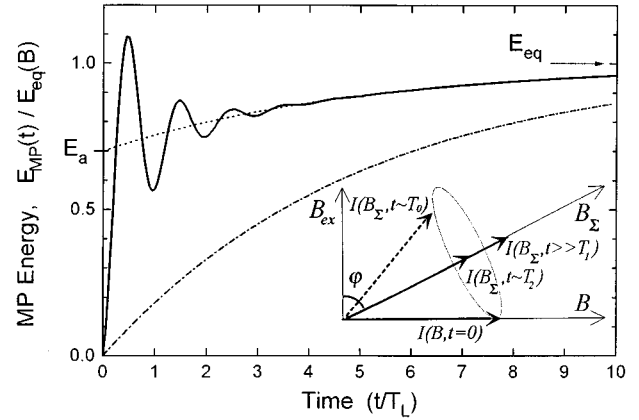


FIG. 1. Behavior of the magnetic polaron energy at external magnetic field $B = 1.4B_p$ (solid line). Parameters used for calculation $B_p = 1$ T, $T_2 = 0.75T_L$, and $T_1 = 5T_L$. Dashed-dotted line shows the polaron formation with T_1 , i.e., without account of the mechanism. The inset displays the precession motion of \mathbf{I} in $\mathbf{B}_\Sigma = \mathbf{B} + \mathbf{B}_{ex}$.

magnetic-ion spins begin to precess with Larmor frequency $\omega_L = \mu_{BG_{Mn}} B_\Sigma / \hbar$ around the direction of the effective field $\mathbf{B}_\Sigma = \mathbf{B} + \mathbf{B}_{ex}$ (see inset of Fig. 1). All spins start their precession simultaneously, and at the very beginning, they move coherently with respect to each other. The precession of the total magnetic moment around \mathbf{B}_{ex} leads to the “coherent” MP state with oscillating energy and with the periodical transmission of energy between the exchange and the Zeeman reservoirs. This initial “coherent” stage of the magnetic polaron formation has been observed recently by means of resonant Raman spectroscopy in $\text{Cd}_{0.98}\text{Mn}_{0.02}\text{Te}/\text{Cd}_{0.76}\text{Mg}_{0.24}\text{Te}$ quantum-well structures.¹⁶

(ii) The component of the spin \mathbf{I} transversal to the field \mathbf{B}_Σ relaxes within the time T_2 . The transversal relaxation time T_2 corresponds to the dephasing time of the coherent precession of all magnetic-ion spins inside the hole orbit. After T_2 the MP reaches its stationary state with the average ion spin $\mathbf{I}(B_\Sigma, t \approx T_2)$ directed along the effective field \mathbf{B}_Σ .

(iii) Further relaxation to the equilibrium MP state by an increase of the \mathbf{I} value up to $I(B_\Sigma, t \gg T_1)$ is inevitably connected with energy transfer to the spin-spin reservoir or to the lattice. This process is determined by the time of longitudinal relaxation of the magnetic-ion spins T_1 .

Let us now estimate typical times related to the different stages of the MP formation. A considerable part of the MP energy is gained within the characteristic time $T_0 = T_L/4 = \pi/(2\omega_L)$. This time can be estimated from the value of exchange field B_{ex} , as the effective field B_Σ is not less than B_{ex} . Measured energies of MP’s allow us to obtain the value of about 1 T as an estimation for B_{ex} .¹⁴ Therefore, we estimate $T_0 \sim 10$ ps. This time corresponds to the typical time of the mechanism of MP formation suggested in this paper. In the strongly inhomogeneous exchange field of the hole, a spread of precession frequencies is as large as a mean value of the precession frequency. Therefore, the coherence among the magnetic-ion spins is destroyed within one period of Larmor precession, and energy oscillations are damped out within typical times of the order of the transversal relaxation

time T_2 , which is of the same order as T_0 . The longitudinal relaxation time T_1 should be of the order of the MP formation time at zero field, i.e., 100–200 ps.^{15,23}

On that account, one can introduce two characteristic MP energies gained with characteristic times T_0 and T_1 , respectively: (i) E_a , gained adiabatically by the energy transfer to the Zeeman reservoir only; and (ii) the equilibrium energy E_{eq} determined by the energy transfer to the Zeeman and spin-spin reservoirs and to the lattice. We note that the Zeeman reservoir has a finite capacity at $B \neq 0$. The behavior of the MP energy with time at an external magnetic field \mathbf{B} is shown in Fig. 1 by a solid line. It demonstrates all typical features of the polaron formation scenario suggested above. At $B = 1.4B_p$ chosen for this example, 70% of the MP energy is gained by the fast adiabatic process, i.e., $E_a = 0.7E_{eq}$. A dashed-dotted line given for comparison shows the polaron formation with time T_1 , when the mechanism is not considered and the polaron formation is determined by the energy transfer to the spin-spin reservoir only.

Now we are going to link E_{eq} and E_a with parameters that can be measured experimentally. Both E_{eq} and E_a are exchange parts of the total energy of the system ‘‘hole+magnetic ions,’’ i.e., scalar products of the hole exchange field \mathbf{B}_{ex} and the magnetic moment of all ions inside the hole orbit \mathbf{I} . In the equilibrium case, \mathbf{I} is determined by the effective field \mathbf{B}_Σ ,¹⁹ so that

$$E_{eq}(B) = \mu_B g_{Mn} [\mathbf{B}_{ex} \cdot \mathbf{I}(\mathbf{B} + \mathbf{B}_{ex})] = \mu_B g_{Mn} \sin\varphi I(B_\Sigma) B_{ex}. \quad (4)$$

Taking into account that $\varepsilon_F(B) = 2\mu_B g_{Mn} I(B) B_{ex}$ we get

$$E_{eq}(B) = \sin\varphi \frac{1}{2} \varepsilon_F(B_\Sigma), \quad (5)$$

where $\varepsilon_F(B)$ is the Zeeman splitting of the heavy-hole sublevels in the magnetic field applied in the Faraday geometry ($\mathbf{B} \parallel \mathbf{z}$). Note that $\varepsilon_F(B)$ can be measured experimentally.

In order to calculate the adiabatically gained energy E_a , one must use the value of the ionic spin equal to the projection of $\mathbf{I}(B)$ on \mathbf{B}_Σ . Thus we obtain

$$E_a(B) = \mu_B g_{Mn} \left(\mathbf{B}_{ex} \frac{\mathbf{B}_\Sigma}{B_\Sigma} \right) \cdot \left(\frac{\mathbf{B}_\Sigma}{B_\Sigma} \mathbf{I}(B) \right) = \mu_B g_{Mn} \cos\varphi B_p I(B). \quad (6)$$

Keeping in mind that in our case ($\mathbf{B}_{ex} \parallel \mathbf{z}$), $B_{ex} \equiv B_p$ and using the connection between $\varepsilon_F(B)$ and $I(B)$, we will get the following expression for E_a :

$$E_a(B) = \frac{B_p B}{B_p^2 + B^2} \frac{1}{2} \varepsilon_F(B). \quad (7)$$

Figure 2 depicts the dependences of the MP equilibrium energy E_{eq} (curve 1) and its fast adiabatic component E_a (curve 2) on a magnetic field calculated according to Eqs. (5) and (7). The experimental dependence $\varepsilon_F(B)$ for $\text{Cd}_{0.93}\text{Mn}_{0.07}\text{Te}$ at a temperature of 1.6 K and $B_p = 1$ T have been used for the calculation. It is clearly seen that E_a approximates E_{eq} at $B \geq 2B_p$, which in turn should result in the decrease of MP formation time from T_1 down to about T_0 with the magnetic field increase. Note that Fig. 1 shows the time evolution of MP formation corresponding to $B = 1.4B_p$ at Fig. 2.

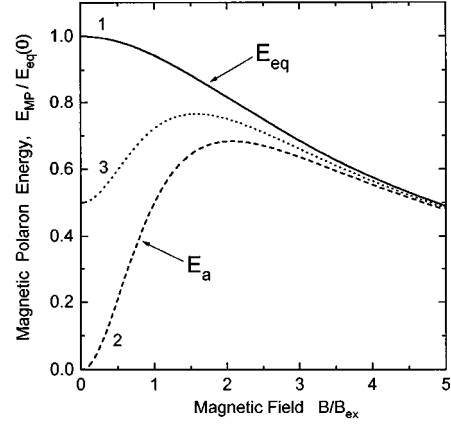


FIG. 2. Magnetic polaron energies as function of external magnetic field: (1) equilibrium MP energy; (2) a part of MP energy gained by the energy transfer to the Zeeman reservoir; (3) modeling of an experimental situation with $\tau_f \approx \tau_0$.

B. General case: Finite splitting of light-hole and heavy-hole states

For the magnetic polaron formed by the heavy-hole, the model considered in the Sec. II A is the zero approximation in the parameter $\eta(B) = \varepsilon_F(B)/\Delta_{lh-hh}$. This parameter determines the mixing between the light-hole and heavy-hole states caused by a deviation of the spin of magnetic ions \mathbf{I} from the z axis.

In the Voigt geometry ($\mathbf{B} \perp \mathbf{z}$), in the lowest orders of the perturbation theory, the shift of the center of gravity of the spin doublet of the heavy-holes, which is analogous to the field-induced shift of the ground state of Van Vleck paramagnets, is proportional to $\varepsilon_F(B)\eta(B) \propto B^2$. Spin splitting of the heavy-hole doublet is proportional to $\varepsilon_F(B)\eta^2(B) \propto B^3$.¹⁹ The effect of the fast energy transfer considered in this paper shows up most distinctly at $B \approx B_p$ and the expected changes of the magnetic polaron energy are about $\varepsilon_F(B)$. Therefore, the value of $\eta(B_p)$ is an important parameter of the problem. If $\eta(B_p) \ll 1$, the admixture of the light-hole wave functions to the heavy-hole ones is negligibly small. In this case, the adiabatic part of MP energy E_a can be calculated by use of Eq. (7).

However, in the situation seen in the experiments reported below, $\eta(B_p) \leq 1$. In this case, the evolution of the spin system in the process of MP formation is no longer limited by the precession of \mathbf{I} at a constant field, as the hole exchange field $\mathbf{B}_{ex}(\mathbf{I})$ is no longer parallel to the z axis. The direction and value of $\mathbf{B}_{ex}(\mathbf{I})$ are determined by the mean hole spin $\langle \mathbf{J} \rangle$, that is found now by averaging the hole-spin operator over the eigenfunction of the spin Hamiltonian:

$$\hat{H} = -\frac{2}{3} \mu_B g_{Mn} B_p (\hat{\mathbf{I}} \cdot \hat{\mathbf{J}}) - \frac{\Delta_{lh-hh}}{2} \hat{J}_z^2, \quad (8)$$

corresponding to the minimum energy E_0 . Here \hat{J}_α are spin component operators of the hole with $J = \frac{3}{2}$ ($\alpha = x, y, z$). Therefore, E_0 corresponds to the least root of an algebraic four-power equation.

The adiabatic part of the MP energy equals the difference of E_0 taken at initial and final states with the spin of magnetic ions $\mathbf{I}_i(B)$ and $\mathbf{I}_f(B)$, respectively:

$$E_a(B) = E_0[\mathbf{I}_i(B)] - E_0[\mathbf{I}_f(B)]; \quad (9)$$

here

$$\mathbf{I}_i(B) = \frac{1}{2} \frac{\varepsilon_F \mathbf{B}}{B_p B}.$$

For a numerical calculation of $\mathbf{I}_f(B)$ we use the same assumptions as in Sec. II A.

(i) After the fast relaxation, $\mathbf{I}_f(B)$ is oriented parallel to the direction of the field $\mathbf{B} + \mathbf{B}_{\text{ex}}(\mathbf{I}_f)$ acting on it.

(ii) Sums of the exchange and magnetic energies of the system “hole+magnetic ions” at the initial and the final states are equal:

$$-\mu_{B\text{S}} g_{\text{Mn}}(\mathbf{I}_i \mathbf{B}) + E_0(\mathbf{I}_i) = -\mu_{B\text{S}} g_{\text{Mn}}(\mathbf{I}_f \mathbf{B}) + E_0(\mathbf{I}_f). \quad (10)$$

Therefore, the endpoint of vector \mathbf{I}_f belongs to the surface given by the condition of energy conservation. The value of \mathbf{I}_f should be minimal.²⁴ Under these conditions \mathbf{I}_f is determined numerically from Eq. (10) and afterwards $E_a(B)$ can be calculated from Eq. (9).

III. EXPERIMENT AND DISCUSSION

An idea of the experimental identification of the suggested mechanism could be discussed on the base of Fig. 2, where the equilibrium and adiabatical parts of the polaron energy are shown by solid and dashed lines, respectively.

A peculiarity of the exciton magnetic polaron is that the time for its formation is limited by the exciton lifetime, which is about 100–200 ps in II-VI semiconductor QW's at low temperatures.^{10,15,25} As a result, the MP shift of the luminescence line taken under cw excitation ($E_{\text{MP}}^{\text{cw}}$) is smaller than the equilibrium polaron energy if the polaron formation time (τ_f) is equal or longer than the exciton lifetime (τ_0).^{15,23} Curves shown in Fig. 2 also reflect the dependence of the MP shift on B for structures with different values of ratio τ_f/τ_0 : (1) $\tau_f \ll \tau_0$, in this case $E_{\text{MP}}^{\text{cw}}(B)$ is just equal to the equilibrium polaron energy $E_{\text{eq}}(B)$ (shown by a solid line); (2) $\tau_f \gg \tau_0$, $E_{\text{MP}}^{\text{cw}}(B=0) \approx 0$ and with rising field $E_{\text{MP}}^{\text{cw}}(B) = E_a(B)$, i.e., $E_{\text{MP}}^{\text{cw}}(B)$ just reflects the part of the MP energy formed by the fast adiabatic process (see a dashed line); (3) $\tau_f \approx \tau_0$, this is an intermediate case between (1) and (2). The MP shift at $B=0$ corresponds to a part of E_{eq} , but at magnetic fields the fast adiabatic process shifts it rapidly towards E_{eq} (see the dotted line). It is remarkable, that in cases (2) and (3) the MP shift increases in external fields. It cannot be explained in any model that considers equilibrium MP states only, as the magnetic susceptibility decreases in magnetic fields. Therefore, the experimental observations of such an increase can document the reduction of the MP formation time from T_1 down to T_0 by the in-plane magnetic field applied.

Experiments have been performed on a $\text{Cd}_{0.93}\text{Mn}_{0.07}\text{Te}/\text{Cd}_{0.64}\text{Mn}_{0.07}\text{Mg}_{0.29}\text{Te}$ heterostructure with 80-, 45-, and 18-Å-thick single QW's. The structure was grown by molecular-beam epitaxy on (100)-oriented

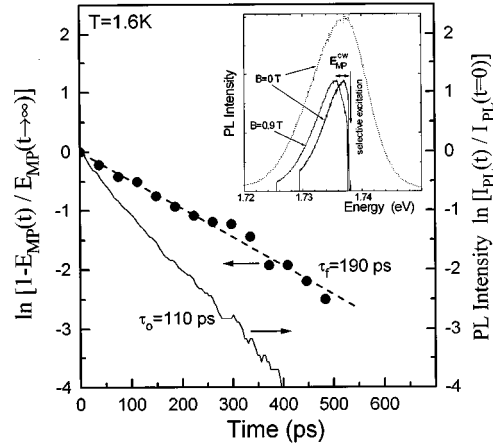


FIG. 3. Magnetic polaron formation (circles) and the photoluminescence decay (solid line) in an 80-Å-thick $\text{Cd}_{0.93}\text{Mn}_{0.07}\text{Te}/\text{Cd}_{0.64}\text{Mn}_{0.07}\text{Mg}_{0.29}\text{Te}$ QW. The inset displays photoluminescence line taken under selective (solid lines) and nonselective (dotted line) cw excitation with and without magnetic field applied perpendicular to the z axis.

$\text{Cd}_{0.96}\text{Zn}_{0.04}\text{Te}$ substrate.²⁶ The MP energies at external magnetic fields and $T=1.6$ K were determined by selective cw excitation of localized excitons.^{9,10} Under these excitation conditions the nonmagnetic spectral diffusion is suppressed and the polaron energy is measured from the shift of the luminescence line with respect to the excitation energy. The photoluminescence (PL) spectra of the 80-Å-thick QW taken under nonselective and selective excitation are shown in the inset of Fig. 3.

At zero magnetic field the dynamics of the MP formation was measured from the evolution of the polaron line shift after 5-ps excitation pulses.^{15,23} A streak camera with time resolution of 15 ps was used in this experiment. The polaron shift plotted in logarithmic scale is shown in Fig. 3 by circles together with the decay of the spectrally integrated PL in the 80-Å-thick QW. For this QW $\tau_f=190$ ps and $\tau_0=110$ ps, which results in a strong dynamical effect on the MP energy. The equilibrium MP energy, determined as a saturation value at longer delays $E_{\text{eq}}(B=0) = E_{\text{MP}}(t \rightarrow \infty, B=0) = 8$ meV, exceeds strongly the polaron shift measured under cw excitation $E_{\text{MP}}^{\text{cw}}=1.2$ meV. We note here that this experimental situation is very close to the case (2) shown in Fig. 2.

As expected, in the Faraday geometry ($\mathbf{B} \parallel \mathbf{z}$) the MP shift decreases effectively and is not any more measurable at fields above 0.2 T (shown by triangles in Fig. 4).¹⁰ At magnetic fields applied in the Voigt geometry ($\mathbf{B} \perp \mathbf{z}$) we found a very pronounced increase of the MP shift up to 2.8 meV at $B=0.7$ T (see Fig. 4 and inset of Fig. 3). We stress here that the increase of the MP shift in magnetic fields applied is quite an unusual appearance. It is definitely caused by the dynamical effect, namely, by a decrease of τ_f/τ_0 ratio. As τ_0 is not modified by magnetic fields we associate this effect with the reduction of τ_f with the magnetic-field increase. A possible mechanism for reduction of τ_f is suggested in the Sec. II of this paper. In the following we estimate its contribution for the 80-Å-thick QW.

In the 80-Å-thick QW a nonvanishing Zeeman splitting of exciton states at the Voigt geometry evidences that the com-

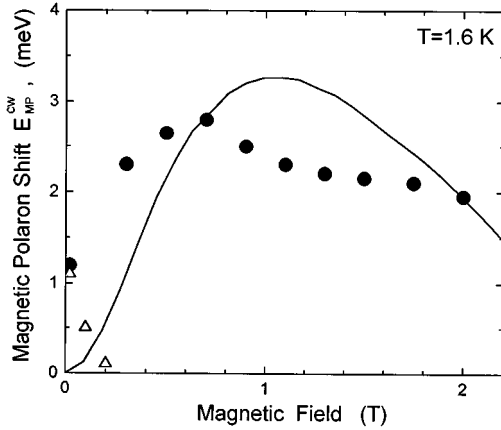


FIG. 4. Magnetic polaron shift taken under cw excitation in an 80-Å-thick Cd_{0.93}Mn_{0.07}Te/Cd_{0.64}Mn_{0.07}Mg_{0.29}Te QW as a function of magnetic field applied parallel (triangles) and perpendicular (circles) to the z axis. Solid line shows the results of the model calculation with $B_p = 0.7$ T.

ponents of the heavy-hole spin (J_x, J_y) are not equal to zero. In this QW, the experimentally measured dependence $\varepsilon_F(B)$ determined from the PLE spectra is described well with the phenomenologically modified Brillouin function with parameters for effective spin $S_0 = 1.3$ and effective temperature $T_0 = 4.7$ K.²⁷ The value of the exchange field in the magnetic polaron was found out from the experimentally determined parameters, according to the approach suggested in Ref. 14:

$$B_p = \frac{2E_{eq}}{[d\varepsilon_F(B)/dB]_{B=0}}. \quad (11)$$

For the studied QW B_p amounts to 0.7 T.

In the 80-Å-thick QW the value $\varepsilon_F(B_p = 0.7$ T) = 14 meV, which is not negligibly small in comparison with $\Delta_{lh-hh} = 24$ meV. That means that condition $\eta(B_p) \ll 1$ is not satisfied and the dependence $E_a(B)$ should be found numerically following the procedure described in the Sec. II B. The values of the magnetic moment at the initial state were found from the experimentally determined parameters: $I_i(B) = \varepsilon_F(B)/2\mu_{B\text{Mn}}B_p$. The result of the calculation is presented by the solid line in Fig. 4. It reproduces all trends of the experimental behavior shown by closed circles. The magnetic polaron shift increases in low fields, achieves its maximum at about 1 T and then falls down at higher fields. The increase in fields below 1 T is caused by the reduction of the magnetic polaron formation time, which allows for MP's to

gain a larger part of equilibrium MP energy during the polaron lifetime τ_0 . It is in line with the scenario suggested in Sec. II for $\tau_f \gg \tau_0$ and illustrated by the curve 2 in Fig. 2. The decrease of the polaron shift in magnetic fields above 1 T is related to the suppression of the equilibrium MP energy.

We note here that all parameters for the model calculation presented in Fig. 4 are determined experimentally. To avoid introduction of additional parameters the calculation was run for the condition $\tau_f \gg \tau_0$, i.e., the adiabatical part of the MP energy was computed, which dominates the polaron energy in the 80-Å-thick QW in magnetic fields above 0.3 T. We believe that the good agreement between the experimental results and the calculated behavior of the MP energy shown in Fig. 4 proves the adequate description of the observed phenomenon.

A similar behavior was found for 45- and 18-Å-thick QW's, where the dynamical effect on the MP energy is considerable ($\tau_f > \tau_0$). We stress here that all structures without dynamical effect on the MP energy (i.e., with $\tau_f \ll \tau_0$ at $B = 0$) show the monotonic decrease of the polaron energy in magnetic fields,⁹ which is in good agreement with the scenario proposed by curve 1 in Fig. 2.

In conclusion, we have suggested a mechanism of magnetic polaron formation in QW structures, which is controlled by the strength of external magnetic field. This mechanism is determined by the fast energy transfer (within about 10 ps) from the carrier-magnetic-ions exchange reservoir to the Zeeman reservoir. The underlying physics relates to reduced symmetry and is close to that responsible for other spin-dynamic effects observed in semimagnetic low-dimensional structures.^{16,21} An unusual increase of the magnetic polaron shift at magnetic fields applied parallel to the quantum-well plane is observed experimentally in the quantum-well structures with the strong dynamical effect. This behavior is modeled well by an accounting of the mechanism. The strong reduction of the polaron formation time provided by this mechanism can affect the hierarchy of relaxation processes by moving the polaron formation time from the range of polaron lifetime down to the range of dephasing and spin-relaxation times of carriers. This effect allows to scan in time by magnetic fields the spin organization process and can be used for the study of basic properties of strongly correlated spin systems.

ACKNOWLEDGMENTS

This work was supported by the Deutsche Forschungsgemeinschaft through SFB 410 (Würzburg) and through SFB 385 (Marburg) and by the Volkswagen Foundation.

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