Exciton spin relaxation in diluted magnetic semiconductor $Zn_{1-x}Mn_xSe/CdSe$ superlattices: Effect of spin splitting and role of longitudinal optical phonons

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(Received 9 October 2002; published 26 March 2003)

Exciton spin relaxation in diluted magnetic semiconductor (DMS) structures based on ZnMnSe is closely examined as a function of exciton spin splitting in an external magnetic field. A drastic increase in spin relaxation is observed when exciton spin splitting exceeds the longitudinal optical (LO) phonon energy. Direct experimental evidence has been provided from (1) spin injection from the DMS to an adjacent nonmagnetic quantum well that can be modulated by the LO-assisted spin relaxation and (2) hot exciton photoluminescence within the DMS where a spin flip is accompanied by the emission of one LO phonon.

DOI: 10.1103/PhysRevB.67.125313 PACS number(s): 78.20.Ls, 78.55.Et, 72.25.−b, 75.50.Pp

I. INTRODUCTION

Spin relaxation is one of the fundamental physical processes in solids which governs spin orientation (or polarization) of carriers and electronic excitations such as excitons. It has gained increasing interest in recent years in light of its predominant role in determining magnetic properties of spintronic materials and in potential applications of future spintronic devices. While spin relaxation is generally known to be caused by physical processes admixing spin states, exact mechanisms are rather complicated and sensitive to many electronic and structural parameters such as band structure, doping, strain, dimensionality, etc.[1–7] Three spin relaxation mechanisms have been found to be relevant for electrons and holes, namely, the Elliott-Yafet,[8,9] D'yakonov-Perel',[10] and Bir-Aronov-Pikus (BAP)[11] mechanisms. The former two mechanisms are both of spin-orbit interaction origin induced by host ions (or impurities) and a lack of inversion symmetry, respectively. The third mechanism is due to the exchange interaction between electrons and holes. Spin relaxation of excitons occurs when either electron or hole spins, or both (for a BAP mechanism), relax according to the above three mechanisms. In addition to these relaxation channels the long-range exciton exchange interaction can be important and can even become dominant.[12]

In diluted magnetic semiconductors (DMS's), additional spin scattering is introduced as a result of strong interactions between magnetic ions and carriers or excitons.[13–16] Due to the strong sp-d exchange interaction between free carriers and magnetic ions, DMS's exhibit interesting properties that have made them a promising material system as a spin aligner (SA) (or a spin polarizer)—an essential element of a spintronic device through which a desired spin orientation can be obtained. The recent successful demonstration of spin injection from such a SA (Refs. 17, 18) across a semiconductor heterointerface has raised hope for future all-semiconductor spintronic devices. DMS's and related spintronic structures have also provided a unique playground to investigate spin relaxation in a system where separation of spin levels can exceed the energy of longitudinal optical (LO) phonons. This regime has never been accessible in a nonmagnetic semiconductor as an unrealistically high magnetic field is required to reach a similar situation.

The aim of this work is to investigate exciton spin relaxation in ZnMnSe based DMS's and its possible physical mechanism, over the range where the energy of exciton spin splitting crosses the LO phonon energy. Taking advantage of the new opportunity provided by spin injection structures, spin relaxation in DMS is closely examined by photogenerated spin injection from the DMS to an adjacent quantum well (a spin detector) and also by hot exciton photoluminescence (PL) in the DMS.

II. EXPERIMENT

The layered structures employed to study effects of spin splitting on exciton spin relaxation contain three important regions (see Fig. 1), i.e., a DMS SA, where spin relaxation is under investigation, a spin detector (SD) by a 70 Å thick ZnCdSe quantum well (QW), and an undoped ZnSe spacer (4–5 nm) inserted between the SA and SD. The results presented in this paper were obtained from the DMS SA made of a ten-period $Zn_{0.96}Mn_{0.04}Se$ (4 nm)/CdSe (0.8 monolayer) superlattice (SL). Similar effects on exciton spin relaxation were also observed in a thin $Zn_{0.95}Mn_{0.05}Se$ DMS layer (15 nm thick). All structures were prepared by molecular beam epitaxy (MBE) on GaAs substrates. The magnetooptical experiments were performed in the Faraday configuration at 2

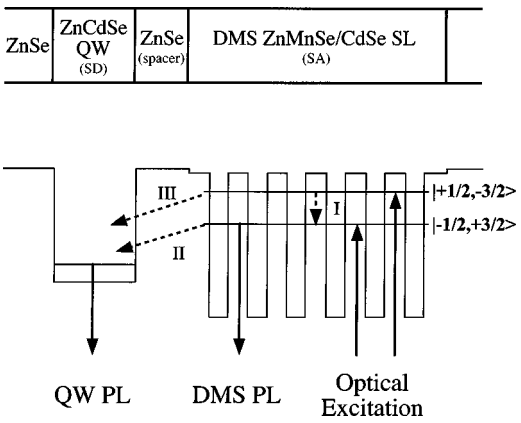

FIG. 1. (a) Schematic diagrams of the sample structure and the physical processes investigated. Process I denotes spin relaxation within the hh exciton states of the DMS, and processes II and III spin injection to the QW from the two spin sublevels of the DMS hh exciton.

K, in an applied magnetic field up to 5 T. PL was excited by a dye laser, and was spectrally dispersed by a grating monochromator and detected by a GaAs photomultiplier. PL polarization is defined as the difference between the intensity of the two circularly polarized light components and is given below in percentage by $100(\sigma^+-\sigma^-)/(\sigma^++\sigma^-)$, where σ^+ and σ^- are the PL intensities measured at left and right circular polarization, respectively.

III. RESULTS

Two types of magnetooptical experimental approaches, namely, spin injection and hot photoluminescence (PL), were employed to investigate the exciton spin relaxation process. In both approaches, a spin orientation of the heavy hole (hh) exciton in the DMS is prepared by optical excitation of the appropriate spin state (Fig. 1).

A. Optical spin injection experiment

The first type of experiments is based on optical spin injection from the DMS hh exciton to the QW. Figure 2 shows characteristic spectra obtained from the optical spin injection experiments, where the intensity and polarization of the QW PL is measured as a function of excitation photon energy within the range of the $|-\frac{1}{2},+\frac{3}{2}\rangle$ and $|+\frac{1}{2},-\frac{3}{2}\rangle$ spin states of the DMS hh exciton. The excitation light is linearly polarized in the SL plane, with an equal probability of exciting the σ^+-active $|-\frac{1}{2},+\frac{3}{2}\rangle$ spin state and the σ^--active $|+\frac{1}{2},-\frac{3}{2}\rangle$ spin state. It is therefore referred as unpolarized below and in the figure captions. In the absence of external magnetic fields, the QW PL is unpolarized as expected when the spin sublevels that lead to different polarizations are degenerate for both DMS and QW. In an applied magnetic field, a weak, negative QW PL polarization is observed under the optical excitation below the energy of the lowest $|-\frac{1}{2},+\frac{3}{2}\rangle$ spin state of the DMS (thus without spin injection from the DMS). This is due to the intrinsic properties of the QW, i.e., the σ^--active $|+\frac{1}{2},-\frac{3}{2}\rangle$ spin state of the QW hh exciton lies lowest in energy and becomes favorably populated at 2 K in magnetic fields. When the excitation photon energy is resonant with the σ^+-active $|-\frac{1}{2},+\frac{3}{2}\rangle$ spin state of the DMS exciton, the QW PL polarization undergoes a sharp change in sign due to the population inversion between the two spin sublevels of the QW hh exciton induced by spin injection from the DMS. When the excitation photon energy is tuned to the σ^--active $|+\frac{1}{2},-\frac{3}{2}\rangle$ spin state of the DMS exciton, on the other hand, the spin injection induced change in the QW PL polarization becomes more complicated. This is because it is determined not only by the spin injection from this spin level directly to the QW (process III in Fig. 1) but also by a competing process of spin relaxation to the lower $|-\frac{1}{2},+\frac{3}{2}\rangle$ spin state of the DMS exciton (process I—shown by the dash arrow in Fig. 1) before being injected to the QW through process II. The measured QW PL polarization is thus governed by the relative efficiency of the two competing processes, which can be affected by the spin splitting of the DMS hh exciton. Therefore the QW PL polarization under

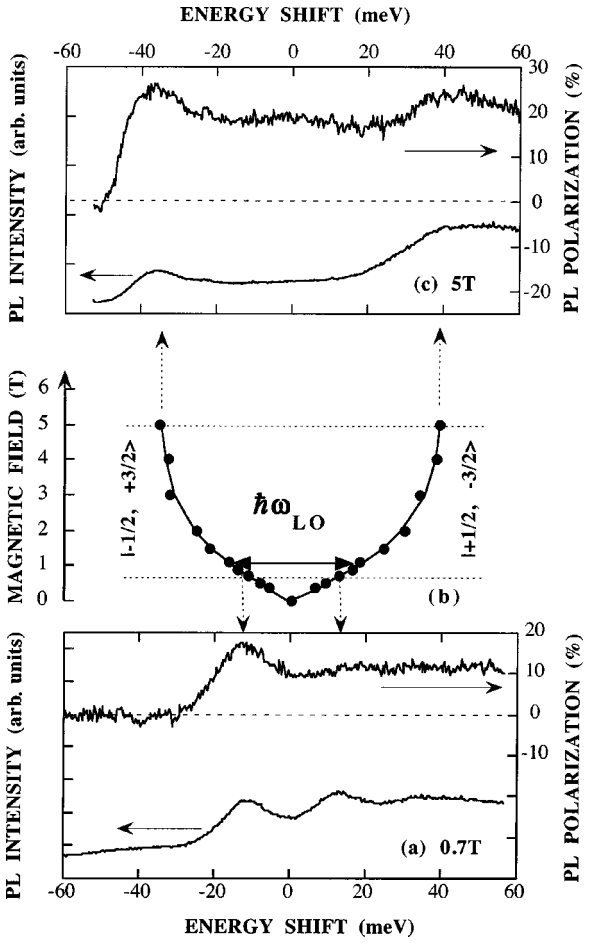

FIG. 2. (a),(c) Intensity and polarization of the QW PL as a function of photon energy of unpolarized excitation light scanning through the DMS hh exciton energy, at a magnetic field of 0.7 T (a) and 5 T (c). The energy shift is measured with respect to the DMS hh exciton energy at 0 T, i.e., 2.736 eV. (b) Measured Zeeman splitting of the DMS hh exciton. The field positions at which the spectra in (a) and (c) were obtained are indicated by the dashed lines.

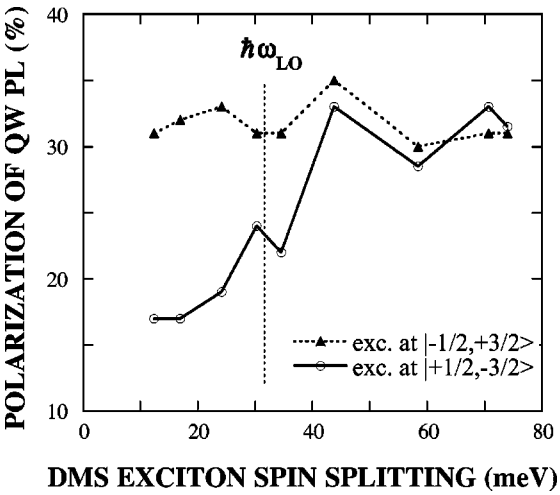

FIG. 3. The corrected polarization of the QW PL is shown under the resonant, unpolarized optical excitation at the upper-lying σ^--active $|+\frac{1}{2},-\frac{3}{2}\rangle$ state (open circles) and at the lower-lying σ^+-active $|-\frac{1}{2},+\frac{3}{2}\rangle$ state (filled triangles) of the DMS hh exciton. The corrected polarization of the QW PL is obtained after subtracting the intrinsic PL polarization of the QW. The saturation value (about 32%) of the QW PL polarization with a large spin splitting, at high magnetic fields, corresponds to complete spin relaxation of the DMS hh exciton. The solid and dotted curves are just guides for the eye.

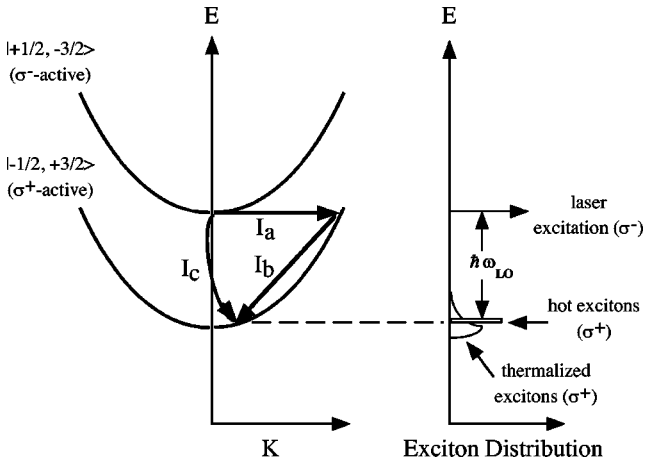

FIG. 4. Schematic diagram of the two optically active hh exciton bands of the DMS in *k* space. Process Ia denotes a nearly inelastic spin scattering, Ib an LO-assisted, spin-preserving exciton kinetic energy relaxation, and Ic depicts a single quantum process of LO-assisted elastic spin relaxation. The exciton distribution in energy under optical pumping in the hot PL experiment is also illustrated.

the resonant excitation at the upper $|+\frac{1}{2},-\frac{3}{2}\rangle$ spin state of the DMS exciton can be taken as a measure of spin relaxation within the two spin states of the DMS hh exciton. This can be demonstrated by the experimental observation shown in Fig. 2. When spin relaxation is much faster than spin injection, i.e., process I dominates over process III, a complete spin polarization corresponding to the lower spin state of the DMS hh exciton is expected such that spin injection is dominated by the process II. This has led to the experimental fact [Fig. 2(c)] that the polarization of the QW PL is predominantly σ^+ at a high magnetic field (thus a large spin splitting of the DMS exciton) when optical excitation is tuned at the upper σ^--active spin level of the DMS exciton, which is identical to that when optical excitation is tuned at the lower σ^+-active spin level. At a low magnetic field (thus a small spin splitting of the DMS exciton), however, a complete spin polarization is not realized. This is evident from the apparently weaker polarization when optical excitation is tuned at the upper spin level of the DMS as compared to that when optical excitation is tuned at the lower spin level [see Fig. 2(a)]. The observation can be interpreted by a reduction of spin relaxation such that spin injection from the upper spin level (process III) can also occur, which competes with the spin relaxation process I and compensates the spin polarization provided by the process II.

In order to closely examine the observed strong dependence of spin relaxation on spin splitting of the DMS exciton, a detailed study of the spin relaxation by monitoring the QW PL polarization is undertaken as a function of the spin splitting of the DMS exciton. The results are summarized in Fig. 3. The values are corrected by subtracting other contributions unrelated to the spin injection. As can be clearly seen, spin relaxation is a sensitive function of the spin splitting and exhibits a sharp increase when the energy separation of the two spin states crosses the LO phonon energy ($\hbar\omega_{\mathrm{LO}} \approx 32$ meV), depicted by the open circles in Fig. 3. Under the condition when the spin splitting reaches maximum, the DMS hh exciton spin is fully relaxed to the lowest spin state before being injected to the QW so that the same QW PL polarization is obtained under the resonant excitation of both spin levels of the DMS hh exciton [Figs. 2(c) and 3]. The resonant optical excitation at the lower spin level of the DMS exciton (filled triangles in Fig. 3), on the other hand, gives rise to a nearly constant polarization of the QW PL independent of the spin splitting. This is not surprising as the spins are readily at the lower spin level within the range of spin splitting at the low temperature (2 K) and spin relaxation plays a negligible role in this case. The limited degree (about 32%) of the QW PL polarization induced by spin injection from the DMS that has reached a complete spin relaxation is likely due to spin scattering during spin injection across the heterointerfaces and subsequent energy relaxation within the QW.[19]

B. Hot photoluminescence experiment

In the second experimental approach, a hot PL experiment designed to examine the possible LO-assisted exciton spin relaxation is employed (see Fig. 4 for an illustration of the principle). In this experiment, the photon energy of the excitation light with the matching σ^- polarization is tuned exactly at the energy of the upper spin state (at $K=0$) of the DMS hh exciton to ensure that only the spin orientation corresponding to that state is selectively generated. When the spin splitting is larger than the LO phonon energy the resonantly excited excitons of the upper spin state can first undergo a LO-assisted spin flip if it is the most efficient one among all available energy relaxation processes, generating a

large number of hot excitons in the lower spin band exactly at one LO energy below the excitation photon energy (Fig. 4). The excitons can then further relax down within the lower spin band branch, through spin-preserving acoustic-phonon assisted energy relaxation, which leads to a thermalized distribution of the excitons at the bottom of the spin band. In principle two PL features related to the lower spin band should be seen, i.e., a broad PL band corresponding to the thermalized excitons and a sharp hot PL line arising from the nonequilibrium distribution of the hot excitons exactly at one LO phonon energy below the excitation energy (Fig. 4). To observe the pronounced hot PL line, it is required that the efficiency of the LO-assisted spin relaxation exceeds that of other exciton decay processes and the latter should be faster than the spin-preserving acoustic-phonon assisted relaxation. This requirement is similar to that for a normal hot PL experiment concerning kinetic energy relaxation within a single exciton band,[20] except that in our case the LO-assisted energy relaxation occurs between two excitonic bands involving an exciton spin flip. In our samples, the observation of the sharp hot PL is further facilitated by the spin injection process as an additional nonradiative decay channel of the DMS exciton, such that the normal broad PL band from the DMS exciton can be largely suppressed with respect to the hot PL line.

A typical hot PL spectrum, obtained under the resonant σ^- optical pumping at the upper spin level of the DMS exciton, is shown in Fig. 5(c) when the spin splitting of the two spin levels exceeds the LO phonon energy. In addition to the broad PL band arising from the thermalized excitons of the lower spin state, a sharp hot PL line[21] can be clearly seen exactly at one LO phonon energy below the pumping energy. The polarization of the hot PL line, obtained after subtracting the contribution from the broad PL band, has the same sign as the lower spin state, which is opposite to that of the pumping light. This change of sign clearly shows that energy relaxation from the optically pumped upper spin state to the lower spin state is accompanied by an exciton spin flip via the emission of one LO phonon, providing direct evidence for the involvement of the LO phonon in exciton spin relaxation. The observed LO assisted spin relaxation is in excellent agreement with the results from the spin injection experiments described above. When the spin splitting of two spin levels of the DMS hh exciton is less than the LO phonon energy, on the other hand, the observed hot PL line as expected retains the polarization of the pumping light [Fig. 5(a)]. In this case the hot PL line is due to the LO phonon replica of the PL arising from the upper spin state of the DMS hh exciton that is resonantly excited by the pumping light.

Unfortunately the exact rate of the LO-assisted exciton spin relaxation cannot be determined from the present study by the cw magnetooptical spectroscopy. Further time-resolved magnetooptical studies are needed. However, the range of the observed LO-assisted spin relaxation rate can be estimated from the hot PL experiments to lie between the spin-preserving LO-assisted momentum relaxation (ps or sub ps) and the decay rate of the DMS exciton (around tens of ps).

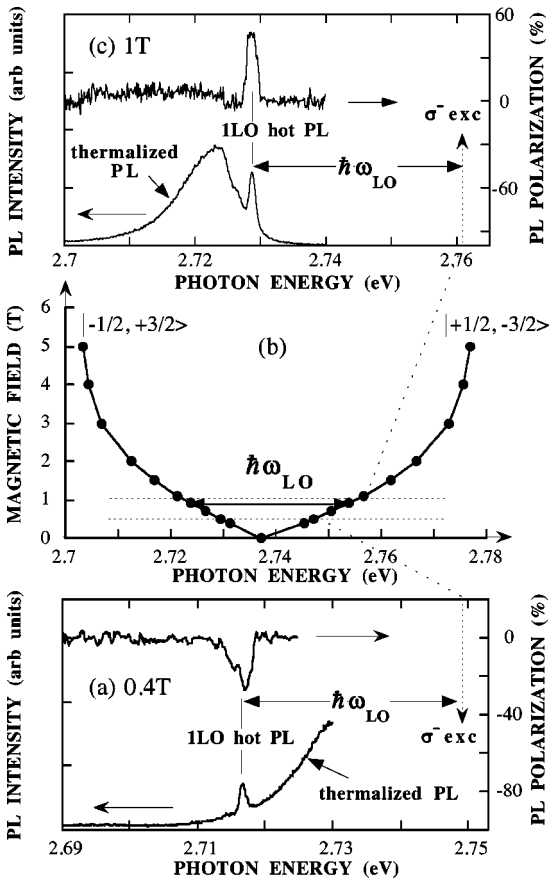

FIG. 5. Intensity and polarization of the DMS 1 LO hot PL under the resonant σ^- optical pumping at the upper-lying $|+\frac{1}{2},-\frac{3}{2}\rangle$ state, at a magnetic field of 0.4 T (a) and 1 T (c). At these two fields the spin splitting of the DMS hh exciton is smaller or larger than the LO phonon energy, respectively. The broad PL band arises from the thermalized distribution of the lower exciton band, over which the 1 LO hot PL line is superimposed. The polarization of the 1 LO hot PL is obtained after subtracting the polarization of the broad PL band. (b) Measured Zeeman splitting of the DMS hh exciton. The field positions at which the spectra in (a) and (c) were obtained are schematically shown by the dashed arrows.

IV. DISCUSSION

Below we shall discuss possible physical mechanisms responsible for the fast LO phonon assisted spin relaxation from the $|+\frac{1}{2}, -\frac{3}{2}\rangle$ state to the $|-\frac{1}{2}, +\frac{3}{2}\rangle$ state of the DMS hh exciton.

Before discussing these mechanisms we note that we use a widely accepted terminology when we talk about the exciton *spin* relaxation. A more precise term is the *angular moment* relaxation, because the hole moment $J=\frac{3}{2}$ is a combination of the valence electron spin $\frac{1}{2}$ and its orbital moment 1. On the other hand, dynamics of the so combined angular moment is the same as of a $\frac{3}{2}$ spin. This is because, due to the strong spin-orbit coupling, only quantum transitions within a manifold of the $\frac{3}{2}$ hole states are relevant, while the split-off states with $J=\frac{1}{2}$ can be ignored. Therefore, below we will refer to the widely accepted term ''exciton spin relaxation.''

A. Spin relaxation mediated by the long-range exciton exchange interaction

The exciton spin can change as a result of simultaneous flips of electron and hole spins, as well as due to a change in the spin state of one of the particles. The former process takes place due to the *e-h* exchange interaction.[12] The long-range part of this interaction gives rise to direct transitions between the $|+\frac{1}{2}, -\frac{3}{2}\rangle$ and $|-\frac{1}{2}, +\frac{3}{2}\rangle$ hh states. This was found[12] to be the main mechanism for the exciton spin relaxation in GaAs/AlGaAs quantum wells. This relaxation, however, was found to be efficient if both exciton states are degenerate or nearly degenerate. In our case an LO phonon could compensate the energy difference between $|+\frac{1}{2}, -\frac{3}{2}\rangle$ and $|-\frac{1}{2}, +\frac{3}{2}\rangle$ states. Hence, a combination of the long-range *e-h* exchange interaction with LO phonon emission could lead to a direct $|+\frac{1}{2},-\frac{3}{2}\rangle\rightarrow|-\frac{1}{2},+\frac{3}{2}\rangle$ relaxation, as shown in Fig. 4 (process Ic). We believe, however, that this mechanism is unlikely to be responsible for the observed spin relaxation for the following two reasons. First, in GaAs/AlGaAs quantum wells the long-range exchange interaction results in exciton spin relaxation times in the range of tens of ps and longer.[12] Participation of the LO phonon in our case can only further slowdown such a relaxation process, which could lead to a value inconsistent with the observed efficient LO-assisted spin relaxation. Secondly, the observed strong enhancement of spin relaxation with increasing magnetic field also disagrees with the opposite trend predicted by the theory[12] if the long-range exciton exchange interaction is the dominant mechanism. Caution must be exercised, however, in comparing our results with the theoretical prediction made for the nonmagnetic semiconductors as the two exciton bands of the DMS in our case have very different K vectors at a given energy that is not the case in nonmagnetic semiconductors with a small spin splitting.

B. Spin scattering via magnetic ions

As we mentioned above, the other path from $|+\frac{1}{2}, -\frac{3}{2}\rangle$ to $|-\frac{1}{2}, +\frac{3}{2}\rangle$ lays via independent electron and hole spin flips. An efficient mechanism for the fast (within several ps) flip of the electron or hole spins involves their strong exchange interaction with Mn magnetic impurities. After a collision with such an impurity the particle spin changes its projection onto the spin quantization axis by $\Delta m=\pm 1$, while the corresponding projection of the Mn^{2+} spin changes by $\Delta m(Mn^{2+})=\mp 1$. In such processes the energy transferred to or from the magnetic spins is rather small because of the small Zeeman splitting of the Mn^{2+} ions. On the other hand, the energy difference between $|+\frac{1}{2}, -\frac{3}{2}\rangle$ and $|-\frac{1}{2}, +\frac{3}{2}\rangle$ exciton states can be compensated by LO phonon emission. One can consider two possible mechanisms involving the emission of one LO phonon (see Fig. 4). The first model is a two-step process. It invokes a nearly elastic spin scattering (process Ia in Fig. 4), which induces the $|+\frac{1}{2},-\frac{3}{2}\rangle\rightarrow|-\frac{1}{2}, +\frac{3}{2}\rangle$ transition with only a little energy change, followed by an LO-assisted spin-preserving kinetic energy relaxation (process Ib). In the second model the change of the exciton spin and the phonon emission occurs as one quantum process (Ic), very similar to the relaxation via the long-range *e-h* exchange interaction considered above except that the driving force for spin flips is now the interaction with magnetic ions.

Now we will consider the first model in more detail. The relaxation rate in such a two-step process is controlled by the slowest step. The second step (Ib in Fig. 4) is the energy relaxation via LO phonons within the $|-\frac{1}{2}, +\frac{3}{2}\rangle$ band. This process is very fast in II-VI materials (typically in the range of ps or even sub-ps in ZnMnSe DMS). The process Ia, in its turn, can consist of several intermediate steps. Their array always includes an electron spin flip from $+\frac{1}{2}$ to $-\frac{1}{2}$. If hh-lh hybridization effects are ignored, in order to complete transition of the hole spin state from $-\frac{3}{2}$ to $+\frac{3}{2}$, as many as three hole spin flips are necessary. Due to strain, however, the light hole exciton bands are about 30 meV higher than the hh bands, such that when the Zeeman splitting of the exciton is near the LO energy the magnetic field is not strong enough to bring about the hh-lh crossing. Therefore, there are no available lh states for an almost elastic transition from $-\frac{3}{2}$ to $-\frac{1}{2}$. Hence, the hh-lh mixing must assist the entire transition $|+\frac{1}{2},-\frac{3}{2}\rangle\rightarrow|-\frac{1}{2},+\frac{3}{2}\rangle$.

As is known,[22] in quantum wells each of the exciton states $|\pm\frac{1}{2},v\rangle$, where $v=\pm\frac{3}{2},\pm\frac{1}{2}$, can be expanded in terms of products of zone center electron and hole Bloch states. The latter are labeled by the hole's angular moment projection $m=\frac{3}{2},\frac{1}{2},-\frac{1}{2},-\frac{3}{2}$. When the hh-lh mixing effect is weak the term with $m=v$ strongly dominates in such an expansion. Otherwise, $m\neq v$ components are also represented. Each of these components contributes to the total wave function of the *e-h* relative motion with its respective amplitude $\Psi^{v}_{pm}(\mathbf{r},z_h)\chi(z_e)$, where $\mathbf{p}$ is the total exciton two-dimensional (2D) momentum and $\mathbf{r}$ is the *e-h* relative 2D coordinate, while z_h and z_e are the carrier coordinates in epitaxy growth direction. For simplicity we will assume that only the lowest conduction electron subband with the wave function $\chi(z_e)$ contributes to the exciton eigenstate. Using the perturbation theory one can write the rate of transition from $|+\frac{1}{2},v\rangle$ to $|-\frac{1}{2},v\rangle$ after an electron spin flip on a magnetic impurity as[23]

$$\frac{1}{\tau_s}=\frac{m_{\mathrm{ex}}^{v'}}{2\hbar^3}xN_0\alpha^2|F^{vv'}(\mathbf{p},\mathbf{p}')|^2 \times\int|\chi(z)|^4dz\sum_l(J^+_{l,l+1})^2\rho(E_l), \quad (1)$$

where $m_{\mathrm{ex}}^{v'}$ is the exciton mass in the v' band, x is the molar fraction of Mn^{2+} impurities, α is the strength of the exchange interaction between conduction electrons and Mn^{2+} ions ($N_0\alpha=0.26$ eV for ZnMnSe), $J^+_{l,l+1}$ is the matrix element of the raising operator for the Mn^{2+} angular moment $J=\frac{5}{2}$, so that l runs from $-\frac{5}{2}$ to $+\frac{5}{2}$. We have ignored interaction between ion spins. Therefore, these spins are distributed independently according to the thermodynamic weight $\rho(E_l)=e^{-E_l/k_BT}/Z$ of each spin state l having the Zeeman energy $E_l=g_{\mathrm{Mn}}\mu_B B$, where B is the external magnetic field strength. For $B=1$ T a typical distance between energy levels E_l is of the order of the thermal energy k_BT at $T=2$ K in our experiment. The form factor $F^{vv'}(\mathbf{p},\mathbf{p}')$, which is a

function of exciton momenta before and after scattering, is defined as

$$F^{vv'}(\mathbf{p},\mathbf{p}') = \sum_{m=\pm 3/2,\pm 1/2} \int \Psi^{v}_{\mathbf{p}m}(\mathbf{r},z)\Psi^{v'*}_{\mathbf{p}'m}(\mathbf{r},z) \times \exp[-i(\mathbf{p}-\mathbf{p}')\mathbf{r}]d^2r\,dz\,\frac{d\varphi_{\mathbf{p}'}}{2\pi}, \qquad (2)$$

where integration over $\varphi_{\mathbf{p}'}$ means integration over $\mathbf{p}'$ directions in the plane of the quantum well. This formfactor is normalized so that $F^{vv'}(\mathbf{p},\mathbf{p}')=\delta_{vv'}$. When $|\mathbf{p}-\mathbf{p}'|a \geqslant 1$, where a is the exciton radius, $F^{vv'}(\mathbf{p},\mathbf{p}')$ decreases. We assume that excitons in the upper $|+\frac{1}{2}, -\frac{3}{2}\rangle$ band are thermalized. Hence, the initial momentum $\mathbf{p}$ is determined by small energies near the bottom of this band. At the same time, after a scattering into some of the lower energy exciton bands, almost entire Zeeman split energy is transformed into the exciton kinetic energy $E_{v'}(p')$. For example, in the vicinity of the threshold $E_{+3/2}(p') \approx 30$ meV. Hence, $p' \gg p$ for the $|+\frac{1}{2},-\frac{3}{2}\rangle \rightarrow |-\frac{1}{2},+\frac{3}{2}\rangle$ scattering, as well as for $|+\frac{1}{2}, -\frac{3}{2}\rangle$ scattering into the optically inactive state $|-\frac{1}{2}, -\frac{3}{2}\rangle$. The final momentum is so large that $pa \geqslant 1$. This leads to decreasing of the form factor. On the other hand, at large p' hh-lh mixing effects become more important, so that $F^{+3/2-3/2}$ is not small.

Since the sum in Eq. (1) is of the order of unity in the broad range of temperatures and $\int|\chi(z)|^4dz \sim 1/L$, where L is the quantum well width, we get for $L=100$ Å, $x=0{,}05$, and $N_0=4\times10^{22}$ cm^{-3}

$$\tau_s \sim 4|F^{vv'}(p')|^{-2} ps.$$

One can see from this equation and Eq. (2) that the fastest scattering process is from $|+\frac{1}{2}, -\frac{3}{2}\rangle$ state into the optically inactive $|-\frac{1}{2}, -\frac{3}{2}\rangle$ exciton. In this case $F^{vv'}$ is expected to be not much less than 1 because $v=v'$ and hh-lh mixing is not important. We, however, cannot say now how small the form factor will be for the transition into the $|-\frac{1}{2}, +\frac{3}{2}\rangle$ state. On the other hand, as soon as $|-\frac{1}{2}, -\frac{3}{2}\rangle$ states are rapidly populated, transitions from these states into the $|-\frac{1}{2}, +\frac{3}{2}\rangle$ exciton can be induced by the Elliott-Yafet spin relaxation mechanism[8,9] via hole scattering on charged impurities.

The $|-\frac{1}{2}, -\frac{3}{2}\rangle$ to $|-\frac{1}{2}, +\frac{3}{2}\rangle$ transition, via the Elliott-Yafet mechanism or the hole spin-flip scattering on magnetic impurities, again requires the lh-hh mixing. This mixing, in its turn, can be diminished by a large hh-lh splitting (about 30 meV for the hh-lh exciton splitting in our samples). A previous experimental study[6] has shown a drastic reduction in exciton spin relaxation by three orders of magnitude from 5 ps in unstrained ZnMnSe to greater than 1 ns in strained ZnMnSe (with 2 or 4 % of Mn). The hh-lh splitting in Ref. 6 is known to be degenerate and 13 meV in the unstrained and strained samples, respectively, whereas the Zeeman splitting of the hh exciton states was kept at 8 meV in both cases. This reduction is likely to be caused by slow spin flips (such as process Ia in Fig. 4) and not by the subsequent acoustic phonon assisted momentum relaxation (process Ib) as the latter is known to be faster than 1 ns [300 ps (Ref. 24)] in their samples. If this is the case, accelerating process Ib from the acoustic-phonon assisted relaxation to the much faster LO-assisted relaxation when the spin splitting crosses over the LO energy would not affect the spin relaxation rate as it is determined by the slowest process Ia. This is not consistent with our experimental finding and therefore seems to indicate the first model unlikely to be responsible in our case, assuming that the hh-lh mixing is the dominant mechanism for the observation given in Ref. 6. It should be pointed out, however, that the exact mechanism for the spin relaxation slowdown in Ref. 6 is to our knowledge not completely understood and a direct, unambiguous connection to the reduction in the hh-lh mixing has yet to be established. Also, the observed relaxation slowdown is too big to be associated with the hh-lh mixing. It should also be noted that the results of Ref. 6 were obtained from bulk samples. It is not clear to what extent quantum confinements like in our samples would affect the conclusion that requires further theoretical studies. Nevertheless, one can not exclude such a possibility and could consider it for our samples with a greater hh-lh splitting.

Within the second model, the concerned spin relaxation of the DMS hh exciton is suggested to be accomplished by an inelastic process involving a direct LO-assisted spin flip (process 1c in Fig. 4). Though this model can explain the experimental findings from both spin injection and hot PL experiments, it is a higher order process requiring some magnetic mediator between the LO phonons and the carrier spins such as lattice deformations associated with the LO phonons that could modulate the exchange interaction between carriers and magnetic ions. It should be pointed out that in the past such a higher order spin relaxation process has not been adequately addressed theoretically and experimentally. It should also be said that the exact role of phonons in spin flip is still largely unknown in semiconductors and metals as the load of computation becomes formidable. For example, only until very recently was phonon-assisted spin relaxation of electrons in a metal calculated.[1]

V. CONCLUSIONS

In conclusion, we have observed a strong effect of spin splitting on spin relaxation of the hh exciton of the DMS ZnMnSe/CdSe SL leading to a significant increase in spin relaxation when the exciton spin splitting exceeds the LO phonon energy. Experimental evidence for the involvement of the LO phonons in promoting spin relaxation has been provided from the spin injection and hot PL experiments. Possible physical processes involved in the LO-assisted spin relaxation have been discussed in terms of either a nearly elastic spin scattering followed by the LO-assisted kinetic energy relaxation of the hot excitons or a high-order LO-assisted spin flip, mediated by the long-range exciton exchange interaction or strong interactions with magnetic ions. The observed efficient LO-assisted exciton spin relaxation in the DMS is rather unusual, which can not be explained by existing theories, and therefore calls for future in-depth theoretical and experimental investigations. The present experimental finding also points to a way to manipulate spin relaxation, important in obtaining active spintronic devices.

ACKNOWLEDGMENTS

The financial support by the Swedish Research Council, the Wenner-Gren Foundation, the Swedish Royal Academy of Sciences, RFBR Grants No. 00-02-16997 and 02-0217643, the INTAS Grant No. 01-2375, the Program of the Ministry of Science of RF ''Physics of Solid State Nanostructures,'' and Vokswagen Foundation is greatly appreciated. We are grateful to Dr. I.G. Ivanov for his expert assistance with the dye laser.

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