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Magnetic-field-dependent type-II transition induced by resonant electron tunneling from a diluted magnetic semiconductor quantum well into self-assembled quantum dots

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Magnetic-field-dependent type-II transition has been observed between electrons in self-assembled quantum dots (QDs) of CdSe and heavy holes (hhs) in a diluted magnetic semiconductor quantum well (DMS-QW) of $Zn_{1-x-y}Cd_xMn_ySe$, originating from an electron tunneling from the DMS-QW into the QDs. The electron tunneling is resonantly assisted by LO-phonon scattering in the QDs. As a result, the type-II emission energy depends on a magnetic field, since the electron energy in the DMS-QW is a function of field strength. The magnetic-field dependence of intensity of the type-II emission suggests a level-crossing field of the hh states between the QD and DMS-QW. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2360779]

I. INTRODUCTION

Self-assembled quantum dots (SAQDs) of compound semiconductors have extensively been studied because of their potential applications to efficient optical devices such as QD lasers. In addition to this, spin states of an electron in the SAQD are well known as one of the important candidates for the realization of spintronic devices and quantum computing.[1,2] The energy levels of the spin-polarized electron and hole are completely discrete in magnetic fields in the SAQD, originating from strong quantum confinement effects, and therefore it enables us to manipulate the individual spin-polarized carriers. In SAQDs, a spin-relaxation time of the electron was also clarified to be as long as several nanoseconds, and the mechanisms responsible for such a long relaxation time were discussed.[3–6] The long relaxation time of electron spin has an important advantage for the manipulation of the spin states. Thus, it is necessary to investigate the spin dynamics of carriers or excitons in the SAQDs. One possible way to the manipulation of the electron spin is the use of a diluted magnetic semiconductor quantum well (DMS-QW). In the DMS-QW, carriers or excitons can be fully spin polarized due to the giant Zeeman effects in magnetic fields, and the direction of spins can be easily controlled by the direction of an external magnetic field.[7–9] Therefore, coupled structures of the SAQDs with the DMS-QW offer a good opportunity for the manipulation of carrier spins in the SAQDs, if the spin-polarized carriers are efficiently injected from the DMS-QW into the SAQDs via quantum tunneling. The spin injection into a nonmagnetic QW has been reported from a II-VI DMS or III-V ferromagnetic semiconductor as a spin aligner.[10,11] The dynamics of the spin injection is recently studied using coupled QW structures composed of a II-VI DMS-QW and a nonmagnetic one, by time-resolved circularly polarized photoluminescence (PL).[12–15] The results show that the spin injection originates from individual tunneling of a spin-polarized electron and heavy hole (hh) with different effective masses, rather than an exciton picture. In addition, the spin injection into SAQDs of CdSe has recently been reported through a diffusion process of excitons from a II-VI DMS.[16] However, the spin injection into the QD via quantum tunneling is sufficiently not examined yet. The tunneling process for manipulating the electron spin is especially important, since the spintronic devices and quantum computing using the QDs will be developed on the basis of the coupled QD structures. In the SAQDs, the density of states of the carriers is discrete and therefore resonant tunneling is strongly required to inject the spin-polarized carriers efficiently into the QDs through a quantum process. Therefore, we have fabricated and studied a coupled SAQD system with a DMS-QW. As a result, an experimental evidence for the resonant electron tunneling is clearly given by the observation of a magnetic-field-dependent type-II transition, which arises from a spin-polarized indirect exciton composed of an electron injected from the DMS-QW into the SAQD and a spin-polarized hh remained in the DMS-QW.

II. EXPERIMENTAL PROCEDURES

A coupled SAQD structure with a DMS-QW was prepared by molecular beam epitaxy. The structure fabricated is illustrated in Fig. 1. A CdSe layer with a thickness of 1.5 monolayers was grown with a ZnSe-buffer layer on a GaAs(100) substrate. SAQDs of CdSe were subsequently formed by a proper annealing process. After the formation of the QD layer, a tunneling barrier of ZnSe with a thickness of 5 nm was deposited, followed by a magnetic well (MW) of $Zn_{0.68}Cd_{0.22}Mn_{0.10}Se$-DMS with a thickness of 10 nm. Finally, a cap layer of ZnSe was deposited on it. In addition, a single layer of CdSe-SAQDs or of the MW was also examined. Micro-PL (m-PL) observations at 4.2 K revealed sharp spectra with a spectral width less than 0.4 meV that was the instrumental width, using a mesa-shaped QD layer with a

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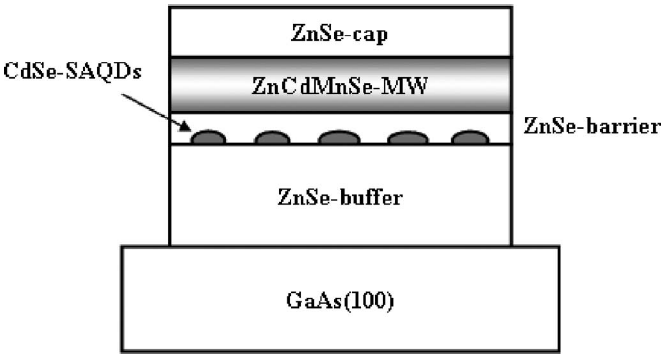

FIG. 1. Schematic drawing of a coupled CdSe-QD structure with a MW of DMS-$Zn_{1-x-y}Cd_xMn_ySe$.

diameter of 60 nm. It indicates excitonic PL from the individual CdSe dots with strong quantum confinements. The m-PL in magnetic fields with the Faraday geometry revealed a negative g value of -1.7 for excitons in the individual CdSe QDs. Time-resolved PL with a circularly polarized plate was measured for the plane sample of the coupled structure in magnetic fields. A second-harmonic light of a mode-locked Ti:sapphire laser with a wavelength of 390 nm was used as an excitation light source, where the photoexcitation was performed above the energy of the ZnSe-buffer, ZnSe-barrier, and ZnSe-cap layers. The circularly polarized time-resolved PL spectra were detected using a streak camera with a time resolution of 15 ps.

III. RESULTS AND DISCUSSION

Figure 2 shows circularly polarized cw-PL spectra and the degree of circular polarization at 5 T in a single SAQD layer of CdSe (a), nonresonantly coupled SAQDs (b), and resonantly coupled SAQDs (c), respectively. The exciton energy of the QD emission band was controlled by changing the averaged QD size. In the resonant sample, two sharp PL peaks overlap with a broad emission spectrum due to the QD ensemble. From the following discussion, these spectral peaks are identified due to a direct exciton recombination in the MW (E^{MW}), a type-II transition ($E^{\text{type-II}}$), and a QD emission band ($E^{\text{CdSe-QDs}}$). First, the σ^+ polarization (left circular) is defined for the exciton recombination of the lower spin state in the DMS-QW ($m_j=-1/2,+3/2$), as it is seen in (b). The CdSe-SAQDs show only a broad PL band due to the size distribution [(a) and (b)]. The Zeeman splitting of excitons in individual QDs is less than 1 meV at 5 T, which was directly confirmed by m-PL under magnetic fields using a mesa-shaped sample. Thus, only the intensity of the broad emission is different between both circular-polarized PL spectra, where the intensity of the σ^--polarized spectrum is higher in the CdSe-QDs since the exciton g value in the CdSe-QD is negative. The excitonic PL band of the QDs overlaps in part in the resonant sample with the excitonic PL of the MW (c). The exciton PL spectrum in the MW is known from the significant field dependence. This shows typical giant Zeeman shifts of excitons, which can be expressed by a Brillouin function and is discussed later. In addition to these spectra, a PL peak due to a type-II transition is observed only in this resonantly coupled QD sample. The type-II transition has

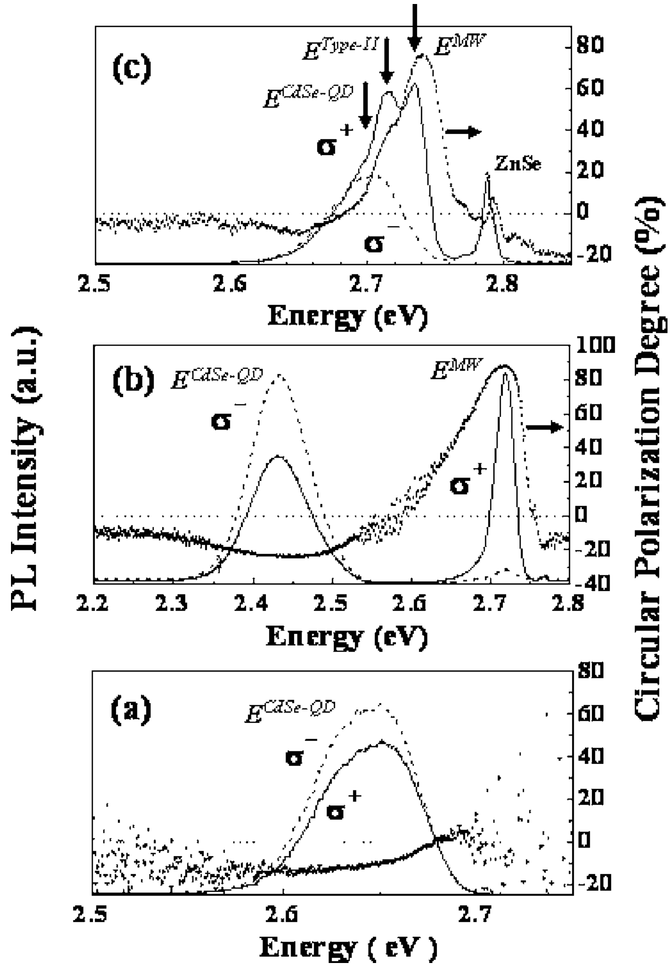

FIG. 2. Circularly polarized (σ^+: a solid line, σ^-: a broken line) cw-PL spectra and the circular polarization degree (a closed circle) at 5 T in a single SAQD layer (a), nonresonantly coupled SAQDs (b), and resonantly coupled SAQDs (c), respectively. Spectral peaks due to a direct exciton in the MW (E^{MW}), a type-II transition ($E^{\text{Type-II}}$), and a QD emission band ($E^{\text{CdSe-QDs}}$) are shown by arrows in the resonant sample.

experimentally been confirmed by the following observation of time-resolved PL corresponding to this spectral peak.

Figure 3 shows a circularly polarized excitonic PL spectrum with a σ^+ probe at 5 T, for the resonantly coupled QDs with the MW. The measurement was performed at 2 K by using a streak camera with a time window of 0–9 ns. The exciton PL spectra of the MW and QDs are shown by solid lines. The PL spectrum of a QD emission band is directly known from a PL spectrum with a σ^- probe. The PL peak due to a type-II transition is shown by a broken line. The result for the time-resolved measurement is shown in Fig. 4. As can be seen, only in the σ^+ geometry, a long-lived emission is clearly seen, where the decay characteristic can be expressed by a simple exponential function with a time constant of 3.5 ns. On the other hand, circularly polarized PL intensities and the circular polarization degree are shown as a function of time in the single SAQDs without the MW, in Fig. 5. The direct recombination of excitons in the CdSe-SAQDs shows a lifetime of 240 ps. The time evolution of the negative polarization (σ^-) indicates the spin relaxation inside the dot since the exciton g value is negative. Time dependences of the circularly polarized PL intensity are fitted by

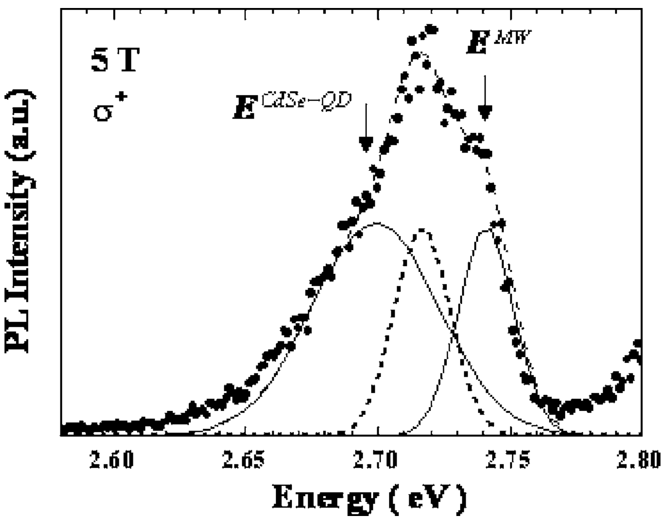

FIG. 3. σ^+-polarized excitonic PL spectrum at 5 T in the resonantly coupled QD structure with MW, where the spectrum was time integrated in the region from 0 to 9 ns using a streak camera. The exciton PL spectra of the MW and QDs are shown by solid lines. The PL component due to the type-II transition is shown by a broken line.

rate equations taking individual electron- and hh-spin relaxations into account. From these fittings, we deduce time constants for the electron- and hole-spin relaxations such as 5 and 1.5 ns, respectively. This transient behavior of the circularly polarized PL is observed at PL components due to the QDs in the coupled samples. The direct exciton recombination in the QD is also observed on the decay curves with the time region less than 1 ns in Fig. 4, where the intensity contribution of the type-I transition is dominant. The lifetime of direct exciton in the MW is in the range of several tens of picoseconds depending on the magnetic field, and therefore it can be separated from the transient behavior of the direct exciton in the QD.

To understand the mechanism responsible for the type-II transition observed, the field dependence of the type-II emission energy is plotted in Fig. 6, as well as that of the direct exciton energy in the MW. Both field dependences can be expressed by Brillouin functions using the same parameters, indicating the typical giant Zeeman effects of excitons. In the calculation, exchange constants of $N_0\alpha$ of 0.26 and $N_0\beta$ of -1.26 were employed for an electron and hole, respectively, which were obtained for $Zn_{0.68}Cd_{0.22}Mn_{0.10}Se$.[17] The energy of the type-II transition follows the same field dependence as that of the exciton energy in the MW, with decrements of 1-LO-phonon energy of the CdSe QD (indicated by a broken line), where the LO-phonon energy of 27 meV is confirmed for the CdSe QD by using photoluminescence excitation (PLE) measurements. For the detailed discussion, the electron and hh energies in the MW are calculated as a function of magnetic field (b). The origin of the vertical energy axis is

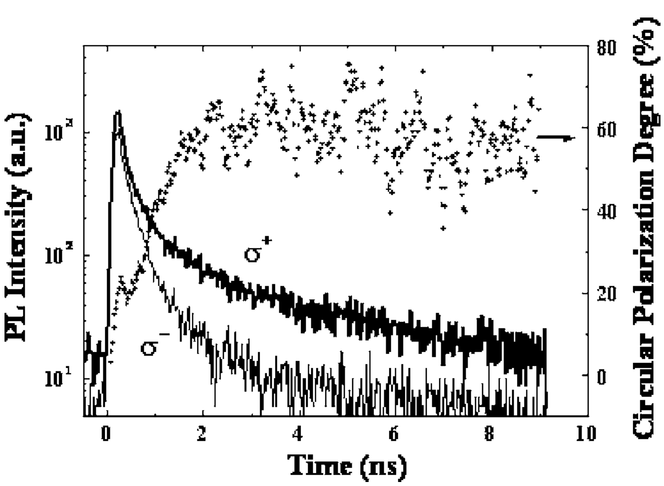

FIG. 4. Time dependences of σ^+ (a thick line)- and σ^- (a thin line)-polarized PL intensities and the circular polarization degree P (a solid circle) at 5 T in the resonantly coupled QD structure, for the PL-energy range of 2.70–2.72 eV corresponding to the type-II transition. The P value is defined as $P=(I_{\sigma+}-I_{\sigma-})/(I_{\sigma+}+I_{\sigma-})$ with the circularly polarized PL intensities, $I_{\sigma+}$ and $I_{\sigma-}$.

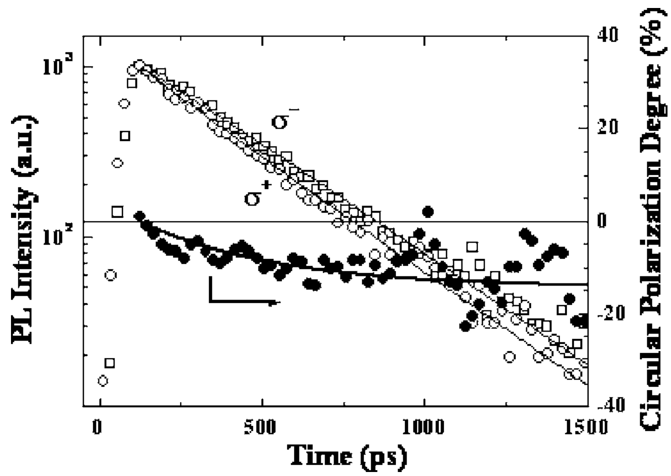

FIG. 5. σ^+ (an open circle)- and σ^- (an open square)-polarized PL intensities and the circular polarization degree (a solid circle) as a function of time, in the single QD layer. The solid lines are fitted calculations using rate equations.

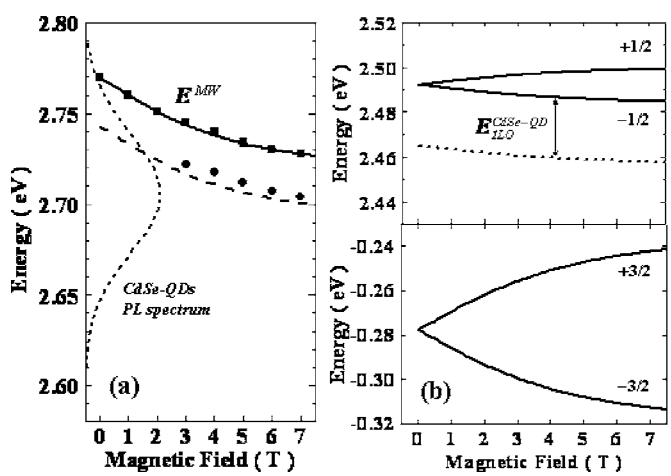

FIG. 6. PL-peak energies of direct excitons in the MW (a solid square) and in indirect excitons (a solid circle) as a function of magnetic field (a). The QD spectral band is also shown by a dotted line. A fitted calculation for the direct exciton energy in the MW using a Brillouin function is shown as a function of magnetic field by a solid line. The energy with the decrement of 1-LO-phonon energy from the Brillouin fitting is also shown by a broken line. The calculated electron and hh energies in the MW as a function of magnetic field (b), where the origin of the vertical axis is set to be 0 at the top of valence band of bulk CdSe.

set to be 0 at the top of the valence band of bulk CdSe, and the valence-band offset of CdSe to ZnSe is set to be 0.30. It should be noted that the exciton effect such as a binding energy is not included in the calculation, since such an effect is rather difficult to apply for our coupled structure. First, the exciton binding energy depends largely on the shape and dimensions of the quantum structures. Especially, the exciton binding energy in the quantum dot is under discussion and the explicit function is given only for the case of spherical shape.[18,19] Moreover, the picture of the indirect exciton is not discussed yet for the coupled QD system with a two-dimensional QW. Therefore, we would discuss about phenomenological aspects for our experimental results and above calculation is used as a reference to understand the phenomena only. If we assume the system of CdSe QD with the ground state of electron at 2.459 eV at 5 T, where the energy of this electron ground state is 1-LO energy lower than the down-spin-electron level $(-1/2)$ in the MW, the corresponding hh $(\pm 3/2)$ energy in this QD is calculated as -0.254 eV assuming a spherical shape of the dot. On the other hand, the down-spin hh $(+3/2)$ energy in the MW is -0.248 eV at the same 5 T and becomes lower than that of the hh state in that QD. Therefore, the band alignment for this coupled structure at 5 T suggests that the down-spin hh $(+3/2)$ in the MW cannot penetrate into the QD, while the electron can be injected via LO-phonon-assisted resonant tunneling. This situation results in the formation of an indirect exciton at the interface between the QD and MW, and therefore a type-II transition can be induced with a relatively low rate of the transition probability.

From the above discussion, the spin-polarized electron can efficiently tunnel directly from the down-spin state in the MW to that in the QD via LO-phonon scattering. As a result, the down-spin electron transforms from a direct exciton state in the MW into an indirect one in the QD, where the indirect exciton binding energy is supposed to be small. In the QDs, the exciton Zeeman shift is almost negligible even in high magnetic fields such as 5 T, since the energy shift was less than 1 meV. The down-spin electron injected in the dot relaxes to the lower energy (up-spin) state since the g value is negative. However, such an electron-spin relaxation time is as long as 5 ns, which is equivalent to the lifetime of the indirect exciton (3.5 ns). Therefore, the energy difference between the exciton PL in the MW and the type-II transition is approximately equal to the 1-LO-phonon energy. Here, the QDs having excess electrons injected from the DMS-QW are selectively replaced by changing the magnetic field since the electron injection is induced by a LO phonon with a constant energy, where the energy of the down-spin-electron level in the DMS-QW depends on the magnetic field. It should be noted that the magnetic-field dependence of the type-II transition showed a slight discrepancy from a Brillouin function, as shown in Fig. 6(a). It implies a magnetic-field-induced change in the indirect exciton binding energy responsible for the type-II transition. The energy of the indirect exciton can depend on the magnetic field, since the hh wave function in the MW depends on the magnetic field due to the shallow potential depth, while the field dependence of the electron wavefunction can be negligible. A possible scenario for this type-II transition at 5 T is schematically illustrated in Fig. 7.

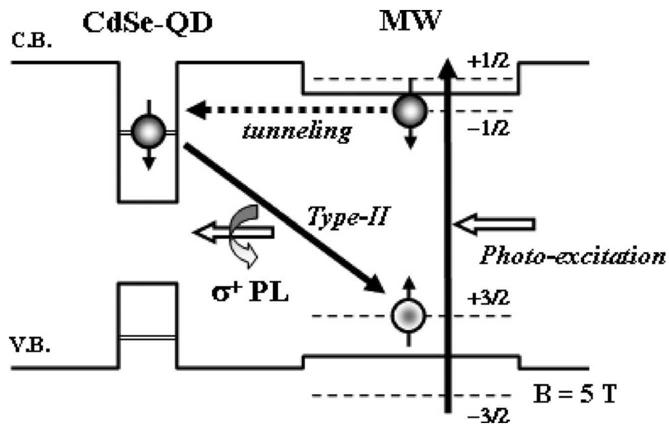

FIG. 7. Schematic band alignment of the resonantly coupled QD structure with MW at 5 T, where the electron energy in the QD is assumed to be 1-LO-phonon-energy lower than the down-spin-electron level in the MW. Related carrier-spin dynamics and the type-II transition are also illustrated.

The down-spin electrons $(-1/2)$ in the MW can efficiently be injected into the QDs by resonant tunneling due to LO-phonon scattering. On the other hand, the up-spin holes $(-3/2)$ in these QDs can migrate from the MW into the QDs, since the hh energy of the up-spin state is higher than the barrier energy at 5 T. The amount of migrated up-spin hh is a function of ratio between the migration time and the spin-flip time in the MW. During the migration, the hole spin can be immediately relaxed because of its ultrafast relaxation time (in the time region of subpicoseconds) due to the subband mixing.[20] Also, the spin-flip time of the hole is generally known to be as fast as several picoseconds or less.[21] As a result, a certain number of holes generated at the up-spin state in the MW fall into both up- and down-spin states in the QDs.

To inquire into the situation closely, time-integrated spectra with the time window from 2.5 to 10 ns are shown as a function of magnetic field in Fig. 8, where the type-I transition with the lifetime of 240 ps in the QDs is completely negligible. As can be clearly seen, the type-II transition of indirect excitons appears at 3 T and shows the giant Zeeman shift toward a lower energy side with increasing magnetic field. Therefore, we conclude that the down-spin hh level $(+3/2)$ of the MW starts across the hh levels of the CdSe QDs at 3 T. This is a direct observation of the magnetic-field-induced type-II transition in the coupled QD system with the DMS-QW. The observation of the present type-II transition is important, because it is a compelling experimental evidence for the electron injection from the MW into the QD via 1-LO-phonon-assisted resonant tunneling. The spin state of the injected electron is not directly known from the type-II transition since only the down-spin hhs exist in the MW in magnetic fields due to the giant Zeeman splittings, and the electrons with the down-spin state in the dot can selectively recombine with these spin-polarized hhs in the MW. Our direct measurements for the electron-spin relaxation deduce a time constant of 5 ns for the single QD layer, which is comparable with the lifetime of the type-II transi-

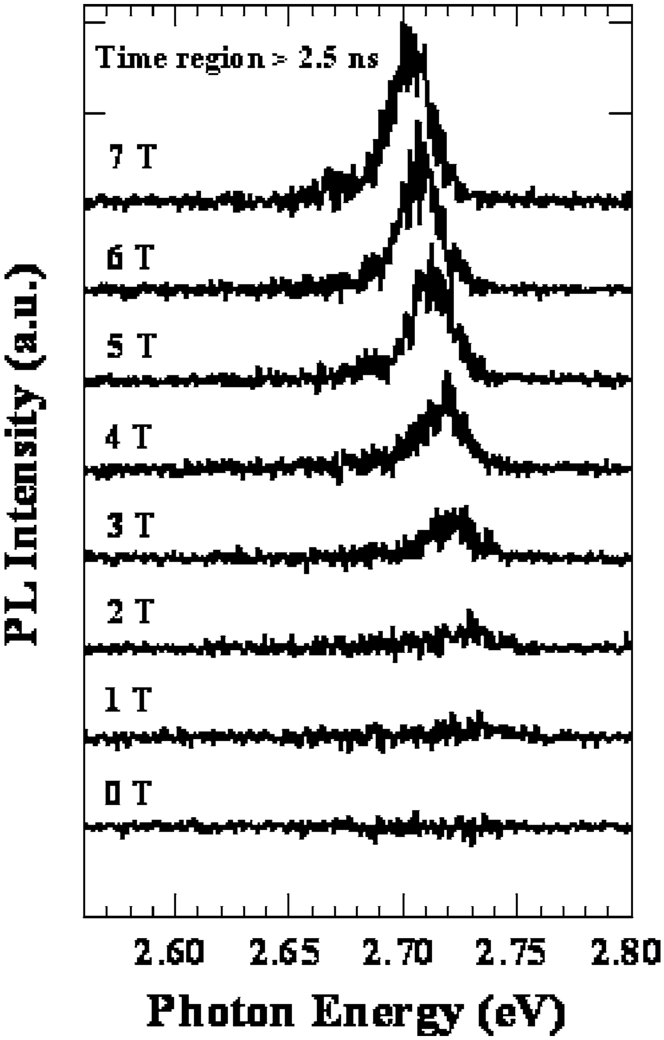

FIG. 8. Time-resolved excitonic PL spectra integrated with the time window of 2.5–10 ns, as a function of magnetic field. The type-II transition is shown to appear at 3 T.

tion. Therefore, the spin state of the electron injected from the down-spin state of the MW can be highly preserved during the indirect exciton recombination.

Finally, we discuss the dynamics of the electron-spin injection. In Fig. 4, we observe the fast decay component of excitonic PL with the lifetime of 240 ps for σ^+ polarization. This indicates the type-I transition due to radiative recombination of an electron and a hh inside the QD. From the PL intensities with the σ^+ and σ^- polarizations, the circular polarization degree P up to 20% is obtained at the initial stage after the excitation. A time constant of the rise of circular polarization is 20 ps. After this rise, the polarization starts to increase gradually, originating from the type-II transition. Therefore, this fast rise of polarization in the coupled QDs can be attributed to the type-I transition inside the QD, where the excess hhs can be migrated from the higher energy (up-spin) state in the MW since the up-spin energy is higher than the barrier energy. These characteristics of the circularly polarized PL are quite different from those of QDs without a DMS-QW. As shown in Fig. 5, excitonic PL shows that a negative value of P rises gradually up to 15% toward 700 ps after the excitation. The rise time of the negative P gives electron- and hole-spin relaxation times of 5 and 1.5 ns, respectively, in the single CdSe-QD layer. Therefore, in the present coupled QDs with the MW, the appearance of positive P values with the fast rise time evidences the recombination of direct excitons with down-spin electrons and holes. This means that the spin-polarized electron is injected from the MW. We consider rate equations in order to understand the double exponential decay characteristic observed in the coupled structure. A three-level system is assumed for the electron with one excited state and two ground states, corresponding to the type-I and type-II transitions. On the other hand, a two-level system is assumed for the hh, corresponding to the type-I transition inside the dot. The PL intensity can be expressed by the number of minority carrier. The decay curve of the PL can be expressed by a double exponential function. It explains the spin-polarized exciton dynamics observed by time-resolved PL for the resonantly coupled QD system.

IV. SUMMARY

We have observed a magnetic-field-dependent type-II transition with a lifetime of 3.5 ns in self-assembled QDs of CdSe coupled with a DMS-QW. The type-II transition is induced by an electron injected from the DMS-QW into the QD and a spin-polarized hh in the DMS-QW. The magnetic-field dependence of the type-II transition energy indicates that the electron tunneling is resonantly assisted by the energy relaxation involving LO-phonon scattering. The level-crossing field of the hh states between the QDs and DMS-QW is experimentally suggested from the field-dependent indirect exciton PL. The spin state of the injected electron is also discussed from the time dependence of the degree of circular polarization for the PL intensity.

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