Spin polarization of self-assembled CdSe quantum dots in ZnMnSe

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Nearly complete spin polarization (SP) of carriers was observed in photoluminescence (PL) experiments on CdSe self-assembled quantum dots (QDs) embedded in ZnMnSe at 5 K in magnetic fields above 1 T. The redshift of the PL from CdSe QDs in the presence of the magnetic field was less than that of ZnMnSe, but still sufficiently large to account for the high SP. At low fields, however, the SP of carriers in CdSe QDs is observed to be smaller than in the surrounding ZnMnSe. This, along with time-resolved PL data, suggests that the SP in CdSe QDs is mainly due, *not* to the injection of SP carriers from ZnMnSe, but rather due to the Zeeman splitting of internal QD states.
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In magnetic semiconductors, electrons and holes experience large spin splittings due to the strong exchange interaction between the band carriers and magnetic ions.[1,2] The spin splitting energy can be measured from the redshift of the luminescence or from the Raman shift associated with spin flip of electrons in the presence of an external magnetic field.[3,4] When the spin splitting is larger than thermal energy, most carriers will relax to the lower energy spin state. Spin-polarized carriers in nonmagnetic quantum wells can then be obtained by the injection of the spin-polarized carriers from magnetic semiconductors, which has already been demonstrated using both the II-VI-based magnetic semiconductor $Zn_{1-x}Mn_xSe$,[5,6] and the III-V system $Ga_{1-x}Mn_xAs$.[7] In multiple-quantum-well structures whose barriers consist of magnetic semiconductors, significant redshift of the luminescence was observed under external magnetic field due to the wave-function penetration of carriers into the magnetic barriers.[8]

Spin states in quantum dots (QDs) are of interest for their potential role in single electron transistors[9] and quantum computing,[10] and the spin polarization (SP) and spin dynamics studies in semiconductor QDs are thus important. In this context, a QD system embedded in a magnetic semiconductor provides an attractive opportunity to manipulate spin states by magnetic fields. Using light excitation above the bandgap of the magnetic barrier in the presence of an external magnetic field the photocarriers generated in the barrier will be spin-polarized, and may retain their SP as they transfer to the QDs. On the other hand, carriers within the QDs are also expected to experience Zeeman splitting thus causing carrier spin alignment within the dots. The Zeeman splitting in self-assembled II-VI QDs has been studied earlier,[11–13] but the specific contributions of the various SP mechanisms in QDs has not been explicitly addressed.

In this letter, we study the SP of carriers in CdSe QDs embedded ZnMnSe as measured by the degree of circular polarization of the photoluminescence (PL) in an applied magnetic field, with an eye on resolving the issue of whether the spin polarization in the QDs is due to the transfer of the spin-polarized carriers generated in $Zn_{1-x}Mn_xSe$, or due to the Zeeman splitting of exciton states within the QD itself. While at high magnetic fields, a nearly complete SP can be achieved in the CdSe QD system, we find that at relatively low fields, the SP ratio in CdSe QDs is considerably smaller than in the surrounding ZnMnSe. The reduced SP in QDs may imply spin scattering at the QD interface, or fast spin reorientation within the QDs. On the other hand we find, as argued subsequently, that we can satisfactorily describe the observed SP of carriers in the CdSe QDs, as well as the carrier dynamics observed in this system, in terms of the Zeeman splitting of the CdSe QD ground state.

$Zn_{1-x}Mn_xSe$ ($x=0$, 0.05, and 0.15) layers were deposited on (100) GaAs substrates by molecular-beam epitaxy, and self-assembled CdSe QDs were subsequently formed by depositing a nominal thickness of 2.0 monolayers of CdSe. The QDs were then capped with 50-nm-thick $Zn_{1-x}Mn_xSe$ layers. We will refer to these structures as the $CdSe/Zn_{1-x}Mn_xSe$ system. The samples were mounted in an optical cryostat equipped with a superconducting magnet. The excitation source for time-integrated and time-resolved PL was a frequency doubled Ti:sapphire laser at a wavelength of around 390 nm. Time-resolved PL measurements were carried out with a streak camera. The incident laser beam was linearly polarized and a quarter-wave plate was

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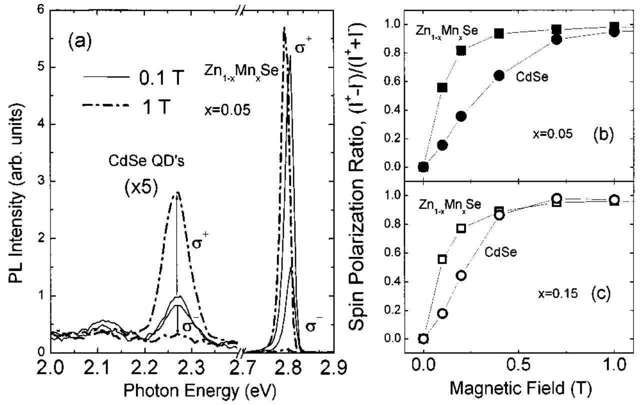

FIG. 1. (a) PL spectra of CdSe/$Zn_{1-x}Mn_xSe$ ($x=0.05$) for two opposite circular polarizations at 5 K for $B=0.1$ T (solid) and 1 T (dash-dot curves). The SP ratios as a function of magnetic field for PL from CdSe and from $Zn_{1-x}Mn_xSe$ are compared for $x=0.05$ and for $x=0.15$ in (b) and (c), respectively.

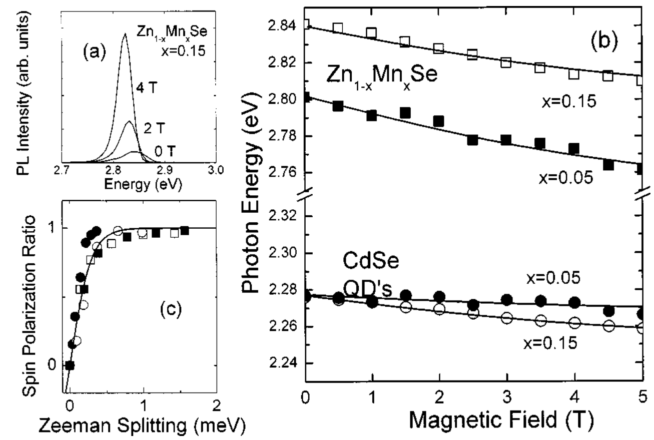

FIG. 2. (a) PL spectra of ZnMnSe exciton emission for various magnetic fields. (b) PL peak energy for $Zn_{1-x}Mn_xSe$ and CdSe as a function of magnetic field at 5 K. The continuous curves represent theoretical Brillouin function fits, as discussed in the text. (c) SP ratio plotted as a function of the Zeeman splitting energy.

placed in front of the streak camera in order to measure the degree of SP.

Figure 1(a) shows the PL spectra of the CdSe/$Zn_{1-x}Mn_xSe$ ($x=0.05$) sample for two opposite circular polarizations, σ^+ and σ^-, at $T=5$ K at external magnetic fields of 0.1 T and 1 T. Peaks at around 2.15, 2.3, and 2.8 eV are the PL peaks associated with intra-Mn transitions, quantum-confined ground state transition in the CdSe QDs, and $Zn_{1-x}Mn_xSe$ excitons, respectively. Both the PL from $Zn_{1-x}Mn_xSe$ and from CdSe QDs exhibit a strong circular σ^+ polarization at 1 T (dash-dot curves), but at 0.1 T (solid curves) the SP effect for CdSe QDs is significantly less than that for $Zn_{1-x}Mn_xSe$. In Figs. 1(b) and 1(c), we show the SP ratio of CdSe QDs (circles) and $Zn_{1-x}Mn_xSe$ (squares) in CdSe/$Zn_{1-x}Mn_xSe$ samples for $x=0.05$ and 0.15, respectively, obtained from the circular polarization ratio $(I^+-I^-)/(I^++I^-)$ of the PL, where I^+ and I^- are the PL intensities for the two opposite circular polarizations. It is clear that the SP ratio of CdSe QDs increases more slowly with increasing magnetic field than that of $Zn_{1-x}Mn_xSe$.

Since we excite the system above the bandgap of ZnMnSe, most photocarriers are initially generated in ZnMnSe. As already noted, the relatively high SP ratio in CdSe QDs may be either due to the transfer of the spin-polarized carriers from ZnMnSe into CdSe QDs, or due to the spin splitting of the carriers in CdSe QDs themselves. The smaller polarization ratio of CdSe QDs compared to ZnMnSe at low fields implies that at least part of the spin information is lost before the carrier recombination occurs in CdSe QDs.

The spin splitting energies for excitons in ZnMnSe and for the quantum-confined ground state in CdSe QDs can be obtained from the PL redshift, as seen in Fig. 2(a). Figure 2(b) shows the change of the PL peak energy for the two samples at 5 K. Ideally, the Mn ions are expected to be located outside of the CdSe QDs, but the carrier wave functions in CdSe QDs penetrate into the ZnMnSe barriers and experience a fairly large spin splitting. At 1 T, the redshift is a few meV, significantly larger than the thermal energy of the carriers at 5 K. The relatively large redshift of the PL peak in CdSe QDs may thus suggest that the large SP ratio observed in CdSe QDs arises primarily from the spin splitting of the carrier states within the CdSe QDs, rather than from the transfer of spin-polarized carriers from ZnMnSe.

The Zeeman splitting of the bandgap ΔE_g in Mn-based magnetic semiconductors is a function of external magnetic field H and temperature T, and can be expressed as[1,12,13]

$$\Delta E_g=\frac{5}{2}\gamma\overline{x}(\alpha-\beta)N_0B_{5/2}\left[\frac{g_{\mathrm{Mn}}\mu_{\mathrm{B}}H}{k_{\mathrm{B}}(T+T_{\mathrm{AF}})}\right],$$

where α and β are the sp-d exchange integrals for the conduction and valence-band electrons, respectively, N_0 is the density of cations, $B_{5/2}$ is the Brillouin function B_J for $J=5/2$, and g_{Mn}, μ_{B}, k_{B} have their usual meanings. The effect of weak antiferromagnetic (AF) interaction between Mn ions is empirically taken into account by replacing x by $\overline{x}$, the effective Mn concentration ($<x$), and by replacing T by $T+T_{\mathrm{AF}}$ ($T_{\mathrm{AF}}>0$). $\gamma(<1)$ indicates the fraction of overlap of the QD wave function into its magnetic barriers. Theoretical fitting curves shown in Fig. 2(b) were obtained with $\overline{x}=0.037$ and 0.027 for $Zn_{1-x}Mn_xSe$ for $x=0.05$ and 0.15, respectively, and $\gamma\overline{x}=0.007$ and 0.019 for CdSe/$Zn_{1-x}Mn_xSe$ for $x=0.05$ and $x=0.15$, respectively.[14] Note that the value of γ has to be as large as 0.7 for $x=0.15$ in order to explain the Zeeman splitting solely by the wave-function overlap. The unusually large value of γ may, in addition to wave-function leakage, be due to some interdiffusion of Mn ions into the CdSe QD interior, especially for $x=0.15$.

Figure 2(c) shows the SP ratio as a function of Zeeman splitting extracted from Figs. 1(b) and 1(c). Reasonable agreement of the curves for ZnMnSe and QDs indicates that SP is due mainly to the Zeeman splitting of the internal QDs. As we will discuss later, injection of spin-polarized carriers from ZnMnSe is not the dominant mechanism for SP in QDs.

Figure 3 shows the decay of PL from ZnMnSe for the two CdSe/$Zn_{1-x}Mn_xSe$ samples. Also shown for comparison is the PL decay for $Zn_{1-x}Mn_xSe$ ($x=0.05$) epilayer without the CdSe QDs. The carrier lifetime with CdSe QDs is clearly shorter, probably due to the efficient transfer of $Zn_{1-x}Mn_xSe$ carriers into the QDs. In the presence of an applied magnetic field, the carrier lifetime was increased for all three samples due to the suppression of the carrier decay channel into the intra-Mn transitions.[15,16] In the inset, we show for compari-

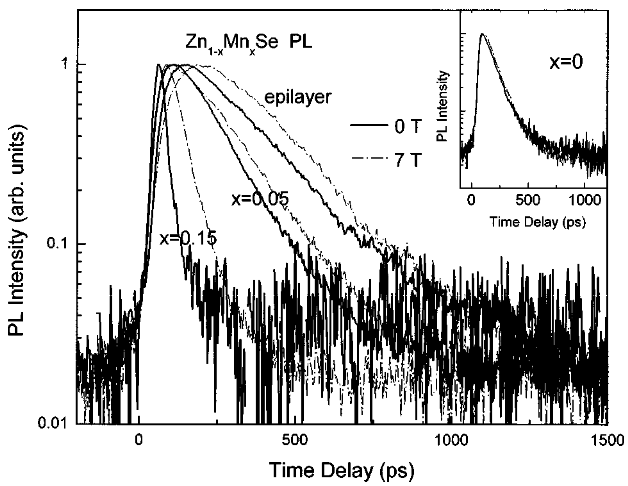

FIG. 3. The PL intensity from ZnMnSe as a function of time delay for $B=0$ (solid) and 7 T (dash-dot curves) for CdSe/$Zn_{1-x}Mn_xSe$ samples as well as for a $Zn_{1-x}Mn_xSe$ "control" epilayer ($x=0.05$) without CdSe QDs. The carrier lifetime appears to be reduced by the presence of the CdSe QDs. The inset shows the PL decay for a nonmagnetic CdSe/ZnSe sample.

son the PL decay from CdSe QDs embedded in ZnSe ($x=0$), where no magnetic field effect is observed.

When we excite the PL above the bandgap of ZnMnSe, photocarriers are initially generated in ZnMnSe and then transfer into the CdSe QDs. The SP ratio for the CdSe QD emission was in this case smaller than that for $Zn_{1-x}Mn_xSe$ at low magnetic fields, as seen in Fig. 1. In order to determine whether some of the carriers in ZnMnSe retain their spin information during the transport into the QDs and affect SP in the QDs, we measured the PL lifetime under external magnetic fields for two circular polarizations by placing a quarter-wave plate in front of a streak camera. In Fig. 4, we show the PL decay for CdSe QDs embedded in $Zn_{1-x}Mn_xSe$ ($x=0$, 0.05 and $x=0.15$) for the σ^+ polarization at 5 K. The carrier lifetime is found to clearly decrease with increasing Mn concentrations, indicating the influence of the Mn ions

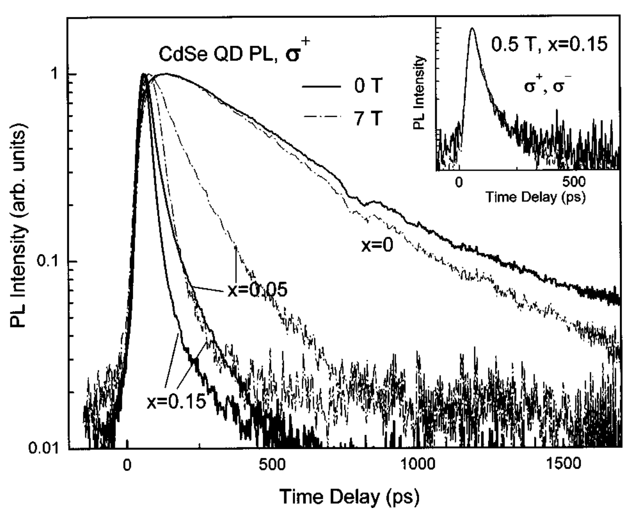

FIG. 4. The decay of CdSe PL intensity for CdSe/$Zn_{1-x}Mn_xSe$ ($x=0$, 0.05, 0.15) samples for the σ^+ polarization at $B=0$ (solid) and 7 T (dash-dot curves). The inset compares the PL traces for σ^+ (solid) and σ^- (dash-dot-curves) polarizations for $x=0.15$ at 0.5 T.

on the carriers in the QDs. In the inset, we show the PL traces of σ^+ and σ^- polarizations for $x=0.15$. Due to the drastic decrease of the PL intensity for the σ^- polarization in high magnetic fields, the PL traces for σ^+ and σ^- polarizations were compared at a relatively low magnetic field of 0.5 T, but then no difference could be detected. This appears to indicate that new spin equilibrium is obtained on a time scale much shorter than the recombination lifetime in QDs either due to spin scattering at the QD interface, or due to fast spin reorientation within the QDs.

In conclusion, the SP ratio for CdSe QDs was found to be clearly less than that for $Zn_{1-x}Mn_xSe$ at low magnetic fields. We are thus led to attribute the observed SP of the CdSe QDs as arising primarily from the Zeeman splitting of QD states either due to Mn interdiffusion or due to the wave-function overlap of the internal QD states with the surrounding magnetic barrier—a process in which the carrier wave function "feels" fewer Mn ions than are actually present in the barrier material. This suggests that the effect of spin-conserving injection of carriers from ZnMnSe barrier in determining the SP in QDs is, by comparison, much weaker than this internal QD process.

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