Magneto-optical studies of ensembles of semimagnetic self-organized Cd(Mn)Se/Zn(Mn)Se quantum dots

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Ensembles of Cd(Mn)Se/Zn(Mn)Se semimagnetic self-organized quantum dots with different Mn content have been studied by photoluminescence and Raman scattering under strong magnetic fields in Faraday and Voigt geometries and with selective excitation. Electron spin-flip transitions have been observed in Voigt geometry in the structures with large Mn content, characterized by a large magnetic shift of photoluminescence bands. Narrow exciton peaks completely $\sigma^-\sigma^+$ polarized have been observed under selective excitation in the Faraday geometry in the structures with medium and small Mn content. A number of effects due to *s*,*p*-*d* exchange interaction and exciton magnetic polaron formation manifested themselves in the structures with the smallest Mn content, where no Zeeman shift of the photoluminescence bands was observed.

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I. INTRODUCTION

Semimagnetic quantum dots (QDs) have attracted attention due to their interesting physical properties and potential for device applications in spintronics.[1] III-V and II-VI systems of different design have been studied: namely, nanocrystals with small radii in organic and glass matrices, characterized by a strong-confinement regime of electrons, and disc-shaped QDs developing during molecular-beam-epitaxy growth of a few monolayers of one semiconductor on another one with a different lattice constant. They are known as self-organized QDs and consist of ensembles of QDs with a large dispersion of sizes and composition, giving broad spectral bands. To overcome this impediment and achieve significantly better spectral resolution in order to study single QDs, different methods providing spatial resolution were used lately, and many interesting results for single QDs were obtained. However, of great interest also are studies of QD ensembles, especially using methods of selective excitation both spectrally and by polarization, which allow one to address separately subensembles of QDs.

In the present work we study magnetoluminescence and resonant Raman scattering processes in strong magnetic fields for a variety of CdMnSe/ZnSe and CdSe/ZnMnSe structures with different nominal thicknesses of QD layers and different concentrations of Mn ions. We have succeeded in observing resonant electron spin-flip (S-F) transitions in semimagnetic QDs and found that they obey the usual Raman S-F selection rules. By measurements with selective excitation both spectrally and by polarization we observed a narrow peak in Faraday geometry and interpreted it as an exciton transition related to a narrow subensemble of quantum dots. We have also drawn attention to the fact that in the structures with very small Mn concentration, when no shift of the photoluminescence (PL) band could be observed in high magnetic field, the presence of Mn could be verified by several specific effects such as PL intensity increase in the magnetic field, magnetic-field-induced circular polarization, and collective multiple Raman scattering at Mn ions. These effects can be regarded as fingerprints for Mn presence in the low-dimensional structures.

II. EXPERIMENT

CdMnSe/ZnSe and CdSe/ZnMnSe samples were grown by molecular-beam epitaxy on GaAs (001) substrates at $T = 280\,^\circ$C. The nominal thickness of the CdSe or CdMnSe layer, where QDs were formed during growth, was 2 or 2.8 monolayers (ML). Well defined ZnCdMnSe disk-shaped QDs possess average lateral dimensions of 10 nm and are extended by 7–10 ML in the growth direction, as has been established by high-resolution transmission electron microscopy on analogous structures without Mn.[2] The average Cd content in the dots was approximately 20%, according to Raman scattering by longitudinal optical (LO) phonons. The main parameters of all structures under study are presented in Table I. The most important property of diluted magnetic structures is giant Zeeman splitting of the conduction and valence bands under applied magnetic fields. The splitting is caused by the *s*,*p*-*d* exchange interaction between band carriers and *d*-shell electrons of the magnetic ion (usually Mn^{2+}). The magnitude of the splitting is defined by the effective concentration of Mn^{2+} ions, x_{eff}. This effect is revealed in the shift of a photoluminescence (PL) band versus magnetic field applied in the Faraday geometry under nonresonant excitation. The values of the shift (ΔE) obtained at the magnetic field 6 T are presented in Table I for all samples. We divide the structures into three main groups according to the Mn concentration. Mn was introduced either into the layers with QDs or into the ZnSe barrier layers. In two samples (#B1 and #C1) ZnMnSe barriers were separated on both sides from the QD layers by thin ZnSe spacers of 1.6 or 1.5 nm thickness, respectively. In this case only the tail of the exciton wave function reaches the magnetic layer, and the effect of *s*,*p*-*d* exchange interaction is decreased. It is equivalent to the decrease of actual Mn concentration by an order of magnitude, as can be seen from comparison of magnetic properties in samples #B1 and #B2, as well as #C1 and #C2. The structures were not intentionally doped but had a residual conductivity of *n* type.

The structure was cooled by pumped He vapor in a cryostat with a superconducting solenoid. Magnetic fields up to 6.5 T could be applied in the Faraday (B along the growth axis) or in the Voigt (B parallel to the sample plane) geometries. PL and

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TABLE I. Structures, composition, nominal thickness d of the QD layer, nominal Mn concentration, position of the PL maximum, and PL shift at a magnetic field of 6 T.

Group	No.	Composition	d of CdSe layer	PL maximum at $B = 0$ T (eV)	PL shift at $B = 6$ T (meV)
A	A1	CdMnSe/ZnSe 15% Mn	2 ML	2.51	38
	A2	CdSe/ZnMnSe 15% Mn	2 ML	2.48	44
	A3	CdSe/ZnMnSe 7%	2.8ML	2.38	35
B	B1	CdSe/ZnSe/ZnMnSe 11% Mn ZnSe spacer 1.6 nm	2 ML	2.509	10
	B2	CdSe/ZnMnSe 1% Mn	2 ML	2.57	7
C	C1	CdSe/ZnSe/ZnMnSe 11%Mn ZnSe spacer 1.5 nm	2.8 ML	2.33	No shift
	C2	CdSe/ZnMnSe 1% Mn	2.8 ML	2.33	No shift

Raman spectra were recorded in the back-scattering geometry with the propagation direction of incident and emitted light normal to the sample plane. Spectra were excited by Ar+ laser lines and a 532 nm solid-state laser, and were dispersed by a double grating spectrometer (dispersion 0.5 nm/mm) with photon counting electronics. Sample #A2 was also measured using a Dilor spectrometer with a triple monochromator and a CCD camera. Linear prism polarizers and quarter-wave plates were used for polarization measurements.

III. RESULTS AND DISCUSSION

A. Electron spin-flip Raman scattering

Resonant electron spin-flip Raman scattering was observed for group A structures with the largest concentration of Mn. Let us first discuss the magnetic shift of PL bands of these structures in applied magnetic fields. As an example we show in the inset to Fig. 1 PL spectra of #A1, measured in the Faraday geometry at $B = 0$ and 6 T, excited nonresonanly at 2.6 eV with $\sigma^+\sigma^+$ circular polarization. In the spectrum, a broad PL band is superimposed with narrow lines attributed to resonant Raman scattering by LO phonons. The large width of the PL band is an intrinsic feature of the QD ensembles due to the dispersion of QD sizes as well as the Cd and Mn content in the dots. In Fig. 1 the shift of the PL band versus magnetic field is presented. It amounts to 38 meV at $B = 6$ T. The maximal error of determination of the PL band energy is not larger than 2.5 meV, as is seen from the data scattering.

The PL band can be due to the recombination of the heavy excitons and/or negatively charged trions in the singlet state because CdSe/ZnSe-type structures have a residual n-type conductivity. The shift of the heavy exciton or trion PL band is given by the expression[3]

$$\Delta E = \Delta E_c + \Delta E_v = -\tfrac{1}{2}\left[x_{\text{eff}} N_0(\alpha - \beta)\left\langle S_z^{Mn}\right\rangle\right], \qquad (1)$$

where x_{eff} is the effective concentration of Mn^{2+} ions that take part in the exchange interaction with the carriers, αN_0 and βN_0 are the exchange integrals for electrons and holes, respectively, and $\langle S_z^{Mn}\rangle$ is the thermal average of the Mn spin projection along $\boldsymbol{B}$ and is given by the modified Brillouin function B_S for $S = 5/2$,

$$\left\langle S_z^{\text{Mn}}\right\rangle = -(5/2)B_{5/2}\left[\frac{5\mu_B B}{k_B(T + T_{\text{AF}})}\right]. \qquad (2)$$

The effective concentration of Mn^{2+} is less than nominal one due to antiferromagnetic pairing of Mn^{2+} ions. Parameter T_{AF} also reflects the Mn^{2+} pairing. There exists also a possibility that an exciton magnetic polaron (EMP) may be formed. An EMP is the region of local magnetization of the Mn^{2+} ions in the exchange field of the heavy hole.[4] In order to take into account the possible formation of EMP one has to realize that PL shift in this case occurs in the combined magnetic field $B_\Sigma = B + B_{\text{EMP}}$, where B_{EMP} is the exchange field of the EMP.

Let us approximate the PL shift in Fig. 1 with Eqs. (1) and (2), where $B \equiv B_\Sigma$, and introduce four fitting parameters, as was, for instance, done in Ref. 5: $P1 = 1.25x_{\text{eff}}(\alpha - \beta)N_0$, $P2 = T + T_{\text{AF}}$, $P_3 = B_{\text{EMP}}$, and $P_4 = E_0$, where E_0 is the energy of the PL maximum at $B_\Sigma = 0$. The difference

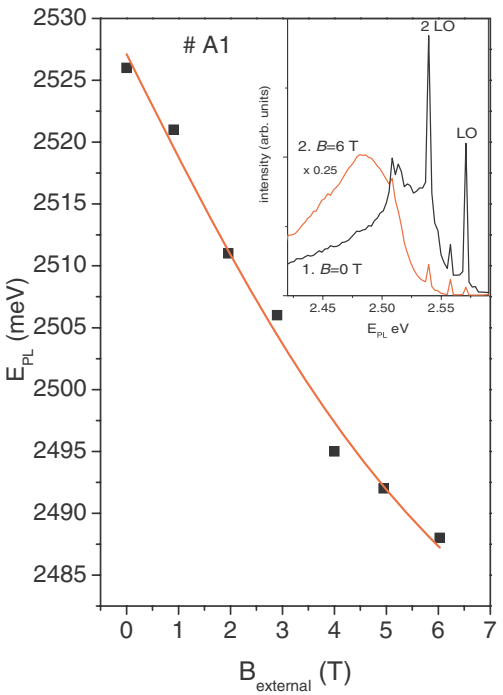

FIG. 1. (Color online) #A1. PL shift versus B. Faraday geometry, $E_{\text{exc}} = 2.6$ eV. The inset shows PL spectra in Faraday geometry at $B = 0$ and 6 T, $E_{\text{exc}} = 2.6$ eV, and $\sigma^+\sigma^+$ circular polarization.

energy $E_{\mathrm{EMP}} = E_0 - E_{B=0}$ is the so-called energy of the exciton magnetic polaron. As the result of the fitting we find $P1 = 64.27$ meV, $B_{\mathrm{EMP}} \cong 0$ T, $T + T_{AF} = 12$ K, and $E_0 = 2528$ meV. Thus, in this structure the EMP does not form before exciton recombination because evidently its formation time is larger than the exciton lifetime. Let us remark that in structures #A2 and #A3 the fitting of the PL shift also shows that $B_{\mathrm{EMP}} \cong 0$ T. In CdMnSe-type structures the energy of the PL maximum is larger than the inner transition of the Mn ion from the $^6A_1$ to the $^4T_1$ state. There is an effective Auger recombination of the exciton which leads to a decrease of its lifetime.

In the Voigt geometry the PL shift in the magnetic field is significantly less than in the Faraday one. The difference in the shifts is related to the magnetic anisotropy of the valence band and was studied earlier in quantum wells.[6,7] It is due to the fact that in the Voigt geometry magnetic moment of the heavy hole is mainly directed along the growth axis z, i.e., perpendicular to the magnetic field, and thus the heavy hole band is not subject to the Zeeman splitting.

If in the Voigt geometry the excitation laser energy is in the region of the PL band (resonant excitation), we observe new peaks in the spectrum in crossed linear polarizations $\sigma\pi$ (the excitation polarization σ is normal to the magnetic field and the detection polarization π is along it). Most extensively these peaks were studied in the #A1 structure and are shown in Fig. 2. Note that in Fig. 2 the usual Raman representation is used: Raman shift, i.e., the difference between the laser energy and the detected energy, is shown on the x axis. The spectra in Figs. 2(a) and 2(b) relate to excitation energies of 2.54 and 2.497 eV, respectively, and are shown for the magnetic field 3 T. Let us focus here on the broad peak designated as S-F. One can conclude that this peak is due to Raman scattering process because it has the same value of Raman shift under different

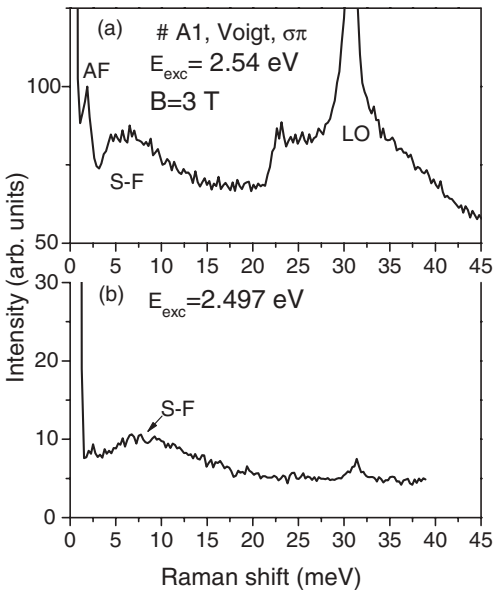

FIG. 2. #A1. Electron S-F peak at $B = 3$ T, Voigt geometry, and crossed $\sigma\pi$ linear polarization. (a) $E_{\mathrm{exc}} = 2.54$ eV and (b) $E_{\mathrm{exc}} = 2.497$ eV. "LO" denotes the peak of Raman scattering by longitudinal optical phonon of ZnSe. "AF" is Raman scattering by antiferromagnetic Mn-Mn pairs.

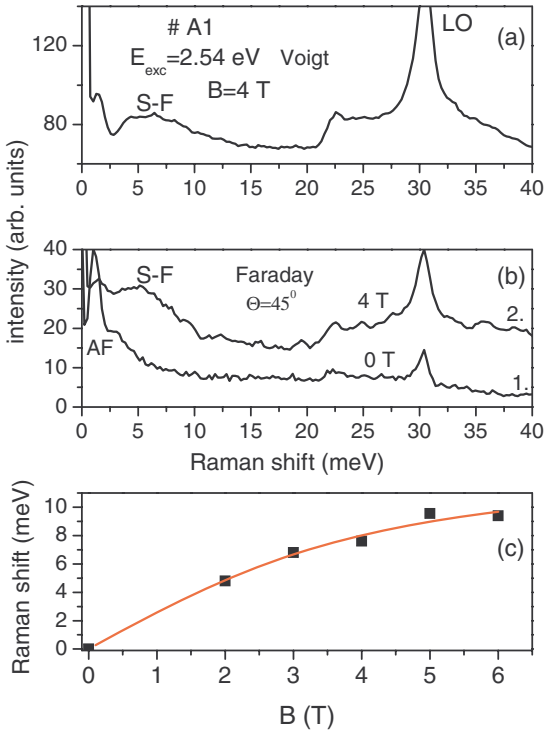

FIG. 3. (Color online) #A1. Spectra at $B = 4$ T and $E_{\mathrm{exc}} = 2.54$ eV. (a) Voigt geometry with crossed linear polarizations $\sigma\pi$. (b) Faraday geometry in a tilted magnetic field $\Theta = \angle(Bz) = 45°$, $\sigma\pi$ polarization: 1. $B = 0$ T, 2. $B = 4$ T. (c) Raman shift of the S-F peak vs B.

excitation energies. The peak is observed only under magnetic field in the Voigt geometry with crossed linear polarizations of the exciting and scattered light. The peak does not appear in the Faraday geometry. The Raman shift of this peak increases with the increase of the applied magnetic field according to the behavior of the Brillouin function, as can be seen in Fig. 3(c). The polarization selection rules are indeed those of the spin-flip Raman transitions in back-scattering configuration $(e_{\mathrm{i}} \times e_{\mathrm{s}}) \times e_{\mathrm{B}} \neq 0$, where e_{i}, e_{s}, and e_B are the polarization vectors of incident and scattered light and the unit vector in the direction of the magnetic field, respectively.[8] The S-F peak was also observed with the same Raman shift in Faraday geometry under a tilted magnetic field, as is shown in Fig. 3(b) for $\angle(Bz) = 45°$ as compared to Fig. 3(a). This means that it has an isotropic g factor, as is appropriate for electronic spin-flip transitions. All these observations corroborate the assignment of the S-F peak as being due to Raman electronic spin-flip scattering between the $\pm 1/2$ spin sublevels of the conduction band or a donor-level split in the applied magnetic field. The dependence of the Raman shift on the magnetic field can be approximated by the well known equation[9] describing the Zeeman splitting of the conduction band in diluted magnetic semiconductors:

$$\delta E_c = x_{\mathrm{eff}} \alpha N_0 \langle S_z^{Mn} \rangle. \quad (3)$$

The solid line in Fig. 3(c) is drawn according to Eq. (3).

Electronic spin-flip resonant Raman scattering was also observed in the structures #A2 (Fig. 4) and #A3 (not shown).

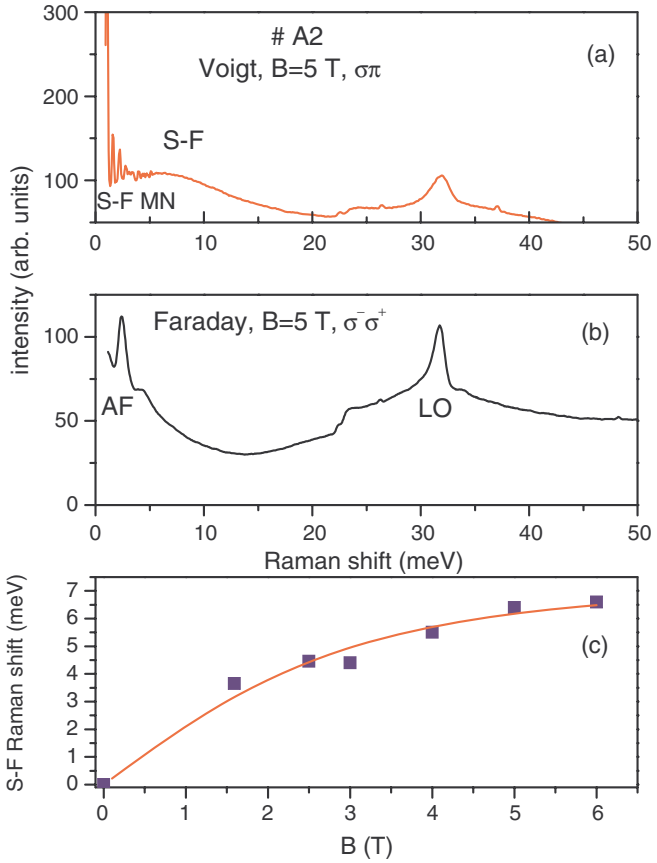

FIG. 4. (Color online) #A2. Spectra under $E_{exc} = 2.497$ eV. (a) Voigt geometry at $B = 5$ T, $\sigma\pi$ polarization. (b) Faraday geometry at $B = 5$ T, $\sigma^-\sigma^+$ polarization. (c) Raman shift of the S-F peak vs B. "S-F Mn" denotes the peaks originating from multiple spin-flip scattering in the Mn ion (see text).

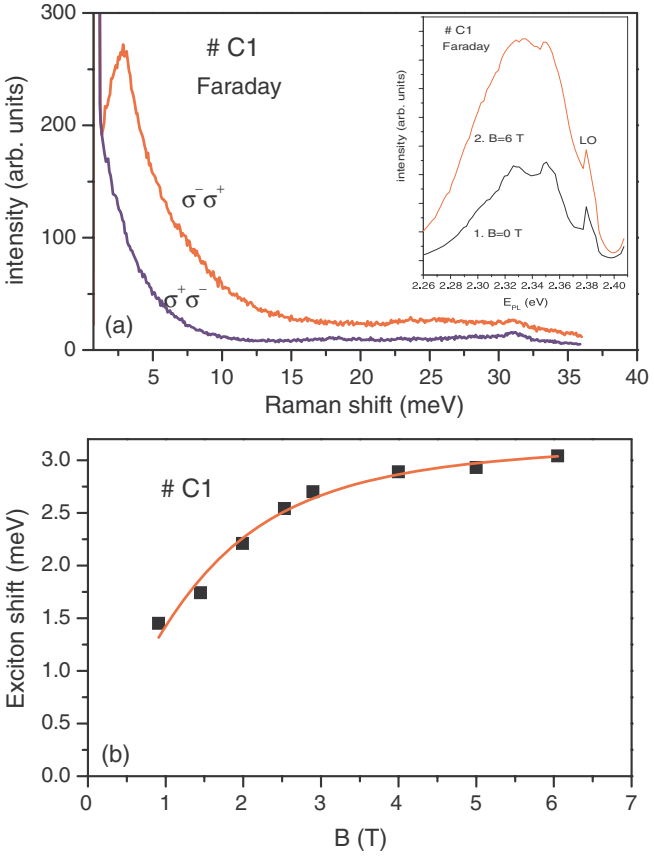

FIG. 5. (Color online) #C1. (a) Selectively excited exciton peak in Faraday geometry. $E_{exc} = 2.331$ eV, $B = 3$ T, and polarizations $\sigma^-\sigma^+$ and $\sigma^+\sigma^-$. (b). Raman shift of the $\sigma^-\sigma^+$ peak vs magnetic field. In the inset, PL spectra at $B = 0$ T (1) and 6 T (2) under nonresonant excitation. Faraday geometry, $E_{exc} = 2.41$ eV, and $\sigma^+\sigma^+$ polarization.

We did not observe the electronic spin-flip peaks in group B and C structures, which have smaller Mn content. This may be explained by the fact that due to the relatively small Mn content the splitting of the conduction band is small and the position of the S-F peak is close to the exciting laser energy, and may be obscured by the scattered laser light.

Let us remark that the electron spin-flip Raman scattering is an important effect that provides directly the magnetic splitting of the conduction band, from which the electron g factor can be obtained. S-F scattering has been observed earlier in semimagnetic bulk and quantum well samples but never, to our knowledge, in semimagnetic QDs. One could assume that for zero-dimensional structures Raman selection rules are relaxed, but we have shown that in the studied self-organized QDs they obey the usual selection rules for S-F Raman scattering.

The narrow peaks in Figs. 2–4 are related to magnetic Raman transitions in the system of Mn ions, which we have observed in our earlier work.[10] The narrow peak, designated as AF, does not shift with B and is due to Raman spin-flip scattering in antiferromagnetic Mn^{2+} pairs, originating from the transition $(S = S1 + S2 = 0, m = 0) \rightarrow (S = 1, m = 0)$. From the position of this peak the antiferromagnetic exchange constant $J_{NN} = -1.053$ meV was evaluated. The intensity of the AF peak decreases as B increases. Several equally spaced narrow lines best observed in Fig. 4(a) at $B = 5$ T are due to multiple spin-flip scattering within a Zeeman split Mn^{2+} ground state.[10] The theory of this collective Raman process was given in Ref. 11.

B. Excitation-selective exciton transitions related to a narrow subensemble of quantum dots

We have observed a rather narrow emission peak in the Faraday geometry under resonant excitation close to the maximum of the PL band. This peak is completely $\sigma^-\sigma^+$ circularly polarized as shown in Fig. 5(a) for the structure #C1. No peak was observed in $\sigma^+\sigma^-$ polarization. The FWHM $\cong 5$ meV at $B = 3$ T of this peak is more than an order of magnitude smaller than that of the PL band under nonresonant excitation [shown in the inset to Fig. 5(a) under $E_{exc} = 2.41$ eV]. In fact, the band width is comparable with the values observed for single semimagnetic QDs, where it is determined by magnetic fluctuations.[12] The FWHM increases with B. In our earlier work[10] a similar peak was observed in one of the samples and interpreted as forbidden electronic spin-flip Raman scattering. We thought that this transition could be observed in the forbidden Faraday geometry due

to relaxation of the selection rules in QDs. But at present we instead consider another interpretation supported by the results obtained in the present paper for a number of structures. In Sec. I we described electronic S-F Raman scattering. The peaks were observed only in the Voigt geometry in crossed linear polarizations of the incident and scattered light in accordance with the usual S-F Raman scattering selection rules. No relaxation of the selection rules in QDs was found. Therefore we think that the selectively excited $\sigma^-\sigma^+$-polarized peak is an exciton transition from the lowest $J = +1$ magnetic sublevel of the exciton that was excited selectively to the $J = -1$ level. Its polarization is in accordance with semimagnetic structures where the ground exciton level has $+1$ angular moment.

The magnetic shift of this peak gives directly the Zeeman splitting of the exciton. The shift is shown in Fig. 5(b). It has a Brillouin-like behavior from which the effective concentration of Mn of about 0.5% was found. Similar selectively excited exciton transitions were also observed in the structures #B1, #B2, and #C2. By such selective excitation (both spectrally and by polarization) one can efficiently probe narrow subensembles from the largely inhomogeneous ensemble distribution of QDs.

Large Zeeman splitting of the conduction and valence bands in semimagnetic structures is always regarded as their most important property, and is revealed as an exciton shift in the applied magntic field in the Faraday geometry. However, when the concentration of Mn is very small there may be no shift even in strong magnetic fields, as was the case for structures #C1 and #C2 (see the inset to Fig. 5).

In spite of this, a number of effects due to the presence of Mn, *s,p-d* exchange interaction, and excitonic magnetic polaron formation manifest themselves in these structures. They include:

(i) Strong increase of the PL intensity in an applied magnetic field, especially in the Faraday geometry. This effect is due to effective Auger recombination of the exciton with excitation of an electron from the $^6A_1$ to the $^4T_1$ state in the *d* shell of the Mn ion. Under increasing magnetic field this process gets suppressed due to selection rules and therefore exciton intensity increases strongly.[13]

(ii) Resonant multiple spin-flip Raman scattering in the Voigt geometry of the magnetically split ground state of the Mn ion.

(iii) Magnetic-field-induced circular polarization, ρ_m. This effect is due to both Zeeman splitting of the conduction and valence bands and the process of exciton magnetic polaron formation. As an example we present in Fig. 6 for sample #C2 the measurement of circular polarization under linear excitation at $E = 2.33$ eV (see the PL spectrum in the inset). The solid curve represents a calculation performed in the model of the thermodynamic magnetic fluctuations in the system of Mn spins.[14] From the fitting to experimental results for ρ_m we get polaron parameters $E_{EMP} = 1$ meV, $B_{EMP} = 1.82$ T, $(dE_z/dB)_{B=0} = 1.1$ meV/T, and $T = 7$ K. To achieve the fitting at large values of B we had to use as a saturation value $\rho_m = 0.65$.

Let us note that these specific effects (i), (ii), and (iii) can be regarded as fingerprints for the presence of Mn ions in low-dimensional structures.

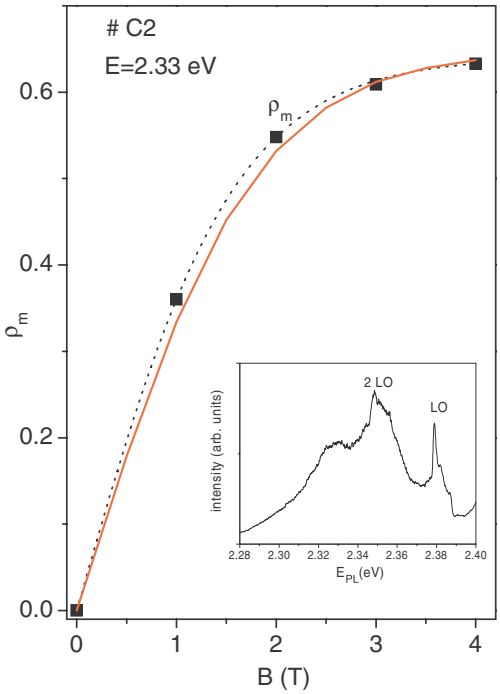

FIG. 6. (Color online) #C2. Dependence of the degree of circular polarization on the magnetic field under linearly polarized excitation, $E_{exc} = 2.41$ eV and $E = 2.33$ eV. Dots represent experimental data, dotted curves serve to guide the eye, and the solid curve is the calculation (see text). In the inset: PL spectrum in Faraday geometry, $E_{exc} = 2.41$ eV, polarization $\sigma^+\sigma^+$, and $B = 0$ T.

IV. CONCLUSION

We have studied PL and Raman scattering spectra in strong magnetic fields for ensembles of Cd(Mn)Se/Zn(Mn)Se semimagnetic self-organized QDs. A number of structures with different concentrations of Mn ions and different designs were studied. We have observed strongly polarized PL of heavy excitons localized in the quantum dots. These excitons or exciton magnetic polarons form also intermediate states for resonant Raman transitions by magnetic excitations of various types.

We have observed in semimagnetic QDs electronic spin-flip Raman transitions in the Voigt geometry, and have shown that they obey the usual spin-flip selection rules.

Narrow subensemble of semimagnetic QD distribution was probed as exciton transition under selective excitation in the Faraday geometry both spectrally and by polarization. Its Brillouin-like shift in the magnetic field gives directly the splitting of the exciton magnetic levels.

In the structures with the lowest Mn content, where no magnetic shift of the PL band was detected under nonresonant excitation, we have observed a number of specific effects directly related to the presence of Mn ions, *s,p-d* exchange interactions, and excitonic magnetic polaron formation, such as an increase both of PL intensity and magnetic-field-induced circular polarization, as well as multiple Raman spin-flip transitions within the Mn ion.

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