

Supporting Information:

Optically Detected Magnetic Resonance in CdSe/CdMnS Nanoplatelets

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S1. Experimental data for ODMR in samples #2, #3 and #4

Here we show additional experimental data on ODMR in the samples #2, #3 and #4. They are presented in the same form as for sample #1 in the main text.

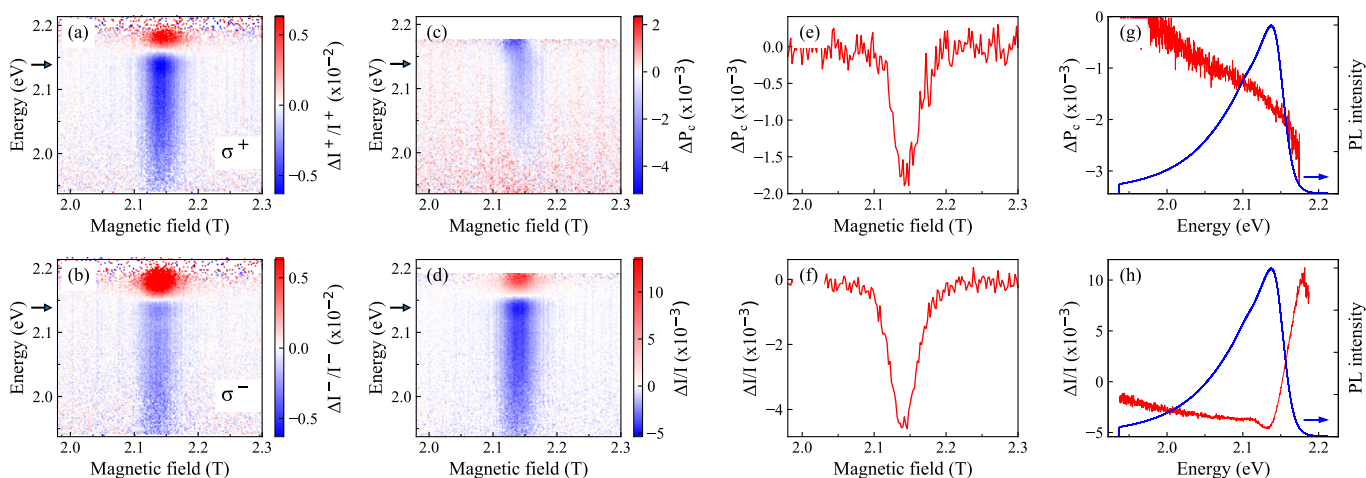


FIG. S1. (a) and (b) Microwave induced variation of σ^- and σ^+ components of the PL intensity in sample #2 as a function of magnetic field. $T = 1.8$ K, microwave frequency $\nu = 59.6$ GHz. (c) Microwave induced change of degree of circular polarization (ΔP_c). (d) Microwave induced change of total PL intensity. (e) ΔP_c as a function of magnetic field at the PL maximum of 2.139 eV. (f) ODMR detected through the total PL intensity at 2.139 eV. Spectral dependence of ODMR signals on the degree of circular polarization (g) and total PL intensity (h) at resonance field $B_R = 2.139$ T.

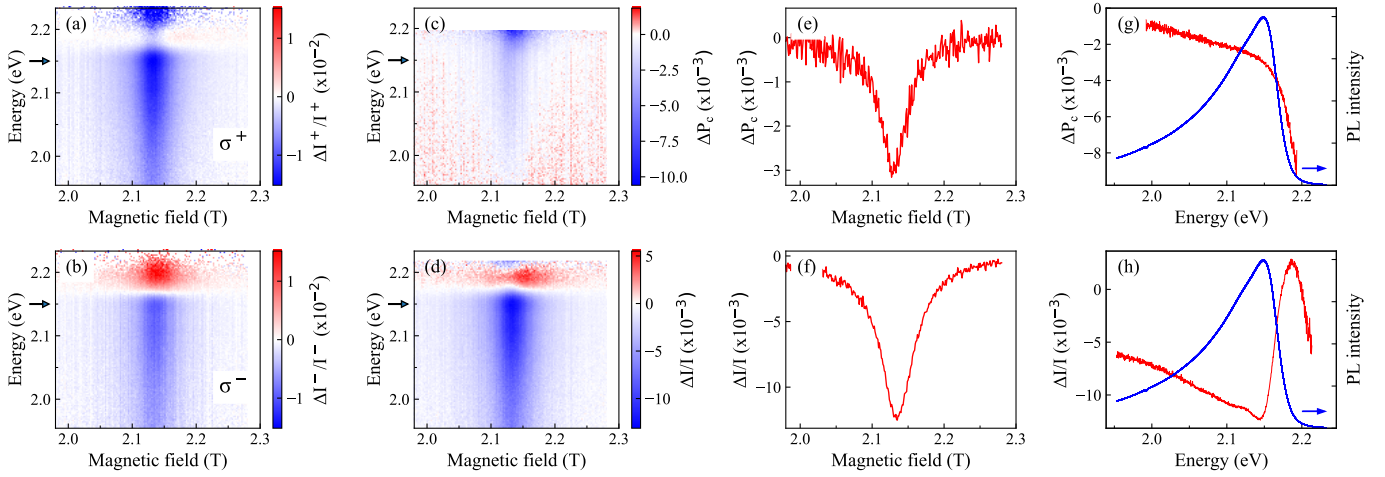


FIG. S2. (a) and (b) Microwave induced variation of σ^- and σ^+ components of the PL intensity in sample #3 as a function of magnetic field. $T = 1.8$ K, microwave frequency $\nu = 59.6$ GHz. (c) Microwave induced change of degree of circular polarization. (ΔP_c) (d) Microwave induced change of total PL intensity. (e) ΔP_c as a function of magnetic field at the PL maximum of 2.150 eV. (f) ODMR detected through the total intensity at 2.150 eV. Spectral dependence of ODMR signals on the degree of circular polarization (g) and total PL intensity (h) at resonance field $B_R = 2.133$ T.

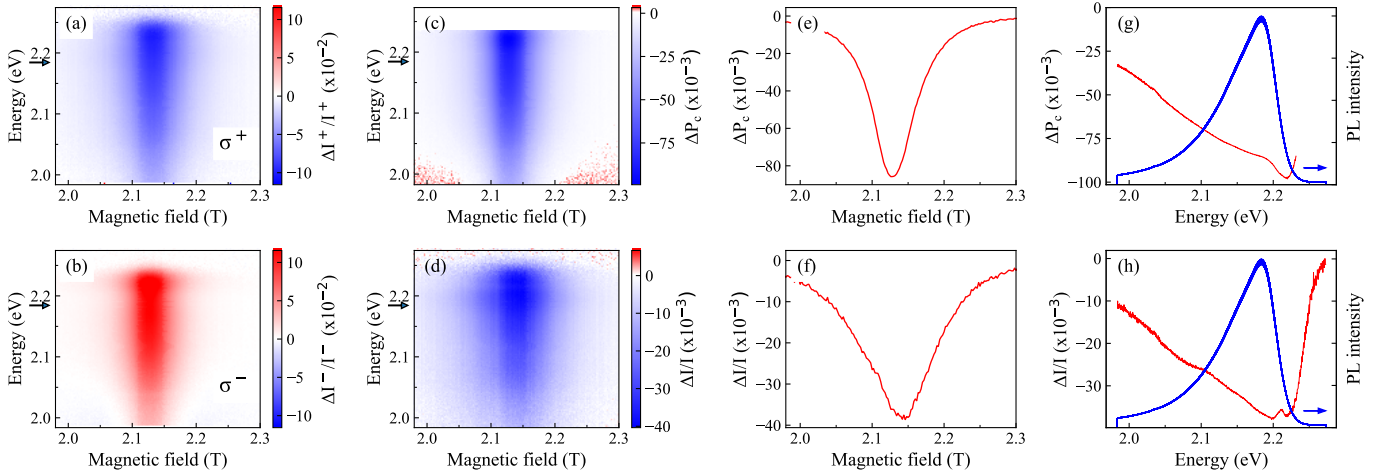


FIG. S3. (a) and (b) Microwave induced variation of σ^- and σ^+ components of the PL intensity in CdSe/Cd_{0.89}Zn_{0.1}Mn_{0.01}S nanoplatelets (sample #4) as a function of magnetic field. $T = 1.8$ K, microwave frequency $\nu = 59.6$ GHz. (c) Microwave induced change of degree of circular polarization. (ΔP_c) (d) Microwave induced change of total PL intensity. (e) ΔP_c as a function of magnetic field at the PL maximum of 2.184 eV. (f) ODMR via total intensity at 2.184 eV. Spectral dependence of ODMR signals on the degree of circular polarization (g) and total PL intensity (h) at resonance field $B_R = 2.132$ T.

S2. Experimental data for ODMR in $\text{Zn}_{1-x}\text{Mn}_x\text{Se}/(\text{Zn,Be})\text{Se}$ quantum wells

ODMR studies of $\text{Zn}_{1-x}\text{Mn}_x\text{Se}/(\text{Zn,Be})\text{Se}$ quantum wells were reported in Ref. [36], where the sample with $x = 0.012$ was studied in detail. Various mechanisms of ODMR detection were identified. In Figure S4 we show resonance profile from this paper for the sample with $x = 0.012$ and for two more samples with $x = 0.004$ and 0.02 . Their parameters are the following:

$x = 0.004$, FWHM = 38 mT, $\Gamma = 5$ mT

$x = 0.012$, FWHM = 38 mT, $\Gamma = 21$ mT

$x = 0.02$, FWHM = 40 mT, $\Gamma = 28$ mT

Values for Γ are given in Fig. 3(d) by open squares.

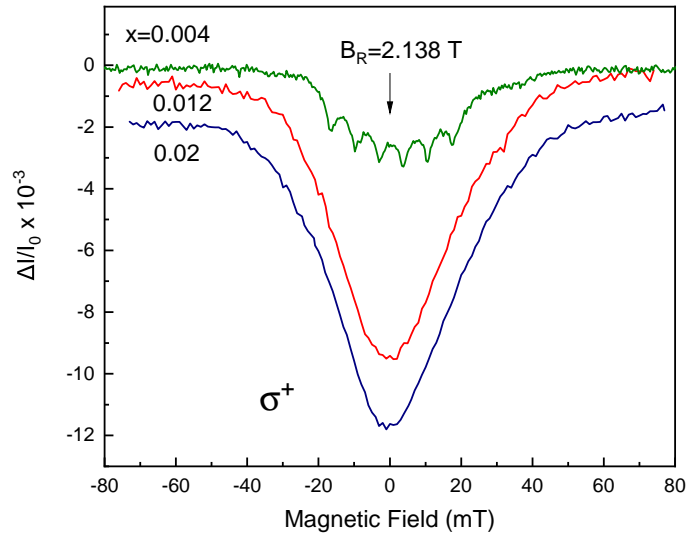


FIG. S4. ODMR resonances in $\text{Zn}_{1-x}\text{Mn}_x\text{Se}/(\text{Zn,Be})\text{Se}$ quantum wells recorded via the degree of circular polarization. In sample with Mn concentration $x = 0.004$ resolved fine structure can be observed (green curve). In samples with $x = 0.012$ (red curve) and $x = 0.02$ (blue curve) only broad line can be observed. ODMR were recorded at $T = 1.8$ K with $\nu = 60.02$ GHz microwave frequency.

S3. Sample preparation

Chemicals: Cadmium acetate dihydrate ($\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$), trioctylamine (TOA), oleylamine (OLA), N-methylformamide (NMF), ammonium sulfide solution (40 – 48 wt. % in water), trioctylphosphine (TOP), oleic acid (OA), hexane, acetonitrile, toluene and manganese(II) acetate were bought from Sigma-Aldrich.

Synthesis of 2 ML CdSe Nanoplatelets: The synthesis of the NPLs was performed according to a previously reported method. [4] The mixture of 860 mg $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$, 1 mL of OA, and 60 mL of TOA was degassed for 1 hour at room temperature. Then, it was heated to 115°C under argon flow. When the temperature reached 115°C , 1 mL of 1M TOP-Se was injected swiftly and the mixture was kept at 115°C for 2 hours. After that, the solution was cooled down to room temperature and centrifuged after addition of ethanol and hexane. The precipitated NPLs were dispersed in hexane.

Synthesis of CdSe/(Cd,Mn)S core/shell NPLs: Here we used a modified procedure of the c-ALD recipe reported previously. [37] 2 ML CdSe NPLs were dispersed in 1 mL hexane and 5 mL of NMF with 40 μL of 40 – 48% aqueous solution of ammonium sulfide – as sulfur shell growth precursor – was added on top of the NPL dispersion and stirred for 2 min. Then, the reaction was stopped by addition of acetonitrile and excess toluene and the mixture was precipitated via centrifugation. The precipitate was redispersed in NMF and precipitated again after addition of acetonitrile and toluene to remove the unreacted precursor. Finally, the NPLs were dispersed in 4 mL of NMF. The cation precursor solution consists of $\text{Mn}(\text{OAc})_2$ and $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ in NMF. For the cation deposition step, 1 mL of cation precursor mixture was added to the NPL dispersion and it was stirred for 45 min in a nitrogen filled glovebox. Then, the reaction was stopped by addition of excess toluene and the mixture was precipitated via centrifugation and dispersed in NMF. The same cleaning step was repeated twice more to get rid of the excess precursors. To increase the number of shells, the steps explained above were repeated until the desired shell thickness was achieved. Lastly, 5 mL of hexane and 100 μL of OLA were added on top of the precipitated NPLs after achievement of the desired shell thickness and the mixture was stirred overnight. To get rid of excess ligands, the dispersion of NPLs was precipitated by addition of ethanol, redispersed and kept in hexane for further usage. The doping levels were obtained using ICP-MS measurements and by taking into account the 2D planar geometry of the NPLs.

Synthesis of CdSe/(Cd,Zn,Mn)S core/shell NPLs: We used a slightly modified c-ALD recipe for the growth of (Cd,Zn,Mn)S shell layer. [37] For the growth of the first sulfur layer, 40 μL of ammonium sulfide was added to the 2 ML CdSe NPLs in 1 mL hexane and 5 mL of NMF and stirred for 2 min. Then, the NPLs was precipitated by centrifugation after the addition of acetonitrile and toluene. The precipitated NPLs were redispersed in NMF and this cleaning steps were repeated twice more for the complete removal of unreacted precursor. Then, 1 ML of sulfur coated 2 ML CdSe NPLs were dispersed in 5 mL of NMF for the growth of the cation layer. The cation precursor mixture contains $\text{Mn}(\text{OAc})_2$, $(\text{Zn}(\text{OAc})_2 \cdot 2\text{H}_2\text{O})$ and $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ dissolved in NMF. 1 mL of cation precursor solution was added to the sulfur coated NPLs and the mixture was stirred for 40 min in a glovebox under nitrogen environment. The NPLs were precipitated by centrifugation after addition of toluene and the (Cd,Zn,Mn)S coated 2 ML CdSe NPLs were re-dispersed in NMF. This cleaning step with toluene was repeated twice more to eliminate the unreacted precursor. The procedures provided above were repeated until the chosen shell thickness was accomplished. After that, the NPLs were precipitated by the addition of toluene, and 4 mL of hexane and 80 μL of OLA was added on top of the precipitated NPLs. The mixture was stirred overnight and the NPLs were precipitated by centrifugation after the addition of ethanol. Finally, the obtained CdSe/(Cd,Zn,Mn)S core/shell NPLs were dispersed in hexane for later usage. The cation concentrations in the shell layers were calculated using ICP-MS measurements and by considering the 2D planar geometry of the resulting NPLs.

S4. Experimental data for CdSe/CdS reference sample

Figure S5 shows additional data on nonmagnetic CdSe/CdS nanoplatelets (sample #0). They have similar structure with 2 monolayer cores and 4 monolayer shells, and were previously studied in ref 22. The spectral position of PL maximum (Figure S5a) is close to samples #1 and #2 (see ref [22] for comparison). The circular polarization degree in a magnetic field is negative (Figure S5b), which reflects the intrinsic electron and hole g -factors. Unlike DMS NPLs, the PL intensity stays constant, when the magnetic field strength increases (Figure S5c). This reflects the absence of Mn-assisted nonradiative recombination channels in the undoped sample.

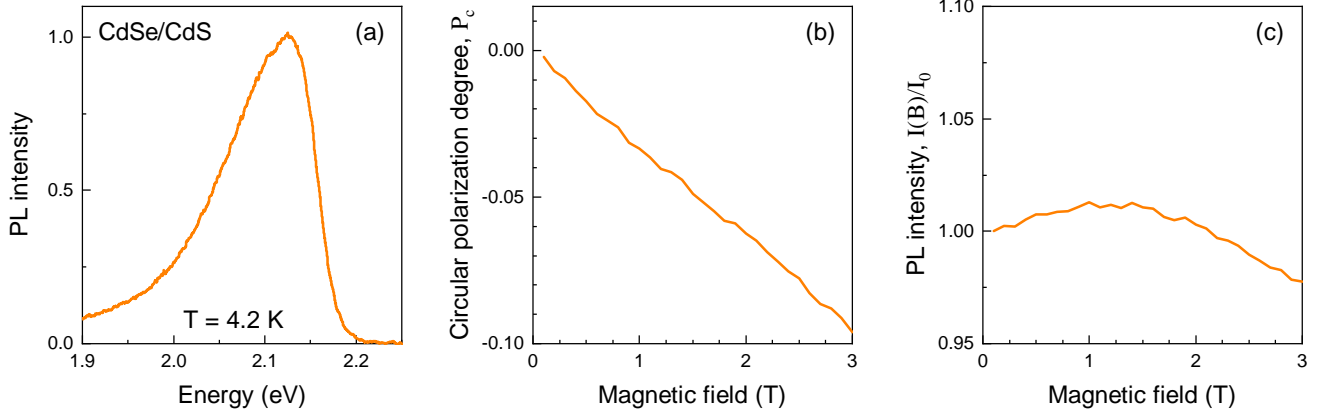


FIG. S5. Additional data on nonmagnetic reference sample #0. (a) Photoluminescence spectrum. (b) Degree of circular polarization as a function of magnetic field. (c) Integral PL intensity on magnetic field normalized to the PL intensity at zero magnetic field, I_0 . In all panels $T = 4.2$ K.