

# Chapter I

## Coherent dynamics of Mn-doped positively charged quantum dots

### I.1 Mn in a II-VI positively charged quantum dot

Cf Optical control of the spin of a magnetic atom in a semiconductor QD, L. Besombes et. al., Sept 2014

#### I.1.1 Spin structure of a positively charged Mn doped quantum dot

Cf XplusMnRes.pptx to detail the e-Mn levels

E included in model for generality (cite Claire paper "Resonant pumping..." [1]). The effects will be discussed in depth later.

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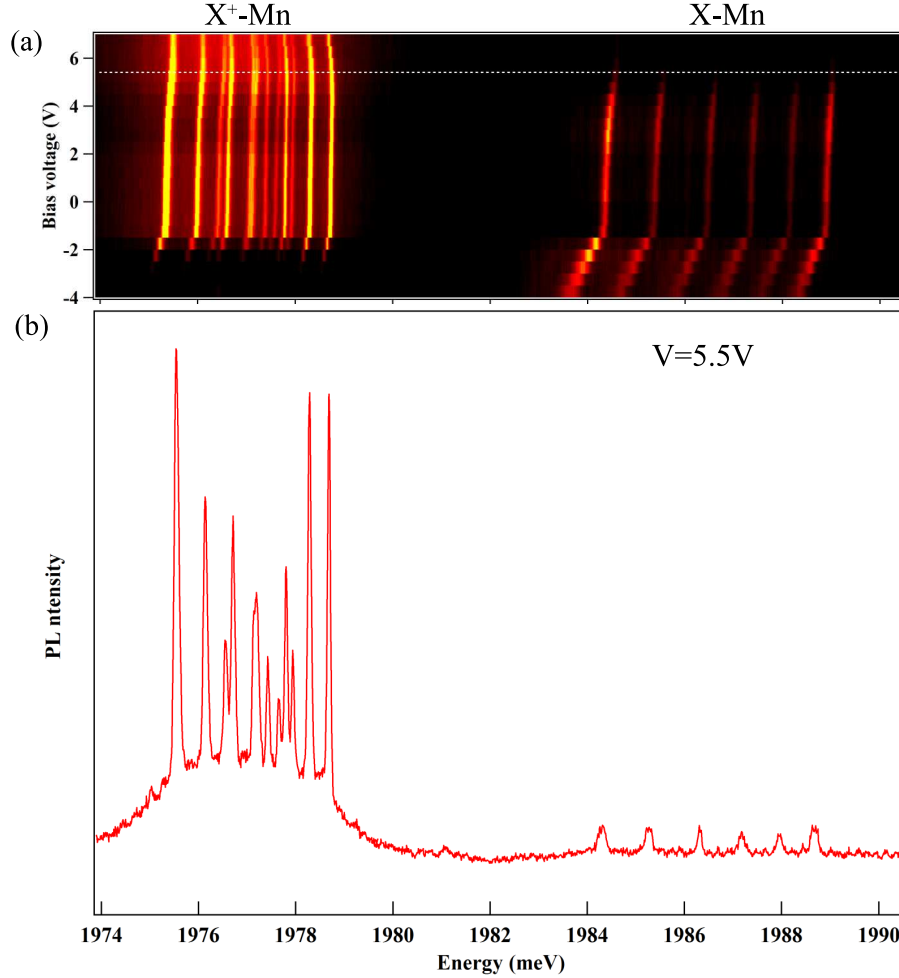


Figure I.1: (a) Color scale plot of the PL intensity of the studied Mn doped QD inserted in Schottky structure showing the emission of the neutral ( $X-Mn$ ) and positively charged ( $X^+-Mn$ ) exciton as a function of energy and bias voltage. (b) PL of the Mn-doped QD under a positive bias voltage of  $V=5.5V$ .

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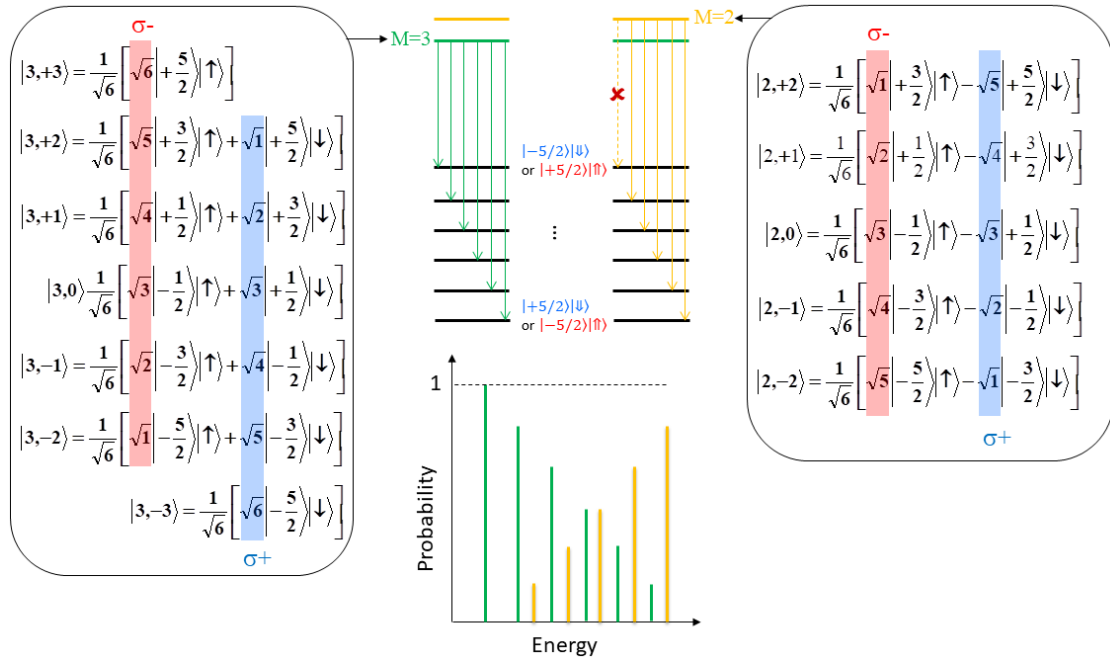


Figure I.2: Electron-Mn spin states for each  $|M, M_z\rangle$ . For each  $M$ , the  $\sigma^-$  (red) and  $\sigma^+$  (blue) probability is highlighted. This probability is directly linked to the intensity of each peak. In the center, the different possible recombination path for  $M = 3$  and  $M = 2$  are presented. A schema of the resulting spectra is drawn below.

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Table I.1: Values of the parameters used in the model of the positively charged Mn-doped QD presented in Fig. I.1.  $I_{eMn}$ ,  $I_{hMn}$ ,  $\frac{\rho_s}{\Delta_{lh}}$ ,  $\theta$ ,  $\eta$  and  $T_{eff}$  are used to model the linear polarization intensity map of Fig. I.3. The other parameters cannot be extracted from the PL measurements and values for typical Mn-doped QDs are chosen for the calculation of the spin dynamics presented in Sec. I.2 and I.3.

$I_{eMn}$	$I_{hMn}$	$\frac{\rho_s}{\Delta_{lh}}$	$\theta$	$\eta$	$T_{eff}$	$g_e$	$g_h$	$g_{Mn}$	$D_0$	$E$
$\mu eV$	$\mu eV$		$^\circ$	$\mu eV$	K				$\mu eV$	$\mu eV$
-175	345	0.09	0	30	20	-0,4	0.6	2	7	1.5

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### I.1.2 Optical $\Lambda$ -level identification

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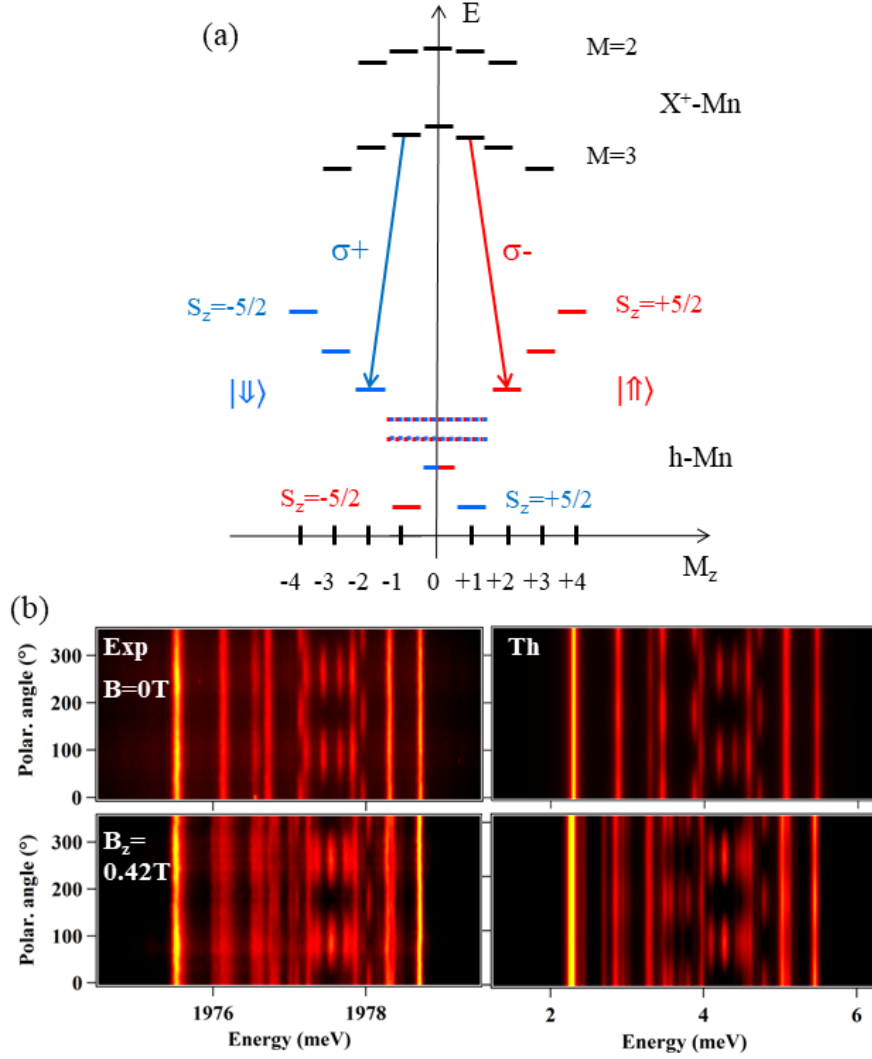


Figure I.3: (a) Energy levels of the ground (h-Mn) and excited ( $X^+$ -Mn) states as a function of their angular momentum ( $M_z$ ). The levels in dotted lines corresponds to the h-Mn states  $|-1/2\rangle|\uparrow\rangle$  and  $|+1/2\rangle|\downarrow\rangle$  coupled by the valence band mixing. Optical recombination towards these levels leads to the linearly polarized lines observed in (b). (b) Experimental (left) and calculated (right) color-scale plot of the linear polarization dependence of the PL of  $X^+$ -Mn at  $B = 0$  T (top) and  $B_\perp = 0.42$  T (bottom). The parameters used in the calculation are listed in Table I.1.

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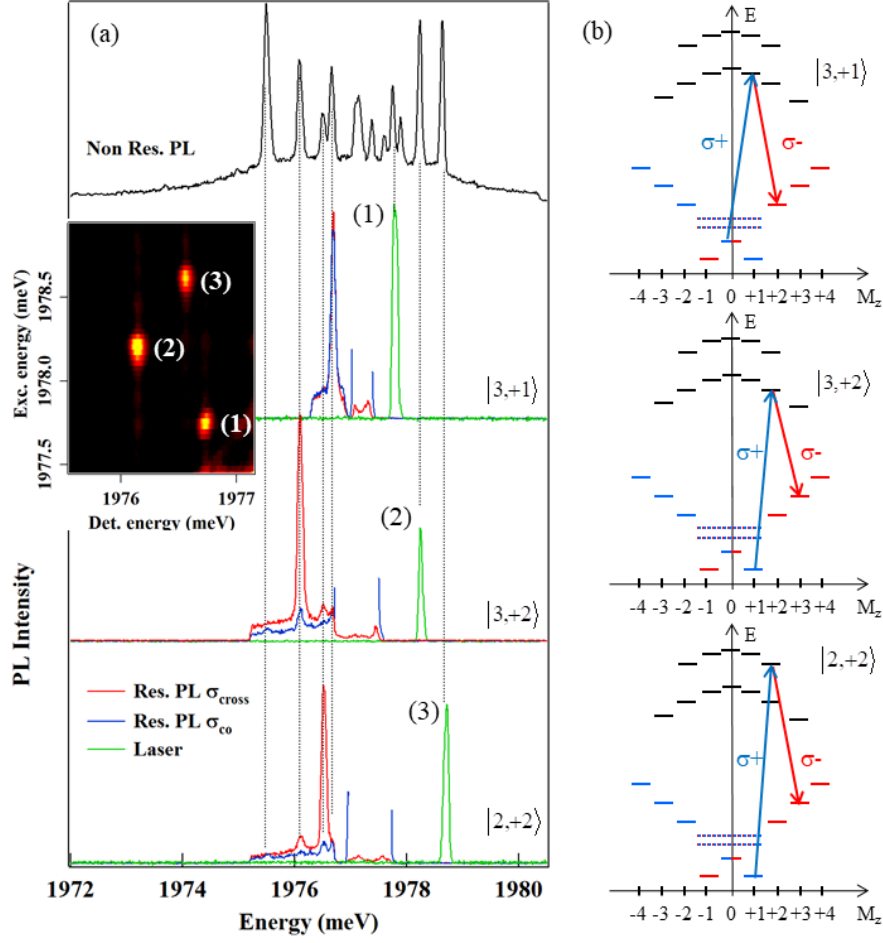


Figure I.4: (a) Non resonant (Non Res.) and resonant (Res.) PL of  $X^+-Mn$ . Co and cross circularly polarized PL spectra are collected for three different energies of the CW resonant laser (green). Inset: intensity map of the cross-circularly polarized PL detected on the low energy side of  $X^+-Mn$  as the CW laser is scanned through the high energy side. (b) Energy levels of  $X^+-Mn$  and identification of the three resonances observed in (a) corresponding to the optical  $\Lambda$  systems associated with the e-Mn states  $|3,+1\rangle$ ,  $|3,+2\rangle$  and  $|2,+2\rangle$ .

## I.2 Spin dynamics under resonant excitation

Cf article 2016/01

### I.2.1 Cycling and escaping the $\lambda$ -level system

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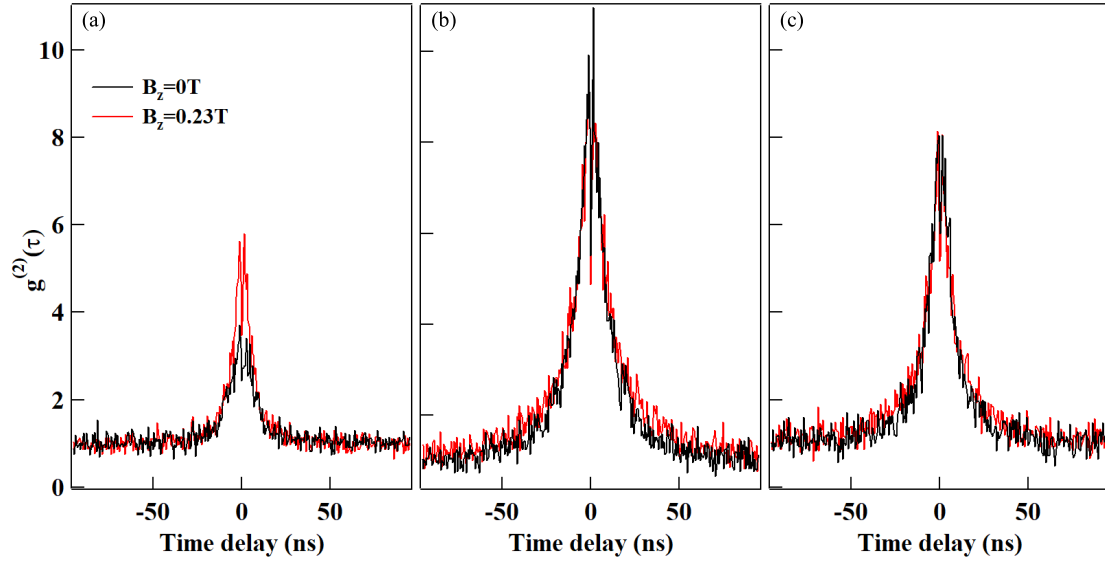


Figure I.5: Auto-correlation of the resonant PL for cross-circularly polarized excitation and detection of the electron-Mn states (a)  $|3, +1\rangle$ , (b)  $|3, +2\rangle$  and (c)  $|2, +2\rangle$ .

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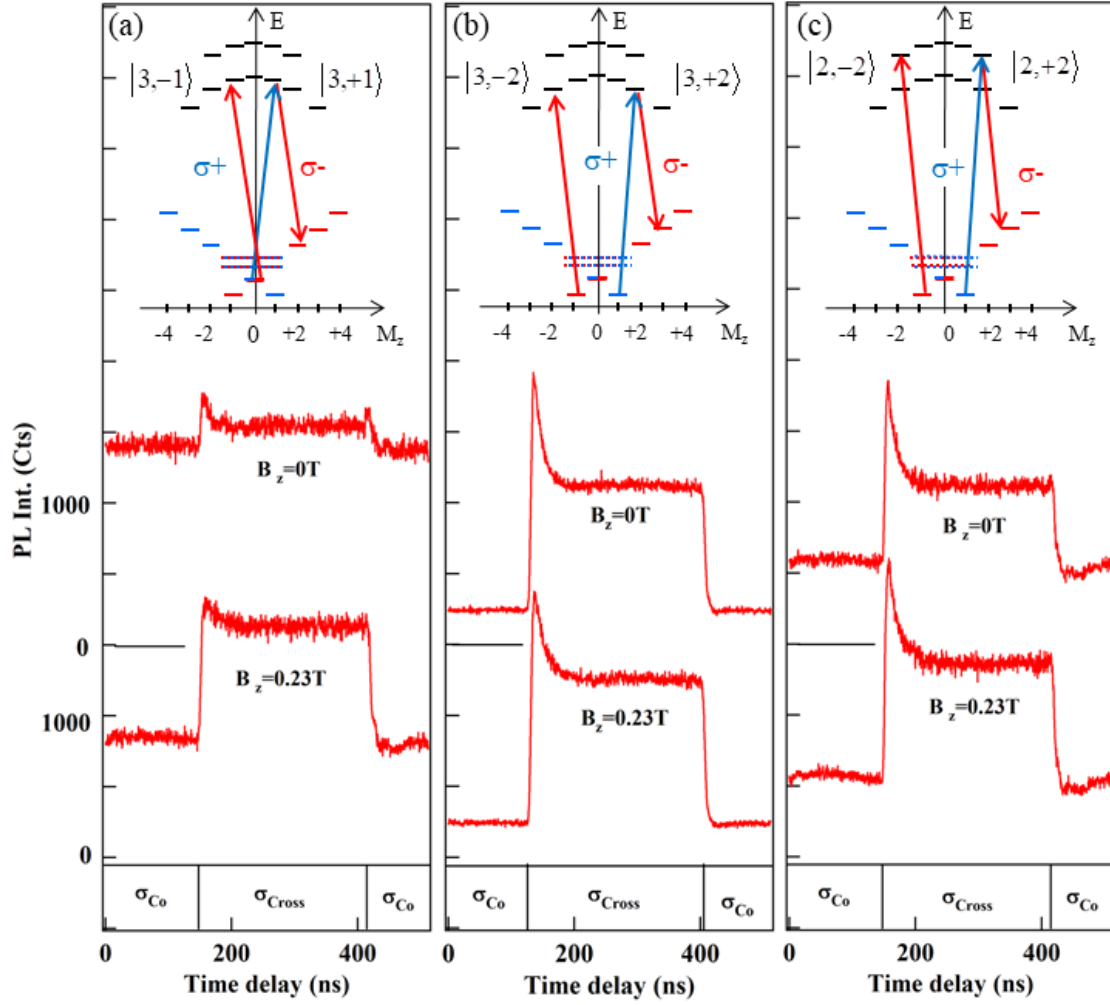


Figure I.6: Resonant optical pumping transients obtained under circular polarization switching of the resonant excitation for the  $\Lambda$  systems associated with (a)  $|3, +1\rangle$ , (b)  $|3, +2\rangle$  and (c)  $|2, +2\rangle$  at zero field and under a weak longitudinal magnetic field  $B_z=0.23\text{T}$ . The insets present the corresponding states which are resonantly excited and detected in  $\sigma-$  polarization.

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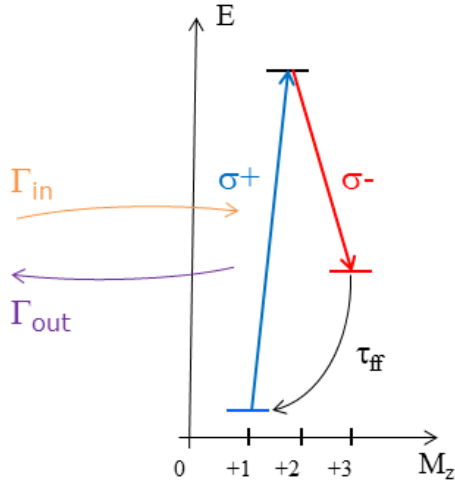


Figure I.7: Schema of the energy levels of the optical  $\Lambda$  system associated with the electron-Mn state  $|3, +2\rangle$  extracted from the full level structure of a positively charged Mn-doped QD (Fig. I.4). The different processes discussed in this section are presented on it.

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## I.2.2 Relaxation mechanism

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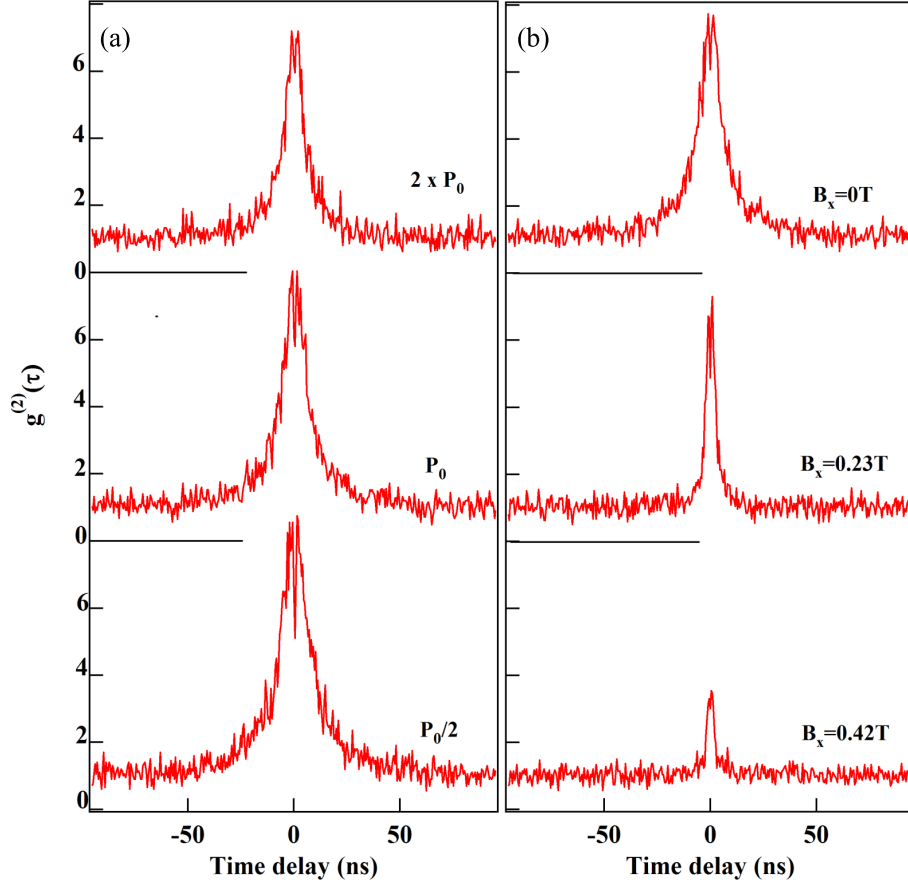


Figure I.8: Excitation power dependence (a) and transverse magnetic field dependence (b) of the auto-correlation of the resonant PL obtained for an excitation on the high energy branch of the  $\Lambda$  level system associated to the e-Mn state  $|2, +2\rangle$ .

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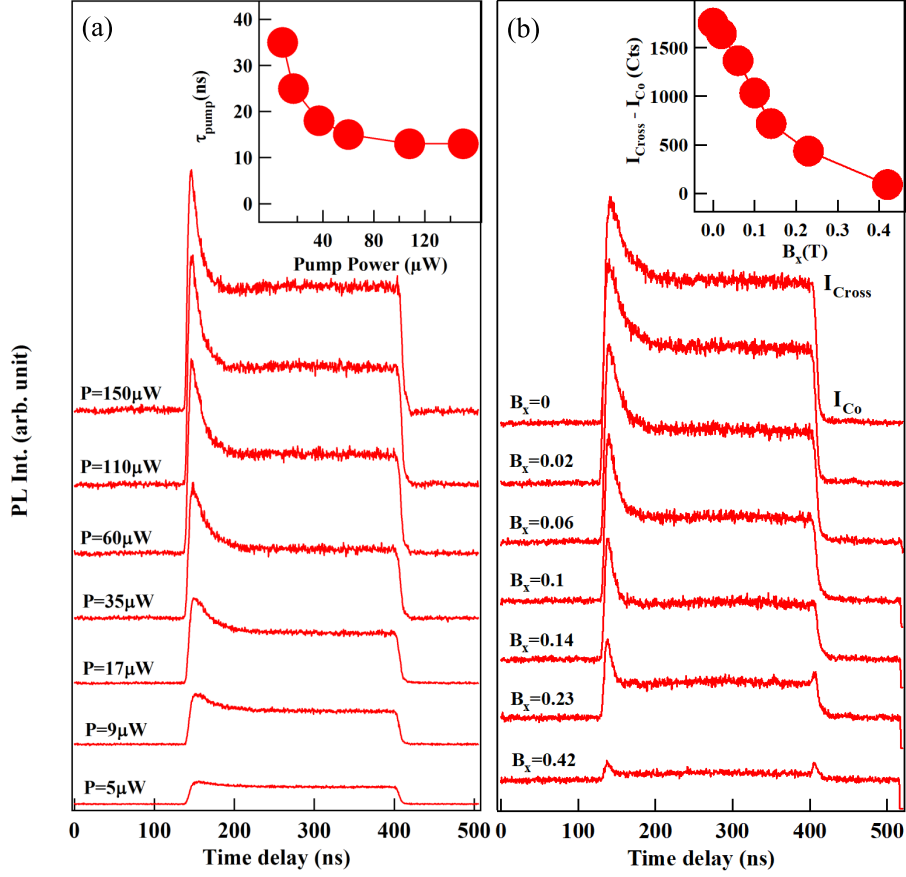


Figure I.9: Excitation power dependence (a) and transverse magnetic field dependence (b) of the optical pumping signal obtained for a resonant excitation on  $|3, +2\rangle$ . Insets: excitation power dependence of the pumping time and transverse magnetic field dependence of the difference of resonant PL intensity between a  $\sigma_{\text{cross}}$  and a  $\sigma_{\text{co}}$  excitation.

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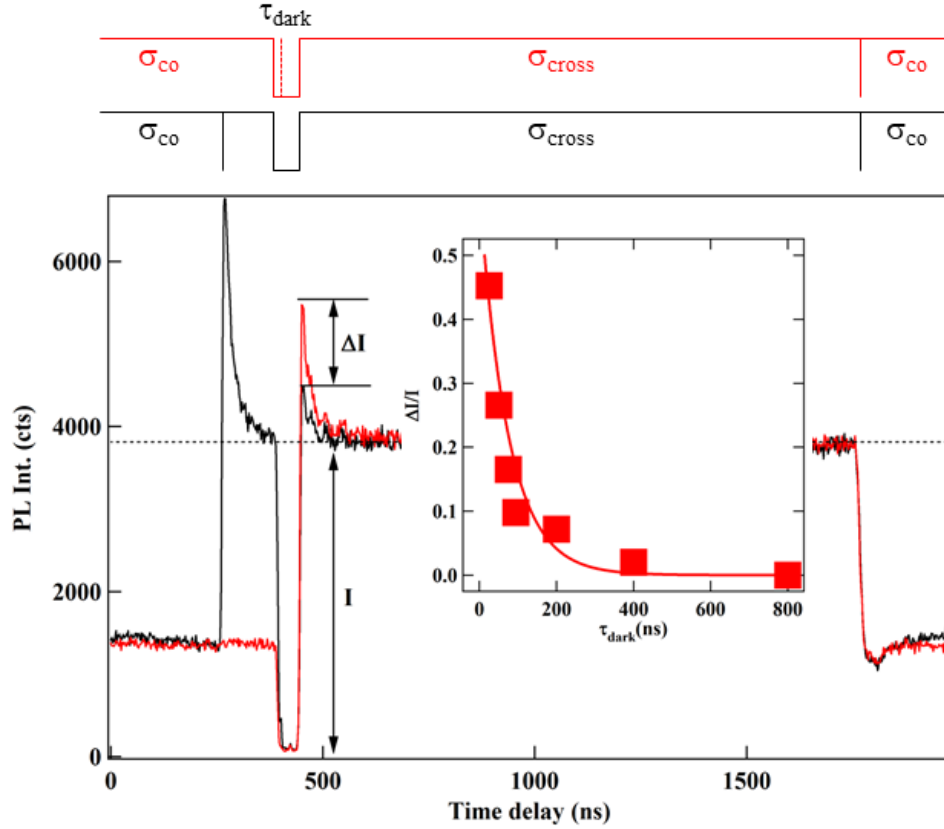


Figure I.10: Optical pumping experiment for an excitation of  $|3, +2\rangle$  with modulated circular polarization. A dark time ( $\tau_{dark} = 50ns$ ) is introduced in the pumping sequence. The polarization switching occurs either before (black) or during (red) the dark time. The black and red diagrams present the corresponding resonant excitation sequences. The inset presents the variation of the ratio  $\Delta I/I$  as a function of  $\tau_{dark}$ . The solid line is an exponential fit with  $\tau_{relax} = 80ns$ .

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Table I.2: Material (CdTe or ZnTe) [2] and QD parameters used in the calculation of the coupled hole and Mn spin relaxation time.

CdTe		
Deformation potential constants	b	-1.0 eV
	d	-4.4 eV
Longitudinal sound speed	$c_l$	3300 m/s
Transverse sound speed	$c_t$	1800 m/s
Density	$\rho$	5860 kg/m <sup>3</sup>
ZnTe		
Deformation potential constants	b	-1.4 eV
	d	-4.4 eV
Longitudinal sound speed	$c_l$	3800 m/s
Transverse sound speed	$c_t$	2300 m/s
Density	$\rho$	5908 kg/m <sup>3</sup>
Quantum dot		
Hole Mn exchange energy	$I_{hMn}$	0.35 meV
hh-lh exciton splitting	$\Delta_{lh}$	15 meV
Hole wave function widths:		
- in plane	$l_{\perp}$	3.0 nm
- z direction	$l_z$	1.25 nm

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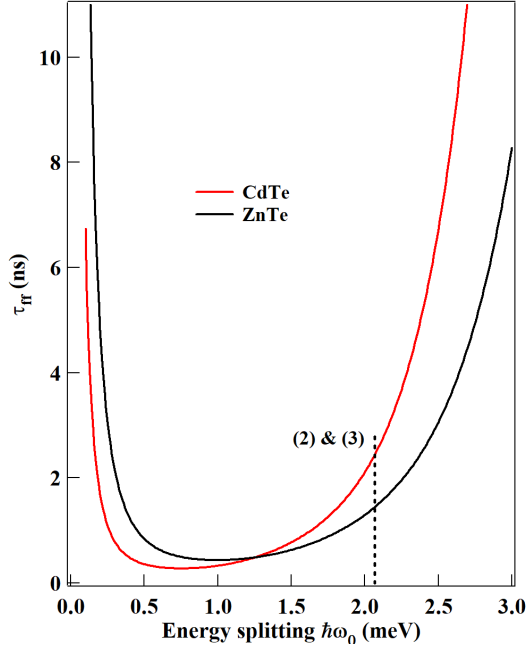


Figure I.11: Relaxation time  $\tau_{ff}$ , between the two Mn-hole ground states of the  $\Lambda$  system calculated with the material and QD parameters listed in Table I.2 and a temperature  $T=7K$ . The vertical line shows the energy splitting in the studied QD of the Mn-hole states involved in the  $\Lambda$  systems considered here (Resonances (2) and (3) identified in Fig. I.4).

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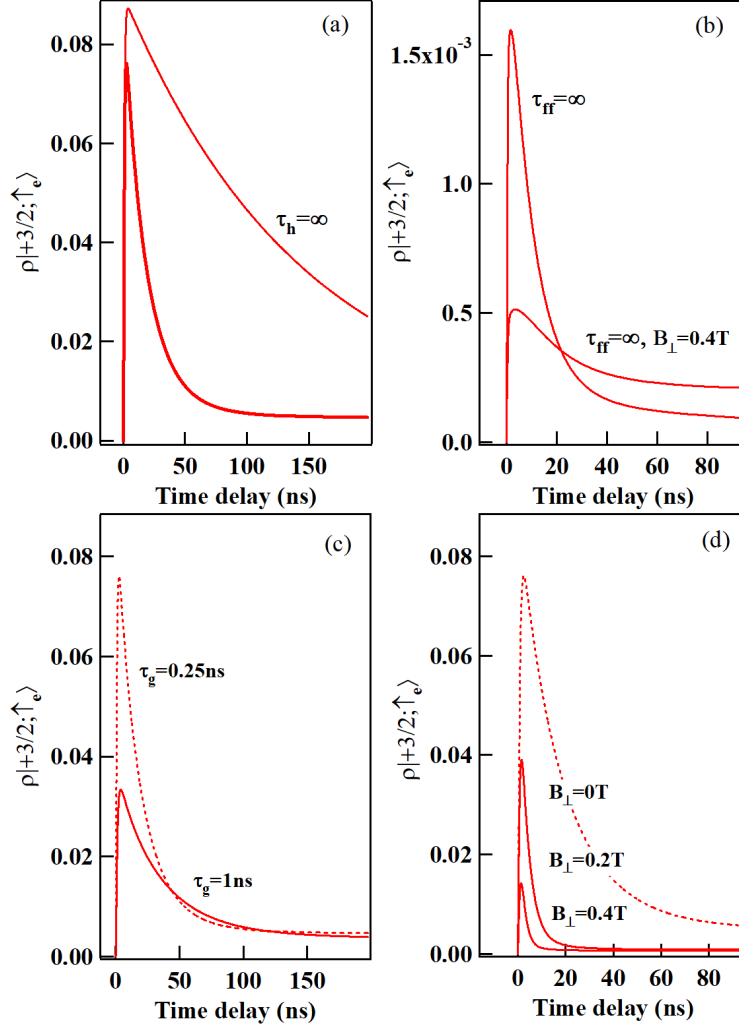


Figure I.12: (a) Calculated time evolution of  $\rho_{|+\frac{3}{2}, \uparrow_e\rangle}(t)$  with the QD parameters listed in Table I.1 and (unless specified)  $\tau_r=0.3\text{ns}$ ,  $\tau_{Mn}=5\text{ }\mu\text{s}$ ,  $\tau_h=10\text{ns}$ ,  $\tau_g=0.25\text{ ns}$ ,  $\tau_{ff}=1.5\text{ ns}$ ,  $T_2^{hMn}=5\text{ ns}$ ,  $T_2^{eMn}=0.5\text{ ns}$ ,  $T=10\text{K}$  and  $B_\perp=0$ . (b) (c) and (d) illustrate the influence of, respectively,  $\tau_{ff}$ ,  $\tau_g$  and  $B_\perp$  on  $\rho_{|+\frac{3}{2}, \uparrow_e\rangle}(t)$ . Note the different vertical scale in (b).

risus. Quisque bibendum pede eu dolor.

Donec tempus neque vitae est. Aenean egestas odio sed risus ullamcorper ullamcorper. Sed in nulla a tortor tincidunt egestas. Nam sapien tortor, elementum sit amet, aliquam in, porttitor faucibus, enim. Nullam congue suscipit nibh. Quisque convallis. Praesent arcu nibh, vehicula eget, accumsan eu, tincidunt a, nibh. Suspendisse vulputate, tortor quis adipiscing viverra, lacus nibh dignissim

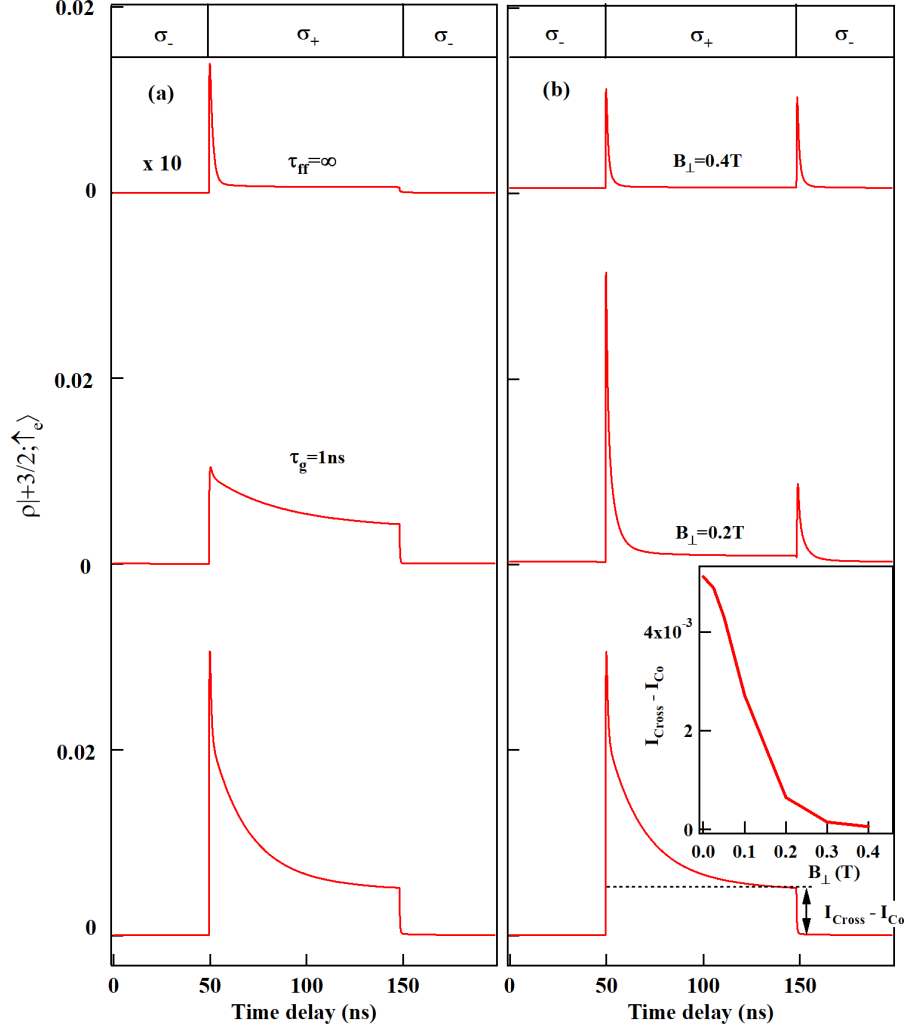


Figure I.13: Calculated resonant optical pumping transients for a  $\sigma-$  detection and an excitation of  $|3, +2\rangle$  and  $|3, -2\rangle$  with modulated circular polarization. The QD parameters for the calculations are those listed in table I.1 and  $\tau_r=0.3$  ns,  $\tau_{Mn}=5$   $\mu$ s,  $\tau_h=10$  ns,  $T_2^{hMn}=5$  ns,  $T_2^{eMn}=0.5$  ns,  $\tau_{ff}=1.5$  ns,  $T=10$  K and  $\tau_g=0.25$  ns. (a) Influence of a variation of  $\tau_g$  and  $\tau_{ff}$ . (b) Influence of a transverse magnetic field  $B_\perp$ . The inset presents the transverse magnetic field dependence of the difference of population for a  $\sigma+$  or a  $\sigma-$  excitation.

tellus, eu suscipit risus ante fringilla diam. Quisque a libero vel pede imperdiet aliquet. Pellentesque nunc nibh, eleifend a, consequat consequat, hendrerit nec, diam. Sed urna. Maecenas laoreet eleifend neque. Vivamus purus odio, eleifend non, iaculis a, ultrices sit amet, urna. Mauris faucibus odio vitae risus. In nisl.

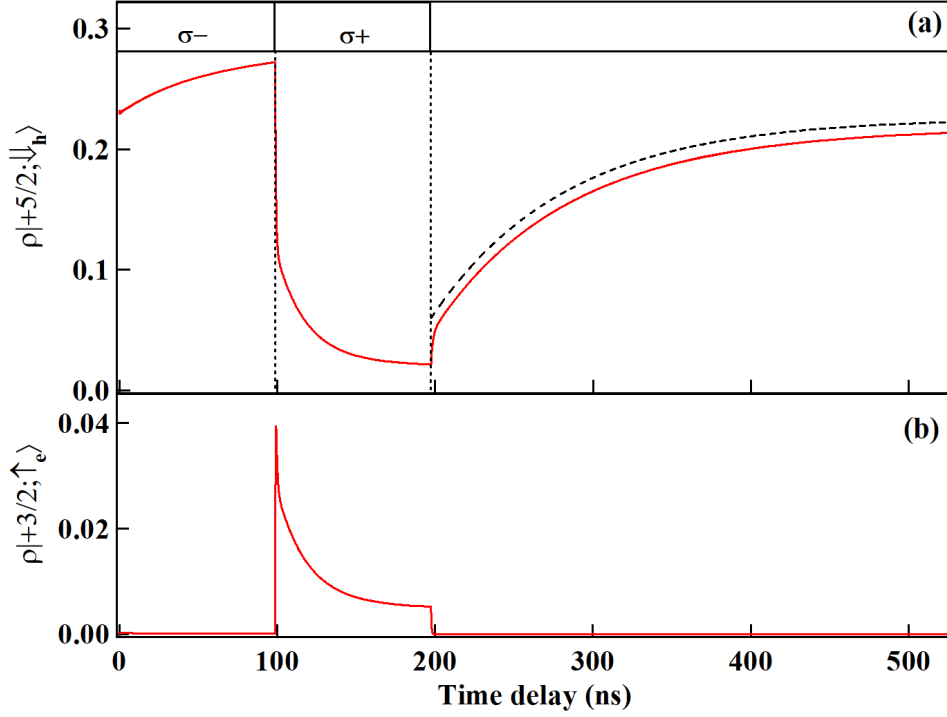


Figure I.14: (a) Calculated time evolution in the dark of the population of the hole-Mn state  $|+\frac{5}{2}, \downarrow_h\rangle$  initialized by a sequence of  $\sigma-/\sigma+$  resonant excitation of  $|3, -2\rangle$  and  $|3, +2\rangle$ . The dashed black line (shifted for clarity) is an exponential fit with a characteristic time  $\tau_{relax}=85$  ns. (b) Corresponding calculated time evolution of the population  $|+\frac{3}{2}, \uparrow_e\rangle$ . The parameters are those of Fig. I.13.

Praesent purus. Integer iaculis, sem eu egestas lacinia, lacus pede scelerisque augue, in ullamcorper dolor eros ac lacus. Nunc in libero.

Fusce suscipit cursus sem. Vivamus risus mi, egestas ac, imperdiet varius, faucibus quis, leo. Aenean tincidunt. Donec suscipit. Cras id justo quis nibh scelerisque dignissim. Aliquam sagittis elementum dolor. Aenean consectetur justo in pede. Curabitur ullamcorper ligula nec orci. Aliquam purus turpis, aliquam id, ornare vitae, porttitor non, wisi. Maecenas luctus porta lorem. Donec vitae ligula eu ante pretium varius. Proin tortor metus, convallis et, hendrerit non, scelerisque in, urna. Cras quis libero eu ligula bibendum tempor. Vivamus tellus quam, malesuada eu, tempus sed, tempor sed, velit. Donec lacinia auctor libero.

Morbi justo. Aenean nec dolor. In hac habitasse platea dictumst. Proin nonummy porttitor velit. Sed sit amet leo nec metus rhoncus varius. Cras ante. Vestibulum commodo sem tincidunt massa. Nam justo. Aenean luctus, felis et condimentum lacinia, lectus enim pulvinar purus, non porta velit nisl sed eros.

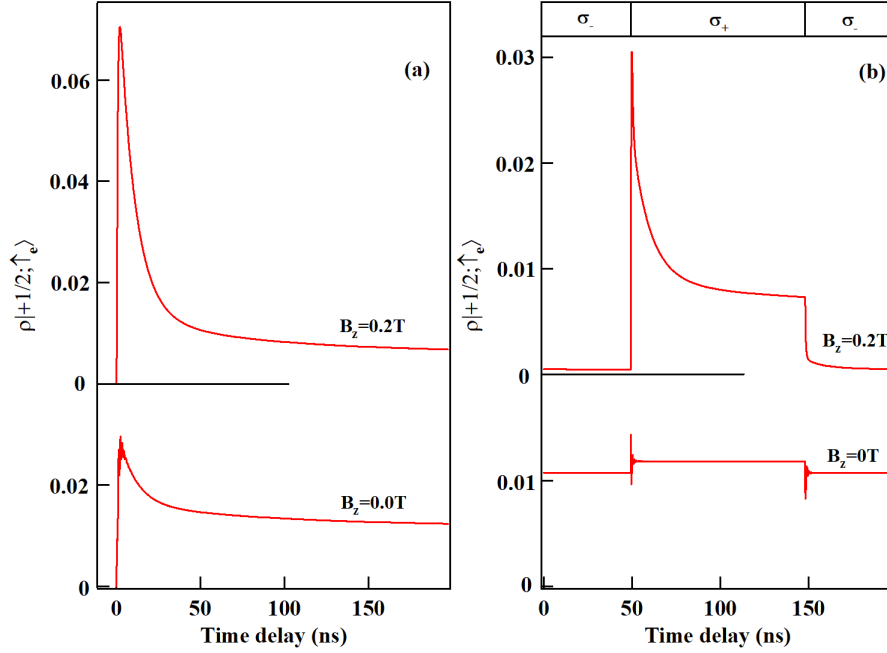


Figure I.15: (a) Calculated time evolution of  $\rho_{|+1/2, \uparrow_e\rangle}$  with  $\rho_{|+1/2, \uparrow_h\rangle}=1$  (Mn-hole spin in the state  $|+1/2, \uparrow_h\rangle$  after a  $\sigma-$  recombination) for a resonant  $\sigma+$  excitation of the coupled electron-Mn states  $|3, +1\rangle$  and  $|3, -1\rangle$  without and with a longitudinal magnetic field. (b) Time evolution of  $\rho_{|+1/2, \uparrow_e\rangle}$  under excitation with modulated circular polarization. The parameters used in the calculations are those of Fig. I.13.

Suspendisse consequat. Mauris a dui et tortor mattis pretium. Sed nulla metus, volutpat id, aliquam eget, ullamcorper ut, ipsum. Morbi eu nunc. Praesent pretium. Duis aliquam pulvinar ligula. Ut blandit egestas justo. Quisque posuere metus viverra pede.

Vivamus sodales elementum neque. Vivamus dignissim accumsan neque. Sed at enim. Vestibulum nonummy interdum purus. Mauris ornare velit id nibh pretium ultricies. Fusce tempor pellentesque odio. Vivamus augue purus, laoreet in, scelerisque vel, commodo id, wisi. Duis enim. Nulla interdum, nunc eu semper eleifend, enim dolor pretium elit, ut commodo ligula nisl a est. Vivamus ante. Nulla leo massa, posuere nec, volutpat vitae, rhoncus eu, magna.

Quisque facilisis auctor sapien. Pellentesque gravida hendrerit lectus. Mauris rutrum sodales sapien. Fusce hendrerit sem vel lorem. Integer pellentesque massa vel augue. Integer elit tortor, feugiat quis, sagittis et, ornare non, lacus. Vestibulum posuere pellentesque eros. Quisque venenatis ipsum dictum nulla. Aliquam quis quam non metus eleifend interdum. Nam eget sapien ac mauris malesuada

adipiscing. Etiam eleifend neque sed quam. Nulla facilisi. Proin a ligula. Sed id dui eu nibh egestas tincidunt. Suspendisse arcu.

a

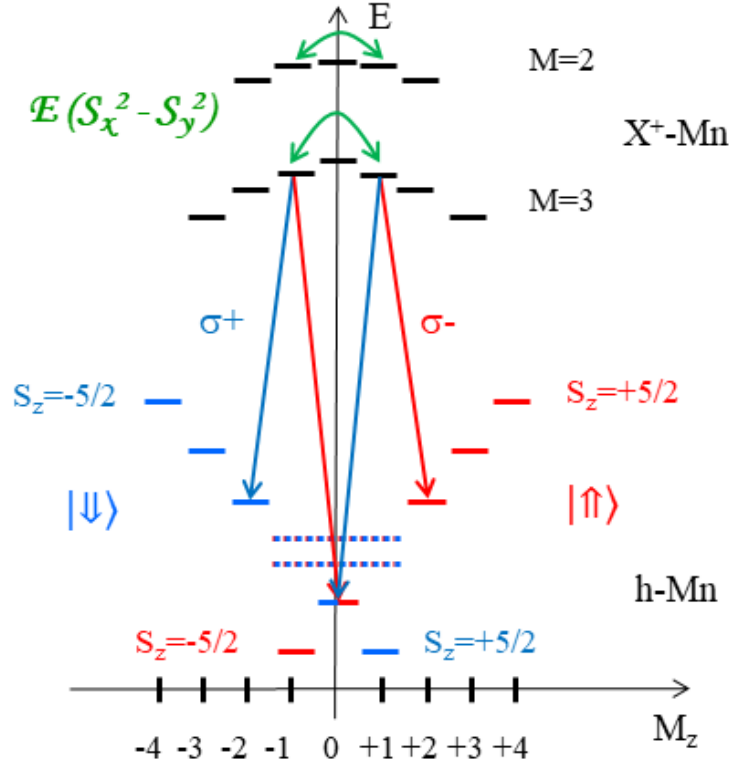


Figure I.16: Energy levels of the ground (h-Mn) and excited (X<sup>+</sup>-Mn) states as a function of their angular momentum ( $M_z$ ). The e-Mn states  $|3, +1\rangle$  and  $|3, -1\rangle$ , as well as  $|2, +1\rangle$  and  $|2, -1\rangle$ , are coupled by the strain anisotropy  $E(S_x^2 - S_y^2)$ . Optical  $\Lambda$  systems associated with  $|3, +1\rangle$  and  $|3, -1\rangle$  are presented.

We saw the influence of strain anisotropy. We will now see a way to extract it more precisely.

### I.3 Influence of the strain anisotropy

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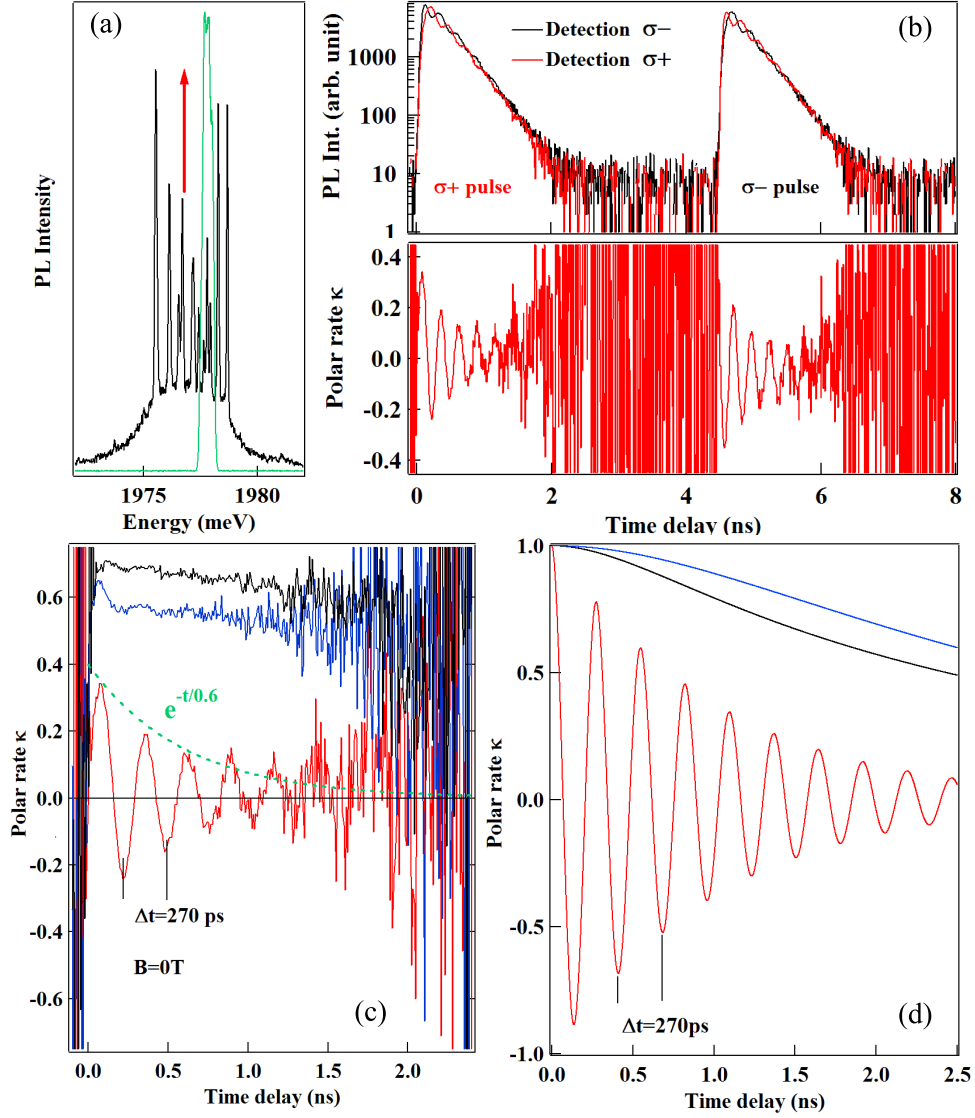


Figure I.17: (a) Configuration of the time resolved PL experiment for an excitation of  $|3,+1\rangle$  (pulsed laser in green). (b) Top panel: Time resolved resonant PL of  $|3,+1\rangle$  with a  $\sigma+/\sigma-$  sequence of laser pulses and a detection in  $\sigma+$  and  $\sigma-$  polarization. Bottom panel: corresponding time dependence of the circular polarization rate  $\kappa = (\sigma_- - \sigma_+)/(\sigma_- + \sigma_+)$ . (c) Time dependence of the circular polarization rate of the resonant PL of the states  $|3,+1\rangle$  (red),  $|3,+2\rangle$  (black) and  $|2,+2\rangle$  (blue). (d) Corresponding polarisation rates calculated with  $D_0 = 7\mu\text{eV}$  [3],  $T_2^{eMn} = 0.6\text{ns}$ ,  $E = 1.8\mu\text{eV}$ , a radiative lifetime  $T_r = 0.3\text{ns}$  and the parameters listed on Table I.1.

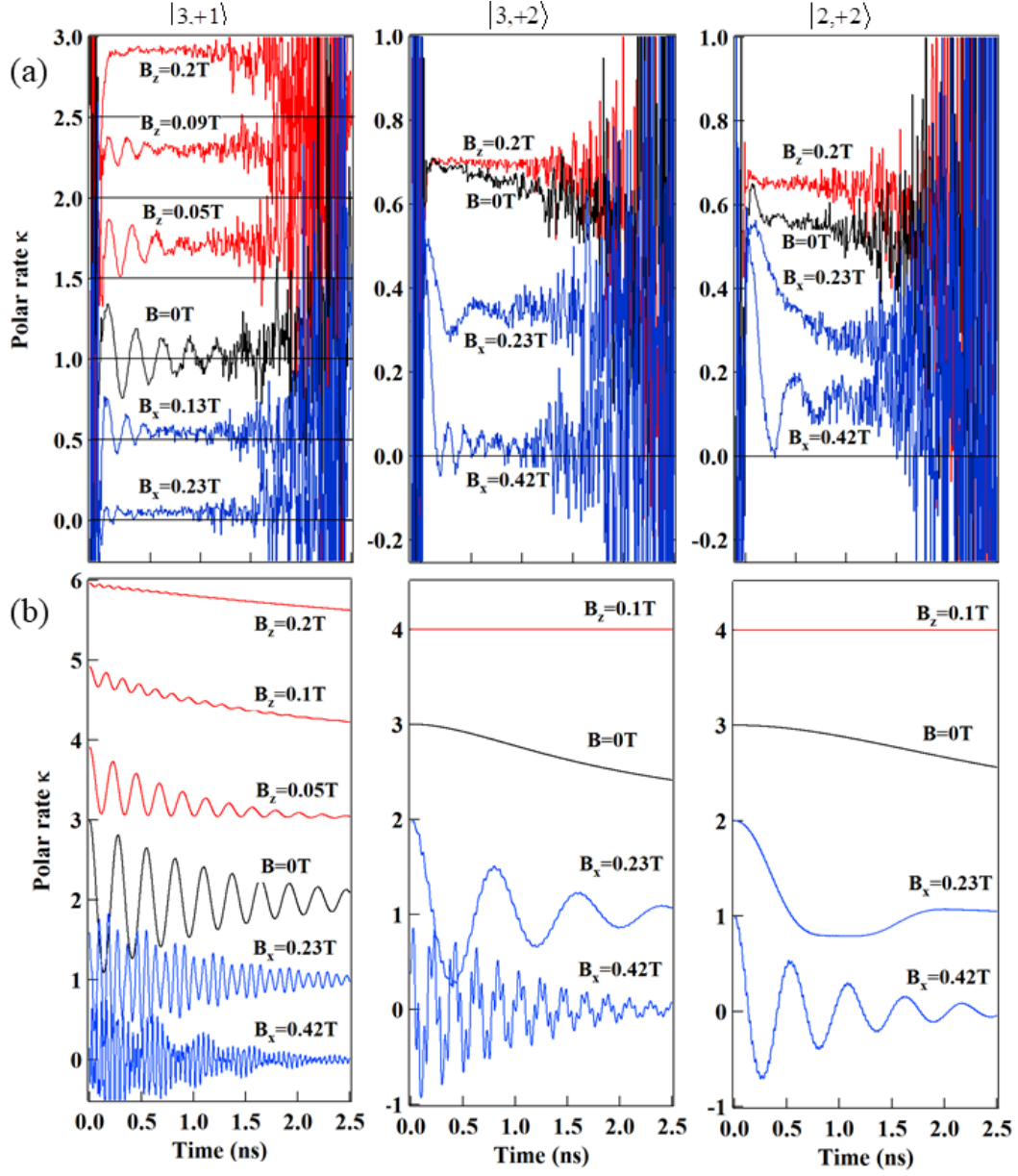


Figure I.18: (a) Influence of a longitudinal ( $B_z$ , red) and a transverse ( $B_x$ , blue) magnetic field on the time dependence of the circular polarization rate  $\kappa = (\sigma_- - \sigma_+)/(\sigma_- + \sigma_+)$  of the resonant PL of  $|3,+1\rangle$ ,  $|3,+2\rangle$  and  $|2,+2\rangle$ . On the top left panel, curves are shifted by 0.5 for clarity. (b) Corresponding time dependence of the circular polarization rate calculated with  $g_{Mn} = 2$ ,  $g_e = -0.4$ ,  $g_h = 0.6$  [3], and the parameters listed on Table I.1. The curves are shifted by 1 for clarity.

vehicula non, enim. Morbi viverra arcu nec purus. Vivamus fringilla, enim et commodo malesuada, tortor metus elementum ligula, nec aliquet est sapien ut lectus. Aliquam mi. Ut nec elit. Fusce euismod luctus tellus. Curabitur scelerisque. Nul lam purus. Nam ultricies accumsan magna. Morbi pulvinar lorem sit amet ipsum. Donec ut justo vitae nibh mollis congue. Fusce quis diam. Praesent tempus eros ut quam.

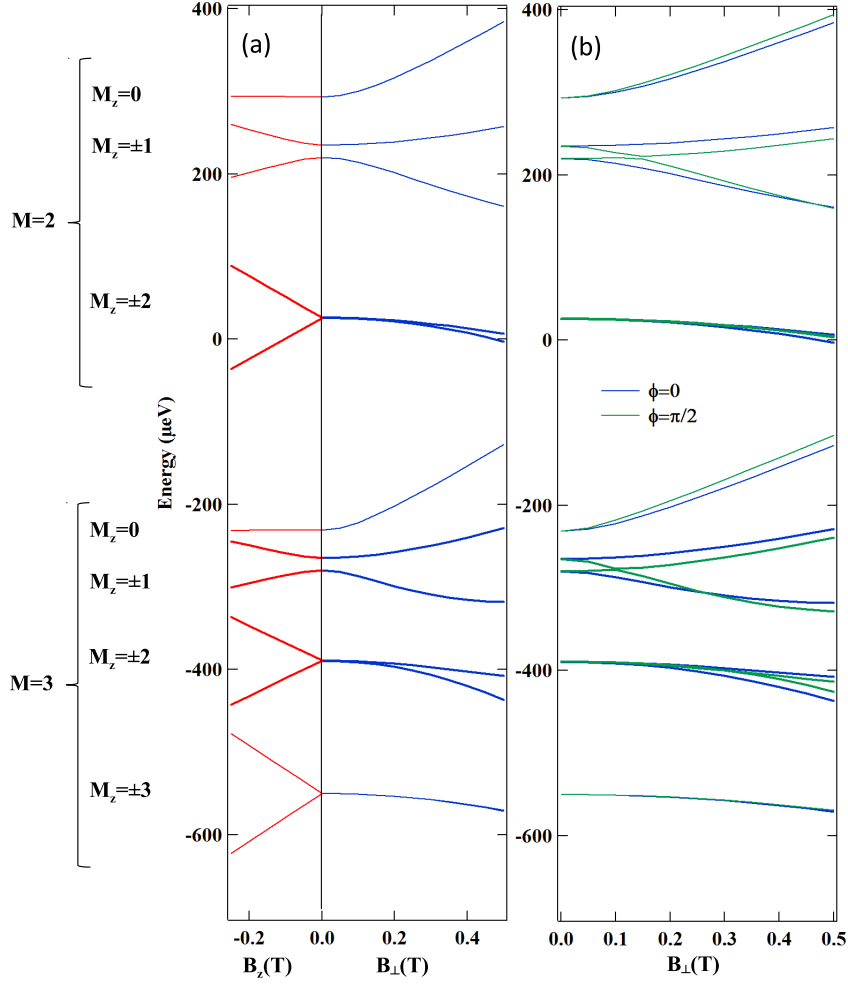


Figure I.19: (Color line) (a) Calculated energy of the electron-M<sub>l</sub> states in a longitudinal magnetic field ( $B_z$ ) and in a transverse magnetic field ( $B_\perp$ ). (b) Energy of the electron-Mn states for two orientations of the transverse magnetic field:  $\phi = 0$  ( $B_\perp = B_x$ )  $\phi = \frac{\pi}{2}$  ( $B_\perp = B_y$ ). The parameters used in the calculations are listed in Table I.2, with the exception of  $E$ , for which the more precise value of  $1.8 \mu\text{eV}$  was chosen.

Donec in nisl. Fusce vitae est. Vivamus ante ante, mattis laoreet, posuere eget, congue vel, nunc. Fusce sem. Nam vel orci eu eros viverra luctus. Pellentesque sit amet augue. Nunc sit amet ipsum et lacus varius nonummy. Integer rutrum sem eget wisi. Aenean eu sapien. Quisque ornare dignissim mi. Duis a urna vel risus pharetra imperdiet. Suspendisse potenti.

# Bibliography

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- <sup>2</sup>S. Adachi, “Properties of group IV, II-V and II-VI semiconductors”, in, edited by Wiley (2005) Chap. 8, p. 178.
- <sup>3</sup>B. Varghese, H. Boukari, and L. Besombes, “Dynamics of a Mn spin coupled to a single hole confined in a quantum dot”, [Phys. Rev. B](#) **90**, 115307 (2014).