

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

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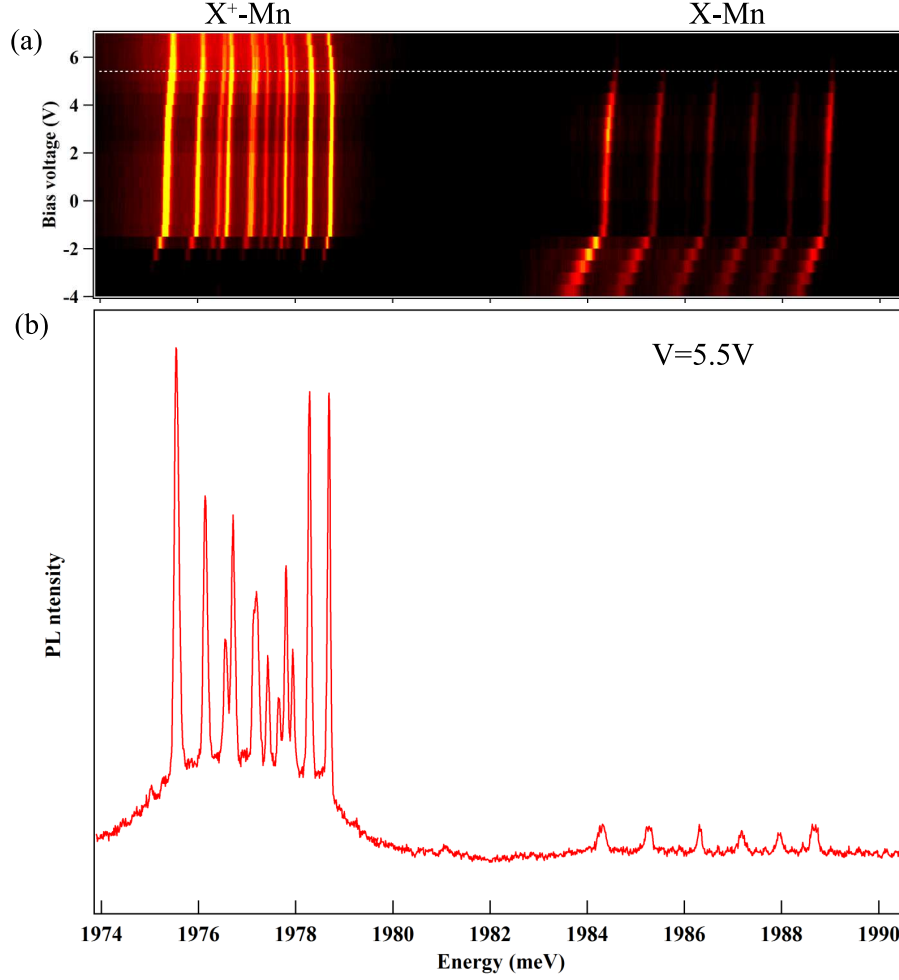


Figure I.1: (a) Color scale plot of the PL intensity of the studied Mn doped QD inserted in Schottky structure showing the emission of the neutral (X-Mn) and positively charged (X⁺-Mn) exciton as a function of energy and bias voltage. (b) PL of the Mn-doped QD under a positive bias voltage of V=5.5V.

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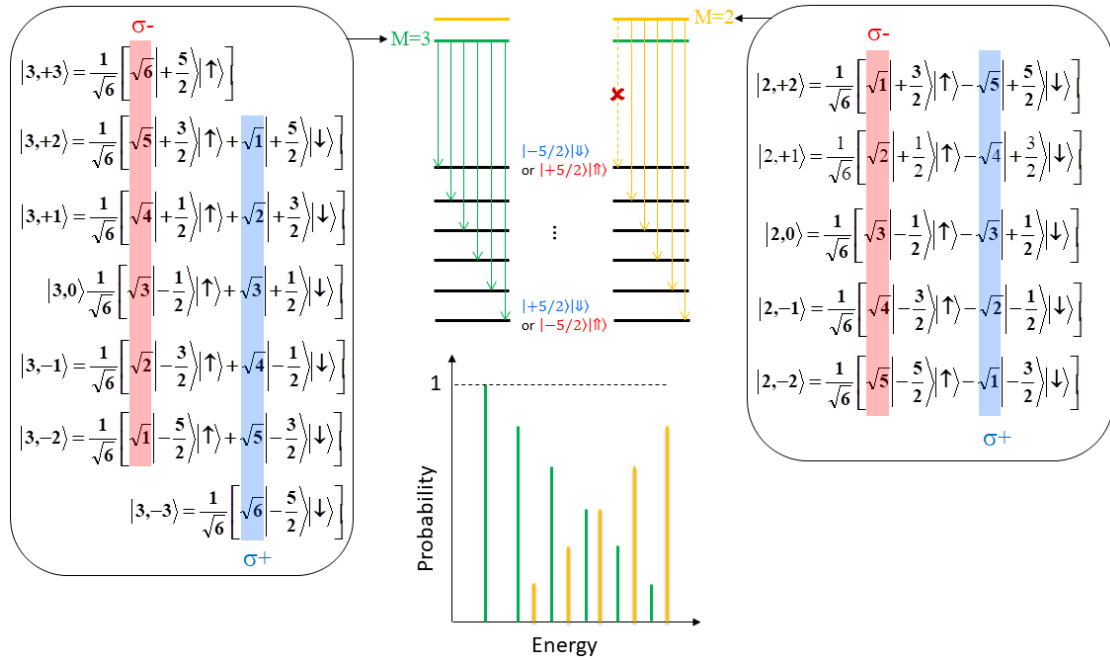


Figure I.2: Electron-Mn spin states for each $|M, M_z\rangle$. For each M , the $\sigma-$ (red) and $\sigma+$ (blue) probability is highlighted. This probability is directly linked to the intensity of each peak. In the center, the different possible recombination path for $M = 3$ and $M = 2$ are presented. 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} , $\frac{\rho_s}{\Delta_{lh}}$, θ , η 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}	$\frac{\rho_s}{\Delta_{lh}}$	θ	η	T_{eff}	g_e	g_h	g_{Mn}	D_0	E
μeV	μeV		$^\circ$	μeV	K				μeV	μeV
-175	345	0.09	0	30	20	-0,4	0.6	2	7	1.5

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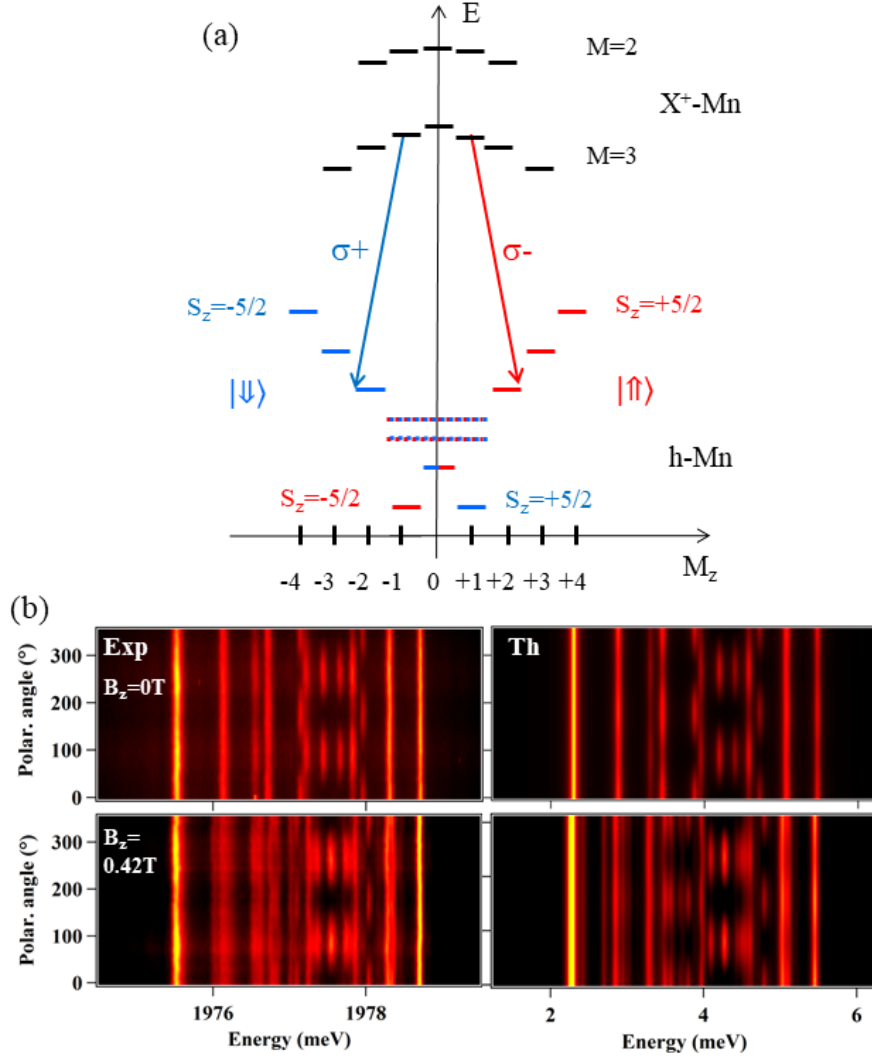


Figure I.3: (a) Energy levels of the ground (h-Mn) and excited (X^+ -Mn) states as a function of their angular momentum (M_z). The levels in dotted lines corresponds to the h-Mn states $| -1/2 \rangle | \uparrow \rangle$ and $| +1/2 \rangle | \downarrow \rangle$ coupled by the valence band mixing. Optical 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.

I.1.2 Optical Λ -level identification

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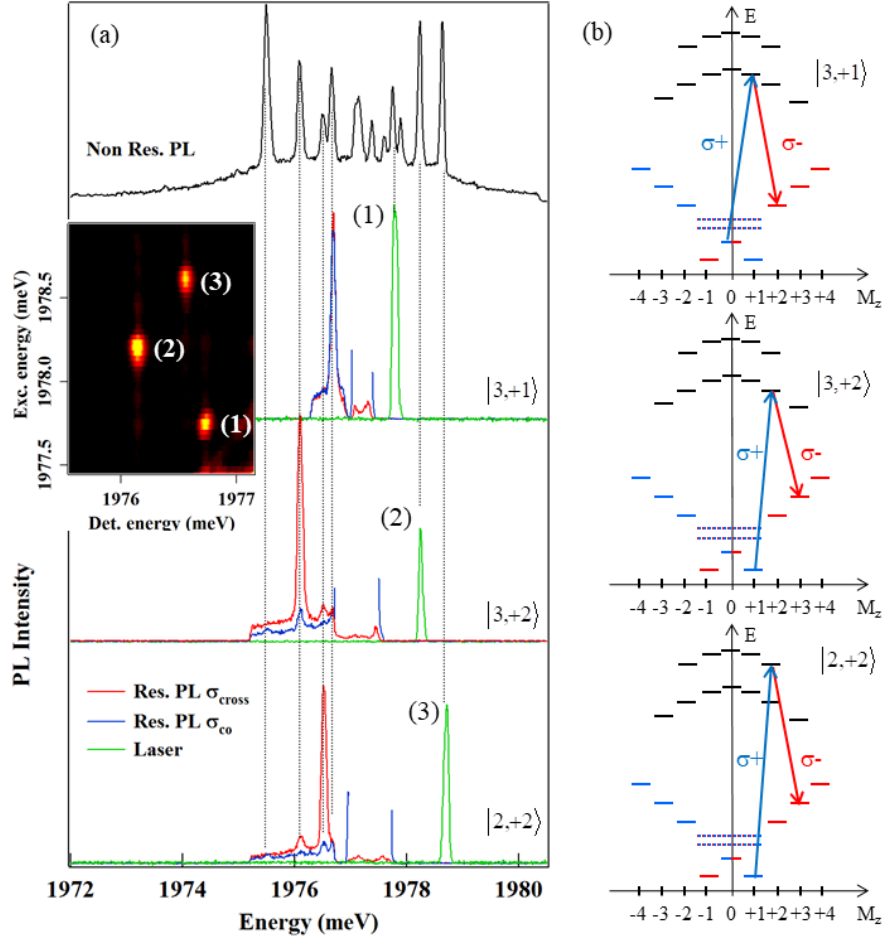


Figure I.4: (a) Non resonant (Non Res.) and resonant (Res.) PL of $X^+-\text{Mn}$. Co and cross circularly polarized PL spectra are collected for three different energies of the CW resonant laser (green). Inset: intensity map of the cross-circularly polarized PL detected on the low energy side of $X^+-\text{Mn}$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

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I.2 Spin dynamics under resonant excitation

Cf article 2016/01

I.2.1 Cycling and escaping the λ -level system

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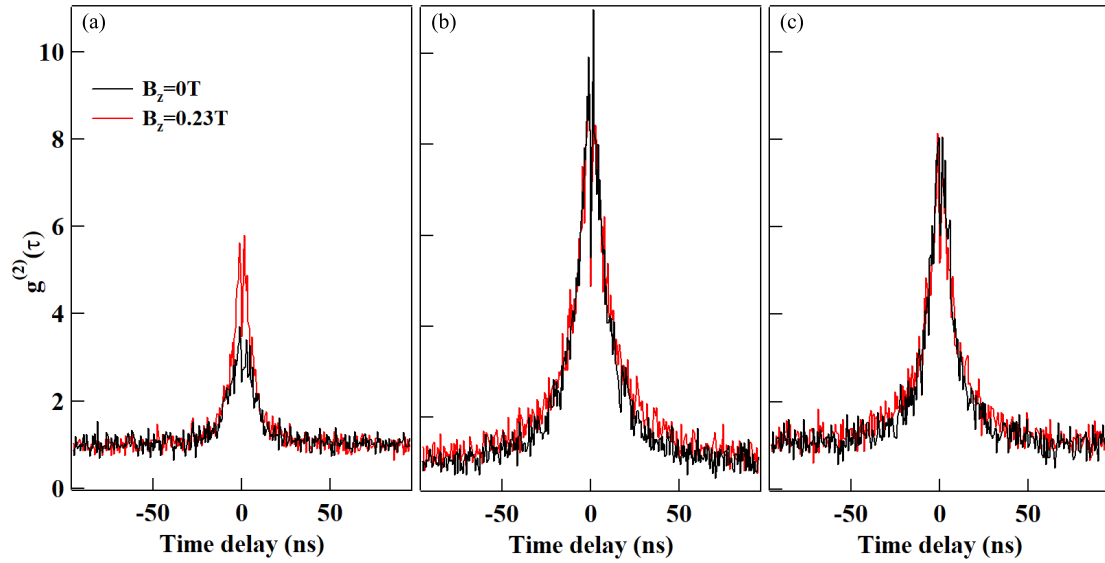


Figure I.5: Auto-correlation of the resonant PL for a cross-circularly polarized excitation and detection of the electron-Mn states (a) $|3, +1\rangle$, (b) $|3, +2\rangle$ and (c) $|2, +2\rangle$.

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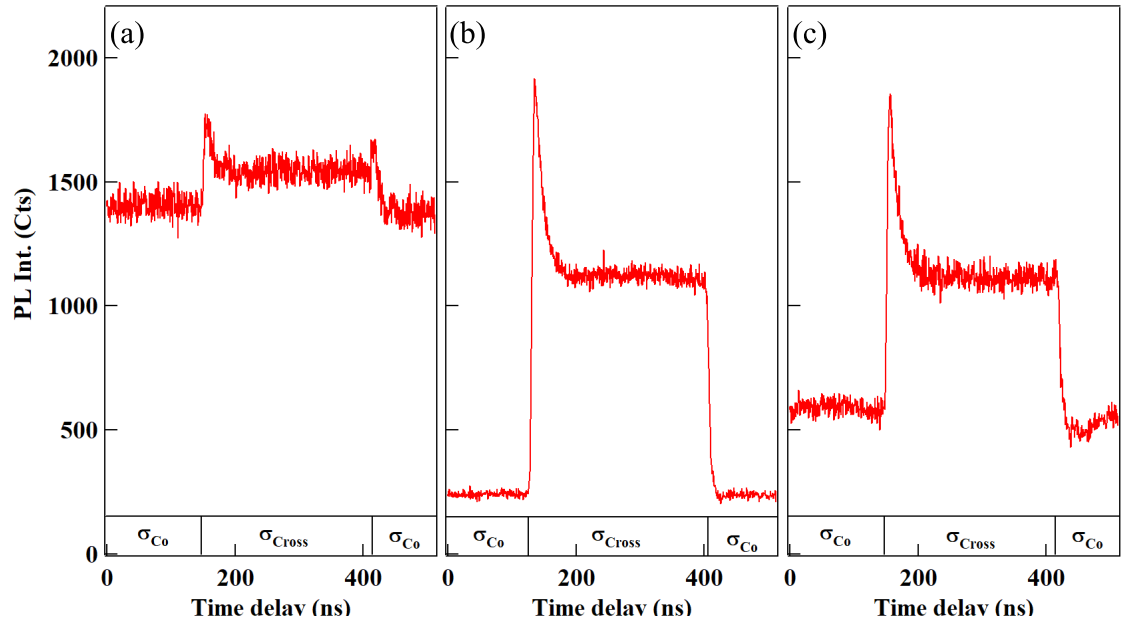


Figure I.6: Resonant optical pumping transients obtained under circular polarization switching of the resonant excitation for (a) $|3, +1\rangle$, (b) $|3, +2\rangle$ and (c) $|2, +2\rangle$ at zero field.

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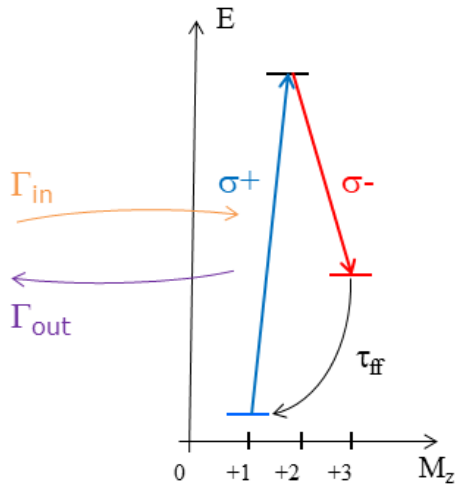


Figure I.7: Schema of the energy levels of the optical A system associated with the electron-Mn state $|3, +2\rangle$ extracted from the full level structure of a positively charged Mn-doped QD (Fig. I.4). The different processes discussed in the section are presented on it.

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I.2.2 Relaxation mechanism

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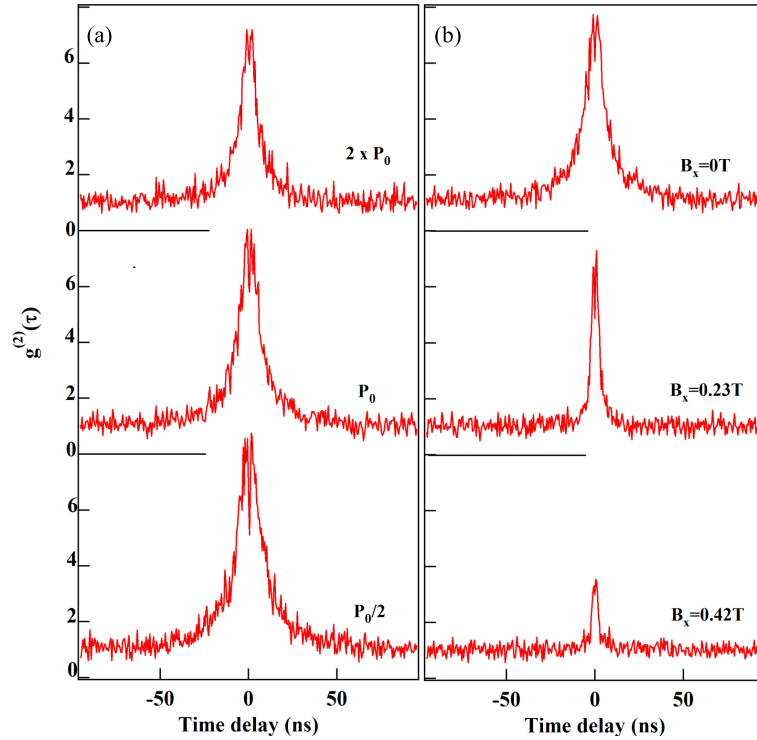


Figure I.8: Excitation power dependence (a) and transverse magnetic field dependence (b) of the auto-correlation of the resonant PL obtained for an excitation on the high energy branch of the Λ level system associated to the e-Mn state $|2, +2\rangle$.

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pretium cursus. Sed sodales. Nam eu neque quis pede dignissim ornare. Maecenas eu purus ac urna tincidunt congue.

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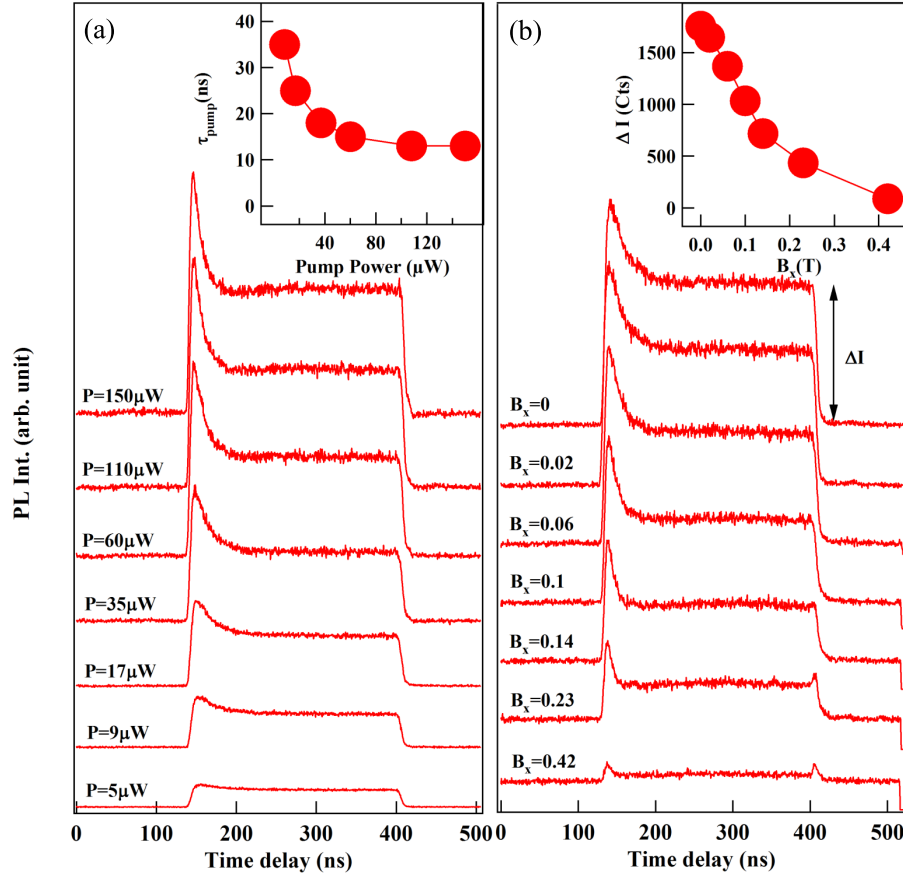


Figure I.9: Excitation power dependence (a) and transverse magnetic field dependence (b) of the optical pumping signal obtained for a resonant excitation on $|3, +2\rangle$. Insets: excitation power dependence of the pumping time and transverse magnetic field dependence of the difference of resonant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

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mentum. Etiam egestas tortor vitae lacus. Praesent cursus. Mauris bibendum pede at elit. Morbi et felis a lectus interdum facilisis. Sed suscipit gravida turpis. Nulla at lectus. Vestibulum ante ipsum primis in faucibus orci luctus et ultrices posuere cubilia Curae; Praesent nonummy luctus nibh. Proin turpis nunc, congue eu, egestas ut, fringilla at, tellus. In hac habitasse platea dictumst.

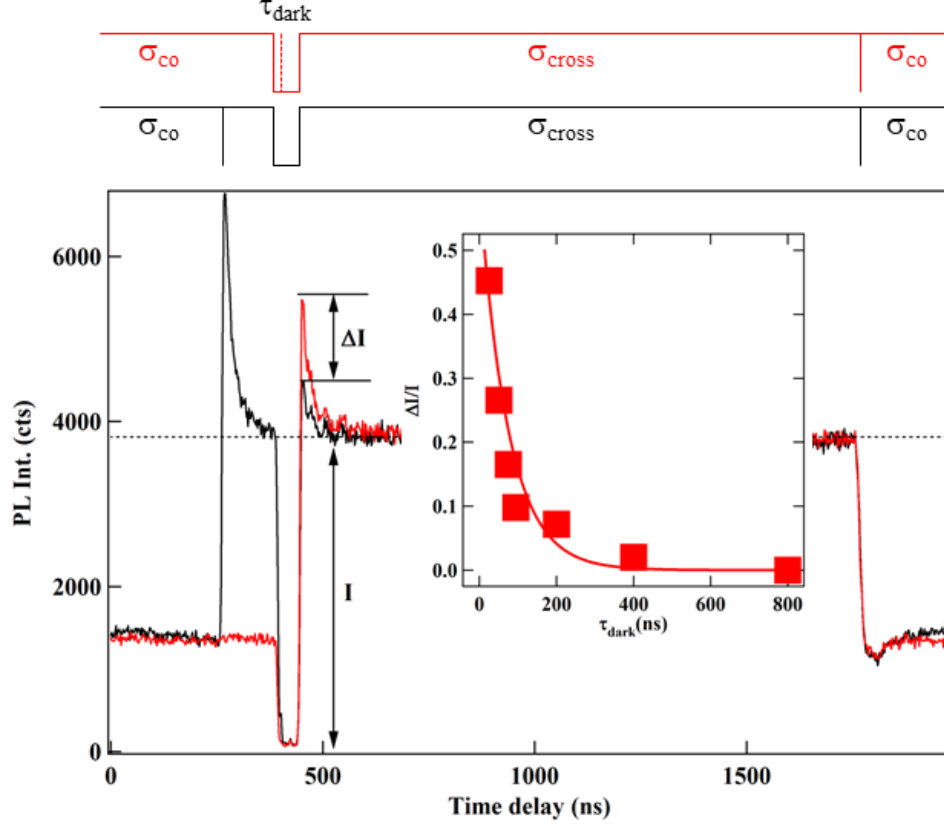


Figure I.10: Optical pumping experiment for an excitation of $|3, +2\rangle$ with modulated circular polarization. A dark time ($\tau_{dark} = 50ns$) is introduced either before (black) or during (red) the change of circular polarization. The black and red diagrams present the corresponding resonant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

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turpis. Fusce augue velit, scelerisque sollicitudin, dictum vitae, tempor et, pede. Donec wisi sapien, feugiat in, fermentum ut, sollicitudin adipiscing, metus.

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Table I.2: Material (CdTe or ZnTe) [1] 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	ρ	5860 kg/m ³
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	ρ	5908 kg/m ³
Quantum dot		
Hole Mn exchange energy	I_{hMn}	0.35 meV
hh-lh exciton splitting	Δ_{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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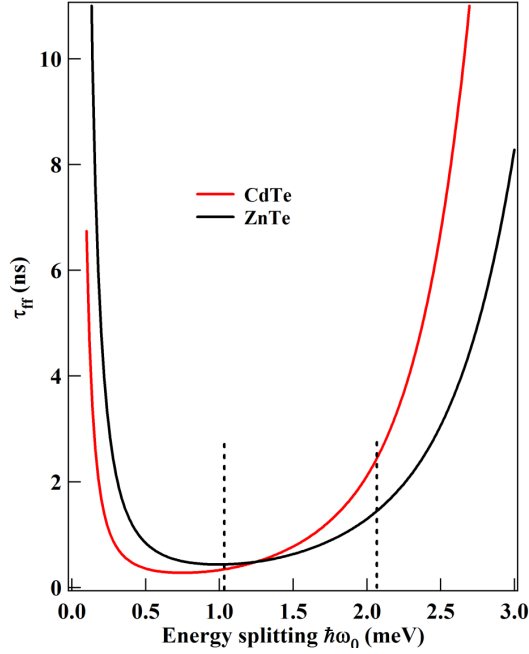


Figure I.11: Relaxation rates between the two h-Mn ground states of the Λ system τ_{ff} calculated with the material and QD parameters listed in table I.2 and a temperature $T=7K$. The vertical lines show the energy splitting of the hole-Mn states involved in the optical Λ systems identified in Fig. I.4.

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Nullam eleifend justo in nisl. In hac habitasse platea dictumst. Morbi nonummy. Aliquam ut felis. In velit leo, dictum vitae, posuere id, vulputate nec, ante. Maecenas vitae pede nec dui dignissim suscipit. Morbi magna. Vestibulum id purus eget velit laoreet laoreet. Praesent sed leo vel nibh convallis blandit. Ut rutrum. Donec nibh. Donec interdum. Fusce sed pede sit amet elit rhoncus ultrices. Nullam at enim vitae pede vehicula iaculis.

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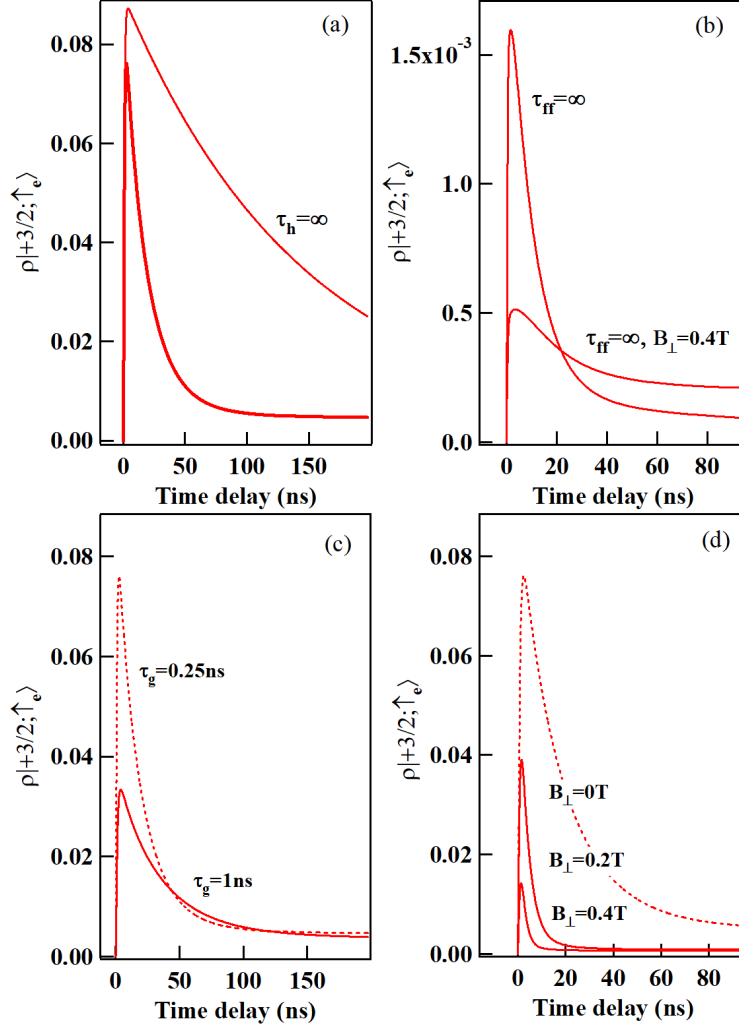


Figure I.12: (a) Calculated time evolution of $\rho_{|+3/2, \uparrow_e\rangle}(t)$ with the QD parameters listed in Table I.1 and (unless specified) $\tau_r=0.3\text{ns}$, $\tau_{Mn}=5\text{ }\mu\text{s}$, $\tau_h=10\text{ns}$, $\tau_g=0.25\text{ ns}$, $\tau_{ff}=1.5\text{ ns}$, $T_2^{hMn}=5\text{ ns}$,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

massa id pede dapibus ultricies. Sed eu leo. In at mauris sit amet tortor bibendum varius. Phasellus justo risus, posuere in, sagittis ac, varius vel, tortor. Quisque id enim. Phasellus consequat, libero pretium nonummy fringilla, tortor lacus vestibulum nunc, ut rhoncus ligula neque id justo. Nullam accumsan euismod nunc. Proin vitae ipsum ac metus dictum tempus. Nam ut wisi. Quisque tortor felis, interdum ac, sodales a, semper a, sem. Curabitur in velit sit amet dui tristique sodales.

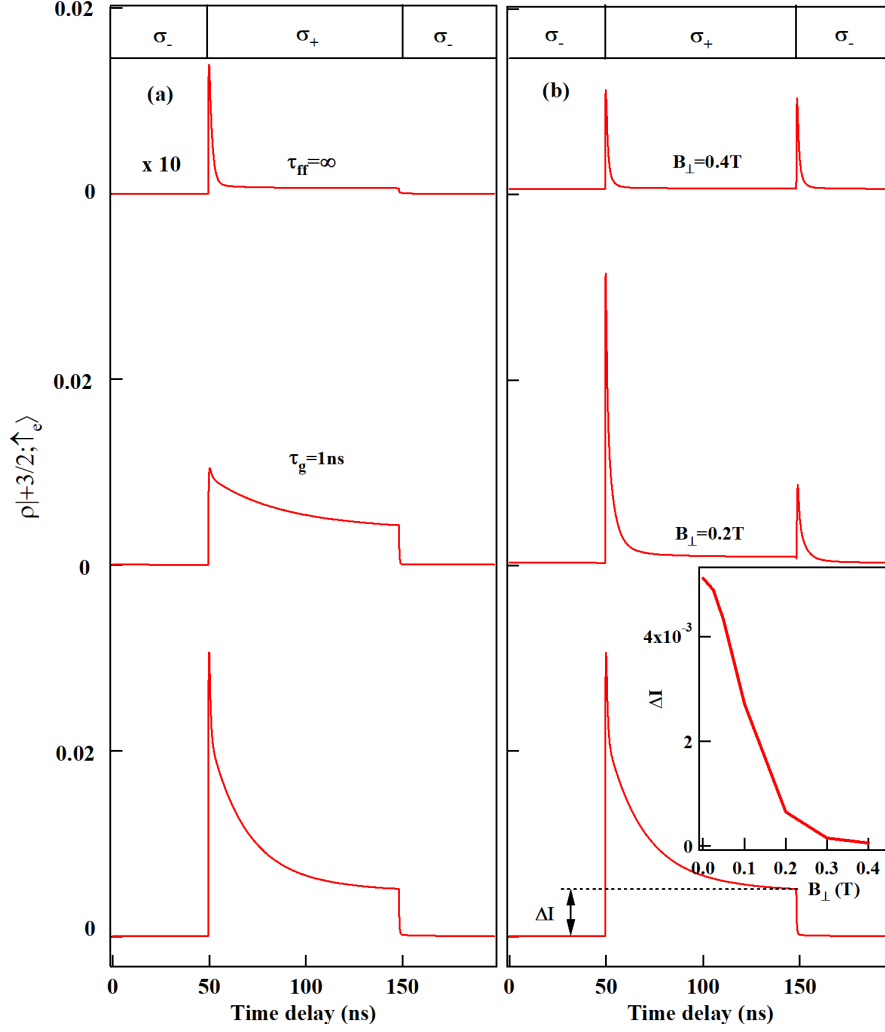


Figure I.13: Resonant optical pumping transients detected in $\sigma-$ polarization under excitation with modulated circular polarization of $|3, +2\rangle$ and $|3, -2\rangle$ calculated with the QD parameters listed in Table I.1 and $\tau_r = 0.3\text{ ns}$, $\tau_{Mn} = 5\text{ }\mu\text{s}$, $\tau_h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

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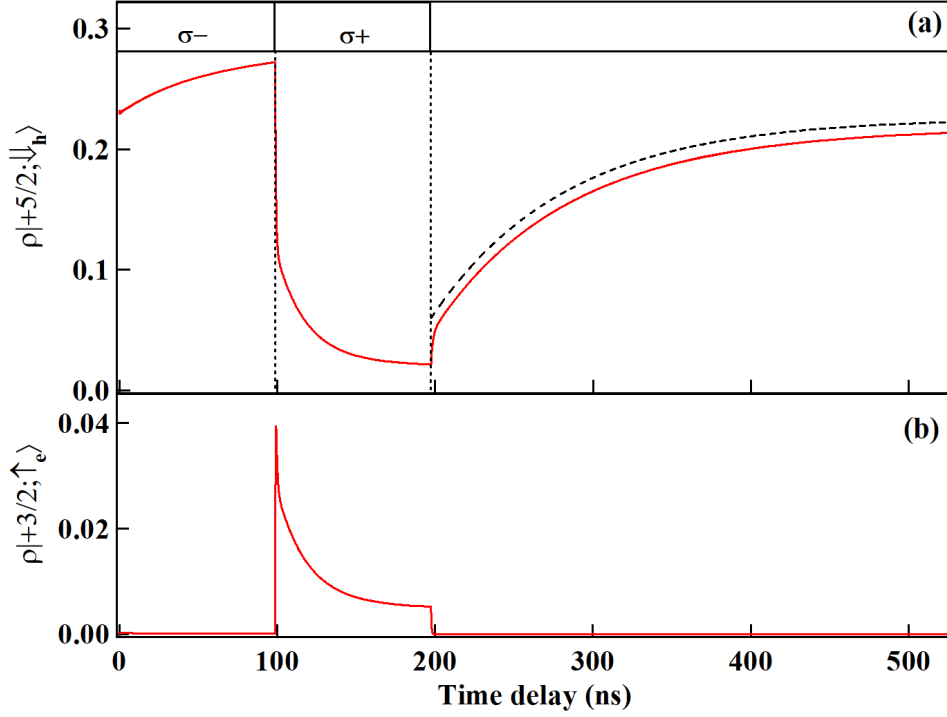


Figure I.14: (a) Calculated time evolution in the dark of the population of the hole-Mn state $|+\frac{5}{2}, \downarrow_h\rangle$ initialized by a sequence of $\sigma-/\sigma+$ resonant excitation of $|3, -2\rangle$ and $|3, +2\rangle$. The dashed black line (shifted for clarity) is an exponential fit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

I.3 Influence of the strain anisotropy

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, tempus sed, velit. Donec lacinia auctor libero.

Sed gravida lectus ut purus. Morbi laoreet magna. Pellentesque eu wisi. Proin turpis. Integer sollicitudin augue nec dui. Fusce lectus. Vivamus faucibus nulla nec lacus. Integer diam. Pellentesque sodales, enim feugiat cursus volutpat, sem mauris dignissim mauris, quis consequat sem est fermentum ligula. Nullam justo

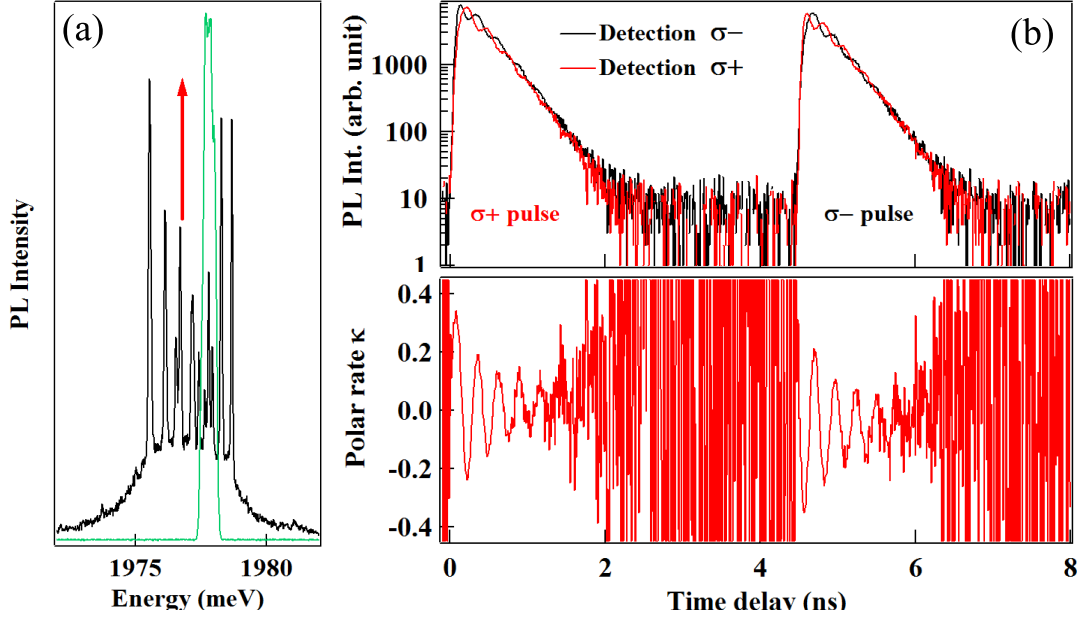


Figure I.15: (a) Configuration of the time resolved PL experiment for an excitation of $|3, +1\rangle$ (pulsed laser in green). (b) Top panel: Time resolved resonant PL of $|3, +1\rangle$ with a σ^+/σ^- sequence of laser pulses and a detection in σ^+ and σ^- polarization. Bottom panel: corresponding time dependence of the circular polarization rate $\kappa = (\sigma_- - \sigma_+)/(\sigma_- + \sigma_+)$.

lectus, condimentum sit amet, posuere a, fringilla mollis, felis. Morbi nulla nibh, pellentesque at, nonummy eu, sollicitudin nec, ipsum. Cras neque. Nunc augue. Nullam vitae quam id quam pulvinar blandit. Nunc sit amet orci. Aliquam erat elit, pharetra nec, aliquet a, gravida in, mi. Quisque urna enim, viverra quis, suscipit quis, tincidunt ut, sapien. Cras placerat consequat sem. Curabitur ac diam. Curabitur diam tortor, mollis et, viverra ac, tempus vel, metus.

Quisque consectetur. In suscipit mauris a dolor pellentesque consectetur. Mauris convallis neque non erat. In lacinia. Pellentesque leo eros, sagittis quis, fermentum quis, tincidunt ut, sapien. Maecenas sem. Curabitur eros odio, interdum eu, feugiat eu, porta ac, nisl. Curabitur nunc. Etiam fermentum convallis velit. Pellentesque laoreet lacus. Quisque sed elit. Nam quis tellus. Aliquam tellus arcu, adipiscing non, tincidunt eleifend, adipiscing quis, augue. Vivamus elementum placerat enim. Suspendisse ut tortor. Integer faucibus adipiscing felis. Aenean consectetur mattis lectus. Morbi malesuada faucibus dolor. Nam lacus. Etiam arcu libero, malesuada vitae, aliquam vitae, blandit tristique, nisl.

Maecenas accumsan dapibus sapien. Duis pretium iaculis arcu. Curabitur ut

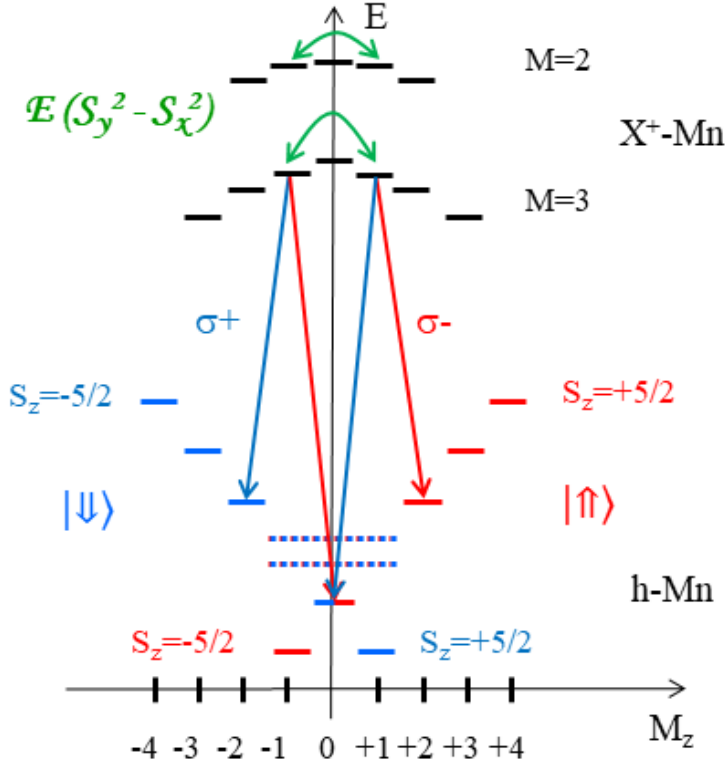


Figure I.16: Energy levels of the ground ($h\text{-Mn}$) and excited ($X^+\text{-Mn}$) states as a function of their angular momentum (M_z). The $e\text{-Mn}$ states $|3, +1\rangle$ and $|3, -1\rangle$, as well as $|2, +1\rangle$ and $|2, -1\rangle$, are coupled by the strain anisotropy $E(S_y^2 - S_x^2)$.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

lacus. Aliquam vulputate. Suspendisse ut purus sed sem tempor rhoncus. Ut quam dui, fringilla at, dictum eget, ultricies quis, quam. Etiam sem est, pharetra non, vulputate in, pretium at, ipsum. Nunc semper sagittis orci. Sed scelerisque suscipit diam. Ut volutpat, dolor at ullamcorper tristique, eros purus mollis quam, sit amet ornare ante nunc et enim.

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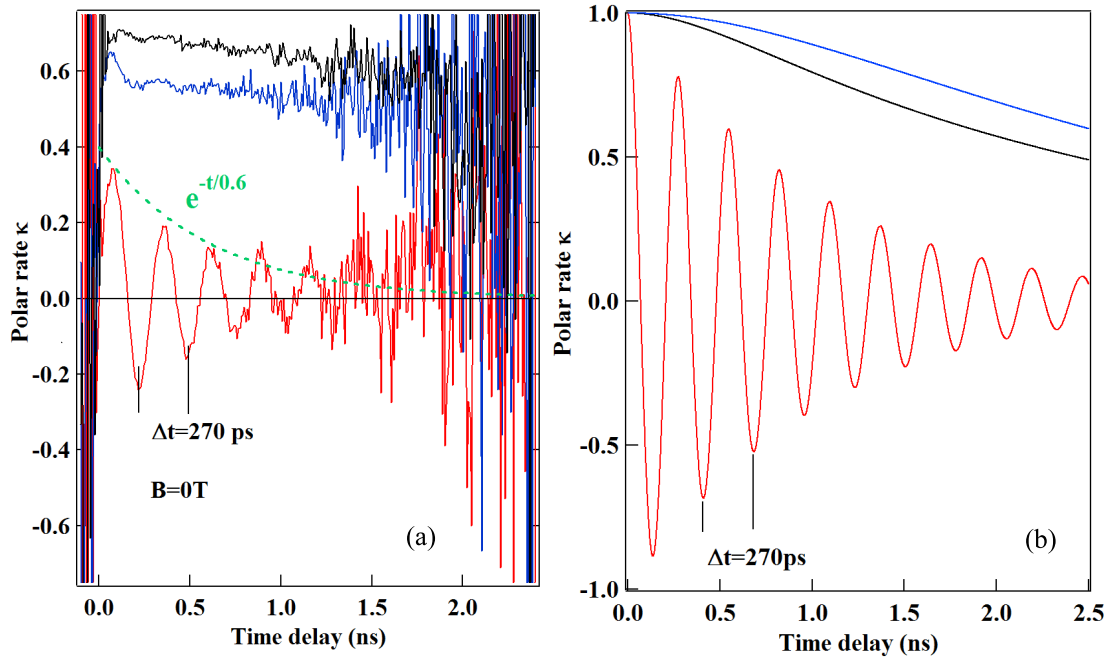


Figure I.17: (a) Time dependence of the circular polarization rate of the resonant PL of the states $|3, +1\rangle$ (red), $|3, +2\rangle$ (black) and $|2, +2\rangle$ (blue). (b) Corresponding polarisation rates calculated with $D_0 = 7\mu\text{eV}$ [2], $T_2^{eMn} = 0.6\text{ns}$, $E = 1.8\mu\text{eV}$, a radiative lifetime $T_r = 0.3\text{ns}$ and the parameters listed on Table I.1.

venenatis placerat. Aenean sed odio. Quisque posuere purus ac orci. Vivamus odio. Vivamus varius, nulla sit amet semper viverra, odio mauris consequat lacus, at vestibulum neque arcu eu tortor. Donec iaculis tincidunt tellus. Aliquam erat volutpat. Curabitur magna lorem, dignissim volutpat, viverra et, adipiscing nec, dolor. Praesent lacus mauris, dapibus vitae, sollicitudin sit amet, nonummy eget, ligula.

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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

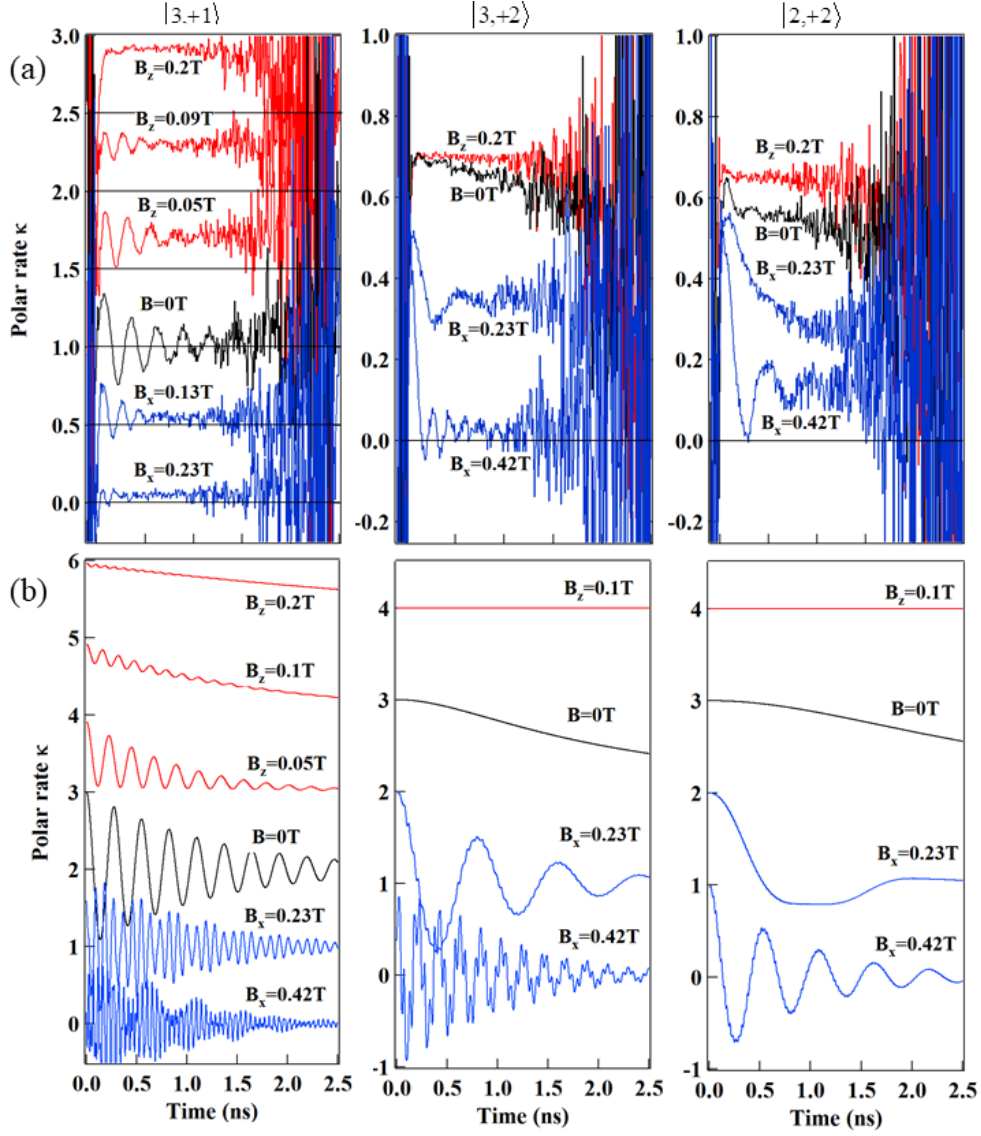


Figure I.18: (a) Influence of a longitudinal (B_z , red) and a transverse (B_x , blue) magnetic field on the time dependence of the circular polarization rate $\kappa = (\sigma_- - \sigma_+)/(\sigma_- + \sigma_+)$ of the resonant PL of $|3, +1\rangle$, $|3, +2\rangle$ and $|2, +2\rangle$. On the top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

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.

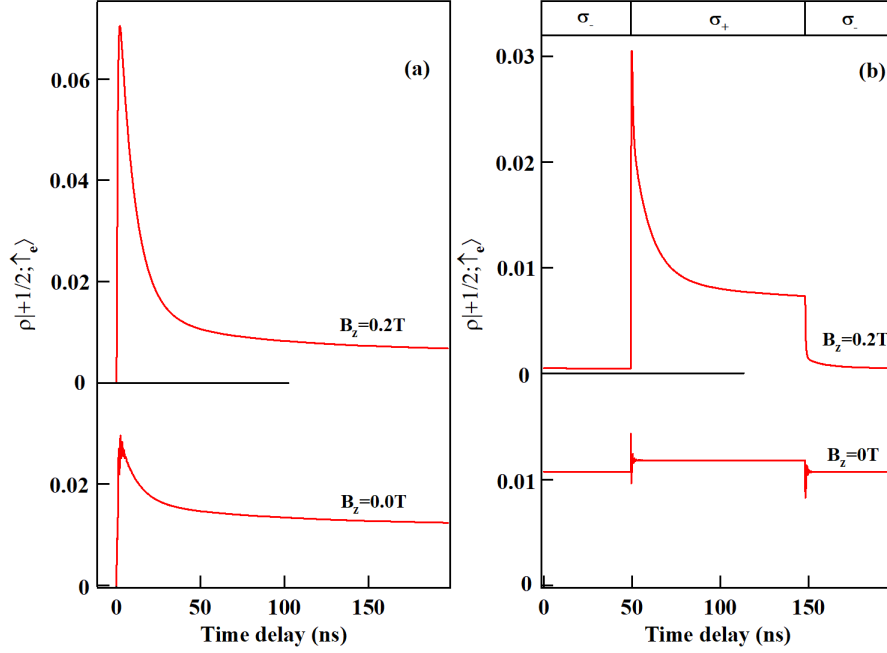


Figure I.19: (a) Calculated time evolution of $\rho_{|+1/2, \uparrow_e\rangle}(t)$ for a resonant excitation of $|3, +1\rangle$ and $|3, -1\rangle$ with the QD parameters listed in Table I.1 without and with a longitudinal magnetic field. (b) Corresponding calculated optical pumping transients under excitation with modulated circular polarization.

Morbi justo. Aenean nec dolor. In hac habitasse platea dictumst. Proin non-ummy 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. 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.

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