Chapter I

Optical control of an individual Cr spin in a CdTe QD

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I.1 Optical pumping and spin dynamics in a Cr doped QD

I.1.1 Resonant optical pumping of the Cr spin

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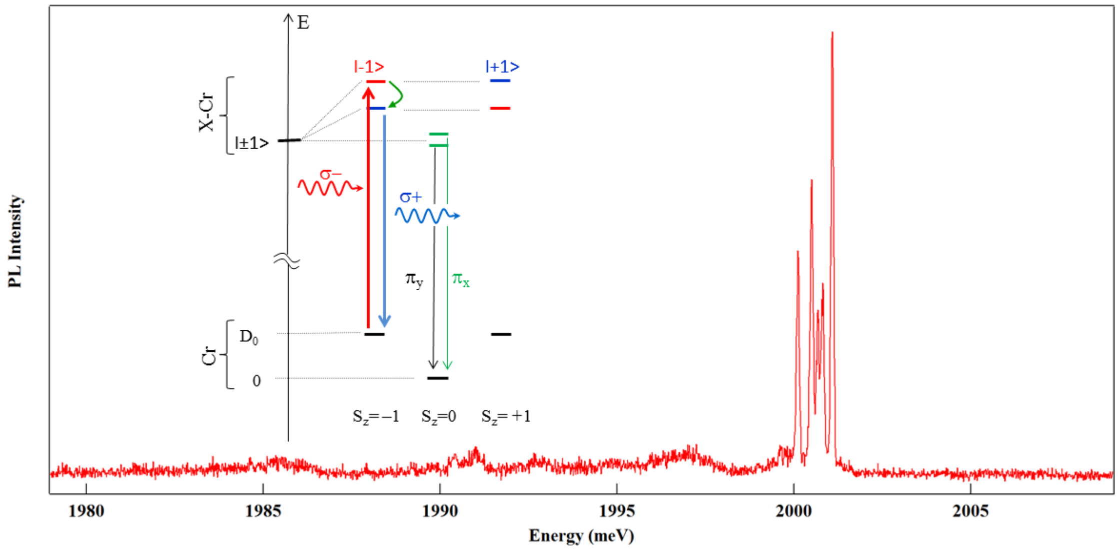

Figure I.1: Low temperature PL spectra of QD2 exciton in co-polarized excitation and detection for B = 0T. Inset: Schematic of the energy levels in a Cr-doped QD and configuration of excitation/detection for resonant optical pumping. The ground states $S_z = 0, \pm 1$ are split by the magnetic anisotropy $D_0 S_z^2$. In the excited state (X-Cr), the exchange interaction with the bright exciton ($| \pm 1\rangle$) split the states $S_z = \pm 1$. The higher energy Cr states, $S_z = \pm 2$, are not displayed.

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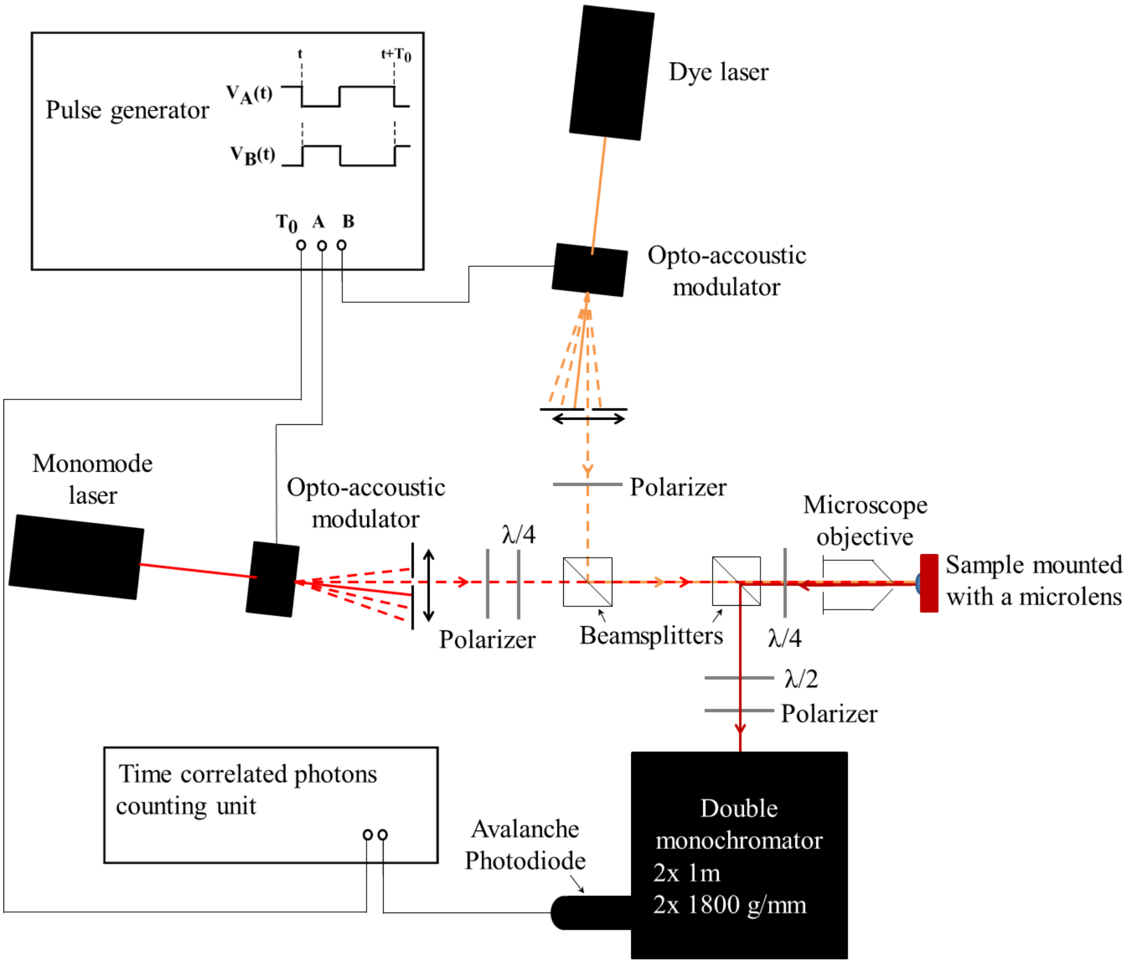

Figure I.2: Schematic view of the micro-spectroscopy set-up used for the time-resolved optical pumping experiment. The monomode laser is a dye laser tuned on resonance with the studied dot transition, acting as the pump. The other dye laser is tuned on a quasi-resonant state, acting as the probe. Both beamsplitters are non-polarizing.

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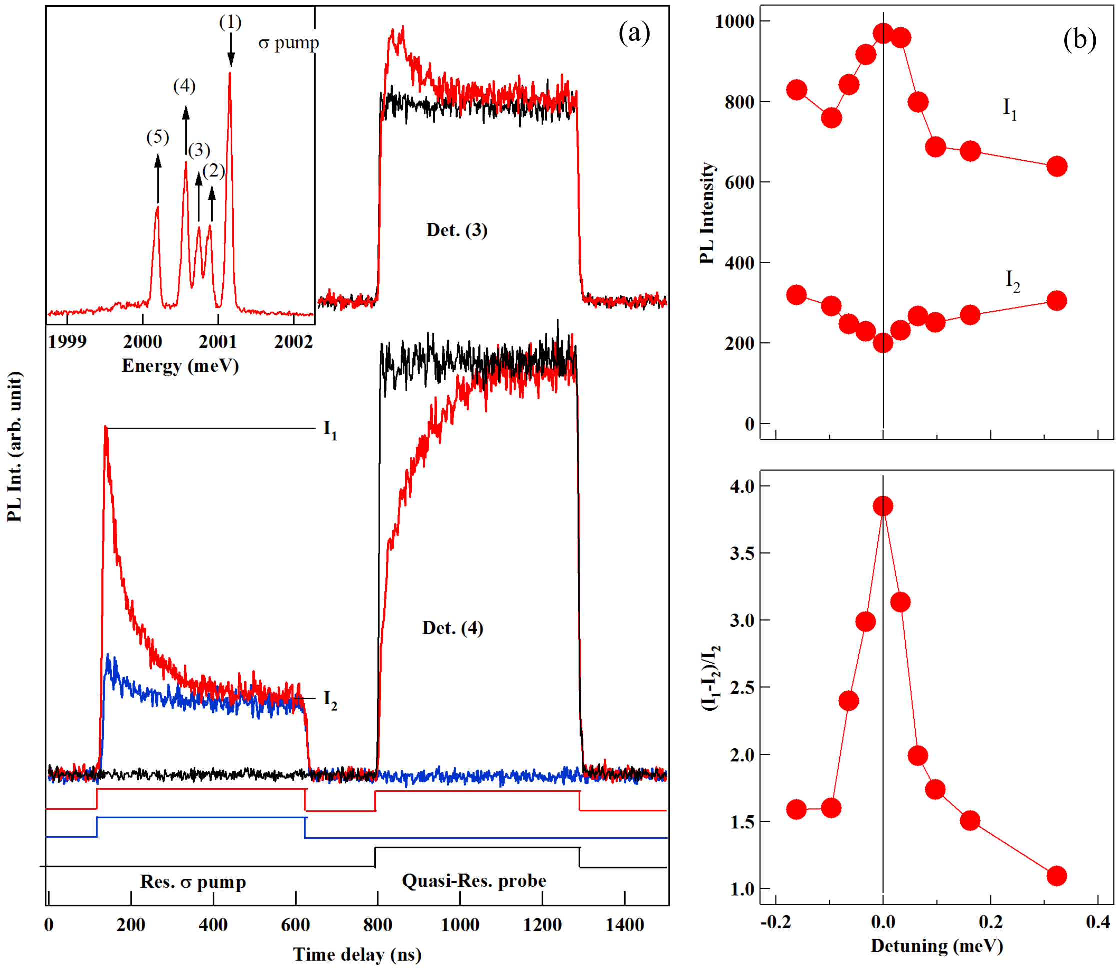

Figure I.3: (a) PL transients recorded in circular polarization on line (3) and on line (4) (as defined in the inset) under the resonant (pump on (1)) and quasi-resonant (probe at $E_{exc.} \approx 2068$ meV) optical excitation sequences displayed at the bottom. Inset: PL of X-Cr and configuration of the resonant excitation and detection. (c) and (d): Energy detuning dependence of resonant PL intensity (I_1, at the beginning and I_2, at the end of the pump pulse) and of the corresponding normalized amplitude of pumping transient $(I_1 - I_2)/I_2$.

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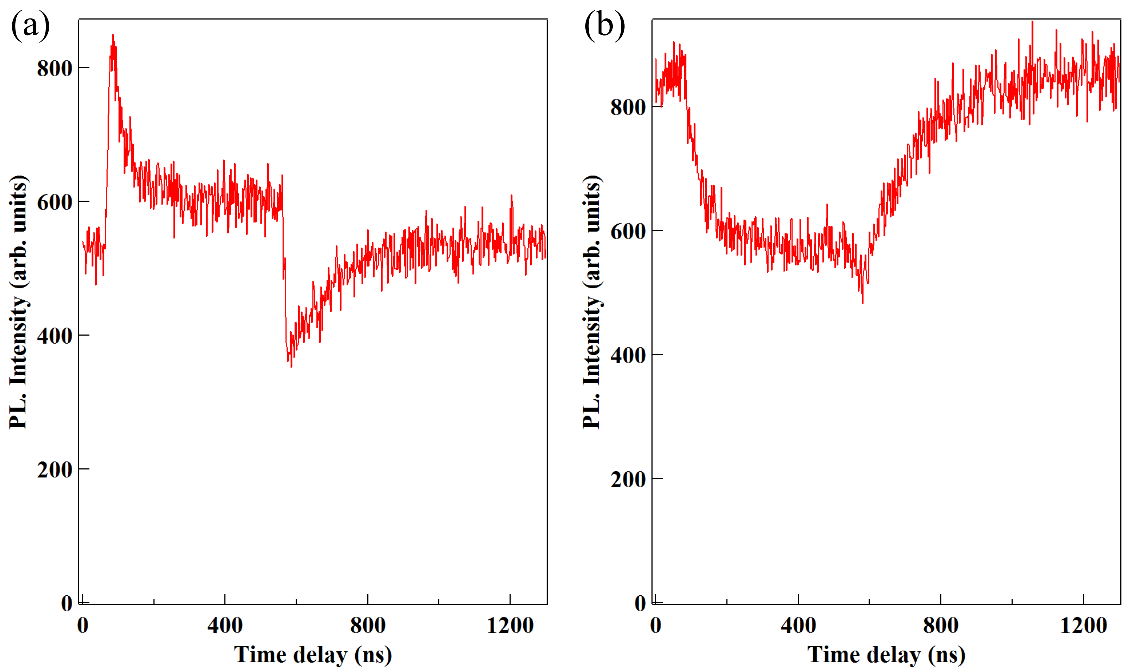

Figure I.4: PL transient recorded when probing with a laser tuned in resonance with (a) the line (1) and detecting on the line (4), and (b) the line (4) and detecting on the line (1). Unlike in Fig. I.3 where the quasi-resonant laser was only turned on for a short amount of time, it was now left on during the entirety of the measurement.

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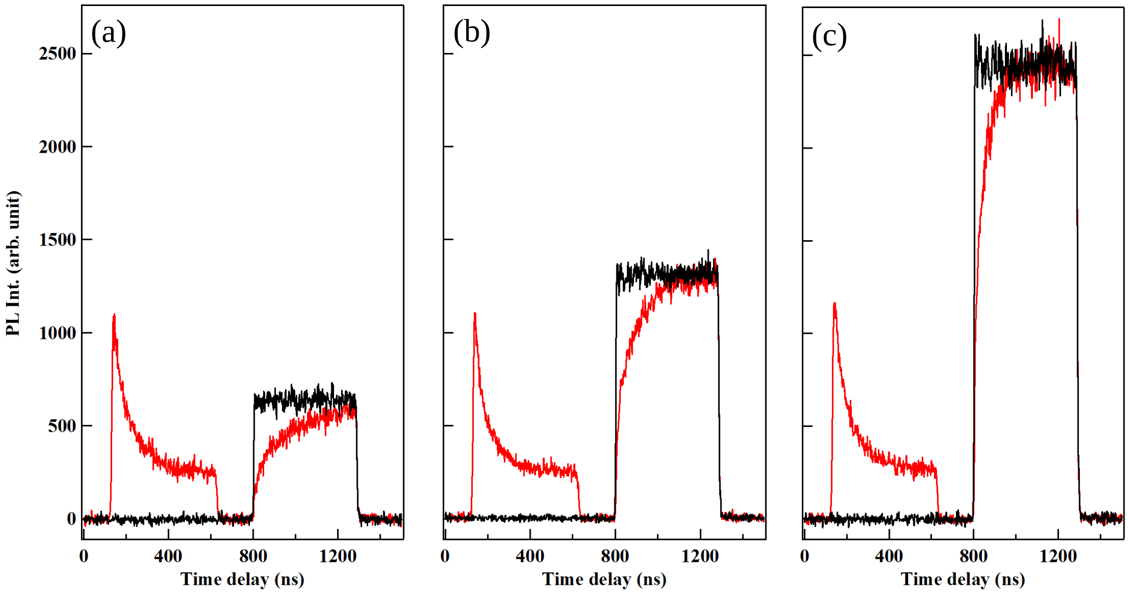

Figure I.5: PL transients measured under variation of the probe power: (a) $P_{probe} = 125\ \mu$W. (b) $P_{probe} = 250\ \mu$W. (c) $P_{probe} = 500\ \mu$W. The pump was either off (dark line) or tuned in resonance with the (1) line at $P_{pump} = 250\ \mu$W.

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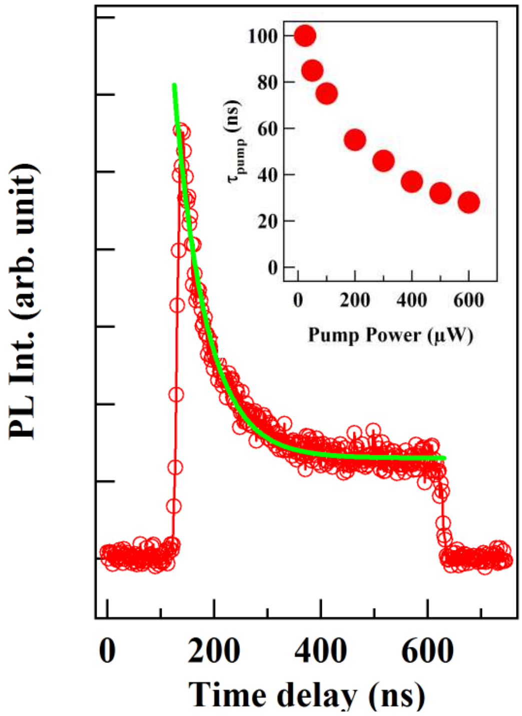

Figure I.6: PL transient measured with the resonant pump pulse for a power $P_{probe} = 240\ \mu$W. The exponential fit is drawn as a green line and gives a characteristic time of $\tau_{pump} = 60$ ns. The inset presents the evolution of τ_{pump} in function of the pumping laser power.

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I.1.2 Dynamics of the Cr spin under optical excitation

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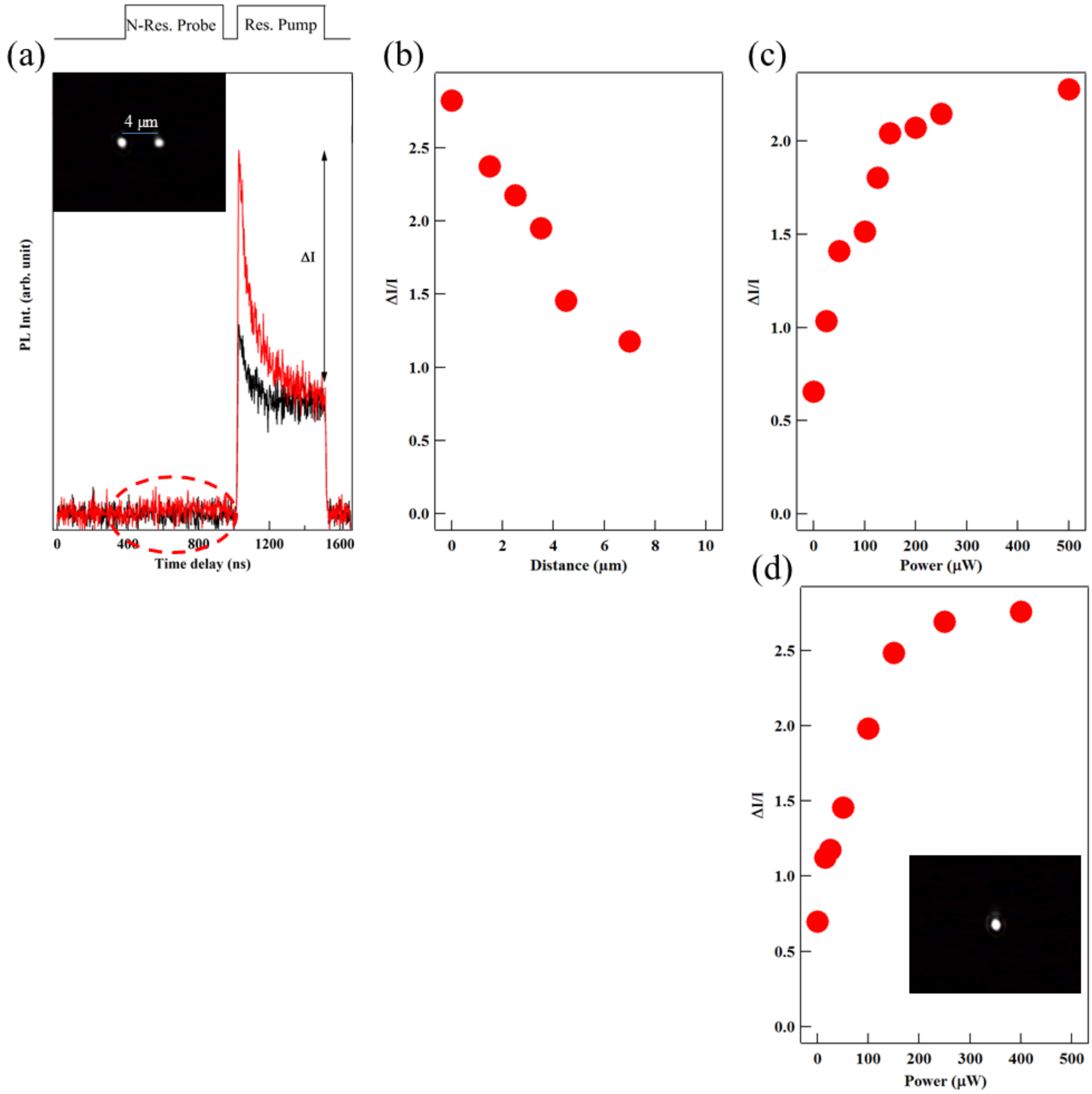

Figure I.7: (a) PL transients recorded for a probing pulse at $d = 4$ μm of the dot for a power $P_{probe} = 250$ μW (red curve) and with no probing pulse (dark curve). The resonant pumping pulse is kept at the dot position. Right to it are presented the evolution of the ratio $\Delta I/I$ function of (b) the distance between the probing pulse and the dot, and (c) the power of the probing pulse for distance between the pulse position and the dot $d = 4$ μm. (d) Evolution of the ratio $\Delta I/I$ function of the power of the probing pulse for a pulse at the dot position, tuned at $E = 2.01$ meV, corresponding to a transparency of the dot and the barriers.

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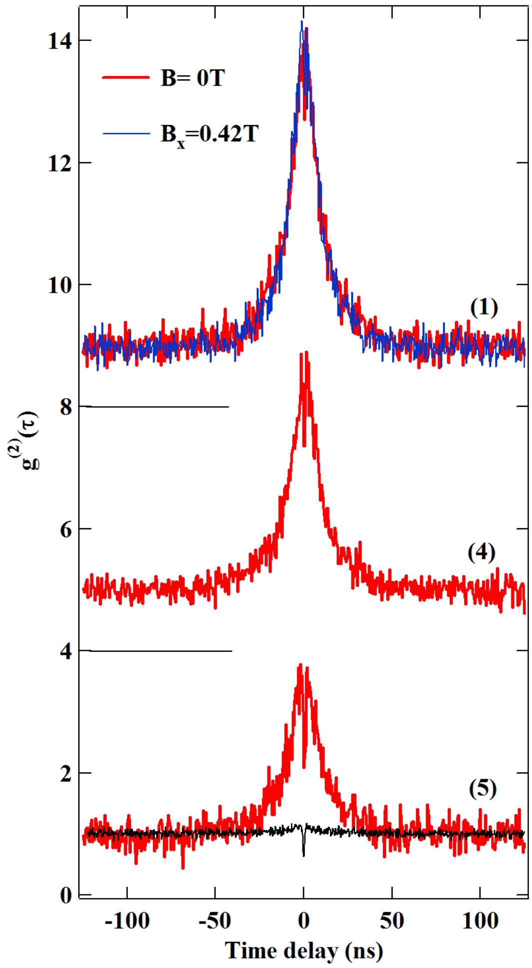

Figure I.8: Auto-correlation of the PL intensity collected in circular polarization on the X-Cr lines (1), (4) and (5) (as defined in Fig. I.3) and compared with the auto-correlation of the exciton in a non-magnetic QD (black line). The curves are shifted for clarity. For line (1), the auto-correlation is also recorded under a transverse magnetic field (blue line).

To observe the time fluctuations of the Cr spin under CW optical excitation, we used the statistics of time arrivals of the photons emitted by a Cr-doped QD given by the second order correlation function $g^{(2)}(\tau)$ of the PL intensity. Fig. **??** shows $g^{(2)}(\tau)$ for the lines (1), (3) and (4) recorded in circular polarization. These signals are compared with the auto-correlation obtained for the PL of a non-magnetic QD which is characteristic of a single-photon emitter with a dip (anti-bunching) at short delays. The width of the anti-bunching is given by the lifetime of the emitter and the generation rate of excitons and its depth is limited by the time resolution of the HBT setup. As illustrated in Fig. **??**, typical non-magnetic CdTe/ZnTe QDs do not present any significant bunching induced by charge fluctuations [1, 2]. A similar auto-correlation on a X-Cr PL line still presents a reduced coincidence rate near zero delay, but it is mainly characterized by a large photon bunching with a full width at half maximum (FWHM) in the 20 ns range. This large bunching reflects an intermittency in the emission of a given line of the QD coming from fluctuations of the Cr spin in a 10 ns timescale as it will be confirmed by cross-correlation measurements.

The amplitude of the bunching reaches 5 for line (1) and is slightly weaker for the lower energy lines. In a simple picture of blinking where the selected QD line can be either in a state ON or OFF, the amplitude of the bunching is given by Γ_{OFF}/Γ_{ON}, ratio of the transition rates from OFF to ON, Γ_{ON}, and from ON to OFF, Γ_{OFF} [3]. An amplitude of bunching larger than 1 is then expected in a multilevel spin system where, after a spin relaxation, multiple spin-flips are usually required to come back to the initial state ($\Gamma_{ON} < \Gamma_{OFF}$). Let us finally note that the bunching signal is not affected by a weak transverse magnetic field ($B_x = 0.42$T in Fig. **??**). This confirms the presence of a large strain induced magnetic anisotropy D_0 which splits the Cr and X-Cr states and blocks their precession in a magnetic field.

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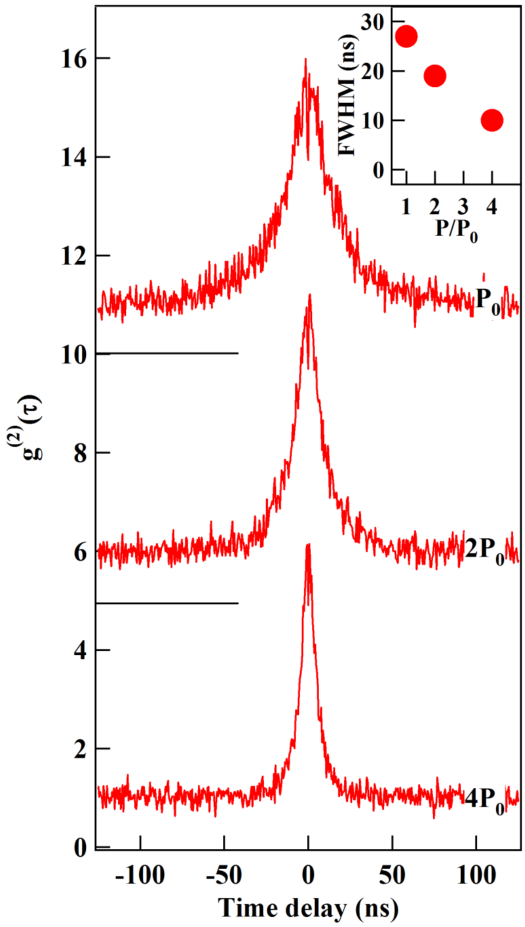

Figure I.9: Auto-correlation of the PL intensity recorded in circular polarization on the high energy X-Cr line (1) for different excitation powers. The inset shows the corresponding FWHM of the bunching signal versus excitation power.

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I.1.3 Spin relaxation in the dark

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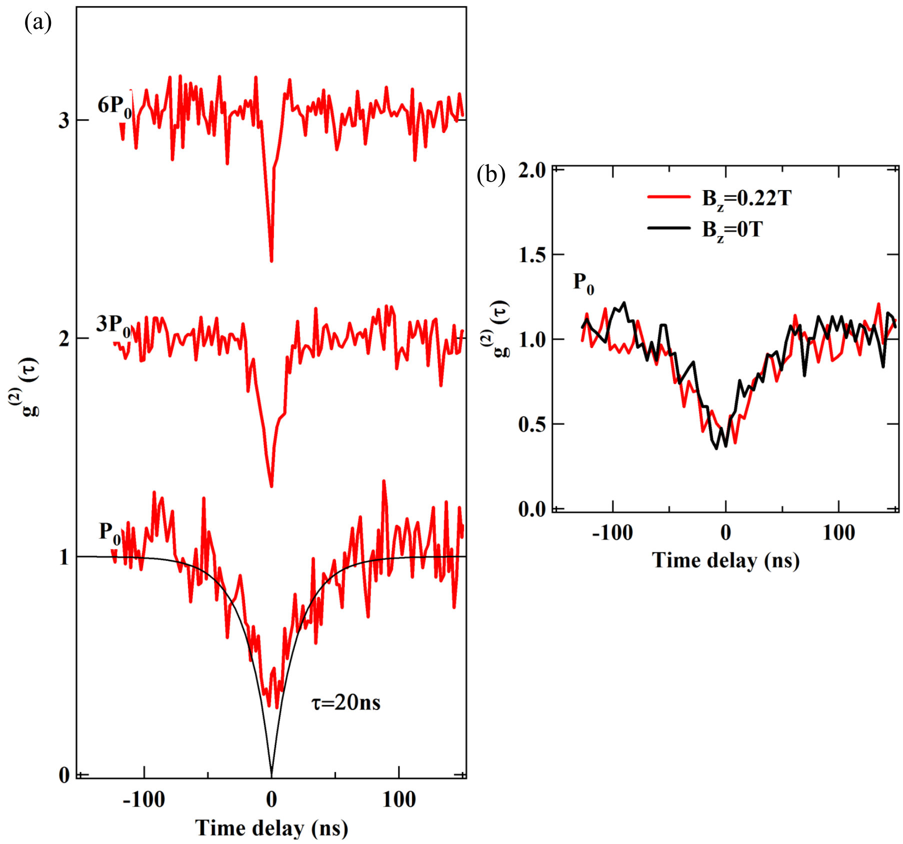

Figure I.10: (a) Correlation signal of the PL intensity of lines (1) and (4) recorded in the same circular polarization (cross-correlation) for three different excitation powers. The curves are shifted for clarity. The black line is an exponential fit with a characteristic time $\tau = 20$ ns. (b) Longitudinal magnetic field dependence of the cross-correlation signal obtained at low excitation power.

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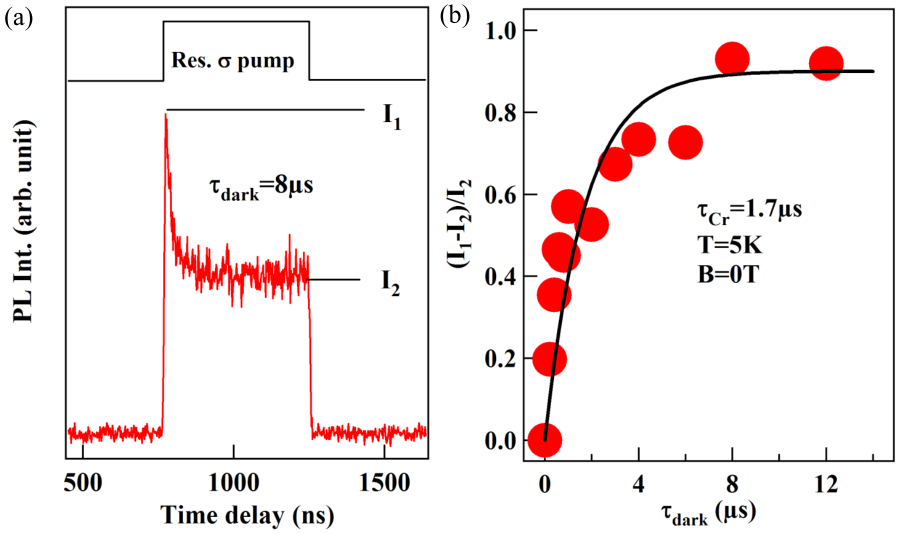

Figure I.11: (a) Time evolution of the PL intensity of line (4) of X-Cr under resonant excitation on line (1) with a circularly polarized excitation pulse. (b) Evolution of the amplitude of the pumping transient $(I_1 - I_2)/I_2$ as a function of the dark time between the excitation pulses. The black line is an exponential evolution with a characteristic time $\tau_{Cr} = 1.7\ \mu$s

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The $\Delta I/I$ evolution over dark time is used as a measure of the relaxation of the Cr spin: after a long enough time, both lines should stabilize at the same value.

I.2 Optical Stark effect on an individual Cr spin

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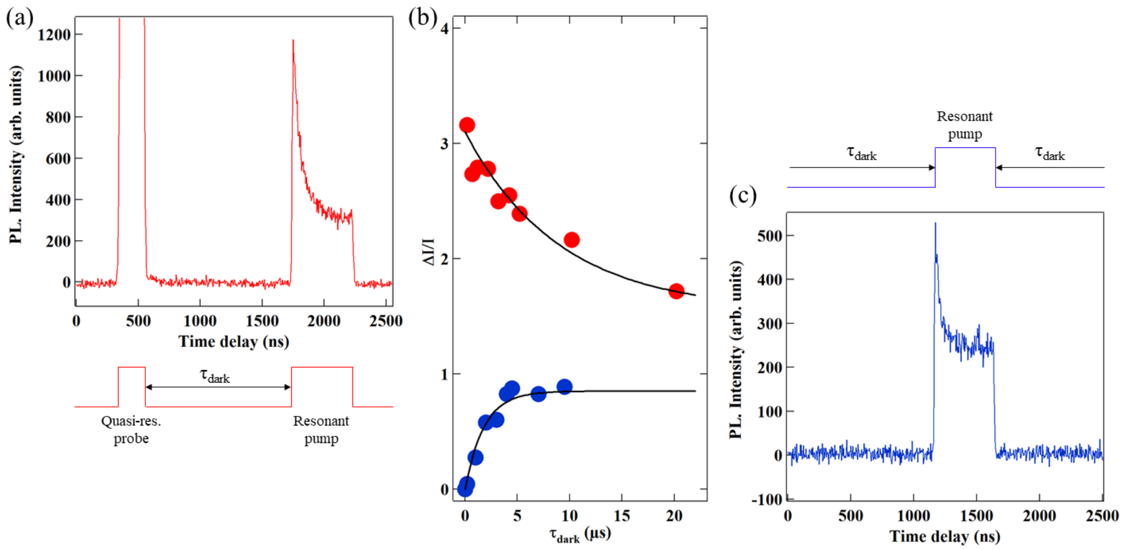

Figure I.12: Comparison of the relaxation of the Cr spin either after the quasi-resonant pulse (red, $E_{pump} = 2.07$ meV) or a resonant pump on the (1) line (blue). (a) Configuration to probe the relaxation after the quasi-resonant pulse. The dark time τ_{dark} between the quasi-resonant probe and the resonant pump is varied and the $\Delta I/I$ as defined in Fig. I.3 is calculated for each τ_{dark}. (b) $\Delta I/I$ in function of the dark time τ_{dark} measured for the relaxation after a quasi-resonant pulse (red) or after a pumping pulse (blue). (c) Configuration to probe the relaxation after a resonant pulse, as presented in Fig. I.11.

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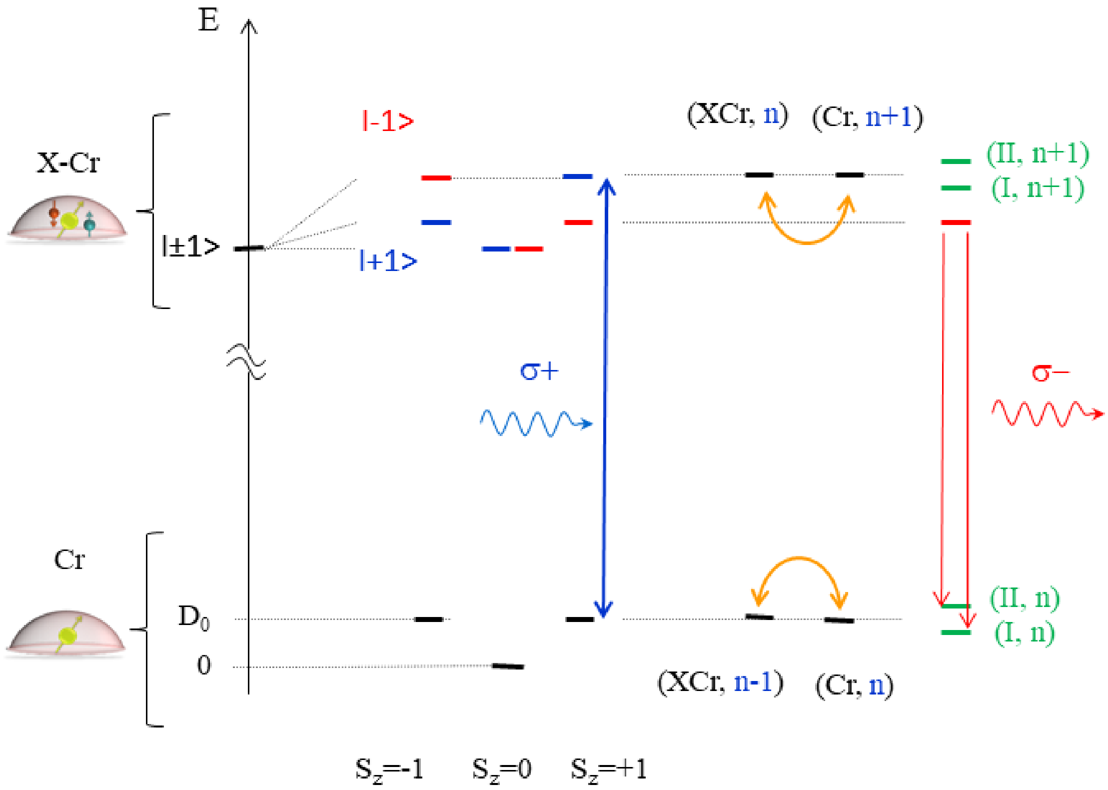

Figure I.13: Energy level of a Cr-doped quantum dot and formation of the dressed-states. The Cr fine structure is controlled by the strain induced magnetic anisotropy. The exchange interaction with the bright exciton further splits the $|S_z = \pm 1\rangle$ levels. We excite resonantly a given transition of the quantum dot with a continuous monochromatic laser (here the $|S_z = +1\rangle \rightarrow |S_z = +1, X_z = +1\rangle$ transition). Considering a mode of n, the levels (Cr, n) and (X-Cr, $n-1$) are coherently coupled through absorption and stimulated emission of photon. The eigenstates of the coupled atom-field system (I, n) and (II, n) are entangled states which, at resonance, have equal contributions of the upper and the lower of the laser-driven transition. The Rabi splitting Ω_r between these two levels can be probed in the cross-polarized PL of the exciton or the biexciton using a second non-resonant probe (as shown on the right part of the diagram). The splitting observed using a third level is the so-called Autler-Townes splitting.

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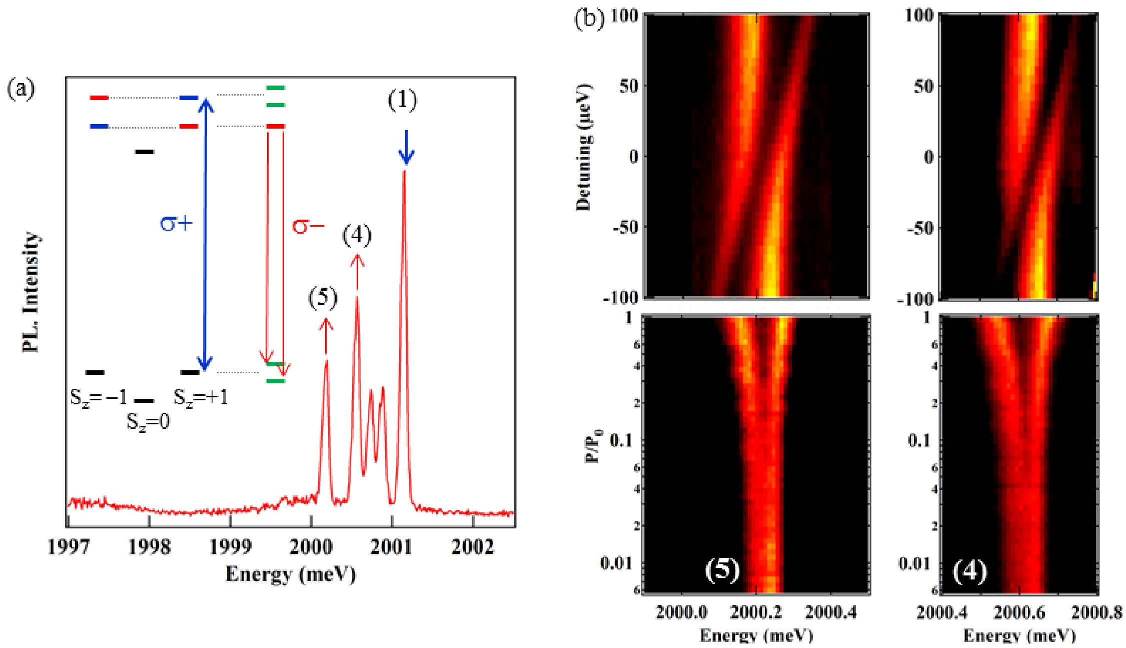

Figure I.14: (a) PL of X-Cr and configuration of excitation in the resonant optical control experiments. The inset illustrate the laser induced splittings in the ground and excited states for a $\sigma+$ excitation on $S_z = -1$. (b) PL intensity maps of lines (5) and (4) for an excitation on (1) as a function of the detuning (top) and of the excitation intensity (bottom). The PL is produced by a second non-resonant laser.

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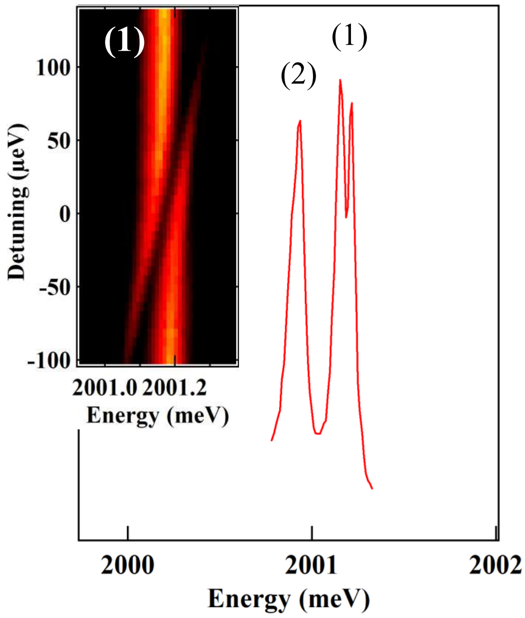

Figure I.15: PL of line (1) and (2) (high energy line) for a laser on resonance with the dark exciton state (5). Inset: PL intensity map of line (1) as a function of the laser detuning around (5).

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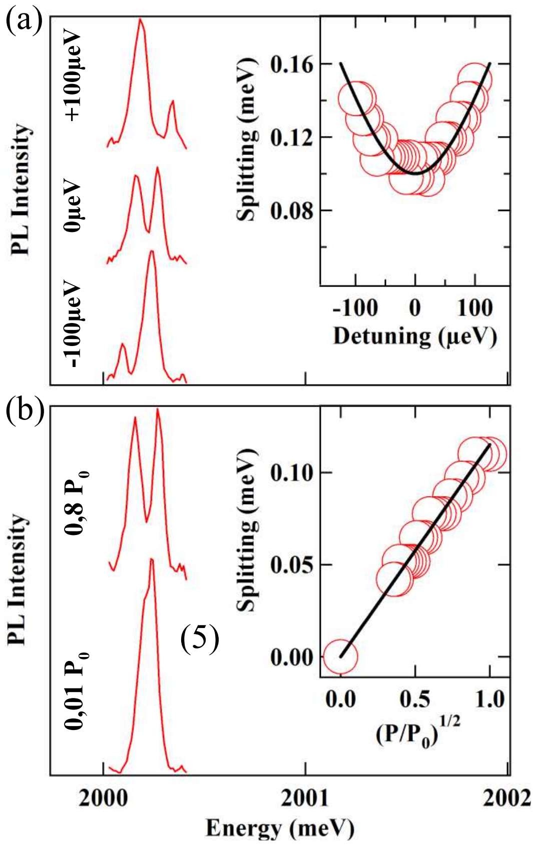

Figure I.16: Line (5) PL spectra taken for (a) no and max detuning in each direction, and (b) low and high power. The insets show the splitting of the PL doublet as a function (a) of the laser detuning and (b) of the excitation intensity. The fit is obtained with $\hbar\Omega_r = 100$ μeV.

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