

# Chapter I

## Dynamics and optical control of an individual Cr spin in a CdTe QD

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### I.1 Resonant optical pumping and spin dynamics in a Cr doped QD

#### I.1.1 Resonant optical pumping of the Cr spin

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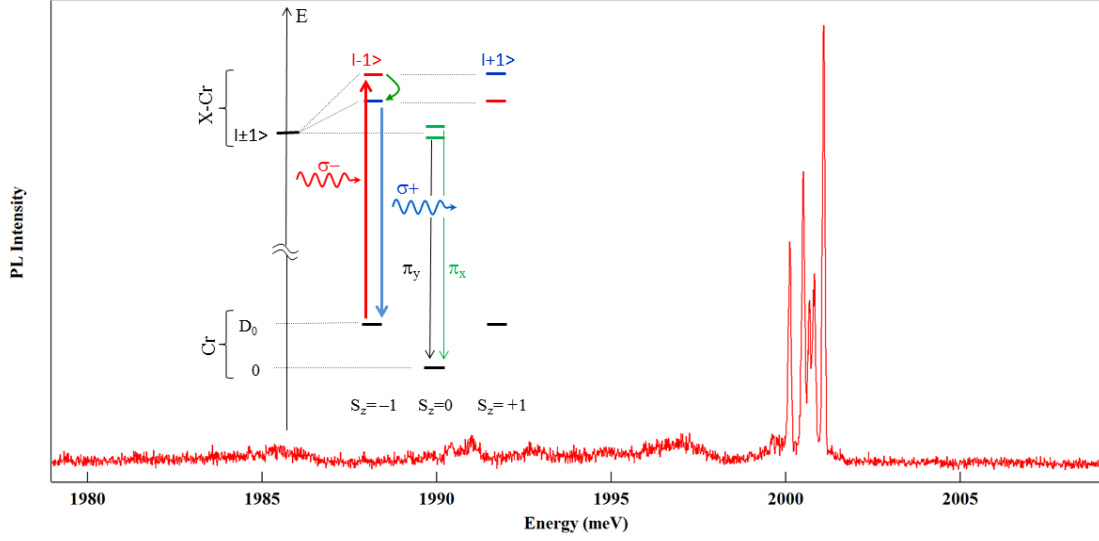


Figure I.1: Low temperature PL spectra of QD2 exciton in linearly polarized excitation and detection for  $B = 0\text{T}$ . No contribution of the charge excitons was found. Inset: Schematic of the energy levels in a Cr-doped QD and configuration of excitation/detection for resonant optical pumping. 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  $S_z = \pm 1$ .

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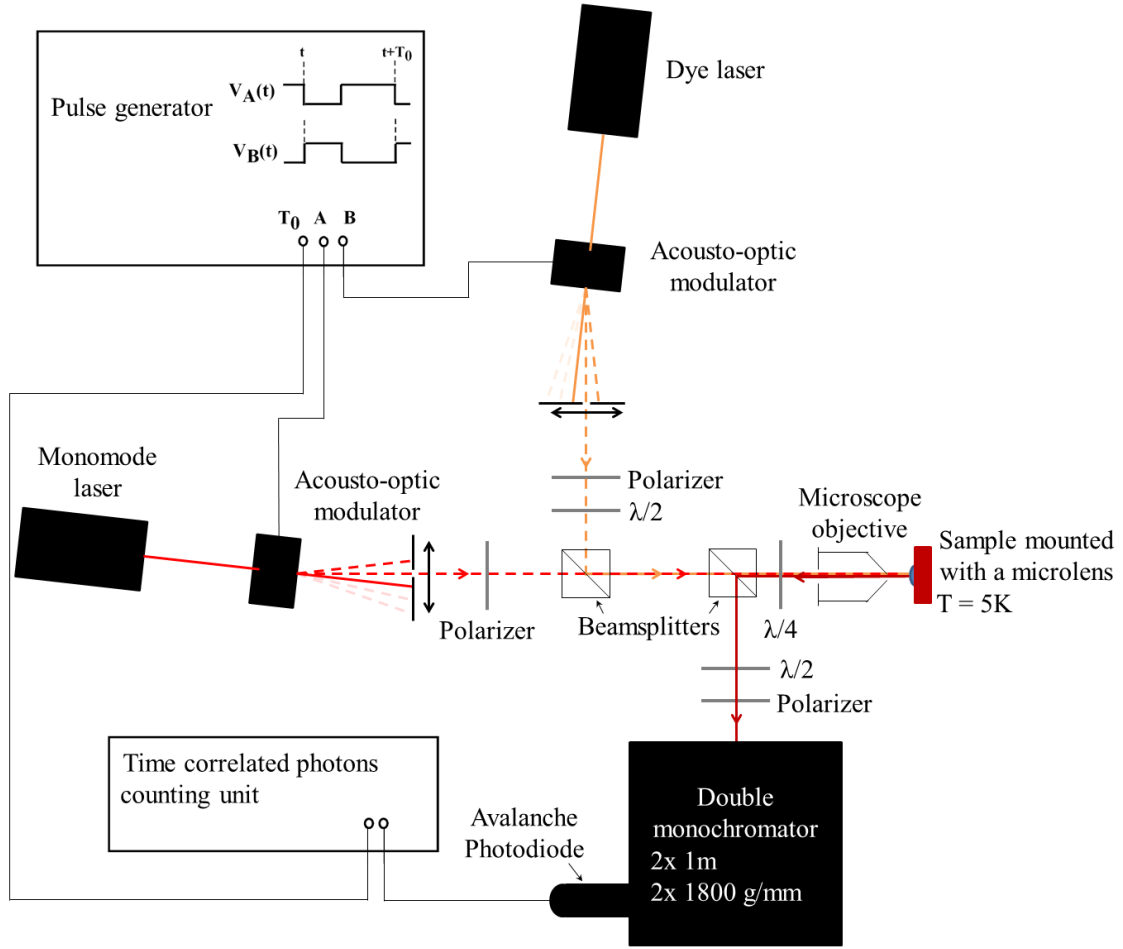


Figure I.2: Schematic view of the micro-spectroscopy set-up used for the time-resolved optical pumping experiment. The monomode laser is a dye laser tuned on resonance with the studied dot transition, acting as the pump. The other dye laser is tuned on a resonant state, acting as the probe. Both beamsplitters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

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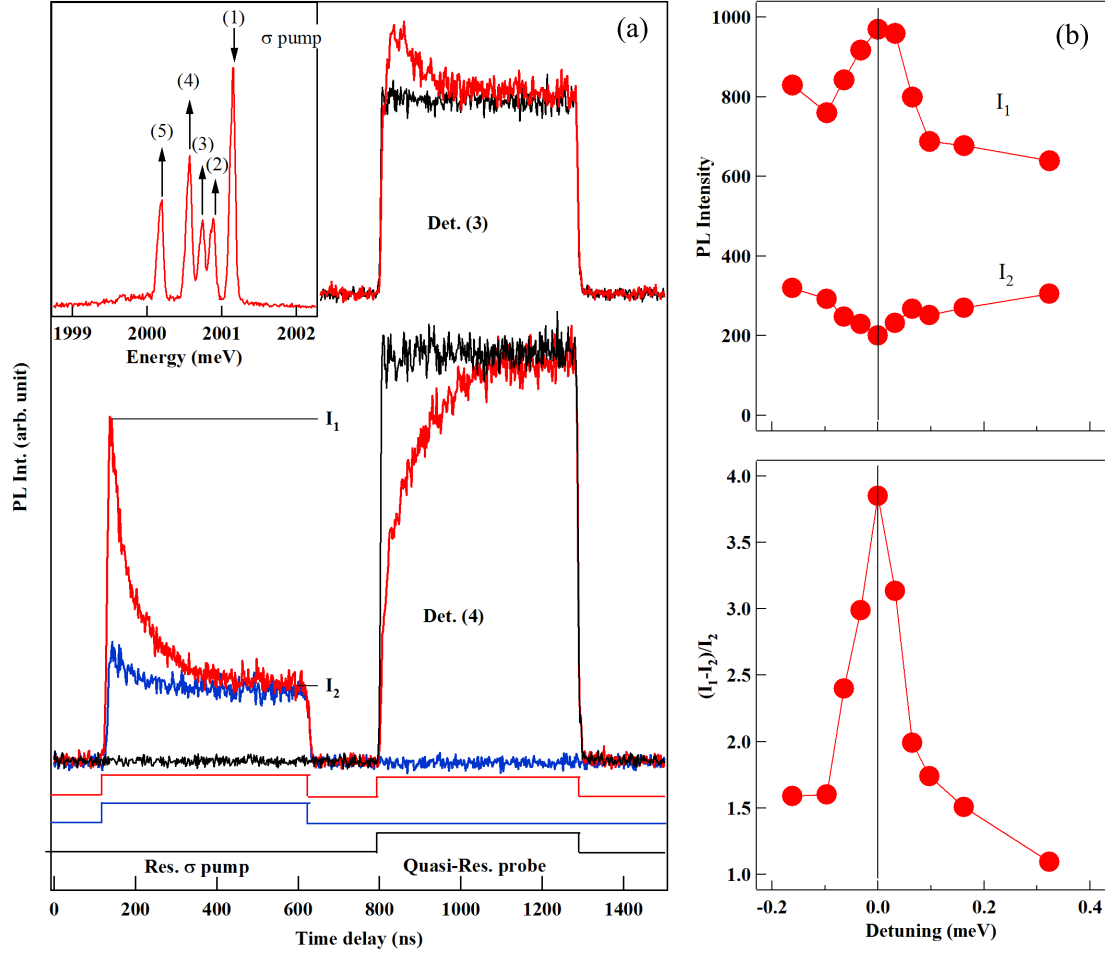


Figure I.3: (a) PL transients recorded in circular polarization on line (3) and on line (4) (as defined in the inset) under the resonant (pump on (1)) and quasi-resonant (probe at  $E_{exc.} \approx 2068$  meV) optical excitation sequences displayed at the bottom. Inset: PL of X-Cr and configuration of the resonant excitation and detection. (b) and (c): 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  $\Delta I/I_2 = (I_1 - I_2)/I_2$ .

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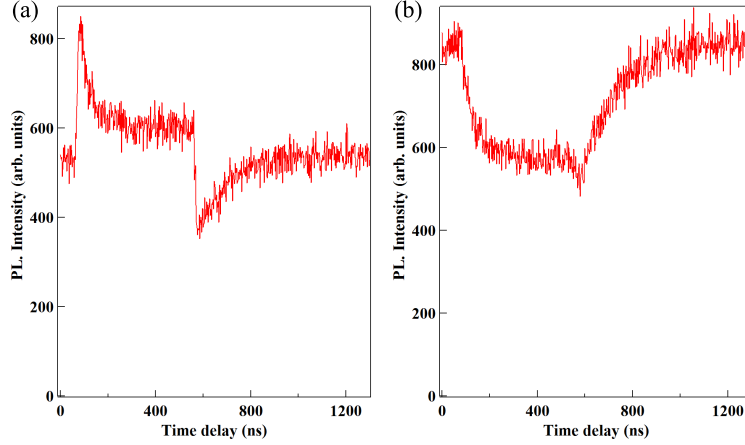


Figure I.4: Optical pumping under a CW probe at  $E = 2004$  meV. (a) Pumping on line (1) and detecting on line (4). (b) Pumping on line (4) and detecting on line (1).

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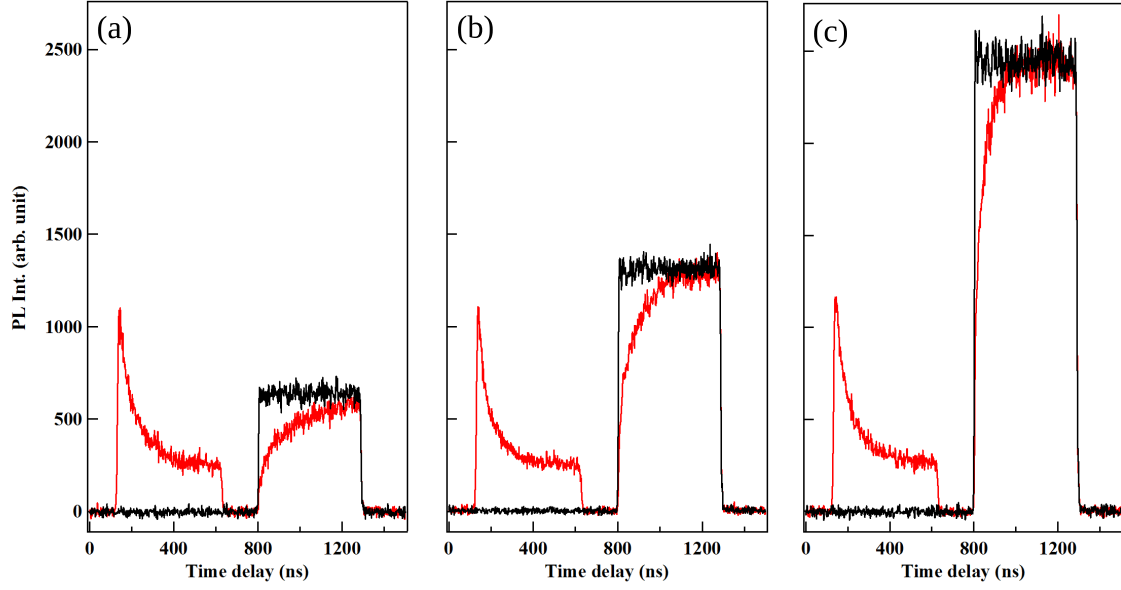


Figure I.5: PL transients measured on line (4) while pumping on line (4) at  $P_{pump} = 250 \mu\text{W}$  (red) or without pumping (dark). (a)  $P_{probe} = 125 \mu\text{W}$ . (b)  $P_{probe} = 250 \mu\text{W}$ . (c)  $P_{probe} = 500 \mu\text{W}$ .

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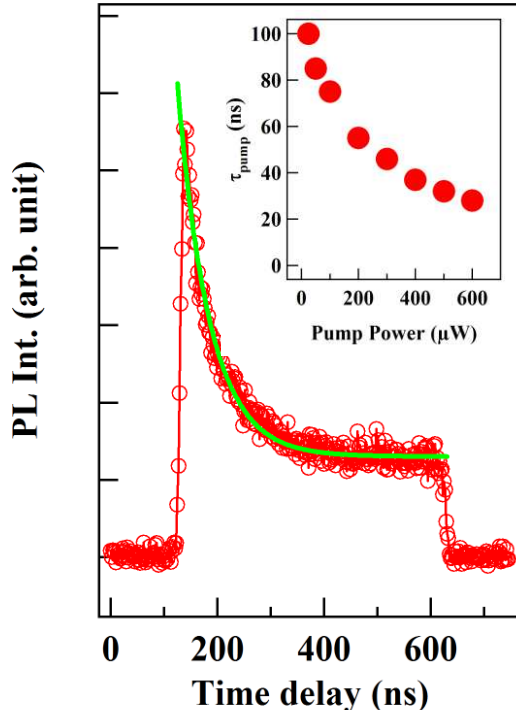


Figure I.6: Detail of the PL transient measured during the resonant pump pulse for a power  $P_{probe} = 250 \mu\text{W}$ . The exponential fit (green) gives a characteristic time of  $\tau_{pump} = 60 \text{ ns}$ . The inset presents the evolution of  $\tau_{pump}$  as a function of the pumping laser power.

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### I.1.2 Dynamics of the Cr spin under optical excitation

We performed auto-correlation of the photoluminescence (PL) intensity emitted by individual lines of an isolated Cr-doped QD to probe the dynamics of the magnetic atom under continuous wave (CW) optical excitation. A Hanbury Brown and Twiss (HBT) setup was used for this purpose. In these start-stop experiments, the detection of the first photon indicates by its energy and polarization that the Cr spin has a given orientation. The probability of detection of a second photon with the same energy and polarization is proportional to the probability of conserving this spin state. The time evolution of the auto-correlation is a probe of the spin dynamics in the Cr-doped QD.

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. I.8 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

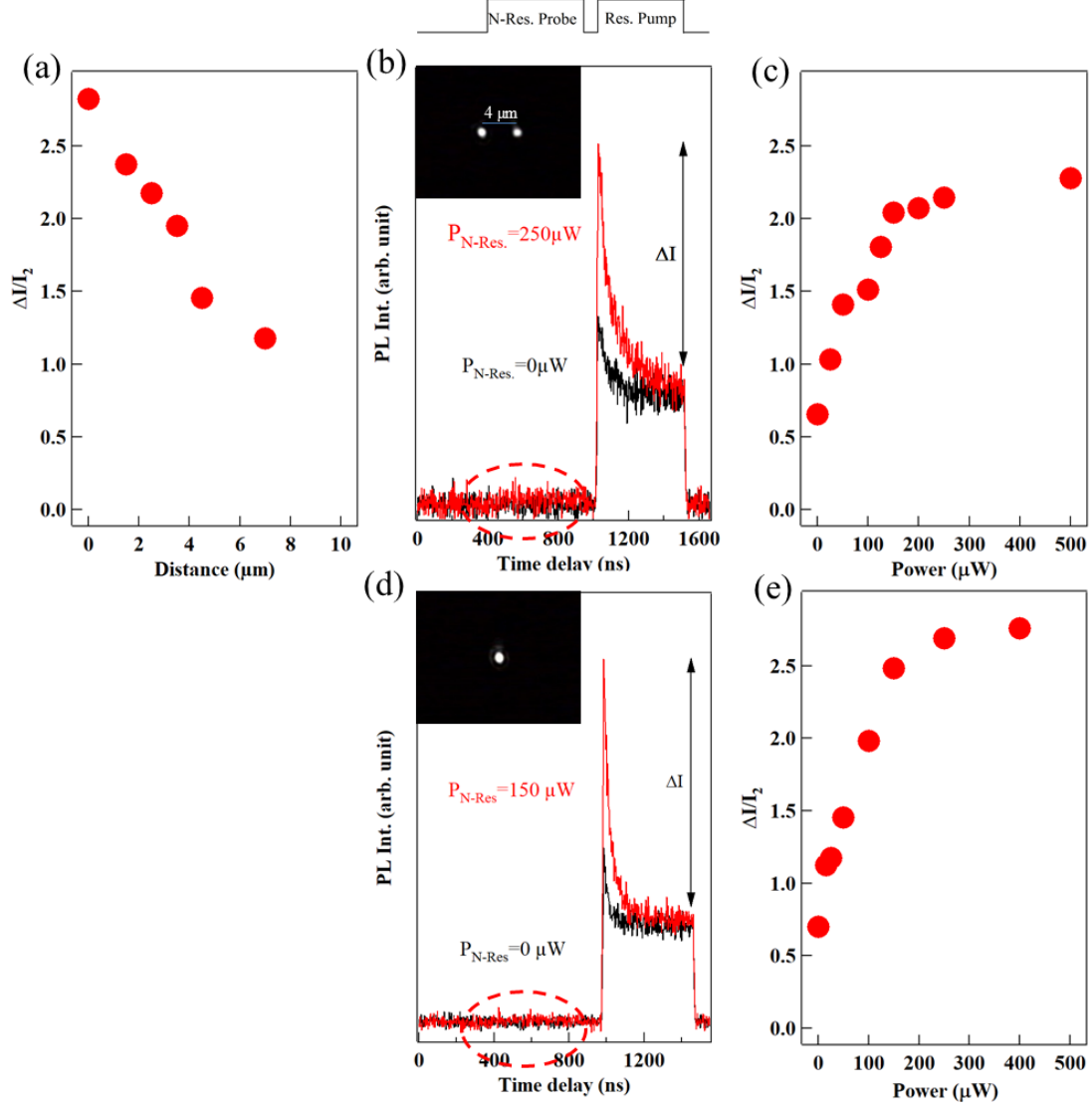


Figure I.7: (a) Evolution of the pumping transient intensity in a pump-probe experiment as a function of the distance between the probe laser and the dot. (b) PL transients recorded for a probe laser at  $d = 4 \mu\text{m}$  of the dot. Red: probe on; black: pump off. (c) Evolution of the pumping transient intensity as a function of the probe power, for a probe laser at  $d = 4 \mu\text{m}$  of the dot. (d) PL transient recorded for a probe laser at  $E_{\text{probe}} = 2010 \text{ meV}$  (energy not absorbed neither by the dot nor the barriers). Red: probe on; black: pump off. (e) Evolution of the pumping transient intensity in function of the probe power, for a probe not absorbed in the sample.

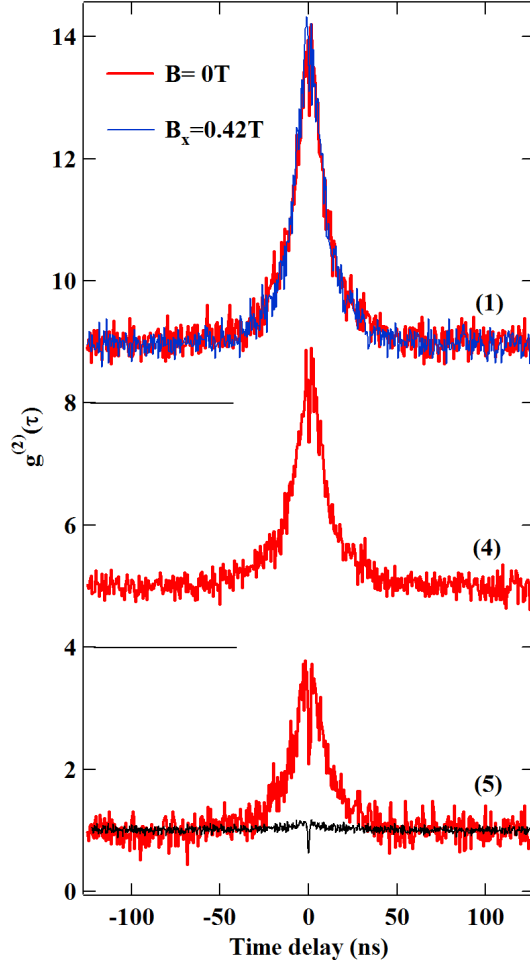


Figure I.8: Auto-correlation of the PL intensity collected in circular polarization on the X-Cr lines (1), (4) and (5) (as defined in Fig. I.3) and compared with the auto-correlation of the exciton in a non-magnetic QD (black line). The curves are shifted for clarity. For line (1), the auto-correlation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

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. I.8, 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  $\Gamma_{OFF}/\Gamma_{ON}$ , ratio of the transition rates from OFF to ON,  $\Gamma_{ON}$ , and from ON to OFF,  $\Gamma_{OFF}$  [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 ( $B_x = 0.42\text{T}$  in Fig. I.8). 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.

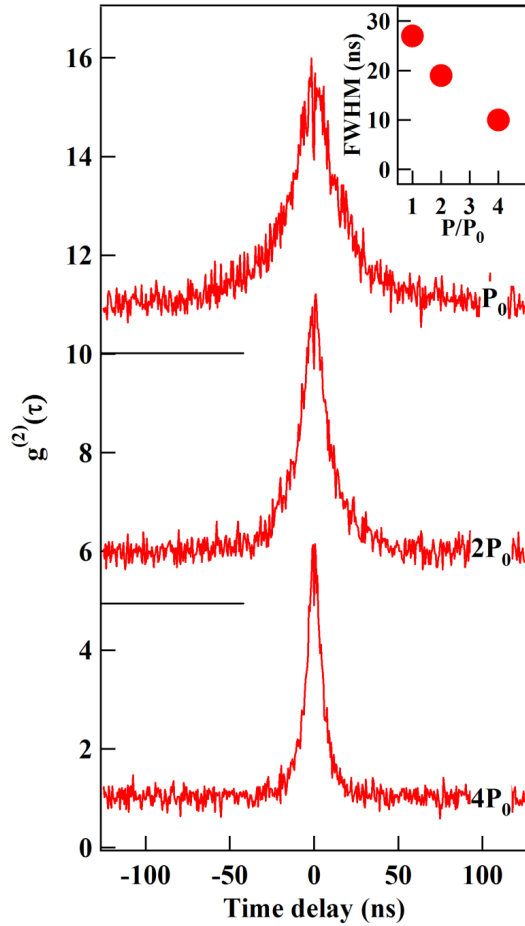


Figure I.9: Auto-correlation of the PL intensity recorded in circular polarization on the high energy X-Cr line (1) for different excitation powers. The inset shows the corresponding FWHM of the bunching signal versus excitation power.

One should note that the observed spin dynamics depends on the optical excitation power. Increasing the excitation power significantly reduces the width of the bunching (Fig. I.9), linked to an increase of the Cr spin fluctuations. Within

the X-Cr complex, the electron-Cr exchange interaction and the hole-Cr exchange interaction in the presence of heavy-hole/light-hole mixing can both induce spin-flips of the Cr. Though weak, the probability of such spin flips increases with the occupation of the QD with an exciton and dominates the spin dynamics in the high excitation regime required for the photon correlation measurements.

The excitation power dependence shows that the measured width of the bunching is not limited by the intrinsic Cr spin relaxation time  $\tau_{Cr}$ . This gives a lower bound for  $\tau_{Cr}$  in the 20 ns range. A shorter value would impose, at low excitation intensity, faster spin fluctuations than observed experimentally. The Cr spin relaxation time is ultimately controlled by the interaction with acoustic phonons and could depend on the optical excitation through the generation of non-equilibrium acoustic phonons during the relaxation of injected carriers [4, 5]. It is however expected to be longer than the observed dynamics [6] and cannot be determined with these measurements which require a large photon count rate.

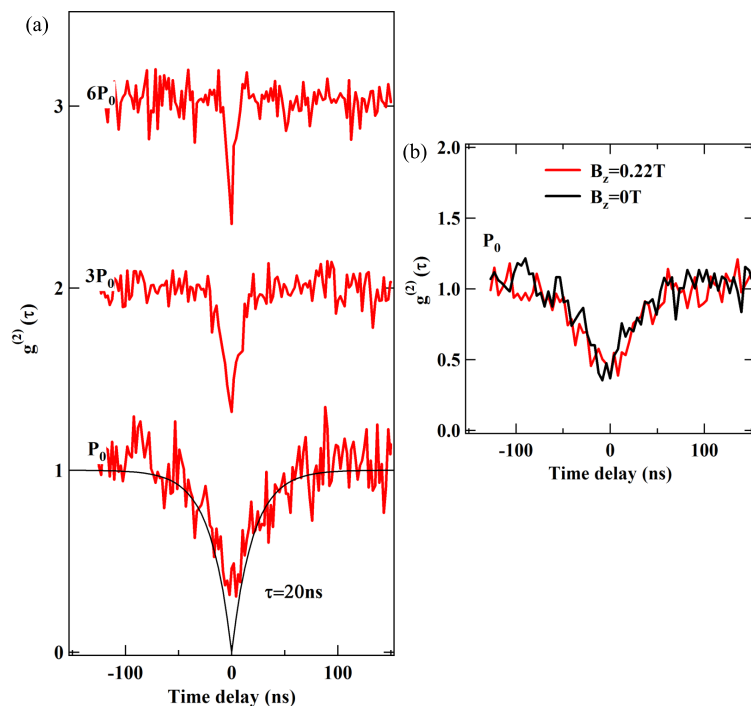


Figure I.10: (a) Correlation signal of the PL intensity of lines (1) and (4) recorded in the same circular polarization (cross-correlation) for three different excitation powers. The curves are shifted for clarity. The black line is an exponential fit with a characteristic time  $\tau = 20$  ns. (b)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

To analyze more in detail the spin relaxation channels, cross-correlation measurements were performed on the PL emitted by the high energy and the low energy lines in the same circular polarization. The cross-correlation shows a large anti-bunching with a FWHM in the 10 ns range and  $g^{(2)}(0) \approx 0.3$  (Fig. I.10(a)). Whereas the auto-correlation probes the probability for the Cr spin to be conserved, this cross-correlation is a probe of the spin transfer time between the spin states  $S_z = +1$  and  $S_z = -1$ . As for the auto-correlation, the cross-correlation strongly depends on the excitation power. At weak excitation, a spin transfer time of about 20 ns is observed. It is accelerated with the increase of the excitation power (Fig. I.10(a)). This transfer time could be controlled by anisotropic in-plane strain which couples Cr spin states separated by two units through an additional term  $E(S_x^2 - S_y^2)$  in the Cr fine structure Hamiltonian [7]. However, even at low excitation power, the measured transfer time is not affected by a longitudinal magnetic field (Fig. I.10(b)). This shows that for such QD the strain anisotropy term  $E$  is weak and is not the main parameter controlling the transfer time between the states  $S_z = \pm 1$ . The spin transfer time is dominated by spin-flips induced by the exciton/Cr interaction.

### I.1.3 Cr spin relaxation in the dark

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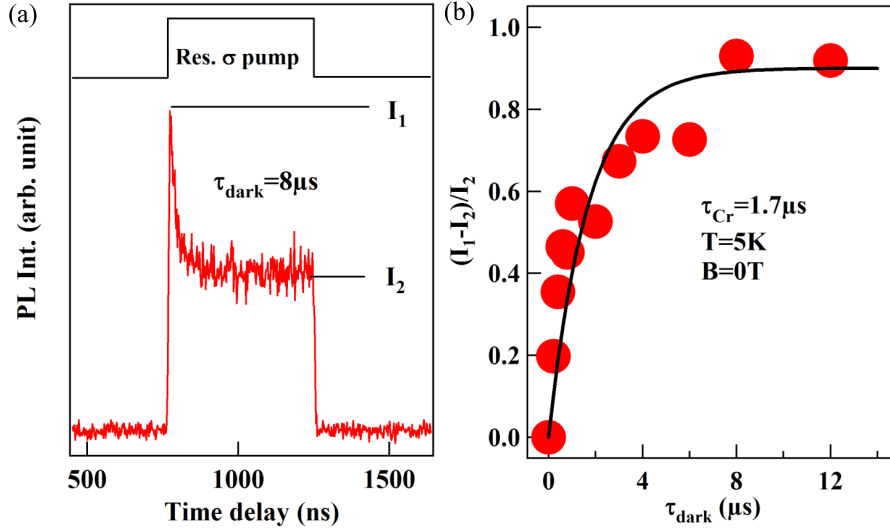


Figure I.11: (a) Time evolution of the PL intensity of line (4) of X-Cr under resonant excitation on line (1) with a circularly polarized excitation pulse. (b) Evolution of the amplitude of the pumping transient  $\Delta I/I_2$  as a function of the dark time between the excitation pulses. The black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

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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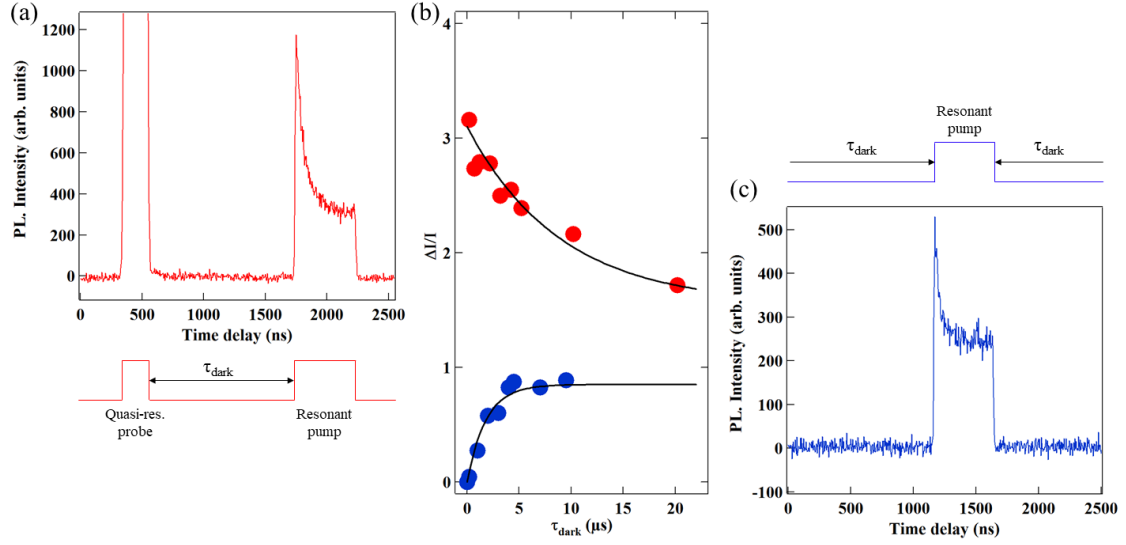


Figure I.12: Comparison of the relaxation of the Cr spin after resonant pumping in the line (1) with (red) or without (blue) a probe pulse ( $E_{\text{probe}} = 2070 \text{ meV}$ ). (a) Time resolved PL of line (4) for a resonant pump on line (1) with a probe pulse. (b)  $\Delta I/I_2$  in function of the dark time  $\tau_{\text{dark}}$  measured for the relaxation between the probe and the pump pulses (red) or between two pump pulses (blue). (c) Time resolved PL of line (4) for a resonant pump on line (1) with no probe pulse.

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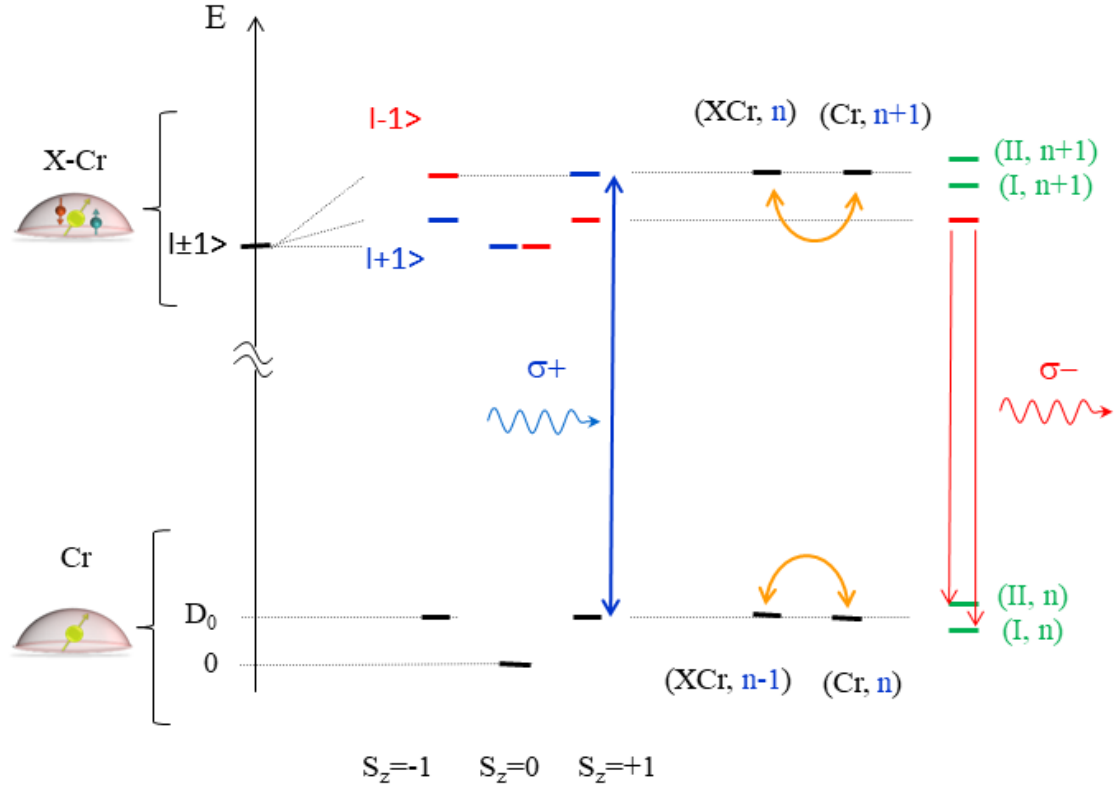


Figure I.13: Energy level of a Cr-doped quantum dot and formation of the dressed-states. We excite resonantly a given transition of the quantum dot with a continuous monochromatic laser (here the  $|S_z = +1\rangle \rightarrow |S_z = +1, X_z = +1\rangle$  transition). Considering a mode of  $n$ , the levels  $(\text{Cr}, n)$  and  $(\text{X-Cr}, n - 1)$  are coherently coupled through absorption and stimulated emission of photon. The Rabi splitting  $\Omega_r$  between these two levels can be probed in the cross-polarized PL 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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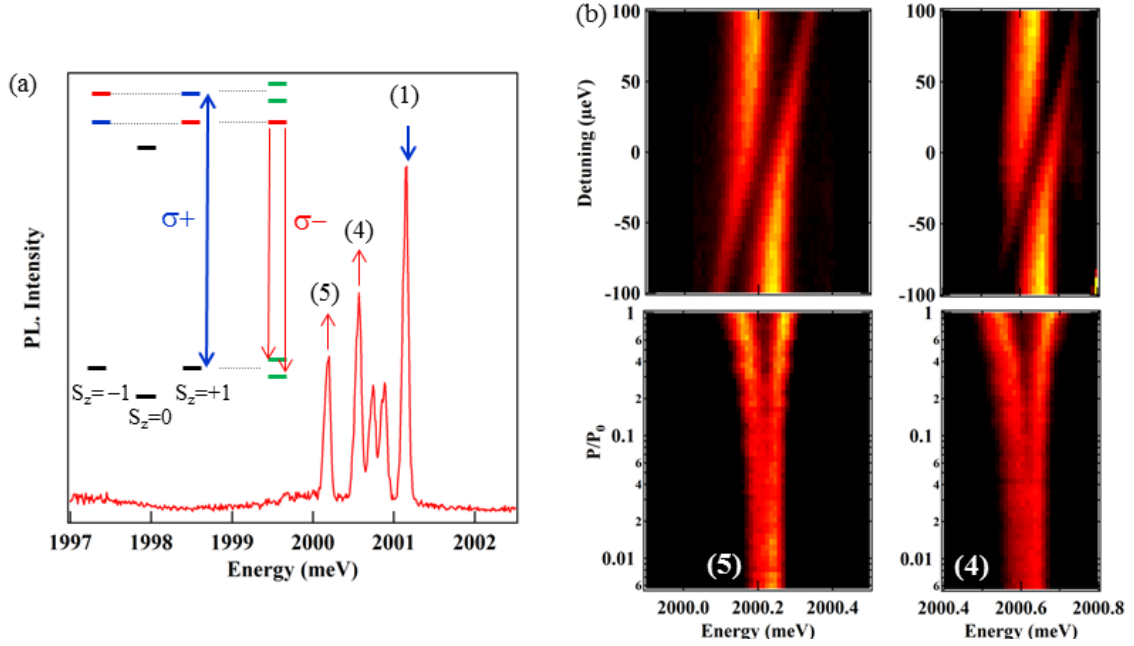


Figure I.14: (a) PL of X-Cr and configuration of excitation in the resonant optical control experiments. The inset illustrate the laser induced splittings in the ground and excited states for a  $\sigma+$  excitation on  $|S_z = +1\rangle$ . (b) PL intensity maps of lines (5) and (4) for an excitation 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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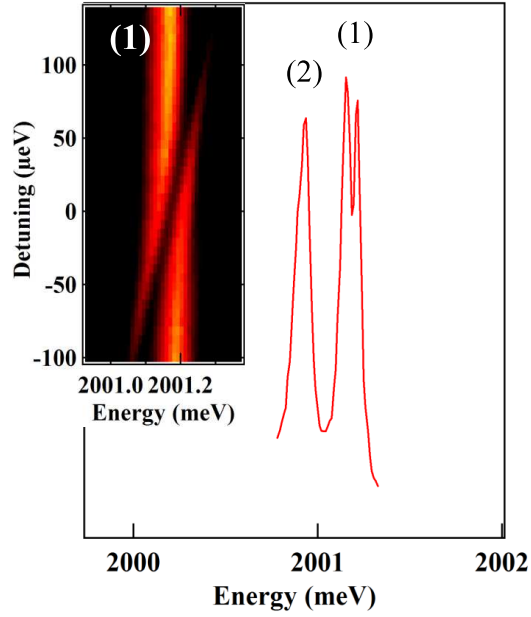


Figure I.15: PL of line (1) and (2) (high energy line) for a laser on resonance with the dark exciton state (5). Inset: PL intensity map of line (1) as a function of the laser detuning around (5).

### I.3 Conclusion

It presents a large bunching revealing a PL intermittency which results from fluctuations of the Cr spin with a timescale in the 10 ns range. Correlation of the intensity emitted by two different lines of the exciton-Cr (X-Cr) complex (namely, cross-correlation), presents an anti-bunching at short delays. A calculation of the time dependence of the spin levels population in Cr-doped QDs shows that the observed spin dynamics is governed by the exciton/Cr interaction. These measurements also provide a lower bound in the 20 ns range for the intrinsic Cr spin relaxation time.

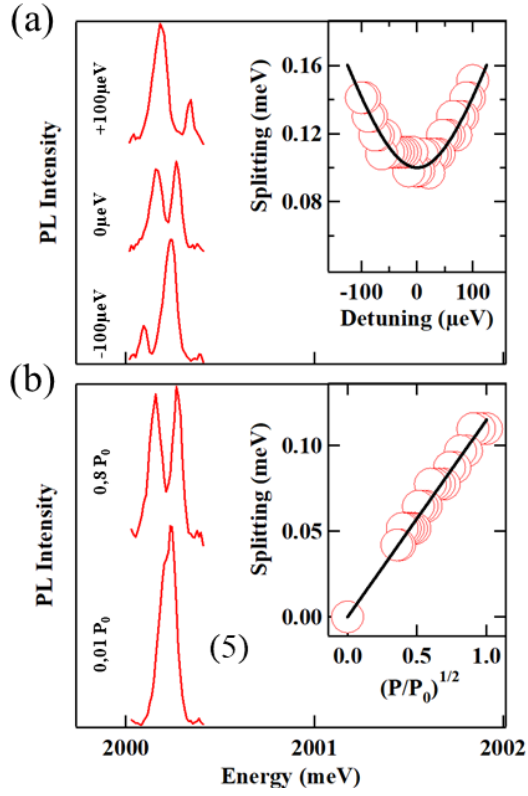


Figure I.16: PL spectra of line (5) for an excitation on line (1) at (a) no detuning and max detuning in each direction, and (b) low and high power. The insets show the splitting of the PL doublet as a function (a) of the laser detuning and (b) of the excitation intensity. The fit is obtained with  $\hbar\Omega_r = 100 \mu\text{eV}$ .

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