Chapter I

Dynamics of a single Cr spin in a ZnTe quantum dot

I.1 Exeperiment configuration

I.1.1 Experimental setup

Figure I.1: Complete experimental setup with three lasers, accousto-modulator, finishing either on the monochromator or the diodes.

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I.1.2 Studied dot

Figure I.2: Studied dot spectra and linear polarization

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Figure I.3: Spectra with different excitation and detection configurations

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I.2 Cr spin time fluctuations

I.2.1 Autocorrelation: conservation of the Cr spin

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.4 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. I.4, 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

Figure I.4: Autocor of X alone in comparison of X-Cr, and autocor of 3 peaks with comparison with and without B field

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. I.4). 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.5: Autocor power variation effect

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I.2.2 Cross-correlation: flipping of the Cr spin

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Figure I.6: Cross-correlation fitted with exponential, and cross correlation under magnetic field.

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Figure I.7: Cross-correlation under power variation

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I.2.3 Power dependency: relaxation paths

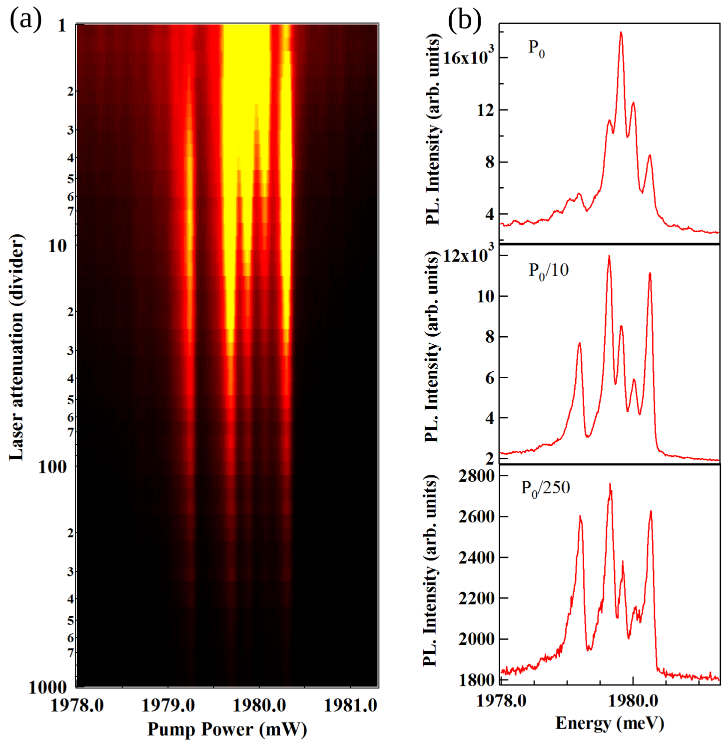

Figure I.8: Excitation power variations on dot334 QD3 and plot of the intensity of each peak

In order to study the different relaxation path of the system, probing of the emission evolution under excitation power was realized, reported on Fig. I.8. As expected, the PL is becoming more intense with the augmentation of the excitation laser, since more exciton are produced and thus injected in the quantum dot. However, this power augmentation response is not the same for each of the peaks. The two central peaks, associated with the $|0\rangle$ states, start at about twice the intensity of the $|\pm 1\rangle$ peaks for the most intense one, seemingly never reaching their maximum under our power range. However, when lowering the excitation power, they diminish quickly, and even seems to disappear at low energy, when exterior peaks still show luminescence. Plotting this evolution shows that the two central peaks exhibit a super-linear evolution. On the other side, the exterior peaks begin by rising in intensity before diminishing in a sub-linear fashion. This is coherent with the usual picture, where high power populating preferentially X^2-Cr states, while low power populates preferentially X-Cr states [?]. Finally, one can notice that the dark exciton exhibit the same behaviour, but the maximum emission intensity is at slightly lower power than the $|\pm 1\rangle$.

Figure I.9: Power variation simulation

The results of this evolution are well reproduced by our spin effective Hamiltonian, using the parameters found for the dot from the magneto-optics and the linear polarization fitting. The model results are presented in Fig. I.9. [NOT SURE, TO BE REDISCUSSED]The super-linear behaviour of the central peaks can be explained by the proximity of dark exciton states. High power excitation can unlock radiative recombination path to states remaining nonradiative at low excitation power [4]. Such states linked to the $|0\rangle$ state make the emission on its peaks present a super-linear behaviour when excited at high power.

I.2.4 Model of the spin dynamics

To identify the main contribution to the observed spin fluctuations, we modelled the auto-correlation of the PL of X-Cr using the full spin level structure of a Cr-doped QD. We calculated the time evolution of the population of the twenty X-Cr states in the excited state of the QD and five Cr states in the ground state by solving numerically the master equation for the corresponding 25 x 25 density matrix

Figure I.10: Simulation of autocorrelation on each peak and cross-correlation |+1> to |-1>

ρ. The time evolution of the density matrix including relaxation and dephasing processes in the Lindblad form is given by $\partial\rho/\partial t = -i/\hbar[\mathcal{H}, \rho] + L\rho$ where $\mathcal{H}$ is the Hamiltonian of the complete system (X-Cr and Cr) and $L\rho$ describes the coupling or decay channels resulting from an interaction with the environment [5, 6]. The energy levels of the Cr are controlled by the magnetic anisotropy $D_0 S_z^2$. The X-Cr Hamiltonian, presented in Ref.[7], contains the energy of the Cr spin states, the carriers-Cr exchange interactions, the electron-hole exchange interaction in a confining potential of low symmetry and the structure of the valence band including heavy-hole/light-hole mixing. D_0 in the Cr Hamiltonian and the parameters in the X-Cr Hamiltonian cannot be precisely extracted from the zero magnetic field PL (Fig. I.2(a)). For a qualitative description of the observed spin dynamics, we use in the model typical Cr-doped QD parameters extracted from magneto-optics measurements presented in Ref. [7]. These parameters give a X-Cr splitting and a dark/bright excitons mixing similar to the one observed in the QD discussed in this article.

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Figure I.11: Autocorrelation simulation with $\tau_C r$ variations

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Figure I.12: Simulation of autocorrelation and cross-correlation under magnetic field

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Figure I.13: Simulation of autocorrelation under power variation

I.3 Preparation the spin of a Cr atom in a quantum dot

I.3.1 Resonant optical pumping of a spin level

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Figure I.14: Pumping on peak 4

Figure I.15: 3 examples of pumping graphs under different excitation and plot of the variation

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Figure I.16: Examples of pumping under detuning with plot of PL maximum intensity variation and of $\delta I/I$

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I.3.2 Spin relaxation

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Figure I.17: Dark time variation of pumping with its plot

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I.4 Optical control of the spin of a Cr atom

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Figure I.18: Map of peak 4 and 5 under augmentation of the excitation power + line shape with plot of the splitting variation with power

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Figure I.19: Map of peak 4 and 5 under augmentation of the detuning + line shape with plot of the splitting variation with detuning

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Figure I.20: Detuning on 1 and 2 while exciting on 5

Chapter II

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