Chapter I

Optical control of an individual Cr spin in a CdTe QD

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 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.2: Schema with energy level illustrating the experiment + experiment result (cf PRB article Fig.1) with detuning -> article picture

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Figure I.3: Continuous probe (heat), modulated pump

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Figure I.4: Heating in gap and far away from the dot

Figure I.5: Magnetic field and power variation of pumping

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I.1.2 Dynamics of the Cr spin under optical excitation

Autocorrelation and cross-correlation

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Figure I.6: Schema of energy level of autocor experiment + autocor on each peak and X alone autocor

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. I.7). 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.7: Variation of autocor under mag field and power variations

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Relaxations through dark states

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In order to study the different relaxation path of the system, probing of the

Figure I.8: Cross-correlation + fit and cross correlation under B field and pw var (cf article picture APL)

Figure I.9: Excitation power variations of the spectra and plot of the intensity of each peak emission evolution under excitation power was realized, reported on Fig. I.9. 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.10: 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.10. [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 non-radiative 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.1.3 Spin relaxation in the dark

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Figure I.11: Schema with energy level illustrating the experiment + experiment result (cf PRB article Fig.1) with detuning -> article picture

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Figure I.12: Continuous probe (heat), modulated pump

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I.2 Optical Stark effect on an individual Cr spin

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Figure I.13: Schema of the stark experiment with energy level (cf article)

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Figure I.14: Splitting under power raising

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Figure I.15: Emission variation with detuning

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