Modelling optical spin pumping of a single Mn atom in a CdTe quantum dot

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Abstract. We present a model for the incoherent spin dynamics of a Mn atom inside a CdTe quantum dot resonantly pumped by a laser light. The relevant quantum states of the system, $2S + 1 = 6$ spin states in the optical ground state and the $24 = (2S + 1) \ast (2S_e + 1) \ast (2S_h + 1)$ are obtained from a effective spin Hamiltonian which describes the basic features of photoluminescence. The model includes the dominant Ising exchange interactions between the spins of the hole, the Mn and the electron but neglects coherent spin-flip terms. The Mn spin dynamics is described with a master equation for the populations of the 30 levels for which dissipative spin-flip processes are permitted. The optical spin pumping observed experimentally is explained if the Mn spin relaxation time in with ($T_{1X}$) exciton is much shorter than ($T_{1G}$), the Mn spin relaxation time without exciton.

1. Introduction
Since their discovery in 2004 [1], quantum dots doped with a single magnetic Mn atom attract great interest, both from theory [2, 3, 4, 5, 6, 7] and experimental sides [8, 9, 10, 11, 12]. As a substitutional impurity in CdTe, Mn$^{2+}$ is known to be a neutral dopant with spin $S = 5/2$ associated to the half filled $d$ shell. Exchange interactions between the $d$ electrons with electrons and holes result in a very characteristic single quantum dot photoluminescence spectra whose landmark are 6 narrow peaks split at zero field by approximately 0.2 meV. This spectrum reveals a very anisotropic exciton-Mn (X-Mn)interaction, which can be in first instance described as an Ising spin coupling between the projection of the Mn and the exciton spin along the growth axis, $z$. The origin of the anisotropic exchange is the interplay between strong spin orbit interaction of valence band holes and strain and confinement induced light-hole heavy-hole (LH-HH) decoupling [4]. In some instances, PL spectra with 7 or 8 peaks are observed. This is understood in terms of spin-flip interactions which mix exciton-Mn states with different value of the total spin along the $z$ axis.

Importantly, the spin state of a single Mn atom affects the PL spectrum of the thousand of atoms that conform the QD. This permits experimental access to the spin of a single atom. In the cases when spin-flip is negligible, so that PL has 6 peaks, the detection of a photon with a given energy and polarization give direct information of the Mn spin state when the photon was emitted [1, 4]. Autocorrelation measurements of the PL gives direct information about the Mn spin relaxation both in dark and interacting with the exciton [10]. Recent optical pumping experiments show that it is possible to polarized the spin of the Mn at zero applied field.
2. The model

The purpose of this paper is to provide a heuristic non-equilibrium model for the Mn spin dynamics driven by laser pumping. We propose a master equation for the occupation of the eigenstates of a simplified single Mn-doped Hamiltonian quantum dot that features the projection along the z axis of the spin $1/2$ electron $S_e$, the pseudospin $J = 3/2, J_z = \pm 3/2$ of the heavy hole and the spin $1/2$ of the Mn:

$$\mathcal{H} = J_{h-Mn} \hat{S}^h_z \hat{M}_z + J_{eh} S^h_z S^e_z + J_{e-Mn} \hat{S}^e_z \hat{M}_z + D \hat{M}_z^2$$

(1)

The first term corresponds to the hole Mn coupling, which is antiferromagnetic ($J_{h-Mn} > 0$). The second to the electron-hole exchange, which is ferromagnetic, so that the dark $\pm 2$ excitons lie below the bright $\pm 1$ exciton doublet. The third term is the electron-Mn exchange.

The master equation model that we use here is an extension of the simple model by Govorov and Kalameitsev (hereafter GK) [2] in their proposal of optical spin pumping of a single Mn in a quantum dot. In GK, a unique rate is assigned to transitions between the 24 exciton levels, complying with principle of detailed balance but neglecting the dependence of the rates on energy and spin change. Whereas a detailed microscopic model will be presented elsewhere [15], here we propose a model for the rates in which transitions are only permitted between states that are connected via the flip of a single spin (the one of either the Mn, the electron or the hole). This rule is certainly present in most of spin relaxation mechanisms and restricts significantly the relaxation pathways. However, we still neglect the dependence of the rates on the energy difference, except for the fact that we use the principle of detailed balance. Thus, our model has 4 elementary rates $\Gamma_e, \Gamma_h, \Gamma_{Mn,X}$ and $\Gamma_{Mn,G}$ for the relaxation of the spin of

Figure 1. (A) Diagram of the 24 exciton-Mn levels and the 6 Mn levels involved in the optical pumping process. (B) Scheme of the optical pumping.
the electron, hole, Mn in the presence of the exciton and Mn in the dark. In addition, $M_z$ conserving transitions between the six ground states and the 12 bright exciton transitions are described with a laser pumping function $g(M_z, X)$ where $X = \pm 1, \pm 2$ labels the excitons. We take $g(M_z, X = \pm 2) = 0$. Mn spin conserving spontaneous emission from bright $\Gamma_B$ and dark $\Gamma_D$ are also included.

![Figure 2](image.png)

**Figure 2.** (A) Calculated photoluminescence transients obtained in $\sigma^-$ polarization on the high $(S_z=-5/2)$ and low $(S_z=+5/2)$ energy line under alternate injection of $\sigma^-$ and $\sigma^+$ excitons. The inset shows the Mn spin population at the end of the $\sigma^-$ pumping sequence. (B) Calculated photoluminescence of $| -1, +5/2 \rangle$ under excitation on $| +1, +5/2 \rangle$. As shown in the inset, the resonant laser excitation empty the state $S_z=+5/2$.

The master equation reads:

$$\frac{dp_N}{dt} = \sum_{N'} \Gamma_{N \rightarrow N'} p_{N'} - \sum_{N'} \Gamma_{N' \rightarrow N} p_{N'}$$

The model permits to consider three excitation modes: (i) resonant, for which $\Gamma(M_z, X)$ is non-zero for only one of the 12 bright excitons states, (ii) unpolarized and non-resonant (or quasi-resonant, using the jargon of the experimental paper [11]) for which $\Gamma(M_z, X)$ is non-zero for all the 12 bright states, and (iii) polarized non-resonant, for which $\Gamma(M_z, X)$ is non zero for the six states of a given bright exciton.

In figure 2 we show the results of the simulation of the optical pumping processed reported by Le Gall et al. [11]. We take $\Gamma_e=\Gamma_h = 1\text{ns}^{-1}$, $\Gamma_B = 4\text{ns}^{-1}$, $\Gamma_D = 0.1\text{ns}^{-1}$, $\Gamma_{Mn,X} = 0.1\text{ns}^{-1}$ and $\Gamma_{Mn,G} = 0.0001\text{ns}^{-1}$. $\Gamma_B$, $\Gamma_D$ and $\Gamma_{Mn,X}$ have been measured using time resolved and photon correlation measurements on single Mn doped QDs [10]. A $\Gamma_{Mn,G}$ larger than a few micro-seconds has been deduced from recent optical pumping experiments [11]. $\Gamma_e=\Gamma_h=\Gamma_X$ in the ns range are typical values of spin relaxation for an exciton under magnetic field [14](the exchange interaction with the Mn act on the exciton as an effective magnetic field of a few Teslas). In addition, recent resonance fluorescence experiments on Mn-doped QDs suggest that $\Gamma_X>\Gamma_{Mn,X}$ [15].
In the work of Le Gall et al., a quantum dot excited state level is pumped with circularly polarized light. This process has some degree of Mn spin selectivity which results in the depletion of the pumped Mn spin transition. The latter is traced by the time evolution of the PL intensity in the low energy peak which corresponds to the Mn spin $+5/2$ and $-5/2$ depending on the polarization of the detected photon. The main result observed experimentally is that the optically pumped Mn spin ground level is depleted. The solution of the master equation (2) with the restricted relaxation rules proposed here is able to account for the observed experimental optical pumping mechanism (Figure 2(a)). As proposed by GV, this still can be understood within our model if the Mn spin relaxation is more efficient in the presence of the exciton than in the ground state ($\Gamma_{Mn,X} >> \Gamma_{Mn,G}$).

A more efficient pumping efficiency could be obtained under resonant excitation on the ground state of the QD. Such process is illustrated in Figure 2(b). The high energy state $+5/2$ is resonantly driven by a $\sigma+$ laser. The initialization of the Mn spin is probed in the resonance fluorescence signal of the low energy state in $\sigma^-$ polarization. The intensity of this signal probe the population of state $+5/2$ and appears after a spin flip of the created exciton without any change in the Mn spin projection. With the same parameters used for the modeling of the non-resonant pumping experiment, we predict a pumping efficiency larger than 95% in a few tens of ns.

In conclusion, we have presented here preliminary simulations to account of Mn spin optical pumping using a model with realistic spin relaxation rules. The results are in good agreement with recently reported experiments of optical orientation under quasi-resonant excitation. The model suggest an efficient optical pumping in a tens of ns range for a resonantly driven Mn-doped QD. This process could be probed in a resonance fluorescence experiment.

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