Dynamical magnetic and nuclear polarization in complex spin systems: semi-magnetic II–VI quantum dots

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Abstract. Dynamical magnetic and nuclear polarization in complex spin systems is discussed on the example of transfer of spin from exciton to the central spin of magnetic impurity in a quantum dot in the presence of a finite number of nuclear spins. The exciton is described in terms of electron and heavy-hole spins interacting via exchange interaction with magnetic impurity, via hyperfine interaction with a finite number of nuclear spins and via dipole interaction with photons. The time evolution of the exciton, magnetic impurity and nuclear spins is calculated exactly between quantum jumps corresponding to exciton radiative recombination. The collapse of the wavefunction and the refilling of the quantum dot with a new spin-polarized exciton is shown to lead to the build up of magnetization of the magnetic impurity as well as nuclear spin polarization. The competition between electron spin transfer to magnetic impurity and to nuclear spins simultaneous with the creation of dark excitons is elucidated. The technique presented here opens up the possibility of studying optically induced dynamical magnetic and nuclear polarization in complex spin systems.

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1. Introduction

There is currently an interest in developing means of localizing and controlling complex spin systems in solid-state devices [1]. This includes electron and/or hole spins in gated [2, 3], self-assembled [4], nanocrystal [5] and carbon nanotube quantum dots [6], nitrogen vacancies in diamond [7] and magnetic impurities in II–VI [8–13] and III–V [14, 15] semiconductors. The complex spin systems involved include heavy valence holes with spin $S_h = 3/2$, nitrogen vacancies with spin $M = 1$, half-filled shell electrons of manganese, Mn$^{2+}$, impurity atom with spin $M = 5/2$ or Mn$^{3+}$ atom with $M = 3/2$ in II–VI semiconductor quantum dots or strongly coupled valence hole-Mn atom in InAs/GaAs quantum dots. Extensive theoretical studies have been carried out, predicting rich phase diagrams for these systems [17–21]. For nitrogen vacancy centers in diamond, carbon nanotube-based quantum dots and magnetic impurities in II–VI semiconductor quantum dots, the complex spin system interacts with only a finite number of nuclear spins. In II–VI materials, a fraction of atoms contain non-zero nuclear spin magnetic moments according to their natural isotope abundances. For example, the natural abundances of isotopes of $^{111}$Cd and $^{125}$Te with nuclear spin $1/2$ are 12% and 7%, respectively. These nuclear spins are distributed randomly, hence they can be considered nuclear spin impurities in the host semiconductor material. The control of the magnetization of complex spin systems is often carried out optically and involves the transfer of photon angular momentum into exciton spin and exciton spin into the spin of the complex spin system [22, 23]. This dynamical magnetic polarization (DMP) process is decohered by photon and nuclear spin baths. Recently, first optical experiments on single magnetic impurities in II–VI quantum dots measured the dynamic evolution of the magnetization process [8–10] with theoretical models of DMP based on rate equations [24, 25].

In this work, we develop a microscopic theory of optically driven DMP of complex spin systems. The theory describes the transfer of spin from exciton to the central spin of magnetic impurity in a quantum dot in the presence of a finite number of nuclear spins using quantum jump approach [26–28]. Note that we consider the manganese localized in the quantum dot as central spin. The exciton is described in terms of the electron and heavy-hole spins interacting via exchange interaction with magnetic impurity, via hyperfine interaction with a finite number of nuclear spins and via dipole interaction with photons. The time evolution
of the exciton, magnetic impurity and nuclear spins are calculated exactly between quantum jumps corresponding to exciton radiative recombination. The collapse of the wavefunction and the refilling of the quantum dot with new spin-polarized exciton as in the recent experiment by Goryca et al [8] is shown to lead to the build up of magnetization of the magnetic impurity as well as nuclear spin polarization. In this model, the build up of magnetic impurity (MI) polarization is a direct consequence of the bright exciton transition to dark exciton due to electron–MI exchange coupling. The dark exciton can be detected by applying a small in-plane magnetic field, e.g. in Voigt geometry. At the quantum jump time, the detection of a photon with the circular polarization equal to the angular momentum of the remaining bright exciton simultaneously occurs with the transfer of angular momentum from dark exciton to MI. Furthermore, the competition between electron spin transfer to magnetic impurity and to nuclear spins simultaneous with the creation of dark excitons is elucidated.

The paper is organized as follows. In section 2, we describe our model. Section 3 describes the quantum jump approach to time evolution of a single MI and a single exciton in the absence of nuclear spins. Section 4 contains the quantum jump approach and the dynamical evolution of MI interacting with a train of excitons in the presence of nuclear spins. In sections 5 and 6, we present the numerical results, discussions, conclusion and summary.

2. The model

We consider a semiconductor quantum dot (QD) containing a complex spin system M, e.g. magnetic ion, coupled with few nuclear spins of the host material, e.g. as in CdTe quantum dots. The quantum dot with MI is attached to a smaller quantum dot with no MI where the electrons and valence holes with definite spin polarization are generated optically by circularly polarized light. This is illustrated in figure 1(a) where circles describe quantum dots, the blue arrow corresponds to electron spin $S_z,e = +1/2$ and the white arrow corresponds to heavy-hole spin $S_z,h = -3/2$ in the smaller dot. The larger dot contains a randomly oriented complex spin $M$, represented by a magenta arrow and a number of randomly oriented nuclear spins represented by small arrows. The DMP process starts with the transfer of spin-polarized exciton from the smaller QD to the larger QD, as illustrated in figure 1(b). As a result of interactions, the spin of electron, MI and nuclear spins undergo a flip-flop process as the wavefunction of the larger dot evolves and forms an entangled state, a linear combination of bright and dark excitons, as shown in figure 1(c). During this process, the smaller dot is refilled with spin-polarized exciton. Simultaneously, the bright exciton decays due to an interaction with the photon field with a random recombination time, resulting in a photon emission and a quantum jump. As a result of this process, the states of the magnetic ion and nuclear spins are modified, the polarization is increased and the larger dot is refilled with spin-polarized exciton and the DMP process continues.

We now quantitatively describe the DMP process. We start with the Hamiltonian describing the quantum dot coupled with the photon bath $H = H_{QD} + H_{phQD} + H_{ph}$. Here, $H_{ph}$ is the photon Hamiltonian, $H_{phQD}$ is the Hamiltonian describing coupling of photons with the exciton in a QD and $H_{QD}$ is the QD Hamiltonian.

The QD Hamiltonian describes the coupling between exciton $X$ and the magnetic moment of the complex spin system, consisting of MI and nuclear spins $I$. It is given by $H_{QD} = H_n + H_I + H_{x} + H_{m} + H_{n}. Here, H_s describes the exciton internal energy, H_n describes the MI internal energy and the remaining terms in $H_{QD}$ represent $X$–MI, $X$–I and MI–I interactions.
Figure 1. Schematic representation of the DMP process. The arrows in the left dot represent a bright exciton. In the right dot, the magenta arrow is MI and small black arrows represent the nuclear spins. In (a), an exciton is created in the smaller dot located in the left. In (b), the exciton tunnels in the large dot located in the right. A photon (the wiggly red line) creates another exciton in the empty dot in (c). The interaction between the bright exciton and manganese and nuclear spins forms an entangled state between bright and dark excitons. In (d), the annihilation of bright exciton led to an emission of a photon and the annihilation of dark exciton led to spin transition in manganese and nuclear spin system.

exchange couplings. The exciton Hamiltonian describes the low-energy quadruplet \( |S_{z,e}, S_{z,h} \rangle \) characterized by quantum spin numbers of an electron, \( S_{z,e} = \pm 1/2 \), and a heavy hole, \( S_{z,h} = \pm 3/2 \) in the QD [31]. The complex spin system is described by a total spin \( \vec{M} = \sum_{i=1}^{N} \vec{u}_i \), where \( N \) is the number of spins \( u = 1/2 \) building up the MI system and \( H_m = \sum_{i<j} J_{ij} u_i \cdot u_j + DM^2_z \) in which \( J_{ij} \) are exchange matrix elements building the total spin \( M \). In quantum dots, one often includes the strain field \( D \) leading to the splitting of different \( M_z \) levels [16]. Similarly, \( \vec{I} = \sum_{i=1}^{N_b} \vec{I}_i \) where \( N_b \) is the total number of nuclear spins.
We assume that exchange coupling constants of MI spins with the environment are identical and the full QD Hamiltonian can be written as [21]

\[
H_{\text{QD}} = H_{\text{m}} + H_{x} + J_{\text{hm}}S_{c,h}M_{z} - J_{\text{em}}\vec{S}_{e} \cdot \vec{M} \\
+ \sum_{n} [J_{ne}\vec{I}_{n} \cdot \vec{S}_{e} + J_{nh}I_{z,n}S_{c,h}] + \sum_{n} \sum_{n' \neq n} J_{nm}I_{n} \cdot \vec{I}_{n'} + \sum_{n} A_{n}\vec{I}_{n} \cdot \vec{M}.
\]

The exciton Hamiltonian \(H_{x} = \Delta_{0}\vec{S}_{c,h} \cdot \vec{S}_{c,h} + \Delta_{1}(S_{h}^{+}S_{h}^{-} + S_{e}^{+}S_{e}^{-})\) describes splitting \(\Delta_{0}/2\) between the low-energy dark exciton doublet \(|\uparrow, \uparrow\rangle = |+1/2, +3/2\rangle, |\downarrow, \downarrow\rangle = |−1/2, −3/2\rangle\) with the total angular momentum \(j_{c} = \pm 2\) along quantization axis, \(\hat{z}\), and higher energy bright exciton (relative to dark exciton) doublet \(|\downarrow, \uparrow\rangle = |−1/2, +3/2\rangle\) and \(|\uparrow, \downarrow\rangle = |−3/2, +1/2\rangle\) with \(j_{c} = \pm 1\). Here, \(\uparrow/\downarrow\) and \(\uparrow/\downarrow\) represent the spin of the electron and the hole [30]. The bright exciton doublet is split by the anisotropic electron–hole exchange interaction characterized by parameter \(\Delta_{1}\) which measures the splitting of the two bright exciton states \(|\pm 1/2, ±3/2\rangle, |−1/2, ±3/2\rangle\). \(\Delta_{1}\) is zero for cylindrical quantum dots and the two bright exciton states correspond to circular photon polarization. Note that in the absence of light holes, \(H_{x}\) is diagonal with or without \(\Delta_{1}\). In this case, the energy splitting of bright and dark excitons are \(\delta = 3\Delta_{0}/2\). The exciton–MI coupling in equation (1) is given as a sum of the ferromagnetic Heisenberg electron–MI exchange \(H_{\text{em}} = −J_{\text{em}}\vec{S}_{e} \cdot \vec{M}\) and anti-ferromagnetic Ising exchange interaction \(H_{\text{hm}} = +J_{\text{hm}}S_{c,h}M_{z}\) [21]. Only electron–MI interaction is responsible for the e–MI spin flip-flop process. The interaction of complex spin MI with nuclear spin associated with the spin complex is denoted here by \(H_{\text{MI}} = A\vec{I}_{\text{M}} \cdot \vec{M}\). This interaction might, for example, describe the coupling of manganese d-shell electron spins with manganese ion nuclear spin [16]. With hole spin strongly aligned along the growth z-direction, the coupling of electron and hole spins to the surrounding nuclear spins of isotopes of QD and barrier material with finite nuclear spin reads \(\sum_{n} [J_{ne}\vec{I}_{n} \cdot \vec{S} + J_{nh}I_{z,n}S_{c,h}] + \sum_{n,n'} J_{nm}I_{n} \cdot \vec{I}_{n'}\), where \(N_{h}\) is the number of nuclear spins in QD and the last term describes nuclear spin interaction. We note that for isotropic QD, the long-range e–h exchange \(\Delta_{1}\) is zero and the heavy-hole spin \(J_{c} = ±3/2\) is preserved.

### 3. Single magnetic impurity, single exciton and no nuclear spin

We start our discussion of DMP by discussing the time evolution of magnetization of X–MI complex interacting with harmonic fields of photons in the absence of nuclear spins. To focus on quantum dynamics in the simplest spin system, we consider MI with \(M = 1/2\) and just two states, \(|\uparrow\rangle = |M_{z} = 1/2\rangle\) and \(|\downarrow\rangle = |M_{z} = −1/2\rangle\), and Hamiltonian \(H_{\text{QD}} = H_{x} + H_{\text{em}}\) where \(\Delta_{1} = 0\) in \(H_{x}\), and \(H_{\text{em}} = −J_{\text{em}}\vec{S}_{e} \cdot \vec{M} + J_{\text{hm}}S_{c,h}M_{z}\). We also consider a continuous wave (CW) laser field with one type of circular polarization, e.g. \(\sigma = +1\), that generates excitons with one type of polarization, \(j_{c} = +1\), corresponding to \(|X_{b}\rangle = |\downarrow, \uparrow\rangle\). Because we neglect the hole spin–flip in the spin flip–flop process of X–MI complex, as discussed in section 2, the dark exciton \(|X_{d}\rangle = |\uparrow, \uparrow\rangle\) with \(j_{z} = +2\) is the only state generated throughout the electron–MI spin–flip. Hence, the space of a single X–MI complex can be spanned by \(|1\rangle = |X_{b}, \downarrow\rangle\), \(|2\rangle = |X_{b}, \uparrow\rangle\), \(|3\rangle = |X_{d}, \downarrow\rangle\), \(|4\rangle = |X_{d}, \uparrow\rangle\), \(|5\rangle = |0, \downarrow\rangle\), \(|6\rangle = |0, \uparrow\rangle\). In this basis, the exciton Hamiltonian is diagonal \(H_{x} = \text{diag}(E_{b}, E_{b}, E_{d}, E_{d}, 0, 0)\). Here, \(E_{b}\) and \(E_{d}\) are the energy of bright and dark excitons measured relative to the vacuum, respectively. In the basis of \(|{1, 4}\rangle\), \(|{2, 3}\rangle\) and \(|{5, 6}\rangle\), the X–MI Hamiltonian is block-diagonal \(H_{\text{sm}} = H_{\text{sm},1} \oplus H_{\text{sm},2} \oplus H_{\text{sm},3}\) where
$H_{\text{em},1} = (-J_{\text{em}} + J_{\text{hm}})/4 \mathbb{1}$, $H_{\text{em},2} = \left( \frac{(J_{\text{em}} - J_{\text{hm}})}{4} - \frac{J_{\text{em}}}{2} \right)$ and $H_{\text{em},3} = 0$, respectively. Here, $\mathbb{1}$ is a $2 \times 2$ unit matrix.

The off-diagonal elements of $H_{\text{em},2}$ describe the mixing of $X_b$ and $X_d$ via spin-$1/2$ MI. Hence, $\ket{\psi(t)} = C_{b\uparrow}(t) \exp(-iE_{bt}/\hbar) \ket{X_b, \uparrow} + C_{d\downarrow}(t) \exp(-iE_{at}/\hbar) \ket{X_d, \downarrow}$ with initial condition $\ket{\psi(t = 0)} = \ket{X_b, \uparrow}$ is one of the solutions of the time-dependent Schrödinger equation $-i\hbar \frac{d}{dt} \ket{\psi(t)} = H_{\text{QD}} \ket{\psi(t)}$. This state describes coherent Rabi-oscillations between bright and dark excitons due to MI spin flip-flop.

Note that in $\ket{\psi(t)}$ there is no mixing with the vacuum, $\ket{0, M_z = \pm 1/2}$, unless we take into account the coupling of bright-exciton with radiation field. In the interaction and rotating wave approximation, the electron–photon coupling is described by the Hamiltonian that does not directly change the state of MI:

$$H_{\text{phQD}}(t) = \hbar \sum_{k, M_z} g^{\pm}_k [b_k^{\dagger}, \rho] X_b, M_z \rangle \langle X_b, M_z | e^{-i(\omega_k - \omega_0)t} + b_k \langle X_b, M_z | \langle 0, M_z | e^{i(\omega_0 - \omega_0)t}],$$

(2)

where $b_k^{\dagger}$ and $b_k$ are creation and annihilation operators of the photon with specific circular polarization $\sigma = +1$, $g^z_k$, and $\omega_k$ are the photon–X coupling constant and photon frequency, respectively, and $\omega_0 = E_b/\hbar$. In the Wigner–Weisskopf approximation, the electron–photon radiation interaction is taken into account perturbatively and $g^{2}_k = \omega_k/(2\epsilon_0 V)p^2 \cos^2 \theta$. Here $\epsilon_0$, $V$, $p$ and $\theta$ are the vacuum dielectric constant, system volume, electron dipole moment element and the angle between the initial and final direction of the absorption and emission of the radiation field. The equation of motion of the QD density matrix, $\rho$, coupled with the thermal bath of photons can be calculated after tracing over photon degrees of freedom. Here, $\rho$ represents the density matrix of a single exciton interacting with a single MI. Assuming that photons are in thermal equilibrium and are weakly coupled with excitons in QDs, the equation of motion for exciton density matrix, $\rho$, can be calculated perturbatively. Up to the second order of perturbation, it is straightforward to show that [28]

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [H_{\text{QD}}, \rho] - \frac{\Gamma}{2} n_B \sum_{M_z} \{\langle 0, M_z | \langle 0, M_z | \rho - 2 \langle X_b, M_z | \langle 0, M_z | \rho \rangle \langle 0, M_z | X_b, M_z | \}

+ \rho \langle 0, M_z | \langle 0, M_z | \rho \rangle \langle X_b, M_z | \langle 0, M_z | + \rho \langle X_b, M_z | \langle X_b, M_z | \}

- 2 \langle X_b, M_z | \langle 0, M_z | \rho \rangle \langle X_b, M_z | \langle 0, M_z | + \rho \langle X_b, M_z | \langle X_b, M_z | \}),$$

(3)

where $n_B = 1/(e^{\epsilon_{0}h_{b}/k_{B}T} - 1)$ is Bose–Einstein distribution function and $\Gamma = \frac{4\omega_k^2}{3\hbar c^3}$ is the transition rate for the spontaneous emission of photons. $\xi$ is the dipole moment matrix element. Note that in equation (3) vacuum can be considered as a shelving state.

The numerical solutions of equation (3) at zero-temperature ($n_B = 0$) are shown in figure 2 for a QD with $E_d = 2$ eV and $\delta = E_b - E_d = 0, 1, 5$ meV. Here, we used $J_{\text{em}} = 1$ meV and $J_{\text{hm}} = 4$ meV. The initial state of MI is completely uncorrelated with half of the spins populated in the up-direction. As it is shown, because of the coupling with the bath of photons, bright-exciton decays to vacuum without flipping the MI spin and mixing with $X_d$, e.g. $|X_b, M_z \rangle \rightarrow |0, M_z \rangle$. However, a coherent Rabi oscillation between $X_b$ and $X_d$ via exchange with MI is responsible for spin-transfer to MI. In figure 2(b), the time evolution of the components of the ensemble-averaged magnetization of MI, $\langle M_\alpha \rangle = \text{Tr}(\rho M_\alpha)$ with $\alpha = x, y, z$ are depicted for $\delta = 0$. As it is shown, $\langle M(t) \rangle$ exhibits under-damped oscillations around a positive field that
Figure 2. Time evolution of density matrix, equation (3), with the initial condition \( \rho_1(t = 0) = \rho_{22}(t = 0) = 0.5 \) and \( \rho_{ij}(t = 0) = 0 \) for other \( i \) and \( j \)'s, corresponding to the initial random state of MI, are shown in (a), (c) and (d) for \( \delta = 0, 1, 5 \) meV, where \( \delta = 3\Delta_0/2 \) is the bright–dark exciton energy splitting. In (b), the expectation value of MI spin for \( \delta = 0 \) is shown. Here, \( |1\rangle = |X_b, \downarrow\rangle, |2\rangle = |X_b, \uparrow\rangle, |3\rangle = |X_d, \downarrow\rangle, |4\rangle = |X_d, \uparrow\rangle, |5\rangle = |0, \downarrow\rangle, |6\rangle = |0, \uparrow\rangle \).

The population of vacuum can be calculated by \( \rho_{\text{vacuum}} = \rho_{55} + \rho_{66} \). The elements of density matrix, not plotted in this figure, are all identical to zero.

decays to zero as a function of time. In figure 2, we find that \( \rho_1(t) = e^{-\Gamma t/2} \) and \( \rho_{55} = (1 - e^{-\Gamma t})/2 \) fit perfectly the numerical solution of \( \rho_1 \) and \( \rho_{55} \) for all \( \delta \)'s. The decay channel of dark-exciton is through a transition to bright-exciton and spin-flip of MI. This process is schematically depicted in the inset of figure 3. A strong dependence of dark-exciton population on \( \delta \) is seen in figure 2.

Consistent with the time evolution of the density matrix, we propose an exciton wavefunction that fits the density matrix via \( \rho(t) = |\psi(t)\rangle\langle\psi(t)| \):

\[
|\psi(t)\rangle = C_{b\downarrow}e^{-\Gamma t/2}|X_b, \downarrow\rangle + C_{b\downarrow}\sqrt{1-e^{-\Gamma t}}|0, \downarrow\rangle + C_{b\uparrow}\cos(J_{em}t/\hbar)e^{-\Gamma t/2}|X_b, \uparrow\rangle + C_{0\downarrow}\sqrt{1-e^{-\Gamma t}}|0, \uparrow\rangle + C_{d\downarrow}\sin(J_{em}t/\hbar)|X_d, \downarrow\rangle + C_{d\uparrow}|X_d, \uparrow\rangle
\]

with \( C_{b\downarrow} = 1/\sqrt{2} \). Note that \( |X_b, \uparrow\rangle \) and \( |X_d, \downarrow\rangle \) coherently oscillate because \( J_{em} \) in off-diagonal elements of \( H_{em} \) mix these two states. Also, from \( \rho_{66}(\Gamma t \gg 1) \rightarrow 1/2 \), we deduce \( |C_{0\downarrow}(\Gamma t \gg 1)| \rightarrow 1/\sqrt{2} \) and finally \( C_{d\uparrow}(t) \rightarrow 0 \) because \( \rho_{44} = 0 \). The rest of the coefficients in \( |\psi(t)\rangle \) can be determined numerically by fitting to the solutions of density matrix that also fulfill the normalization of the wavefunction \( \langle\psi|\psi\rangle = 1 \).
Figure 3. Time evolution of $S_z$ of a train of injected photo-electrons inside QD (circles) and a single MI (stars) with spin $S = 1/2$ and no nuclear spin. The inset shows the $\Lambda$-shape three level optical resonance of bright- and dark-exciton ($X_b$ and $X_d$). The Rabi oscillation between $X_b$ and $X_d$ occurs because of the exchange interaction between the exciton and the system of MI and nuclear spins. The optical selection rule allows decay of $X_b$ to vacuum. However, the population of $X_d$ decreases indirectly through the conversion of $X_d$ to $X_b$.

The results obtained in this section illustrate that a bright-exciton transfers the angular momentum of the CW laser field to the magnetization of the MI during the transient time, $t < t_t \approx 10\Gamma^{-1}$. However, it gradually loses the magnetization to the environment via annihilation of the exciton within the exciton annihilation time $t_t$. Two vacuum states $|0, \uparrow \rangle$ and $|0, \downarrow \rangle$ are equally populated within $t_t$, hence the final MI magnetization is randomized and its ensemble average vanishes. Note that the lack of DMP and the build up of MI magnetization is the consequence of the ground state with random and uncorrelated states of MI. If the annihilation of the exciton is selectively blocked for one type of spin of MI, e.g. by interruption of the decay process by quantum jumps within $t < t_t$, a dramatic change in the dynamics of the system occurs due to the interaction between MI and other excitons in the environment and a final state with non-vanishing MI magnetization appears. As seen in figure 1, the time evolution of the density matrix predicts that the state of MI after first quantum jump ($t < t_t$) is partially spin polarized. Thus, the MI with partial spin polarization interacts with the second exciton tunneling in from the small quantum dot and as a result a net spin polarization builds up. The rest of this paper is devoted to the discussion of the DMP by quantum jumps.

3.1. Quantum jump algorithm

As noted in [27], the detection of photons from a single quantum system requires spontaneous emission due to vacuum fluctuations, i.e. the photon emission is a stochastic process, described by quantum jump approach [26, 27]. The time evolution of the density matrix of a QD interacting with photons is given by equation (3). At zero temperature, $n_B = 0$ and the time evolution of the density matrix is described by a standard Lindblad master equation.
\[
\frac{d\rho}{dt} = -\frac{i}{\hbar}[H_{QD}, \rho] - \frac{\Gamma}{2} \sum_{M_z} \left( |P_{M_z}^\dagger P_{M_z} , \rho\rangle - 2P_{M_z} \rho P_{M_z}^\dagger \right),
\]

where \{\cdots\} is the anti-commutator. Comparing equation (5) with equation (3), we identify quantum jump operator \(\hat{P}_{M_z} = |0, M_z\rangle\langle X_b, M_z|\) and its Hermitian conjugate \(\hat{P}_{M_z}^\dagger = |X_b, M_z\rangle\langle 0, M_z|\) that project the excitonic state onto vacuum and vice versa without flipping spin of MI, hence \(\sum_{M_z} \hat{P}_{M_z}^\dagger \hat{P}_{M_z} = |X_b, \uparrow\rangle\langle X_b, \uparrow| + |X_b, \downarrow\rangle\langle X_b, \downarrow|\). At each instance of time, \(t\), the density matrix can be divided into a series of density matrices, each representing a specific quantum trajectory associated with a sequence of randomly generated quantum jumps in the interval of time \([0, t]\). Hence, the QD density matrix can be calculated by ensemble average of density matrices over all quantum jump trajectories.

The formulation of quantum jump starts from equation (5). In the absence of MI, a recipe for quantum jump algorithm can be found in [26]. For the completeness of our presentation, we first review this algorithm and then generalize it to exciton in the presence of MI and nuclear spins. We consider optical transition in a two-level system consisting of bright exciton and vacuum without considering an intermediate transition to dark exciton. This condition is fulfilled if we disregard the presence of any MI and nuclear spin. Here, the quantum jump operators are \(\hat{P} = |0\rangle\langle X_b|, \hat{P}^\dagger = |X_b\rangle\langle 0|\), hence \(\hat{P}^\dagger \hat{P} = |X_b\rangle\langle X_b|\). Starting at \(t = 0\) with the initial condition \(|\psi(t = 0)\rangle = |X_b\rangle\), we calculate the time evolution of the system in discrete time steps \(\delta t\). In each time step, we evaluate the quantum jump probability by calculating \(\delta q_0 = \Gamma \delta t \langle \psi | \hat{P}^\dagger \hat{P} | \psi \rangle = \Gamma \delta t\) and drawing a random number \(r\). If \(r < \delta q_0\), a quantum jump occurs and \(|\psi\rangle\) collapses to \(|0\rangle\), otherwise \(|\psi(t + \delta t)\rangle = e^{-i\delta t \hat{H}^\dagger} |\psi(t)\rangle\). Here, the generator for the time-evolution operator is a non-Hermitian Hamiltonian \(\hat{H}_{eff} = -i\hbar \Gamma \hat{P}^\dagger \hat{P} / 2\). So, at \(t = 0 + \delta t\), we have \(|\psi(0 + \delta t)\rangle = e^{-i\delta t |X_b\rangle\langle X_b| / 2} |X_b\rangle = e^{-i\delta t / 2} |X_b\rangle + \sqrt{1 - e^{-i\delta t}} |0\rangle\). The last term keeps the norm of \(|\psi\rangle\) constant (if we use the norm of wavefunction as a constraint in our calculation). At this time, \(\delta q_1 = \Gamma \delta t e^{-i\delta t}\). We draw \(r\); if \(r < \delta q_0 + \delta q_1\), then \(|X_b\rangle\rightarrow |0\rangle\) and a photon is detected and calculation is terminated. Otherwise, \(|\psi(\delta t + \delta t)\rangle = e^{-i\delta t |X_b\rangle\langle X_b| / 2} |\psi(0 + \delta t)\rangle = e^{-i\delta t / 2} |X_b\rangle + \sqrt{1 - e^{-i\delta t}} |0\rangle\). In \(n\)th-step, \(\delta q_n = \Gamma \delta t e^{-n\Gamma \delta t}\), thus, we calculate a cumulative quantum jump probability

\[
\delta p_n = \sum_{k=0}^{n} \Gamma \delta t e^{-k\Gamma \delta t} = \Gamma \delta t \frac{1 - e^{-(n+1)\Gamma \delta t}}{1 - e^{-\Gamma \delta t}}
\]

and if \(r < \delta p_n\) quantum jump occurs. As the time advances, the chance for a quantum jump becomes more likely; however, the probability amplitude for \(X_b\) in \(|\psi\rangle\) decreases with the same rate simultaneously. In the \(n\)th-step if there is still no quantum jump, then \(|\psi(n \delta t)\rangle = e^{-i\delta t |X_b\rangle\langle X_b| / 2} |\psi((n-1) \delta t)\rangle = e^{-i\delta t / 2} |X_b\rangle + \sqrt{1 - e^{-i\delta t}} |0\rangle\).

The quantum jump algorithm in the presence of MI is similar to the one in the absence of MI, with a difference that the time evolution of the wavefunction is generated by an effective Hamiltonian \(\hat{H}_{eff} = H_{QD} - i\hbar \Gamma \delta t \hat{P}^\dagger \hat{P} / 2\) that allows an intermediate transition to the dark-exciton due to spin exchange with MI. Therefore, the description of quantum jump process in the presence of MI is based on a three-level system depicted in the inset of figure 3 and consist of \(|0\rangle, |X_b\rangle, |X_d\rangle\) and MI.

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3.2. Conservation of total angular momentum

In this section, we discuss the transfer of spin angular momentum to MI/NS. For illustration, consider a spin-1/2 MI in interaction with a bright exciton with total angular momentum $L_z = -1$ that is created in a quantum dot at time $t = 0$ by, e.g., tunneling from a smaller dot as in the experiment by Goryca et al [8]. In the experiment, because of the imperfection in spin-transfer mechanism, the laser creates a small population of excitons with opposite polarization relative to the polarization of the external field. In our model, without losing the generality, we focus on the DMP mechanism induced only by the majority of the spin-polarized excitons. The exciton is in a pure state with electron and hole spins along quantization axis, $\hat{z}$, 1/2 and $-3/2$, respectively. The spin of MI is initially in a random state $|M_z(t = 0)\rangle = a_0|\uparrow\rangle + b_0|\downarrow\rangle$, where $a_0$ and $b_0$ are two complex random numbers, satisfying the normalization condition, $|a_0|^2 + |b_0|^2 = 1$. Hence, the starting configuration of exciton and magnetic ion is characterized by their expectation value of total spin $\langle L_z \rangle = -1$ and $\langle M_z \rangle = (|a_0|^2 - |b_0|^2)/2$.

The bright exciton and magnetic ion complex begins to evolve in time into a linear combination of bright and dark exciton states entangled with MI because of e–MI exchange interaction. The wavefunction of such a state involves the entanglement between bright and dark excitons with angular momentums $L_z = -1$ and $-2$, respectively, i.e. $|\Psi(t)\rangle = |L_z = -1\rangle(a|\uparrow\rangle + b|\downarrow\rangle) + |L_z = -2\rangle(c|\uparrow\rangle + d|\downarrow\rangle)$. In this situation, clearly the angular momentum of bright exciton is transferred to both dark exciton and the modified angular momentum state of MI. Here, $|a|^2 + |b|^2 + |c|^2 + |d|^2 = 1$. The expectation values of angular momentum of MI and electron are $\langle M_z(t)\rangle = (|a|^2 + |c|^2 - |b|^2 - |d|^2)/2$ and $\langle S_z(t)\rangle = (|a|^2 + |b|^2 - |c|^2 - |d|^2)/2$. In our model, the heavy-hole spin does not flip, thus $\langle L_z(t)\rangle = -3/2 + \langle S_z(t)\rangle + \langle M_z(t)\rangle = -3/2 + |a|^2 - |d|^2$. The conservation of angular momentum implies $\Delta J_z = \langle J_z(t)\rangle - (-1 + (|a_0|^2 - |b_0|^2)/2) = -1/2 + |a|^2 - |d|^2 - |a_0|^2/2 + |b_0|^2/2 = 0$.

At the quantum jump time, the observer detects a bright exciton with angular momentum $L_z = -1$ emitted in the $\hat{z}$-direction, perpendicular to the plane of the quantum dot. In principle, an observer could carry out the measurement of dark exciton by applying a small in-plane magnetic field (Voigt geometry). In Voigt geometry, there would be an obvious transfer of angular momentum from bright exciton to dark exciton. Another alternative is to observe the magnetic field (Voigt geometry). In Voigt geometry, there would be an obvious transfer of angular momentum from bright exciton to dark exciton.

Hence, in our present discussion, we focus on the detection of bright exciton. The quantum jump detection of a photon with $J_z = -1$ implies projection of the exciton–MI state onto the bright exciton state. Both bright and dark excitons are erased simultaneously (there is no electron nor hole left after we observed a photon) even though we observe only bright exciton (due to mechanisms responsible for the collapse of dark exciton including small spin–orbit coupling that opens a channel in transferring angular momentum to MI through the underlying lattice). Because we observe bright exciton, the MI ion state left behind is $|M_z(t)\rangle = a(t)|\uparrow\rangle + b(t)|\downarrow\rangle$. The expectation value of MI, $\langle M_z(t)\rangle = (|a(t)|^2 - |b(t)|^2)^2$, is different from the expectation value of $\langle M_z \rangle$ at $t = 0$. When the quantum dot is refilled, we start with bright exciton and
MI in the normalized state $|M_z(t)⟩ = a(t) |↑⟩ + b(t) |↓⟩$. The expectation value of the total angular momentum is now changed from $t = 0$ and the change is related to the erasure of dark exciton wavefunction and the compensating change in MI spin polarization. This explains the mechanism that MI changes its magnetic moment. We could detect it by detecting dark exciton in Voigt geometry and comparing the intensity of emission from bright and dark excitons.

4. Dynamical evolution of magnetic impurity by a train of excitons in the presence of nuclear spins

As illustrated in figure 1(a), a small quantum dot is continuously refilled by a non-resonant, circularly polarized CW laser. The spin-polarized excitons transfer into the QD containing the complex spin system MI. We assume therefore a train of incoming bright excitons $|X_0⟩ = |↓, 1⟩$ interacting with MI in the quantum dot. Each electron in the exciton transfers spin to MI and creates a superposition of dark and bright excitons entangled with MI and nuclear spins. At the bright exciton recombination time, a $t_r$, a photon is detected, a quantum jump takes place, the dark exciton wavefunction is erased and the MI and nuclear spin complex are left in a modified state. The exciton removal is performed by using the quantum jump projector method [26, 27] described below which yields the modified wavefunction of the MI and nuclear spins. New spin-polarized exciton tunnels into the quantum dot and begins interaction with MI and nuclear spins modified by electron spin of previous exciton.

The basis for a combined exciton–spin system is composed of three groups of basis states: vacuum $|0, M_z, I_{z1}, \ldots, I_{zN_h}⟩$, bright exciton $|X_b, M_z, I_{z1}, \ldots, I_{zN_h}⟩$ and dark exciton $|X_0, M_z, I_{z1}, \ldots, I_{zN_h}⟩$. Only the vacuum and bright exciton group of the states are coupled to the photon field via projectors $P_± = |0, λ⟩ ⟨X_b, λ|$. Here, states $|λ⟩ = |M_z, I_{z1}, \ldots, I_{zN_h}⟩$ describe a total of $N_5 = (2M_z + 1)(2I_z + 1)^N_h$ complex spin MI and nuclear spin states.

The time evolution of the density matrix $ρ = |Ψ⟩ ⟨Ψ|$ in ME, equation (5), can be generalized by $M_z → λ$. As described in section 3.1, the wavefunction $|Ψ⟩$ subjected to stochastic ‘birth–death’ process [29] of recombination and photo-excitation can be used to describe the time propagation of the system coupled with the radiation field and undergoing the quantum jump process. At $t = 0$, we start with the initial state $|Ψ^{n=0}(t = 0)⟩ = |0⟩ |X_0⟩$ of MI and the nuclear spin bath. The index $n$ counts the number of quantum jump events. The state $|X_0⟩ = \sum_λ C_λ^{n=0} |λ⟩$ is a random linear combination of all possible configurations with the coefficients $C_λ^{n=0}$ being uniformly distributed random complex numbers. We note that if we were to compute an expectation value $⟨M_z⟩$ for this random state, we would obtain a finite value. However, averaging over many sets of $C_λ^{n=0}$ yields no initial magnetization.

At $t = 0^+$, a bright exciton created in neighboring QD enters the central QD. The creation of $|X_b⟩$ and annihilation of $|0⟩$ are described by operator $|X_b⟩ ⟨0|$. The initial wavefunction of the injected bright exciton, MI and nuclear spins is an uncorrelated state. However, the Hamiltonian $H_{QD}$ that accounts for the exchange coupling creates quantum correlation in the exciton–MI–nuclei complex and $|Ψ⟩$ evolves into a linear combination of all configurations, including an entangled state between bright and dark excitons. Simultaneously, the bright-exciton decays into vacuum because of coupling with quantized electromagnetic field.

To be consistent with the quantum jump algorithm, we discretize the time $t$ into small steps $δt$. Note that because of the small eh- and MI–nuclear-spin couplings ($J_{ne}$, $J_{nh}$ and $J_{nt}$), the excitons and MI evolve in a frozen-fluctuating field of nuclear spins [32–34]. The eh recombination time, $t_r$, is the smallest time scale in our model, hence $δt ≪ t_r$. 

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The time evolution of the wavefunction of eh–MI–nuclei complex is calculated in the Schrödinger picture [33] by using the relation $|\Psi(t + \delta t)\rangle = \exp(-i H_{\text{eff}}\delta t/\hbar)|\Psi(t)\rangle$. Here, $|\Psi(t)\rangle$ is the wavefunction of the entire system, $H_{\text{eff}} = H_{\text{QD}} - i\hbar(\Gamma/2)\sum_\lambda P_\lambda^\dagger P_\lambda$ where the last term describes the decay of bright exciton due to coupling with photon field. Because $H_{\text{eff}}$ is time independent, we employ the method based on Bessel–Chebyshev polynomial expansion [33] to calculate the time evolution of the wavefunction. Note that $H_{\text{QD}}$ is Hermitian and thus $\exp(-i H_{\text{QD}}\delta t/\hbar)$ is a unitary operator that conserves the norm of wavefunction. This is in contrast with the operator $\exp(-\Gamma \delta t/2 \sum_\lambda P_\lambda^\dagger P_\lambda)$ that is non-unitary and does not preserve the norm of wavefunction; however, because it describes the decay of $X_b$ into vacuum, we build a norm-conserving wavefunction by adding vacuum. Using an iterative procedure to propagate the wavefunction in time we find

$$|\Psi_{n=1}(t)\rangle = \sum_\lambda |\lambda\rangle[C_{X_{b,\lambda}}^{n=1}(t)|X_b\rangle + C_{0,\lambda}^{n=1}(t)|0\rangle + C_{X_{d,\lambda}}^{n=1}(t)|X_d\rangle], \quad (7)$$

where the coefficients $C_{X_{b,\lambda}}^{n=1}(t)$, $C_{0,\lambda}^{n=1}(t)$ and $C_{X_{d,\lambda}}^{n=1}(t)$ are determined numerically. This wavefunction describes a correlated state of bright and dark excitons as well as vacuum. Because of the spin flip-flop process of electron with MI and nuclear spins, the initially formed bright exciton $|X_b\rangle$ mixes with the dark exciton $|X_d\rangle$.

From the Lindblad ME, the quantum jump transition rate is given by $\Gamma_{\text{jump}} = \Gamma\rho_b$. $\rho_b$ represents the population of the bright exciton obtained from the full QD density matrix after tracing over MI and nuclear spin degrees of freedom. The quantum jump probability $\delta\rho_{\text{jump}} = \Gamma_{\text{jump}}\delta t$ is then calculated and compared with a random number $r$ generated between zero and one. If $r < \int_0^\delta t dr\Gamma_{\text{jump}}$, a quantum jump takes place, a photon is recorded and the quantum dot is in the ground state. The elapsed time $t_r$ recorded for this quantum jump is the eh-recombination time. At $t = t_r$, we allow the exciton to annihilate by the spontaneous emission of a photon. The operator that allows annihilation of bright exciton and the creation of vacuum is $|0\rangle\langle X_b|$. Immediately after the annihilation of exciton, a new spin-polarized exciton tunnels into the quantum dot from the neighboring dot. The spin-polarized exciton interacts with the MI spin $M$ and nuclear spins $I$ in a state modified by the previous exciton. At $t = t_r + 0^+$, the second bright exciton $X_b$ is created in the neighboring QD tunnels into the central QD.

Note that there is no type of linear combination between $|0\rangle$ and $\{|X_b\rangle, |X_d\rangle\}$ because there are no Rabi-oscillations between vacuum and excitons. The quantum correlation appears from the time evolution of wavefunction generated by exchange couplings in $H_{\text{eff}}$ right after $t = t_r$.

The annihilation and creation of a bright exciton is described by a projector $Q_{n \to n+1} = |X_b^{n+1}\rangle\langle X_b^n|$. The superscripts refer to the annihilated $n$th and created $(n+1)$th exciton. Note that $Q_{n \to n+1}|X_b^n\rangle = |X_b^{n+1}\rangle$ and $Q_{n \to n+1}|X_d\rangle = Q_{n \to n+1}|0\rangle = 0$, hence the quantum jump operator projects out any correlated state composed of the superposition of bright and dark excitons to a new born bright exciton. The new wavefunction in the QD can then be constructed as $|\Psi(t = t_r^+)\rangle = Q_{n \to n+1}|\Psi(t = t_r^-)\rangle$ where $t_r^\pm = t_r \pm \eta$ and $\eta \to 0$. In this state, $|X_b^{n+1}\rangle$ is initially uncorrelated from MI and nuclear spins. At $t = t_r^+$, it can be expressed as

$$|\Psi_{t_r^+}\rangle = |X_b^{n+1}\rangle \sum_\lambda A_n C_{X_{b,\lambda}}^n(t_r)|\lambda\rangle. \quad (8)$$

We observe that after the quantum jump, the new injected exciton $X_b^{n+1}$ starts with the normalized (factor $A_n$) state of MI and nuclear spins $\sum_\lambda C_{X_{b,\lambda}}^n(t_r)|\lambda\rangle$ which was left over by
the previous bright exciton \( X^b_n \) at the time of radiative recombination. Detecting a photon erased the dark exciton wavefunction and modified the state of both MI and nuclear spins. This is the DMP mechanism discussed here. With the initial condition established, the time evolution of the entangled state of photo-carriers with MI and spin bath can be calculated after updating the coefficients \( C \)’s. At the end, one needs to average over initial conditions. Although the procedure discussed here describes the immediate refilling of central QD after the annihilation of the exciton, we can always implement a waiting time between the recombination and refilling process.

5. Numerical results and discussion

Our approach to DMP is illustrated using parameters based on (Cd,Mn)Te QDs with \( \tilde{J}_{em} = 15 \), \( \tilde{J}_{hm} = 60 \) meV nm\(^3\) corresponding to the exchange coupling in the bulk materials, hence \( J_{em} = \tilde{J}_{em} |\phi_e(R_m)|^2 \) and \( J_{hm} = \tilde{J}_{hm} |\phi_h(R_m)|^2 \). The circular symmetry of quantum dots is implemented by assuming \( \Delta_\gamma = 0 \). Here, \( \phi_{e/h}(R_m) \) is the e/h envelope-wavefunction in the central dot at \( R_m \), the position of MI. We assume \( J_{eh} = 0.6 \) meV [30] and initialize \( J_{ne}, J_{nh}, J_{nn} \) and \( J_{nn'} \) as random numbers with a mean value of the order of 1 \( \mu \)eV. However, we note that the realistic value for nuclear hyperfine interaction is reported within 1 neV [35–37] three orders of magnitude smaller than the energy scales used in our finite size calculation.

Here, we discuss numerical results with nuclear spins, immediately after the refilling of QD by bright exciton. Figures 4–7 illustrate DMP/DNP and the quantum jump trajectories for exciton, MI and nuclear spins. In figure 4(a), the single quantum jump trajectory for spin-1/2 MI is plotted. In figures 5–7, the ensemble average of 20 quantum jump trajectories for spin-1/2 (figure 5), spin-1 (figure 6) and spin-5/2 (figure 7) MI are plotted. The trajectories are time evolution of the initial spin wavefunctions which are generated in a random linear combination of spin configurations. Each curve consists of thousands of time steps and points. For the clarity of the legends, after every hundred points, symbols like circle, star and triangle are superimposed on each curve. As shown, the MI and the average polarization of \( N_h = 15 \) 

Figure 4. Time evolution of \( S_z \) of a train of injected photo-electrons inside QD (circles), a single MI (stars) and the average of \( N_h = 15 \) nuclear-spin polarization (triangles).
nuclear spins gradually build up by a train of injected bright excitons. At $t = t_r$, one pair of eh collapses into vacuum with $\langle S_e,z(t) \rangle < 1/2$, as part of the $e$-spin is transferred to MI. An empty dot instantaneously absorbs the second photo-generated eh pair with total angular momentum $j_z = -1$ which transfers to the spin of MI and nuclei before its removal. We repeat this procedure until the spin polarization of MI and nuclear spins is built-up. The method presented here is limited to a finite number of nuclear spins because of exponentially increasing computational effort with the number of spins. However, a systematic study of the convergence of the numerical results by increasing $N_b$ shows satisfactory outcomes around $N_b = 15$.

We now discuss DMP for MIs with more than one localized electron. We consider two cases of MI with two and five electrons localized in open-shell $p/d$-orbitals. The spin Hund’s rule implies that the total spin of the electronic ground state of MIs is maximum. For two electrons, the spin-triplet manifold $(M = 1)$ is separated from the higher energy spin-singlet state $(M = 0)$ with the singlet-triplet energy gap $E_{M=0} - E_{M=1} = |J_m|$. Here, $J_m$ is the ferromagnetic exchange coupling between two electrons localized in MI. Similarly, the lowest energy state of MI (e.g. Mn) with five d-electrons corresponds to the total spin $M = 5/2$ with a six-fold degeneracy. These states are separated from higher energy spin-manifold with energy gap proportional to $J_m$. Considering $J_m$ few times larger than other exchange couplings avoids mixing ground state with the higher energy excited states of MI.

For a system containing spin-1 MI, we consider an ensemble with equally populated states $M_z = \pm 1, 0$ (1/3 for each $M_z$). Similarly, the ensemble of Mn’s contains $M_z = \pm 5/2, \pm 3/2, \pm 1/2$ with equal population (1/6 for each $M_z$). Thus, the ensemble average over all possible QJ trajectories includes the overall summation $S_z$. In figure 6(a), we show the time evolution of spin-triplet states initially started from $M_z = \pm 1, 0$. We observe that MI with $M_z = -1$ switches polarization to $\langle M_z \rangle \approx +1$ because of the strong interaction with excitons that allow the DMP mechanism to proceed efficiently. It is therefore expected that the polarization of $M_z(t = 0) = +1$ does not alter dramatically, although it is initially decohered by nuclear spins, but we find that it stays polarized with $\langle M_z \rangle \approx +1$ because of the strong interaction with the train of excitons. On the other hand, MI with initial polarization $M_z(t = 0) = 0$ fluctuates around $M_z = 0$. Analogous to its spin-singlet counter part, the spin-triplet $M_z = 0$
Figure 6. (a) Time evolution of the magnetic moment of an ensemble of MIs with two localized p/d-electrons in ferromagnetically ordered spin-triplet $M = 1$ and $M_z = \pm 1, 0$ interacting with a train of injected photo-electrons inside QD, and $N_b = 15$ nuclear spins. Unlike $M_z = \pm 1$ that interacts strongly with excitons, $M_z = 0$ exhibits a weak interaction. The average of three states shows saturation close to $\langle M_z \rangle = \frac{1}{3}(2 \times 1 + 0) = 0.67$ as the ensemble is populated equally among all possible spin-triplet states. (b) Time evolution of an ensemble average of spin of MI (stars), photo-electrons (circles) and $N_b = 15$ nuclear spins (triangles).

Figure 7. Time evolution of the magnetic moment of the individual states of Mn (a) and an ensemble of Mn’s (b) with five localized d-electrons (stars) interacting with a train of injected photo-electrons inside QD (circles), and $N_b = 15$ nuclear spins (triangles). The ensemble is constructed with equally populated $M_z = \pm 5/2, \pm 3/2, \pm 1/2$ among finite number of Mn’s. The average of six states show saturation close to $\langle M_z \rangle = \frac{1}{6}(2 \times \frac{5}{2} + 2 \times \frac{3}{2} + 2 \times \frac{1}{2}) = \frac{9}{6} = 1.5$ as the ensemble is populated equally among all possible spin-triplet states.

weakly interacts with excitons and nuclear spins \cite{38}. Hence, the polarization obtained for this ensemble indicates that the final state is a mixture of all spin-triplet configurations.

\footnote{Under particular conditions, beyond the present model, strong coupling between spin-singlet states and MI leads to the formation of spin texture and molecular states of magnetic polarons. Such possibilities were investigated recently in \cite{38}.}

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with maximum achievable polarization \(\langle M_z \rangle = \frac{1}{3}(2 \times 1 + 0) = \frac{2}{3} = 0.67\) as \(M_z = \pm 1\) equally contribute to the ensemble average of \(\langle M_z \rangle\). Similarly, we can predict that the maximum spin polarization achievable for the ensemble of Mn can be calculated by \(\langle M_z \rangle = \frac{1}{6}(2 \times \frac{3}{2} + 2 \times \frac{1}{2} + 2 \times \frac{1}{2}) = \frac{9}{6} = 1.5\) as shown in figure 7. It is straightforward to show that mixing with higher energy excited states suppresses the magnetic saturation down to \(\langle M_z \rangle = 1.1\) if the final state is a mixture of equally populated all spin multiplicities of five spin-1/2 electrons. The results shown in figure 7 suggest that the saturation of \(\langle M_z \rangle\) occurred between 1.1 and 1.5, which might be interpreted as an indication of leakage of the optically pumped ground state of Mn to its excited states.

6. Summary

In conclusion, dynamical magnetic and/or nuclear polarization in single quantum complex spin systems is discussed for the case of spin transfer from exciton to the central spin of magnetic impurity in a quantum dot in the presence of a finite number of nuclear spins. The exciton is described in terms of the electron and heavy-hole spins interacting with magnetic impurity via exchange interaction, with a finite number of nuclear spins via hyperfine interaction and with photons via dipole interaction. The time evolution of the exciton, magnetic impurity and nuclear spins is calculated exactly between quantum jumps corresponding to exciton radiative recombination. The collapse of the wavefunction and the refilling of the quantum dot with new spin-polarized exciton is shown to lead to a build up of magnetization of the magnetic impurity as well as nuclear spins. The competition between electron spin transfer to magnetic impurity and to nuclear spins simultaneous with the creation of dark excitons is therefore elucidated. The technique presented here opens up the possibility of studying optically induced DMP in complex spin systems.

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