Optical Pumping of Quantum-Dot Nuclear Spins

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Hyperfine interactions with randomly oriented nuclear spins present a fundamental decoherence mechanism for electron spin in a quantum dot, that can be suppressed by polarizing the nuclear spins. Here, we analyze an all-optical scheme that uses hyperfine interactions to implement laser cooling of quantum-dot nuclear spins. The limitation imposed on spin cooling by the dark states for collective spin relaxation can be overcome by modulating the electron wave function.

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One of the principal features that distinguish a quantum dot (QD) from an atom is the completely different role that hyperfine interactions play in the two systems. In contrast to valence electrons of an atom, a quantum dot (conduction band) electron has confinement length scales that extend over many lattice sites. As a result, a single electron spin interacts with $N \simeq 10^3$–10^5 nuclei and the interaction strength with each nucleus is reduced by a factor determined by the probability of finding the electron at that lattice site ($\sim 1/N$). Equivalently, the electron hyperfine field seen by each nucleus will be extremely weak and the nuclei will remain unpolarized. Interactions with this random nuclear-spin orientation present a fundamental decoherence mechanism for an electron spin confined in a quantum dot [1–3].

In this Letter, we propose an all-optical technique to polarize the nuclear spins interacting with a single quantum-dot electron spin. The basic idea is to use the hyperfine coupling to induce a controlled electron-nuclear spin-flip process. This can be achieved by changing the energy of the initial (spin-up) electronic state using the ac Stark effect [4], in order to allow for resonant electron-nuclear spin-flip to take place. When the spin-flip is completed, the electron spin is reflipped into its original state using a laser-induced π pulse followed by spontaneous emission. Starting from a random unpolarized ensemble nuclear-spin state, each step as described above flips one nuclear spin, albeit in a collective way. Resonance condition between the spin-up and spin-down electronic states can also be achieved in subnanosecond time scales using the electric-field dependence of the electron g factor [5], without requiring a laser-induced ac Stark effect.

Before proceeding, we note that partial polarization of nuclear spins using hyperfine interactions and optical pumping is well studied [6]. More recently, dynamical polarization of lattice nuclei (the Overhauser effect) has been demonstrated in interface QDs which form due to monolayer fluctuations of the thickness of a GaAs quantum well [7]: Here, a circularly polarized laser creates electrons in a well-defined spin state, which in turn polarizes the nuclear spins via hyperfine contact interaction. Spin-flipped electrons are then removed from the system by radiative recombination, maintaining a relatively high degree of spin polarization for electrons and the nuclei [7]. One limitation of this dynamic polarization scheme is the fact that in a QD, creation of nuclear-spin polarization eventually makes joint electron-nuclear spin-flip processes energetically forbidden due to the large electron Zeeman energy induced by the nuclear magnetic field. In addition, optical Overhauser effect relies on fast hole-spin relaxation for the removal of the spin-flipped electron by radiative recombination. However, recent experiments [8] demonstrate that hole-spin relaxation is significantly slower in small quantum dots. It is essential to overcome these two difficulties in order to achieve a high level of nuclear-spin polarization in QDs.

We consider a charged quantum dot where a single conduction band electron interacts with $N \simeq 10^4$ (spin-1/2) nuclei. The interaction with the ith nucleus is proportional to the absolute value of the electron wave function squared at that site (α_i, with $\sum_i \alpha_i = N$). The Hamiltonian describing this hyperfine contact interaction is [1]

$$\hat{H}_{\text{int}} = \frac{A}{N}\sum_i \alpha_i \left[\frac{1}{2}\hat{I}_z^i \hat{\sigma}_z + \hat{I}_+^i \hat{\sigma}_- + \hat{I}_-^i \hat{\sigma}_+\right], \qquad (1)$$

where $\hat{I}_k^i$ and $\hat{\sigma}_k$denote the Pauli operators for the ith nucleus and the electron, respectively. $\hat{\sigma}_+ = |\uparrow\rangle\langle\downarrow|$ is the electron spin-flip operator. A is an effective hyperfine interaction constant that takes into account the coupling of all the nuclei in the unit cell; for GaAs $A \simeq 90$ μeV [2].

We assume that the QD is subject to a large constant magnetic field that removes the degeneracy of the electron spin-up and spin-down states; for an electron g factor $g_e \sim 2$, we expect an energy difference of 1 meV with $B \sim 10$ T. The magnetic field is applied along the direction of strong confinement, i.e., $\hat{z}$ for QDs grown by molecular beam epitaxy. For temperatures ($T \sim 3$ K) typical of magneto-optical cryostats, the electron is spin polarized

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(in the spin-up state) and the nuclei are not:

$$|\Psi\rangle = |\psi\rangle_e \otimes |\psi\rangle_N = \hat{\sigma}_+ \prod_j^M \hat{I}_+^j |\phi\rangle, \quad (2)$$

where the product of nuclear-spin operators is over a random set of nuclei. For unpolarized nuclei $M \sim N/2$. $|\phi\rangle = \hat{e}_\downarrow^\dagger \prod_{i=1}^N \hat{n}_{i,\downarrow}^\dagger |0\rangle$, where $\hat{e}_\downarrow^\dagger$ and $\hat{n}_{i,\downarrow}^\dagger$ correspond to the creation operator of the spin-down electron and the ith nucleus, respectively.

We assume that the nuclear spins are in a random but constant state during the time scale over which the electron spin is manipulated. The effective (nuclear) magnetic field [1,2] seen by the QD electron is $B_z^{\rm eff} \sim A/(\sqrt{N} g_e \mu_B)$, where μ_B is the Bohr magneton. The magnetic field along the x and y directions have the same expectation value for unpolarized nuclei.

Interactions with a classical time-dependent laser field are governed by the Hamiltonian

$$\hat{H}_{\rm laser} = \hbar[\Omega_+(t)(\hat{e}_\downarrow \hat{h}_{3/2} + \eta \hat{e}_\uparrow \hat{h}_{3/2}) e^{i\Delta t} + {\rm c.c.}] + \hbar[\Omega_-(t)(\hat{e}_\uparrow \hat{h}_{-3/2} + \eta \hat{e}_\downarrow \hat{h}_{-3/2}) e^{i\Delta t} + {\rm c.c.}], \quad (3)$$

where $\hat{h}_{\pm 3/2}$ denotes a valence band hole state with angular momentum projection $j_z = \pm 3/2$. $\Omega_+(t)$ $[\Omega_-(t)]$ is the time-dependent Rabi frequency of the right- (left-)hand circularly polarized laser field propagating along the z direction and interacting with the strongly allowed QD transition that satisfies the j_z selection rules. Because of heavy-light hole mixing of the valence band states, these selection rules are relaxed in actual QD structures, leading to nonzero but small coupling ($\eta^2 \ll 1$) to optical fields that violate the j_z selection rules [9]. The frequency of the laser field determines the detuning Δ of the optical transition.

In the presence of a large Zeeman splitting, electron-nuclear spin-flip processes are forbidden by energy conservation [Fig. 1(a)]. The first step in the proposed scheme is the application of a red-detuned left-hand circularly polarized (lcp) laser pulse that creates a "spin-state dependent ac Stark effect" that effectively cancels the Zeeman splitting of the electron caused by the external magnetic field [4]. Assuming a detuning $\Delta = 25$ meV and a QD optical oscillator strength $f = 10$, a lcp laser intensity $I_{\rm lcp} \sim 50\ {\rm MW/cm^2}$ is needed to achieve an ac Stark shift of about 1 meV. While this laser is on, electron-nuclear spin-flip process due to $\hat{H}_{\rm int}$ of Eq. (1) is energetically allowed [Fig. 1(b)]. The effective coupling coefficient for spin-flip process for a random initial state is given by

$$g_{\rm spin\text{-}flip} = \left\| \frac{A}{N} \sum_i \alpha_i \hat{I}_+^i \hat{\sigma}_- |\Psi\rangle \right\|, \quad (4)$$

as a direct consequence of the collective enhancement of the transition due to participation of many nuclei. Since $g_{\rm spin\text{-}flip}$ is comparable to $g_e \mu_B B_{\rm eff}$ ($\hbar = 1$) for unpolarized nuclei, we expect significant probability for spin-flip if we leave the laser on for $\tau \sim 1/g_{\rm spin\text{-}flip} <$ 1 nsec [2]. Therefore, choosing a laser pulse width of $\tau \sim 300$ psec $< g_{\rm spin\text{-}flip}^{-1}$ will yield a spin-flip probability of $\sim$0.1. If an electron-nuclear spin-flip event does take place, the state vector of the QD is $|\Psi\rangle_A = \hat{\sigma}_- \sum_i \alpha_i \hat{I}_+^i |\Psi\rangle / \mathcal{N}$, where $\mathcal{N}$ ($\neq N$ in general) is the normalization factor.

After the ac Stark laser is turned off, we turn on a resonant right-hand circularly polarized (rcp) laser field that realizes a π pulse on the quasiforbidden electronic transition $\hat{e}_\downarrow^\dagger |0\rangle \rightarrow \hat{e}_\downarrow^\dagger \hat{e}_\uparrow^\dagger \hat{h}_{3/2}^\dagger |0\rangle$ (with transition amplitude $\propto \eta$) [10], only if an initial electron-nuclear spin-flip process has taken place due to the resonant hyperfine

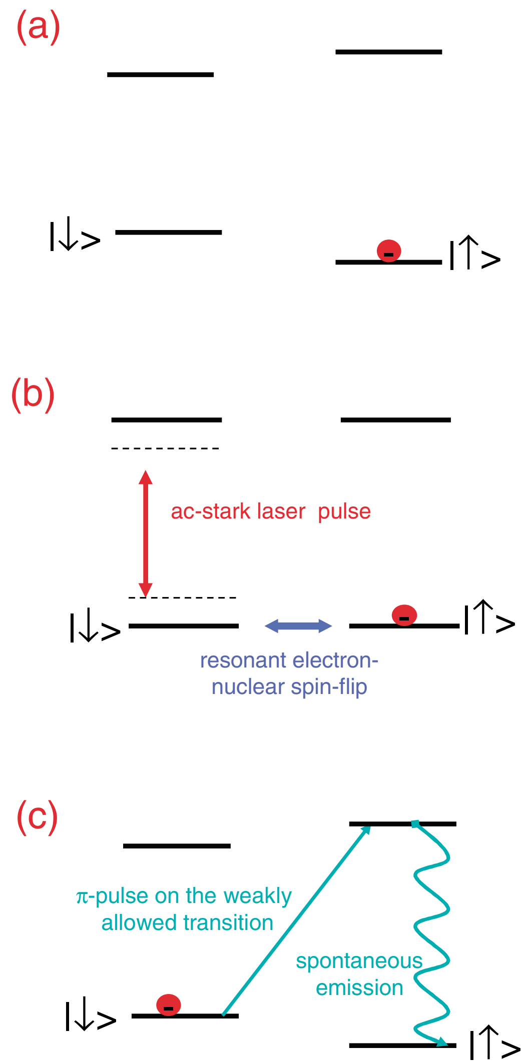

FIG. 1 (color). All-optical manipulation of electron-nuclear spin-flip in a single quantum dot. (a) In the presence of a large Zeeman splitting, electron-nuclear spin-flip events are energetically forbidden. (b) Introduction of a red-detuned laser field can effectively cancel the Zeeman splitting and allow for resonant spin-flip processes due to hyperfine interaction. (c) The spin-flipped electron is repumped into the initial state by a combination of a π pulse, followed by spontaneous emission.

interaction in the presence of the red-detuned lcp laser. If this is the case, the excited trion state $\hat{e}_\downarrow^\dagger \hat{h}_{3/2}^\dagger |\psi\rangle_e$ is populated with probability approaching unity. This excited state will relax down predominantly to the electronic state $|\psi\rangle_e = \hat{e}_\uparrow^\dagger |0\rangle$ by spontaneous emission of a rcp photon with rate $\Gamma_{\rm rad}$, thereby projecting the electron spin onto the initial spin-up state [Fig. 1(c)]. The final state following spontaneous emission is

$$|\Psi\rangle_C = \frac{1}{\sqrt{\mathcal{N}}} \sum_i \alpha_i \hat{I}_+^i |\Psi\rangle. \tag{5}$$

The successive application of two laser pulses followed by spontaneous emission flips a single nuclear spin with probability ~ 0.1 and constitutes the elementary step of the proposed laser cooling scheme for nuclear spins. If electron-nuclear spin-flip due to hyperfine interaction does not take place, then the applied π pulse does not couple an occupied transition and the whole system remains in its initial state $|\Psi\rangle$.

Having discussed the elementary step of flipping of a single nuclear spin collectively, we next turn to the question of the effectiveness of successive applications of this cooling step in achieving large nuclear-spin polarization. First we note that $B_z^{\rm eff}$ will change as the nuclear-spin polarization increases. For unpolarized nuclei $g_e \mu_B B_z^{\rm eff} \sim g_{\rm spin\text{-}flip}$, whereas for nearly polarized nuclei $g_e \mu_B B_z^{\rm eff} \sim A \gg g_{\rm spin\text{-}flip}$. Therefore, the magnitude of the ac Stark shift needs to be adjusted as the nuclear-spin polarization increases. In the proposed scheme, each spin-flip is accompanied by spontaneous emission of a photon and it is in principle possible to estimate the degree of nuclear-spin polarization by counting the spontaneously emitted photons. We can use this information to change the magnitude of the ac Stark shift to ensure resonance condition for electron-nuclear spin-flip processes, for all values of nuclear-spin polarization. Alternatively, we can envision a laser pulse shape that will ensure resonance condition for a sufficiently long interaction time for any $\langle \sum_i \hat{I}_z^i \rangle$.

The principal question that determines a limitation of the proposed scheme is the probability of the nuclear-spin system evolving into a dark state of the Hamiltonian of Eq. (1); i.e., if the nuclear-spin state after n- steps of laser-induced collective spin-flip events ($|\psi\rangle_N^{(n)}$) satisfies

$$\sum_i \alpha_i \hat{I}_+^i |\psi\rangle_N^{(n)} = 0, \tag{6}$$

then the prescribed procedure cannot be utilized to achieve further nuclear-spin polarization with the given $\hat{H}_{\rm int}$. An illustrative example is the case when $\alpha_i = 1, \ \forall i$ and the QD nuclear-spin system is in the first excited state with a single flipped nuclear spin. Of the N states in this manifold, the only state with appreciable coupling ($g_{\rm spin\text{-}flip} \sim A/\sqrt{N}$) to the fully polarized ground state is the completely symmetric state. The other $N - 1$ asymmetric states satisfy Eq. (6).

In the limit of inhomogeneous electron-nuclear spin coupling ($\alpha_i \neq \alpha_j, \ \forall i, j$) that is of practical interest, total nuclear spin $\hat{I}_T^2$ is not conserved and the limitation due to (quasi)dark states will be relevant for the subcollection of nuclear spins for which $\alpha_k \simeq \alpha_l$. A potential remedy in this case is provided by the fact that the spatial wave function of the electron confined in the QD can be modified using external electric fields. This modification will in turn alter the hyperfine interaction coefficients α_i. We can then use the feedback from the measurement of emitted photons to change these coefficients as the cooling progresses: if the number of detected photons falls below a certain predetermined level, this is a good indication that the system has evolved into a quasidark state of $\hat{H}_{\rm int}$ with the current α_i. Based on this information, we can apply an external electric field and change its magnitude or its orientation until we increase the photon detection rate. Alternatively, we can apply a random electric field to ensure that distant nuclei will have $\alpha_i \neq \alpha_j$ for the majority of the elementary cooling steps.

To evaluate the role of dark states in nuclear-spin cooling, we have carried out a numerical simulation of the proposed scheme for a toy system consisting of ten nuclei. When we choose a symmetric Gaussian wave function for the electron, we find that the nuclear polarization saturates at 75% (Fig. 2, solid line). This saturation is due to the dark (singlet) states of pairs of nuclei with identical hyperfine coupling to the electron. We then shift the electron wave function by $0.5a_L$, where a_L is the internuclear separation in this toy model. Since the new hyperfine coupling distribution has a different set of dark states, the polarization increases abruptly and then saturates at a higher level. Further small shifts of the electron wave function result in 95% polarization of the nuclear spins (Fig. 2). In contrast, for a fixed (Gaussian) α_i with $\alpha_i \neq \alpha_j, \ \forall i \neq j$, the nuclear system reaches $> 99\%$ spin polarization in much shorter time scales.

For actual QDs, we will have $\alpha_i \sim \alpha_j$ for nuclei that are nearest neighbors, even when a varying external electric field is applied. This will in turn result in the slowing down of the cooling process. A possible remedy for moving neighboring nuclei out of quasidark states is provided by the first term in $\hat{H}_{\rm int}$ of Eq. (1) which acts to randomize the relative phase between product states with identical $\langle \hat{I}_z \rangle = \sum_i \langle \hat{I}_z^i \rangle$ that make up a dark state. For $\tau \gg 10^{-5}$ sec, we estimate that two product states for which only two neighboring nuclei have differing I_z values can accumulate phases that differ by π. Therefore, we expect the nuclear system to move out of a dark state in ~ 100 μsec and the laser cooling to proceed.

The achievable nuclear-spin temperature is limited by the nuclear-spin diffusion from the (highly polarized) QD nuclei to the (partially polarized) nuclei of the surrounding semiconductor. The physical mechanism for nuclear-spin diffusion could be provided by the (secular) terms in nuclear dipole-dipole interactions which allow for resonant spin exchange between two nuclei while

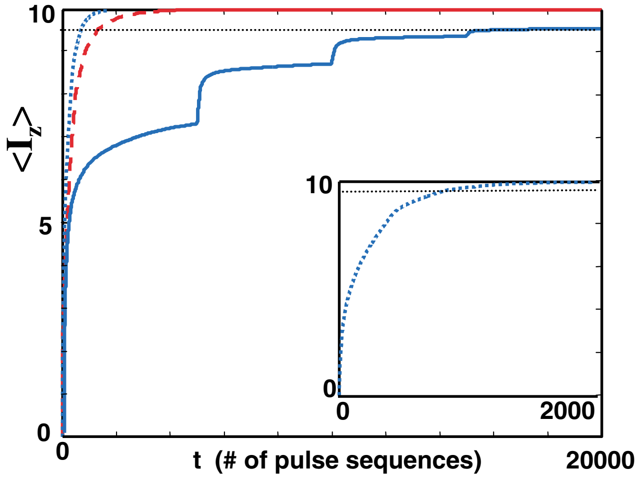

FIG. 2 (color). Optical pumping of a quantum dot with 10 nuclear spins. We assume a Gaussian wave function for the electron and choose its center such that pairs of nuclear spins have identical α. Starting from a completely unpolarized nuclear state with density matrix $\rho_i = 1/2^N$, we find that the nuclear polarization saturates at 0.75 (solid line). This saturation is a clear indication of the influence of dark states. We then shift the electron wave function (after 5000 pulse sequences) in a way to ensure that α's are identical for different sets of nuclei: Since the new coupling distribution has a different set of dark states, the polarization increases abruptly and then saturates at a higher level. Further shifts after 15 000 and 20 000 pulse sequences result in 95% polarization of the nuclear spins (solid line). The dashed line shows $\langle \hat{I}_z \rangle$ when the electron wave function is shifted by the same small amounts, but now after every 50 cooling pulse sequences. The dotted line shows the simulation result for a single Gaussian wave function that ensures $\alpha_i \neq \alpha_j, \ \forall i \neq j$: In this case there are no dark states and the final nuclear spin polarization $\geq 99\%$ is achieved after only 2000 pulses (inset).

preserving $\langle \hat{I}_z \rangle$. For QDs embedded in a semiconductor of a different type, we expect the different g factors for the nuclei in the two semiconductors to largely inhibit spin diffusion into the surrounding material. This should be the case for CdSe/ZnS core-shell nanocrystals and InAs self-assembled QDs. For such QDs, nuclear dipole-dipole interactions with typical time scales $\tau_{\rm nuclear} \sim 10^{-4}$ sec will help the nuclear-spin cooling by transferring the polarization to those QD nuclei which have small wave function overlap ($\alpha_i \ll 1$) with the QD electron. In addition, nuclear spin-flips due to resonant dipole-dipole interaction will also be effective in moving the total QD nuclei system out of dark states.

For electrically defined structures, the semiconductors that make up the QD and (part of) the barrier are identical. In this case, nuclear dipole-dipole interactions can cause spin diffusion into the barrier and limit the effectiveness of laser cooling. A possible remedy for nuclear-spin cooling in such QDs can be obtained from NMR techniques, such as magic angle time-dependent fields, that can be used to inhibit dipolar interactions to a large extent [11]. We also note that recent experiments on electrically defined QDs showed a spin diffusion time of 800 sec — more than 6 orders of magnitude longer than the typical time scale for dipolar interactions [12].

To make a worst case estimate of the spin cooling time we can assume that the QD nuclear-spin system moves into a dark state after each electron-nuclear spin-flip event. Since we estimate the time to move out of a dark state to be 10^{-4} sec, the cooling time for a system of $N = 10^4$ nuclei would be $\sim$1 sec. For nuclear-spin diffusion (i.e., T_1) times exceeding 100 sec, $\geq 99\%$ polarization of the nuclear spins could be possible [13].

In summary, we have described an all-optical method that flips the nuclear spins in a predetermined direction. Successive application of the spin-flip procedure will realize laser cooling of nuclear spins in a zero-dimensional structure. The elementary step of nuclear-spin flip may be used to generate highly entangled states of the nuclei, even before significant nuclear-spin polarization is achieved. It has been shown recently that such states can have completely different signatures for electron spin dynamics, as compared to unpolarized nuclei in a product state [14]. Another promising application of fully polarized nuclear spins is in quantum state storage of an electron spin state in collective excitations of nuclear spins [15].

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