An all-optical scheme to polarize nuclear spins in a single quantum dot is analyzed. The hyperfine interaction with randomly oriented nuclear spins presents a fundamental limit for electron spin coherence in a quantum dot; by cooling the nuclear spins, this decoherence mechanism could be suppressed. The proposed scheme is inspired by laser cooling methods of atomic physics and implements a “controlled Overhauser effect” in a zero-dimensional structure.

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 $N \sim 10^3 - 10^5$ 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. This random nuclear spin orientation presents a fundamental decoherence mechanism for an electron spin confined in a quantum dot. Recently, Khaetskii et al. [1] and Merkulov et al. [2] have analyzed this decoherence mechanism.

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 [3], in order to allow for resonant electron-nuclear spin flip to take place. When the spin-flip is completed, the electron spin is re-flipped into its original state using a laser induced $\pi$-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 between the spin-up and down electronic states can also be achieved in sub-nanosecond time-scales using the electric-field dependence of the electron g-factor [4], 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 [5]. 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 [6]: 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 [6]. 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 [7] 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 quantum dot where a single conduction band electron interacts with $N \sim 10^4$ (spin 1/2) nuclei. We assume that the interaction with the $i^{th}$ nucleus is proportional to the absolute value of the electron wave-function squared at that site ($\alpha_i$, with $\sum_i \alpha_i = N$). The Hamiltonian describing this interaction is the hyperfine contact interaction [1]

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

where $\hat{I}_i^+$ and $\hat{\sigma}_k$ denote the Pauli operators for the $i^{th}$ 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; Merkulov et al. estimate $A$ to be $90 \mu eV$ for GaAs [2].

We assume that the QD is subject to a large constant magnetic field that removes the degeneracy of the spin-up and down states; for an electron g-factor $g_e \sim 2$, we expect an energy difference of $1 \text{ meV}$ with $B = 5 \text{ Tesla}$. The magnetic field is applied along the direction of strong confinement; i.e. $\hat{z}$ for QDs grown by molecular beam epitaxy. For temperatures ($T \sim 3K$) typical of magneto-optical cryostats, the electron is spin-polarized (in the spin-up state) and the nuclei are not:

$$|\Psi\rangle = |\psi\rangle_e \otimes |\psi\rangle_N = \hat{\sigma}_z \prod_j^{M} \hat{I}_j^z |\phi\rangle,$$

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

The electron spin dynamics in a QD takes place on timescales much shorter than that of the nuclear spin system. As a result, we can assume that the nuclear spins are in a random but constant state during the timescale over which the electron spin is manipulated. The effective (nuclear) magnetic field $H_{\text{eff}} = B_{z,\text{eff}} \sim A/(\sqrt{N} \mu_B)$, where $\mu_B$ is the Bohr magneton. The magnetic field along $x$ and $y$ directions have the same expectation value for unpolarized nuclei.

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

$$\hat{H}_\text{laser} = \hbar \left[ \Omega_+ (t) (\hat{e}_j \hat{h}_{3/2} + \eta \hat{e}_j^\dagger \hat{h}_{3/2}) e^{i \Delta t} + c.c. \right]$$

$$+ \hbar \left[ \Omega_- (t) (\hat{e}_j \hat{h}_{-3/2} + \eta \hat{e}_j^\dagger \hat{h}_{-3/2}) e^{i \Delta t} + c.c. \right],$$

where $\hat{h}_{\pm 3/2}$ denotes a valence band hole state with angular momentum projection $j_z = \pm 3/2$. $\Omega_\pm (t)$ ($\Omega_-(t)$) 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. Due to heavy-light hole mixing of the valence band states, these selection rules are relaxed in actual QD structures, leading to non-zero but small coupling ($\eta \ll 1$) to optical fields that violate the $j_z$ selection rules [8]. The frequency of the laser field determines the detuning $\Delta$ 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 (lc$p$) 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 [3]. While this laser is on electron-nuclear spin flip process due to $\hat{H}_\text{int}$ 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

$$g_{\text{spin-flip}} = \frac{\langle \hat{F}_+ \hat{F}_- \rvert \hat{\sigma}_- \rvert \Psi \rangle}{\sqrt{N} \sum_i \alpha_i \hat{F}_+ \hat{\sigma}_- |\Psi\rangle},$$

as a direct consequence of the collective enhancement of the transition due to participation of many nuclei. Since $g_{\text{spin-flip}}$ is comparable to $g_\mu_B B_{z,\text{eff}}$ (for unpolarized nuclei), we expect significant probability for spin-flip if we leave the laser on for $\tau \sim 1/\hbar g_{\text{spin-flip}}$. We estimate the spin-flip time for an electron in an InAs QD to be less than 1 nsec [2]. Therefore choosing a laser pulse-width of $\tau \approx 300\text{psec} < g_{\text{spin-flip}}$ will yield a spin-flip probability $\sim 0.1$. If an electron-nuclear spin flip event does take place, the state-vector of the QD is $|\Psi\rangle_A = \hat{\sigma}_- \sum_i \alpha_i \hat{F}_+ |\Psi\rangle / \sqrt{N}$, where $N$ ($\neq N$ in general) is the normalization factor. Since collective enhancement factor becomes smaller with increasing nuclear polarization, it is desirable to increase $\tau$ as cooling proceeds.

After the ac-Stark laser is turned off, we turn on a resonant right hand circularly polarized (rc$p$) laser field that realizes a $\pi$-pulse on the quasi-forbidden electronic transition $\hat{e}_j^\dagger |0\rangle \rightarrow \hat{e}_j^\dagger \hat{e}_j^{3/2} |0\rangle$ (with transition amplitude $\propto \eta$), only if an initial electron-nuclear spin-flip process has taken place due to the resonant hyperfine interaction in the presence of the red-detuned lc$p$ laser. If this is the case, the excited trion state $\hat{e}_j^\dagger \hat{h}_+^{3/2} |\psi\rangle_e$ is populated with probability approaching unity. This excited state will relax down predominantly to the electronic state $|\psi\rangle_e = \hat{e}_j^\dagger |0\rangle$ by spontaneous emission of a rc$p$ photon with rate $\Gamma_{\text{rad}}$, thereby projecting the electron spin onto the initial spin-up state (Fig. 1(c)). The final state following spontaneous emission is

$$|\Psi\rangle_C = \frac{1}{\sqrt{N}} \sum_i \alpha_i \hat{F}_+ |\Psi\rangle.$$

The successive application of two laser pulses followed by spontaneous emission flips a single nuclear spin with probability $\sim 0.1$ 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 $\pi$-pulse does not couple an occupied transition and the whole system remains in its initial state $|\Psi\rangle$. Residual coupling on the detuned strongly allowed transition $\hat{e}_j^\dagger |0\rangle \rightarrow \hat{e}_j^\dagger \hat{e}_j^{3/2} |0\rangle$ by the rc$p$ laser will result predominantly in Rayleigh scattering down to the electronic state $\hat{e}_j^\dagger |0\rangle$. We note that the energy of the spontaneously emitted photons (which indicate the presence of a nuclear spin flip) and Rayleigh scattered photons (which indicate unchanged nuclear spin state) differ by the electron Zeeman splitting.

Having discussed the elementary step of near-deterministic 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 $B_{z,\text{eff}}^\text{ac-Stark}$ will change as the nuclear spin polarization increases. For unpolarized nuclei $g_\mu_B B_{z,\text{eff}}^\text{ac-Stark} \sim g_{\text{spin-flip}}$, whereas for nearly polarized nuclei $g_\mu_B B_{z,\text{eff}}^\text{ac-Stark} \sim A \gg g_{\text{spin-flip}}$. Therefore, if the magnitude of the ac-Stark shift simply cancels out the Zeeman shift due to the external magnetic field, the spin-flip process will become ineffective as the nuclear spin polarization increases. By adjusting the intensity of the lc$p$ laser, it should be possible 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. Since each spin-flip is accompanied by spontaneous emission of a photon, it is
in principle possible to estimate the degree of nuclear spin polarization by counting the spontaneously emitted photons. Alternatively, we can envision a laser pulse shape that will ensure resonance condition for a sufficiently long interaction time, for any \( \sum_i \hat{I}_i \).

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
\[
\sum_i \alpha_i \hat{I}_i |\psi\rangle_N^{(n)} = 0,
\]
then the prescribed procedure cannot be utilized to achieve further nuclear spin polarization with the given \( H_{\text{int}} \). 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 this manifold, the only state with appreciable coupling \( (g_{\text{spin-nuclei}} \sim A/\sqrt{N}) \) 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 \( \hat{I}^2 \) is not conserved and the limitation due to (quasi) dark states will only be relevant for the sub-collection of nuclear spins for which \( \alpha_i \approx \alpha_j \). 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 \( \alpha_i \). 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 pre-determined level, this is a good indication that the system has evolved into a quasi-dark state of \( H_{\text{int}} \) with the current \( \alpha_i \). Based on this information, we can introduce an external electric field and increase its magnitude (or change its orientation) until we increase the photon detection rate. Alternatively, we can apply a random electric field with coherence time shorter than the separation of successive elementary cooling cycles 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 10 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 lattice constant. 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 results in 95\% polarization of the nuclear spins (Fig. 2). In contrast, for a fixed (Gaussian) \( a_L \) with \( \alpha_i \neq \alpha_j, \forall i \neq j \), the nuclear system reaches > 99\% 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 quasi-dark states is provided by the first term in \( H_{\text{int}} \) of Eq. (1) which acts to randomize the relative phase between product states with identical \( \langle I_2 \rangle = \sum_i \langle I_i^2 \rangle \) that make up a dark state. For \( \tau >> 10^{-5} \) sec, we estimate that two product states for which only two neighboring nuclei have differing \( I_i \) values, can accumulate phases that differ by \( \pi \). Therefore, we expect the nuclear system to move out of a dark state in less than 100 \( \mu \) sec 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 preserving \( \langle I_2 \rangle \). 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 \( \tau_{\text{nuclear}} \sim 10^{-4} \) sec will act to 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 nuclear 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 [9]. 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 eliminate dipolar interactions to a large extent [10]. 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 timescale for dipolar interactions [11].

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, \( > 99\% \) polarization of the nuclear spins could be possible [12].

In summary, we have described an all-optical method
that flips the nuclear spins in a pre-determined direction. Successive application of the spin-flip procedure will realize laser cooling of nuclear spins in a zero-dimensional structure. The elementary step of near-deterministic nuclear spin flip process can 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 [13]. Another promising application of fully polarized nuclear spins is in quantum state storage of an electron spin state in collective excitations of nuclear spins [14].

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FIG. 1. 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 re-pumped into the initial state by a combination of a $\pi$-pulse, followed by spontaneous emission.
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