Collective Nuclear Stabilization by Optically Excited Hole in Quantum Dot

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We propose that an optically excited heavy hole in a quantum dot can drive the surrounding nuclear spins into a quiescent collective state, leading to significantly prolonged coherence time for the electron spin qubit. This provides a general paradigm to combat decoherence by environmental control without involving the active qubit in quantum information processing. It also serves as a unified solution to some open problems brought about by two recent experiments [X. Xu et al., Nature 459, 1105 (2009) and C. Latta et al., Nature Phys. 5, 758 (2009)].

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The hole, the removal of an electron from the fully occupied valence band, is an elementary excitation in semiconductors. Because of the lower orbital symmetry, the hole in the valence band has properties different from the electrons in the conduction band. In a quantum dot (QD), the spin of a localized hole is coupled to the spins of the surrounding atomic nuclei of the host lattice through the long-range dipolar hyperfine interaction, as opposed to the stronger contact hyperfine interaction between the electron spin and the nuclear spins. The hole-nuclear coupled system forms a different paradigm of the central spin model, which has been of interest for a long time in spin resonance spectroscopy, quantum dissipation, and classical-quantum crossover. Currently, the central hole spin in the QD has attracted increasing interest due to its potential usage as carriers of long-lived quantum information and efforts are being directed towards understanding its interactions with the environment, especially the nuclear spins. Recently, a symmetric hysteretic broadening of the absorption spectrum was observed and attributed to a nuclear polarization transient induced by an optically excited hole through a non-collinear term of the dipolar hyperfine interaction.

Compared with the newly initiated hole spin based quantum technology, the electron spin based quantum technology has achieved a higher degree of maturity. One critical obstacle is the short electron spin coherence time $T_2^*$ due to the randomly fluctuating nuclear field produced by the "noisy" nuclear spins. Prolonging the electron spin coherence time is of paramount importance in quantum information processing. For this purpose, two major approaches are under rapid development. One aims at decoupling a general qubit from the environments by repeated pulsed control of the qubit. The other aims at suppressing the fluctuation of the nuclear field, i.e., stabilizing the nuclear spins, by inducing a steady-state nuclear polarization through the electron-driven Overhauser and reverse Overhauser effects. The latter approach, being specifically designed for the QD electron spin, has the merit that once the nuclear spin environment is stabilized, it remains "quiet" for a long time for electron spin manipulation. Intensive research efforts have led to successful nuclear stabilization in QD ensembles and suppression of the fluctuation of the nuclear field difference between two neighboring QDs.

Recently, three groups reported significant nuclear stabilization in single QDs, most relevant for quantum information processing. Vink et al. deduced the stabilization from the observed electron spin resonance locking, attributed to the electron-driven reverse Overhauser effect. Xu et al. and Latta et al. directly observed the stabilization by optical excitation of trion and blue exciton (both containing a hole spin) in the Voigt and Faraday geometries, respectively. Xu et al. also observed the resulting prolonged electron spin coherence time. However, two key observations, the nuclear stabilization observed by Xu et al. and the bidirectional hysteretic locking of the blue exciton absorption peak observed by Latta et al., cannot be explained by existing theories. For a complete understanding of the fundamental physics and future optimization of the nuclear stabilization, a new theory is needed.

In this Letter, we link the hole spin based quantum technology with the electron spin based quantum technology by constructing a theory showing that an optically excited central hole spin can stabilize the nuclear spins and significantly prolong the coherence time of the electron spin qubit. This provides a general paradigm to combat decoherence by environmental control without involving the active qubit. The essential ingredients of this theory include a finite nuclear Zeeman splitting and the non-collinear dipolar hyperfine interaction, through which the fluctuating hole spin stabilizes the nuclear spins by driving them from the "noisy" thermal equilibrium state into a "quiet" collective state, where the strong thermal fluctuation is largely cancelled by the inter-spin correlation. This enables a flexible control of the nuclear fluctuation and hence the coherence time of the electron spin qubit by engineering the hole spin fluctuation spectrum through the highly developed coherent optical techniques. It also provides a unified explanation...
to the two distinct experimental observations: the appearance of the “quiet” state explains the observation of Xu et al. in the single pump experiment [19], while the switch between the “noisy” state and the “quiet” state explains the observation of Latta et al. on the blue exciton [17].

The first step of the hole-driven collective nuclear stabilization is a steady-state nuclear polarization induced by the optically excited hole. The essential physics of this step is captured by the dynamics of a typical nuclear spin-1/2 in a QD coupled to a heavy hole state $|1\rangle$ through the non-collinear dipolar hyperfine interaction $\tilde{\sigma}i\tilde{a}_h(\tilde{I}^+ + \tilde{I}^-)$, where $\tilde{\sigma}i|j\rangle \langle i|$, $\tilde{I}^+ = \tilde{I}^x \pm i\tilde{I}^y$, and $\tilde{a}_h = O(\eta^2)\tilde{a}_h$, with $\eta$ being the heavy-light hole mixing coefficient. An external magnetic field along the $z$ axis gives rise to a finite nuclear Zeeman frequency $\omega_N$ and a continuous wave laser couples the ground state $|0\rangle$ to the excited hole state $|1\rangle$ with Rabi frequency $\Omega_R$ and detuning $\Delta$. The hole dephases with rate $\gamma_2$ and decays back to the ground state with rate $\gamma_1$ [Fig. 1(a)]. The hole dephasing broadens $|1, \uparrow\rangle$ and $|1, \downarrow\rangle$ to Lorentzian distribution $L^{(2)}(E) = (\gamma_2/\pi)/(E^2 + \gamma_2^2)$. In the weak pumping limit, two competing nuclear spin-flip channels $|0, \downarrow\rangle \overrightarrow{\Omega_2} \rightarrow |1, \downarrow\rangle \tilde{a}_h \rightarrow |1, \uparrow\rangle$ (down-to-up channel) and $|0, \uparrow\rangle \overleftarrow{\Omega_2} \rightarrow |1, \uparrow\rangle \tilde{a}_h \rightarrow |1, \downarrow\rangle$ (up-to-down channel) are opened up to leading order [Figs. 1(b) and 1(c)]. For each channel, the transition rate is proportional to the square of the coupling strength times the final density of states of each step determined by the energy mismatch. For the down-to-up channel, the transition rate $W_+ \propto \Omega_R^2 L^{(2)}(\Delta) \times \tilde{a}_h^2 L^{(2)}(\omega_N + \Delta)$, where $\Delta$ is the energy mismatch and $L^{(2)}(\Delta)$ is the final density of states for the first step $|0, \downarrow\rangle \overrightarrow{\Omega_2} |1, \downarrow\rangle$, while $(\omega_N + \Delta)$ and $L^{(2)}(\omega_N + \Delta)$ are corresponding quantities for the second step $|1, \downarrow\rangle \overrightarrow{\Omega_2} |1, \uparrow\rangle$ [Fig. 1(b) or 1(c)]. For the up-to-down channel, the transition rate $W_- \propto \Omega_R^2 L^{(2)}(\Delta) \times \tilde{a}_h^2 L^{(2)}(\omega_N - \Delta)$. The competition between the two channels establishes an intrinsic (i.e., in the absence of other nuclear spin relaxation mechanisms) steady-state nuclear polarization

$$\langle \tilde{I}^z \rangle_0 \propto \frac{W_+ - W_-}{W_+ + W_-} = -\frac{2\Delta\omega_N}{\Delta^2 + \gamma_2^2} + O(\varepsilon^2)$$

(1)

during a time scale characterized by the inverse of the nuclear polarization buildup rate $\Gamma_p = W_+ + W_- = O(a_h^2\Omega_R^2)$, where $\varepsilon \equiv \omega_N/\gamma_2 \sim 0.1$ for a typical QD under a magnetic field $B = 1$ T. The hole-driven intrinsic steady-state polarization $\langle \tilde{I}^z \rangle_0$ shows a sign dependence on the detuning $\Delta$, which is the key to explain the observation of Latta et al. on blue exciton excitation [17]. By contrast, the intrinsic steady-state polarization $\langle \tilde{I}^z \rangle_{\text{eq}}$ due to the electron-driven Overhauser [13] (reverse Overhauser [14]) effect is equal to the nonequilibrium (equilibrium) part of the electron spin polarization and is insensitive to the laser detuning, while the hole-driven nuclear polarization transient $\langle \tilde{I}^z \rangle_{\text{transient}}$ or random shift [8] produces no intrinsic steady-state nuclear polarization.

For the hole-driven evolution of a typical nucleus with spin $I \geq 1/2$ under a general pumping intensity, we single out the slow dynamics of the diagonal part $\hat{P}(t)$ of the reduced density matrix $\hat{\rho}_N(t)$ of this nucleus by adiabatically eliminating the fast motion of the hole and the off-diagonal part and arrive at

$$\dot{\hat{P}} = -W_+ [\hat{I}^-, \hat{I}^+ \hat{P}] - W_- [\hat{I}^+, \hat{I}^- \hat{P}],$$

(2)
valid up to $O(a_h^2)$, where the transition rates for the down-to-up and up-to-down channels, $W_{\pm} = C(\mp \omega_N)$, are determined by the steady-state hole fluctuation $C(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \tilde{I}^{(2)} \rangle e^{-i\omega t}$.

For $I = 1/2$, the motion of the (degree of) nuclear polarization $s(t) \equiv \langle \tilde{I}^z(t) \rangle / I$ follows from Eq. (2) as

$$\dot{s} = -\Gamma_1 s - \Gamma_p (s - s_0),$$

(3)
where $\Gamma_1$ is a phenomenological nuclear depolarization rate for other nuclear spin relaxation mechanisms,

$$\Gamma_p = W_+ + W_- = \frac{4a_h^2}{\gamma_1} \left(\frac{W\gamma_1^2}{(\gamma_1 + 2W)^3}c_1 + O(\varepsilon^2)\right)$$

(4)
is the hole-driven nuclear polarization buildup rate, and

$$s_0 = \frac{W_+ - W_-}{W_+ + W_-} = -\frac{\Delta\omega_N}{\Delta^2 + \gamma_2^2} \frac{\gamma_1}{\gamma_2} c_1 + O(\varepsilon^2)$$

(5)
is the intrinsic (i.e., when $\Gamma_1 = 0$) steady-state nuclear polarization, in agreement with Eq. (4). Here $c_0 \equiv 1/2 + \gamma_2/\gamma_1 + f + W/\gamma_1$ and $c_1 \equiv 1 + [\gamma_1/(2\gamma_2)]f + W/\gamma_1$ are non-negative constants, $f \equiv (\gamma_2^2 - \Delta^2)/(\gamma_2^2 + \Delta^2)$, and $W \equiv 2\pi(\Omega_R/2)^2 L^{(2)}(\Delta)$ is the hole excitation rate. The steady-state nuclear polarization is $s_{\text{eq}} = \Gamma_p s_0/\Gamma_p$. These results agree well with the exact numerical solutions to the coupled motion (see Fig. 2). For a general nuclear spin-$I$, the nuclear polarization $s$ still obeys
Eq. (3) by extending \( s_0 \) to \( s_0^{(I)} \equiv s_0((\hat{I}^x)^2 + (\hat{I}^y)^2)/I \), which reduces to \( 2(I + 1)s_0/3 \) for \( |s_0^{(I)}| \ll 1 \).

The second step of the hole-driven collective nuclear stabilization is the feedback of the nuclear spins on the hole excitation by shifting the energy of the excited hole state \(|0\rangle\) or the ground state \(|1\rangle\). For specificity, we consider a negatively charged QD and identify \(|1\rangle\) with the trion state (still referred to as hole, despite the additional inert electron spin singlet) and \(|0\rangle\) with the spin-up electron state. The hole is coupled to the nuclei in the QD through the non-collinear dipolar hyperfine interaction \( \hat{H}_{11} \equiv \sum_j \hat{a}_{h,j}(\hat{I}_j^+ + \hat{I}_j^-) \). The electron is coupled to the nuclear spins through the diagonal part of the contact hyperfine interaction \( \hat{H}_{00} \equiv \sum_j \hat{a}_{e,j}\hat{I}_j^2/2 \equiv \hat{\sigma}_0\hat{h} \), with the off-diagonal terms involving the electron spin flip being suppressed by the large electron Zeeman splitting. The feedback of the nuclear spins proceeds by shifting the energy of the electron state \(|0\rangle\) by \( \hat{h} \equiv \sum_j a_{e,j}\hat{I}_j^2/2 \) (referred to as Overhauser shift hereafter), which consists of the macroscopic mean-field part \( \hat{h}(t) \equiv \text{Tr} \hat{P}(t)\hat{h} \) and the fluctuation \( \delta \hat{h}(t) \equiv \hat{h} - \hat{h}(t) \), with \( \hat{P}(t) \) being the diagonal part of the reduced density matrix \( \hat{\rho}_N(t) \) of the nuclear spins. Then the laser detuning is changed from \( \Delta \) to \( \Delta \equiv \Delta - \hat{h} \equiv \Delta(t) - \delta \hat{h}(t) \), where \( \Delta(t) \equiv \Delta - \hat{h}(t) \).

This changes the nuclear spin dynamics from Eq. (2) to

\[
\dot{\hat{P}} = -\sum_j [\hat{I}_j^- \hat{I}_j^+ W_j^{(+)\Delta \hat{P}} - \sum_j [\hat{I}_j^+ \hat{I}_j^- W_j^{(-)\Delta \hat{P}}],
\]

where \( W_j^{(+)\Delta \hat{P}} \) are obtained from \( W_j^{(+)\Delta \hat{P}} \) by replacing \( \hat{a}_{h,j}, \omega_N, \) and \( \Delta \) with \( \hat{a}_{h,j}, \omega_N, \) and \( \Delta \), respectively. The feedback is manifested in Eq. (6) as the Overhauser shift \( \hat{h} \). As shown below, the feedback of the macroscopic part \( \hat{h}(t) \) leads to nonlinear, bistable nuclear spin dynamics, experimentally manifested as hysteretic locking of the electronic excitation [17]. The feedback of the fluctuation \( \delta \hat{h}(t) \), introduced by qualitative argument [17] or stochastic assumption [13, 14] in previous treatments, correlates the dynamics of different nuclear spins and drives the nuclear spins from the “noisy” thermal equilibrium state into a “quiet” collective state [6].

To keep the exposition simple, we consider identical nuclei \( I_j = I, \omega_{N,j} = \omega_N, a_{e,j} = a_e, \hat{a}_{h,j} = \hat{a}_h \), so that \( \hat{h} = (N a_e I/2)\hat{s} \) (\( N \) is the number of QD nuclei) is proportional to the nuclear polarization operator \( \hat{s} \equiv \sum_{j=1}^N \hat{I}_j^z/(NI) \). First we consider the feedback of the macroscopic part \( \hat{h}(t) \) by dropping the fluctuation \( \delta \hat{h} \) from Eq. (6) to obtain a mean-field description

\[
\hat{h}(t) = -\Gamma_p(\Delta(t))(\hat{h}(t) - h_0^{(I)}(\Delta(t))), \tag{7}
\]

valid for \( I = 1/2 \) or \( |s_0^{(I)}| \ll 1 \), where \( \Gamma_p(\Delta(t)) \) and \( h_0^{(I)}(\Delta(t)) \) are obtained from \( \Gamma_p \) and \( h_0^{(I)} \equiv (N a_e I/2)s_0^{(I)} \), respectively, by replacing \( \Delta \) with \( \Delta(t) = \Delta - \hat{h}(t) \). Equation (7) shows that the dynamics of \( h(t) \) is highly nonlinear, since \( \Gamma_p \) and \( s_0^{(I)} \) are highly nonlinear functions of \( \Delta \) (see Fig. 2). Consequently for a given detuning \( \Delta \), the steady-state solution \( h^{(ss)}(t) \) may be multi-valued, e.g., two stable solutions (black lines) and one unstable solution (gray line) in Fig. 3a.

For a given detuning \( \Delta \), each stable solution \( h^{(ss)}(t) \) corresponds to a macroscopic (mixed) nuclear spin state with macroscopic polarization \( s_0^{(ss)} \equiv 2I h^{(ss)}/(N a_e I) \). The noise on the electron spin qubit produced by this state is characterized by the fluctuation of \( \tilde{s} \) around its macroscopic value \( s_0^{(ss)} \). To quantize this fluctuation, we define the probability distribution function \( p(s, t) \equiv \text{Tr} \hat{P}(t)\delta(s - \tilde{s}) \) of \( \tilde{s} \). Then, instead of introducing the fluctuation stochastically by assuming that \( \tilde{s} \) experiences a random walk described by a Fokker-Planck equation (applied to nuclear spin-1/2’s only) [13, 14], we treat this feedback quantum mechanically by directly deriving the equation of motion (which also assumes the Fokker-Planck form) of \( p(s, t) \) from Eq. (10), valid for nuclei with arbitrary spin \( I \). The steady-state solution \( p^{(ss)}(s) \) sharply peaks at each macroscopic value \( s_0^{(ss)} \). The fluctuation of \( \tilde{s} \) in the \( \alpha \)th Gaussian state is quantified by the standard deviation of the Gaussian peak at \( s_0^{(ss)} \):
ment [7]. For \( \omega \) tion is maximal at large detunings. 

FIG. 3. (color online). Stable (black lines) and unstable (grey lines) \( h^{(sa)} \) [(a), (d)] and \( \sigma_{\alpha}/\sigma^{eq} \) in stable states [(b),(e)]. (c) and (f): Optical absorption spectra obtained by sweeping \( \Delta \) in different directions (indicated by the arrows). A typical QD with identical nuclear spin-9/2’s and (unit: ns \(^{-1} \)) \( N_{\alpha} = 100, \gamma_1 = \gamma_2 = \Omega_K = 1 \), and \( \omega_N = -0.2 \) [(a)-(c)] or 0.2 [(d)-(f)] is considered. The sharp Lorentzian peaks at \( \Delta = 0 \) in (c) and (f) are absorption spectra in the absence of the nuclei.

\( s^{(sa)} \approx 1 \), and nuclear stabilization by the competition between polarization and depolarization processes [13, 14, 18] by showing that the degree of stabilization as quantified by the smallness of \( \sigma_{\alpha}/\sigma^{eq} \) is the product of the individual contribution \( [1 - (s_{v,0}^{(sa)})^2]^{1/2} \) and the collective contribution \( (1 + dh_{\alpha}^{(sa)}/d\Delta)^{-1/2} \). The individual contribution comes from the suppression of the fluctuation of each individual nuclear spin by the steady-state nuclear polarization. The collective contribution comes from the suppression of the collective fluctuation of all the nuclear spins by the correlation \( \langle I_i^z I_j^z \rangle - \langle I_i^z \rangle \langle I_j^z \rangle \rangle/I^2 \approx -(\sigma^{eq})^2 \) between different nuclear spins, established by the correlated dynamics [Fig. 3c] driven by the feedback from the fluctuation \( \dot{h}_\alpha \) [7]. The collective contribution typically dominates the nuclear spin stabilization, e.g., in the Q (i.e., “quiet”) branch in Fig. 3(b), the nuclear spins are significantly stabilized collectively, especially near the resonance point \( \Delta = 0 \), where the slope \( dh^{(sa)}/d\Delta \) (and hence collective stabilization) is maximal, but the magnitude \( h^{(sa)} \) (and hence individual stabilization) is negligible. This explains the observation by Xu et al. in the single pump experiment [7]. For \( \omega_N > 0 \) [Fig. 3(d)], the nuclear stabilization is maximal at large detunings.

In the Q branch of Fig. 3(a), \( h^{(sa)} \propto \Delta \) always shifts the effective detuning \( \Delta^{(sa)} = \Delta - h^{(ss)} \) towards resonance, while in the N (i.e., “noisy”) branch, \( h^{(sa)} \) is very small. When sweeping the laser frequency in different directions, the nuclear spin state switches between the N branch and the Q branch, leading to bidirectional hysteretic locking of the optical absorption peak [Fig. 3c]. This explains the observation by Latta et al. [13] upon blue exciton excitation when we identify \( |0 \rangle \) as the vacuum and \( |1 \rangle \) as the spin-up exciton. In Fig. 3(d), \( h^{(sa)} \) always tends to repel \( \Delta^{(ss)} \) away from resonance and the absorption peak is shifted hysteretically to finite detunings [Fig. 3f]. Qualitatively similar behaviors are seen in some experimental data under red exciton excitation [21].

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