Title

Intrinsic bidirectional dynamic nuclear polarization by optically pumped trions in quantum dots

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Hole-induced Dynamic Nuclear Polarization in Quantum Dots

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We present a microscopic theory showing that an optically excited heavy hole can induce a steady-state nuclear polarization in a quantum dot. With the preferential direction of the nuclear spin flip set by the energy mismatch instead of thermal relaxation, the resulting nuclear polarization shows a sign dependence on the product of the nuclear Zeeman splitting and the frequency detuning of the pumping laser, leading to experimentally observed bidirectional hysteretic locking or shift of the optical absorption peak, accompanied by a significant suppression of the nuclear fluctuation and hence prolonged electron spin coherence time.

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Electron spins in semiconductor quantum dots (QDs) are promising candidates as qubits for quantum technology [1]. The main practical obstacle is the short spin coherence time, limited to a few nanoseconds by the contact hyperfine interaction with the QD nuclear spins [2], which produce a fluctuating effective magnetic field that randomly shifts the electron Zeeman splitting (referred to as Overhauser shift in literature) and rapidly diminishes its phase coherence [3]. To suppress the nuclear fluctuation and hence the electron spin decoherence, the simplest idea is to completely polarize the nuclear spins through dynamic nuclear polarization (DNP), e.g., 99% polarization yields an order of magnitude suppression [4]. Two DNP mechanisms, the well-known Overhauser effect [5] and the recently proposed reverse Overhauser effect [6], both based on the electron-nuclear contact hyperfine interaction, have been intensively investigated [7]. The highest polarization achieved so far is only $\sim 65\%$ [8].

Recently, significant suppression of the QD nuclear fluctuation has been reported [9–13]. In coherent dark-state spectroscopy in Voigt geometry, Xu *et al*. [11] observe a $\sim$ 6-fold suppression by one pump laser and a unprecedented $\sim 10^3$-fold suppression by two pumps, accompanied by a symmetric hysteretic broadening of the transient dark-state spectra. The hysteretic broadening is attributed to the feedback from a *transient* nuclear polarization induced by the *non-collinear* dipolar hyperfine interaction [14] with the optically excited heavy hole through a semi-phenomenological *third-order* process [15]. A theory of hole-induced suppression of the nuclear fluctuation is lacking at present.[16]

In this Letter, we present a microscopic theory showing that through a *second-order* process, an optically excited heavy hole can induce a *steady-state* nuclear polarization. The preferential direction of the nuclear spin flip is set by the product of the nuclear Zeeman splitting ω_N and the detuning $\Delta \equiv \omega_{\rm eh} - \omega$ between the electron-hole excitation energy $\omega_{\rm eh}$ and the laser frequency ω. This hole-induced DNP is manifested as a bidirectional hysteretic locking of the optical absorption peak onto resonance or as a bidirectional hysteretic shift of the peak away from zero detuning. This sheds light on a puzzling observation of bidirectional hysteretic locking of the neutral exciton absorption peak in Faraday geometry [12]. Through the Fokker-Planck equation, we found a $\sim$ 10-fold

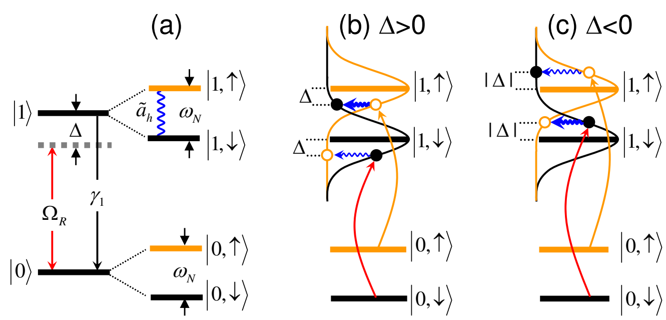

FIG. 1. (color online). (a) Energy levels of the electron, the hole, and a typical nuclear spin-1/2. (b) and (c): Two competing nuclear spin-flip processes for (b) $\Delta > 0$ and (c) $\Delta < 0$, respectively.

suppression of the steady-state nuclear fluctuation, in reasonable agreement with the single pump experiment of Ref. [11].

The essential physics of the hole-induced DNP is captured by a simple model consisting of a heavy hole state $|1\rangle$, an electron state $|0\rangle$, and a typical nuclear spin-1/2 with Zeeman splitting ω_N in an external magnetic field along the z axis in a QD [Fig. 1(a)]. The hole state is optically excited from the electron state by a pumping laser with Rabi frequency Ω_R and detuning $\Delta \equiv \omega_{\rm eh} - \omega$. Due to heavy-light hole mixing, the hole-nuclear dipolar hyperfine interaction contains a non-collinear secular term $\hat{\sigma}_{11}\tilde{a}_h(\hat{I}^+ + \hat{I}^-)$ [11], where $\hat{\sigma}_{ji} \equiv |j\rangle\langle i|$ and $\tilde{a}_h \equiv O(\eta^2)a_h$ with η being the hole mixing coefficient. The hole dephasing broadens $|1,\uparrow\rangle$ and $|1,\downarrow\rangle$ to Lorentzian distribution $L^{(\gamma_2)}(E) = (\gamma_2/\pi)/(E^2+\gamma_2^2)$. In the weak pumping limit, two nuclear spin-flip channels $|0,\downarrow\rangle \xrightarrow{\Omega_R} |1,\downarrow\rangle \xrightarrow{\tilde{a}_h} |1,\uparrow\rangle$ (down-to-up channel) and $|0,\uparrow\rangle \xrightarrow{\Omega_R} |1,\uparrow\rangle \xrightarrow{\tilde{a}_h} |1,\downarrow\rangle$ (up-to-down channel) are opened up to leading order [Figs. 1(b) and 1(c)]. For each channel, the transition rate is *qualitatively* proportional to the square of the coupling strength times the final density of states determined by the energy mismatch between the intermediate state and the initial state. For the down-to-up channel, the transition rate $W_+ \propto \Omega_R^2\tilde{a}_h^2 L^{(\gamma_2)}(\Delta)L^{(\gamma_2)}(\omega_N+\Delta)$, where Δ is the energy mismatch and $L^{(\gamma_2)}(\Delta)$ is the final density of states for $|0,\downarrow\rangle \xrightarrow{\Omega_R} |1,\downarrow\rangle$, while $(\omega_N + \Delta)$ and $L^{(\gamma_2)}(\omega_N + \Delta)$ are corresponding quantities for $|1,\downarrow\rangle \xrightarrow{\tilde{a}_h} |1,\uparrow\rangle$. For the up-to-down channel, the transition rate $W_- \propto \Omega_R^2 \tilde{a}_h^2 L^{(\gamma_2)}(\Delta) L^{(\gamma_2)}(\omega_N - \Delta)$. Without other nuclear relaxation mechanisms, the hole mechanism alone establishes an *intrinsic* steady-state nuclear polarization

$$\langle \hat{I}^z \rangle_0 \propto \frac{W_+ - W_-}{W_+ + W_-} = -\frac{2\Delta\omega_N}{\Delta^2 + \gamma_2^2} + O(\omega_N^2/\gamma_2^2) \tag{1}$$

during a time scale characterized by the inverse of the DNP buildup rate $\Gamma_p \equiv W_+ + W_- = O(\tilde{a}_h^2 \Omega_R^2)$. This polarization shows a striking dependence on $\Delta\omega_N$: for $\Delta\omega_N < 0$, the down-to-up channel involving a smaller energy mismatch $|\omega_N + \Delta|$ dominates and leads to $\langle \hat{I}^z \rangle_0 > 0$; for $\Delta\omega_N < 0$, the down-to-up channel dominates and leads to $\langle \hat{I}^z \rangle_0 < 0$.

Compared with the electron-induced DNP, the direction of the hole-induced nuclear polarization is determined by the energy mismatch instead of thermal relaxation. For a nuclear spin-1/2, the *intrinsic* electron-induced nuclear polarization $\langle \hat{I}^z \rangle_0 = \langle \hat{S}_e^z \rangle - \langle \hat{S}_e^z \rangle_{\rm eq}$ ($\langle \hat{I}^z \rangle_0 = \langle \hat{S}_e^z \rangle_{\rm eq}$) through the Overhauser (reverse Overhauser) mechanism *alone* is equal to the nonequilibrium (equilibrium) part of the electron spin polarization and is insensitive to the pumping frequency.

For the microscopic theory, we consider a negatively charged QD for specificity. We identify $|0\rangle$ with the spin-up electron state and $|1\rangle$ with the trion state (still referred to as *hole*, which is the only active member of the trion). With the spin flip of the electron (hole) suppressed by the large electron (hole) Zeeman splitting, we focus on the energy-conserving term $\hat{\sigma}_{00} a_e \hat{I}^z/2 \equiv \hat{\sigma}_{00} \hat{h}$ of the electron-nuclear contact hyperfine interaction and the non-collinear term $\hat{\sigma}_{11} \tilde{a}_h (\hat{I}^+ + \hat{I}^-)$ of the hole-nuclear dipolar hyperfine interaction. The density matrix $\hat{\rho}$ obeys the Lindblad master equation with spontaneous emission $|1\rangle \to |0\rangle$ (rate γ_1), hole dephasing (rate $\gamma_2 \geq \gamma_1/2$), and nuclear depolarization (rate Γ_1) included. For a typical self-assembled InAs QD containing $N = 10^4$ nuclear spins under a magnetic field $B = 1$ T, we have (units: μs^{-1}) $\Omega_R, \gamma_1, \gamma_2 \sim 10^3$, $\omega_N \sim 10^2$, $a_e \sim 10$, $a_h \sim 0.1 a_e$ [17], and $\Gamma_1 \sim 10^{-6}$. Since $|a_e| \ll |\omega_N|$, the contact hyperfine interaction has a negligible influence on the energy mismatch and hence the nuclear polarization. That $\gamma_{1,2} \gg |\omega_N|$ provides a small parameter $\omega_N/\gamma_{1,2}$ for keeping only the leading order.

In the exact steady state $\hat{\varrho}$ of the electron-hole subsystem in the absence of the nuclear spin, the populations on $|0\rangle$ and $|1\rangle$ are $\varrho_{00} \equiv (1 + W/\gamma_1) d_0$ and $\varrho_{11} \equiv (W/\gamma_1) d_0$, respectively, where $d_0 \equiv \varrho_{00} - \varrho_{11} = \gamma_1/(\gamma_1 + 2W)$ and $W \equiv 2\pi(\Omega_R/2)^2 L^{(\gamma_2)}(\Delta)$ is the optical transition rate between $|0\rangle$ and $|1\rangle$. The symmetric and antisymmetric correlation functions of the population fluctuation $\tilde{\sigma}_{00} \equiv \hat{\sigma}_{00} - \langle \hat{\sigma}_{00} \rangle$ are $C(t - t') \equiv \langle \{\tilde{\sigma}_{00}(t), \tilde{\sigma}_{00}(t')\} \rangle/2$ and $\chi(t - t') \equiv \langle [\tilde{\sigma}_{00}(t), \tilde{\sigma}_{00}(t')] \rangle/2$, respectively, where $\langle \cdots \rangle \equiv \mathrm{Tr}\, \hat{\varrho}(\cdots)$. Their Fourier transforms are evaluated through the quantum regression theorem as

$$C_{\omega=0} = \frac{2}{\gamma_1} \varrho_{11} d_0^2 c_1,$$
$$\chi_{\omega=\omega_N} \approx \frac{\omega_N \Delta}{\Delta^2 + \gamma_2^2} \frac{2}{\gamma_2} \varrho_{11} d_0^2 c_0,$$

where $c_0 \equiv 1/2 + \gamma_2/\gamma_1 + f + W/\gamma_1$ 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 "≈" is used for approximate results up to the first order of $\omega_N/\gamma_{1,2}$ hereafter.

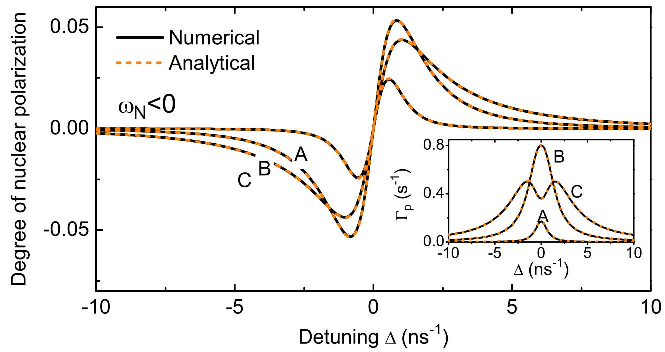

FIG. 2. (color online). $s^{(ss)}$ and Γ_p (inset) from analytical (dashed lines) and numerical (solid lines) results for $\Gamma_1 = 0.2$ s^{-1} and (units ns^{-1}) $\Omega_R = 0.2$ (curve A), 1.0 (curve B), 2.0 (cuve C) and $\gamma_1 = \gamma_2 = 1$, $\omega_N = -0.1$, $\tilde{a}_h = 4 \times 10^{-5}$ (corresponding to a typical $\eta \sim 0.2$).

In the absence of optical pumping, the system is in thermal equilibrium $\hat{\rho}^{\rm eq} = (1/2)(|0,\uparrow\rangle\langle 0,\uparrow| + |0,\downarrow\rangle\langle 0,\downarrow|)$ with the nuclear spin being unpolarized. The (degree of) nuclear polarization $s \equiv 2\langle \hat{I}^z \rangle$ is driven by the optically generated hole,

$$\dot{s} = -\Gamma_1 s - 4\tilde{a}_h\, \mathrm{Im}\, \rho_{1\uparrow,1\downarrow}. \tag{2}$$

In the weak pumping limit ($\Gamma_p \ll \Gamma_1$), perturbation theory gives $\rho^{(ss)}_{1\uparrow,1\downarrow} = A_+ \rho^{\rm eq}_{0\downarrow,0\downarrow} + A_-^* \rho^{\rm eq}_{0\uparrow,0\uparrow}$ with

$$A_\pm = \frac{\Omega_R/2}{\pm\omega_N - i\gamma_1} \frac{\tilde{a}_h}{\Delta \pm \omega_N - i\gamma_2} \frac{\Omega_R/2}{\Delta - i\gamma_2} \tag{3}$$

up to $O(\tilde{a}_h \Omega_R^2)$. Here A_+ (A_-) is the contribution from the down-to-up (up-to-down) channel $\rho^{\rm eq}_{0\downarrow,0\downarrow} \stackrel{\Omega_R}{\to} \rho_{1\downarrow,0\downarrow} \stackrel{\tilde{a}_h}{\to} \rho_{1\uparrow,0\downarrow} \stackrel{\Omega_R}{\to} \rho_{1\uparrow,1\downarrow}$ ($\rho^{\rm eq}_{0\uparrow,0\uparrow} \stackrel{\Omega_R}{\to} \rho_{1\uparrow,0\uparrow} \stackrel{\tilde{a}_h}{\to} \rho_{1\downarrow,0\uparrow} \stackrel{\Omega_R}{\to} \rho_{1\downarrow,1\uparrow}$). The steady-state nuclear polarization is

$$s^{(ss)} \approx -\frac{4\tilde{a}_h^2/\gamma_2}{\Gamma_1} \frac{\Delta\omega_N}{\Delta^2 + \gamma_2^2} \varrho_{11} c_0 \approx -\frac{2\tilde{a}_h^2}{\Gamma_1} \chi_{\omega=\omega_N} \tag{4}$$

up to leading order in $\tilde{a}_h$ and Ω_R. The sign of $s^{(ss)}$ is determined by $-\Delta\omega_N$, in agreement with Eq. (1).

For the evolution of the nuclear spin-1/2 under a general pumping intensity, we note that the motion of the nuclear polarization s characterized by the DNP buildup rate $\Gamma_p = O(\tilde{a}_h^2 C_{\omega=0})$ (estimated from the fluctuation-dissipation theorem) is much slower than the electron-hole subsystem and the nuclear spin coherence $\langle \hat{I}^- \rangle \sim e^{-\Gamma_2 t}$, which is strongly damped by the fluctuating contact hyperfine interaction, with $\Gamma_2 = (a_e^2/8) C_{\omega=0}$. This enables us to identify $s = 2\langle \hat{I}^z \rangle$ as the slow variable and single out its dynamics from the coupled motion through the adiabatic approximation, which essentially assumes that the response of other variables to s is instantaneous. Replacing $\rho_{1\uparrow,1\downarrow}(t)$ in Eq. (2) with its steady-state response $\rho^{(\mathrm{sr})}_{1\uparrow,1\downarrow}(s)$ to a given s yields

$$\dot{s} = -\Gamma_1 s + 2W_+ P_\downarrow - 2W_- P_\uparrow = -\Gamma_1 s - \Gamma_p(s - s_0) \quad (5)$$

valid up to $O(\tilde{a}_h^2)$, where W_+ (W_-) is the transition rates for the down-to-up (up-to-down) channel, $P_\downarrow \equiv (1-s)/2$ [$P_\uparrow \equiv (1+s)/2$] is the spin-down (spin-up) probability of the nucleus,

$$s_0(\Delta) \equiv \frac{W_+ - W_-}{W_+ + W_-} \approx -\frac{\Delta\omega_N}{\Delta^2 + \gamma_2^2}\frac{\gamma_1}{\gamma_2}\frac{c_0}{c_1} \approx -\frac{\chi_{\omega=\omega_N}}{C_{\omega=0}} \quad (6)$$

is the *intrinsic* steady-state nuclear polarization in the absence of other nuclear spin relaxation mechanisms, and

$$\Gamma_p(\Delta) \equiv W_+ + W_- \approx \frac{4\tilde{a}_h^2}{\gamma_1}\varrho_{11} d_0^2 c_1 \approx 2\tilde{a}_h^2 C_{\omega=0} \quad (7)$$

is the hole-induced DNP buildup rate. In the presence of nuclear depolarization, the steady-state nuclear polarization is $s^{(\mathrm{ss})} = \Gamma_p s_0/(\Gamma_1 + \Gamma_p)$, which recovers Eq. (4) in the weak pumping limit ($\Gamma_p \ll \Gamma_1$). The analytical results in Eqs. (6) and (7) agree well with the direct numerical solutions of the density matrix master equation (see Fig. 2).

The adiabatic theory above can be readily generalized to many nuclei of spin higher than 1/2. The only difference is that for many nuclear spins, the large Overhauser shift $\hat{h} \equiv \sum_j a_{e,j}\hat{I}_j^z/2$ of the electron level $|0\rangle$ must be treated non-perturbatively. With the electron-hole motion and off-diagonal nuclear coherences [18] adiabatically eliminated, the diagonal part $\hat{P}$ of the nuclear density matrix $\hat{\rho}_N \equiv \mathrm{Tr}_{\mathrm{eh}}\,\hat{\rho}$ obeys the rate equation

$$\begin{aligned}\dot{\hat{P}} = &-\frac{1}{2}\sum_j W_{j,-}(\hat{\Delta})\left(\hat{I}_j^+\hat{I}_j^-\hat{P} + \hat{P}\hat{I}_j^+\hat{I}_j^- - 2\hat{I}_j^-\hat{P}\hat{I}_j^+\right)\\ &-\frac{1}{2}\sum_j W_{j,+}(\hat{\Delta})\left(\hat{I}_j^-\hat{I}_j^+\hat{P} + \hat{P}\hat{I}_j^-\hat{I}_j^+ - 2\hat{I}_j^+\hat{P}\hat{I}_j^-\right) \quad (8)\end{aligned}$$

up to $O(\tilde{a}_h^2)$. Eq. (8) shows that the jth nuclear spin jumps between adjacent eigenstates of $\hat{I}_j^z$ with rates $\sim I_j(I_j+1)W_{j,\pm}(\hat{\Delta})$ dependent on other nuclear spins through the Overhauser shift $\hat{h}$, where $W_{j,\pm}(\hat{\Delta})$ is obtained from $W_\pm$ by replacing $\tilde{a}_h$, ω_N, and Δ with $\tilde{a}_{h,j}$, $\omega_{N,j}$, and $\hat{\Delta} \equiv \Delta - \hat{h}$, respectively.

For a single nuclear spin-I, by neglecting the Overhauser shift, Eq. (8) gives the *intrinsic* steady-state (degree of) nuclear polarization $s \equiv \langle \hat{I}^z\rangle/I$:

$$s_0^{(I)}(\Delta) \equiv B_I\left(I \ln\frac{1+s_0}{1-s_0}\right) \xrightarrow{|s_0|\ll 1} \frac{2(I+1)}{3}s_0, \quad (9)$$

where $B_I(x)$ is the Brillouin function. For many nuclear spins, to keep the theory simple, we consider identical nuclei $I_j = I$, $\omega_{N,j} = \omega_N, a_{e,j} = a_e, \tilde{a}_{h,j} = \tilde{a}_h$ and hence uniform nuclear polarization $s_j \equiv \langle \hat{I}_j^z\rangle/I_j = \langle \hat{I}^z\rangle/I \equiv s$. When the fluctuation of $\hat{h}$ is much smaller than γ_2, we can replace $\hat{h}$ by its mean-field average $h_{\mathrm{MF}} = A_e I s/2$ and obtain

$$\dot{h}_{\mathrm{MF}} = -\Gamma_p(\Delta_{\mathrm{MF}})\left[h_{\mathrm{MF}} - \frac{1}{2}A_e I s_0^{(I)}(\Delta_{\mathrm{MF}})\right] \quad (10)$$

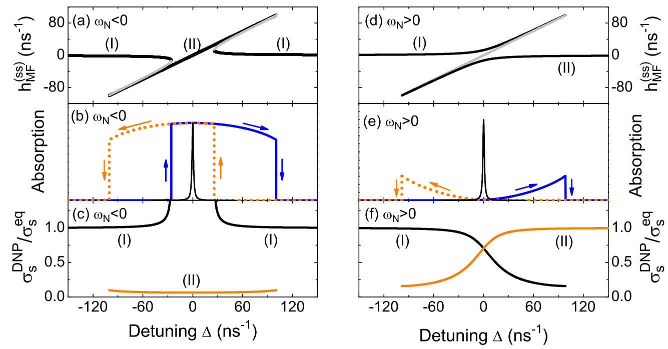

FIG. 3. (color online). (a) and (d): Stable (black lines) and unstable (grey lines) $h_{\mathrm{MF}}^{(\mathrm{ss})}$ vs. detuning. (b) and (e): Optical absorption spectra obtained by sweeping Δ in different directions (indicated by the arrows). (c) and (f): Nuclear fluctuation under hole-induced DNP relative to thermal fluctuation. The calculation is done for a typical QD (e.g., InAs QD) containing $N = 10^4$ identical nuclear spin-9/2's with (unit: ns^{-1}) $A_e = 100$, $\gamma_1 = \gamma_2 = \Omega_R = 1$, and $\omega_N = -0.2$ [(a)-(c)] or 0.2 [(d)-(f)]. The sharp Lorentzian peaks centering at $\Delta = 0$ in (b) and (e) are absorption spectra in the absence of the nuclei.

for $|s_0| \ll 1$, where $A_e \equiv \sum_j a_{e,j}$ is the contact hyperfine interaction constant and $\Delta_{\mathrm{MF}} \equiv \Delta - h_{\mathrm{MF}}$. The steady-state Overhauser shift $h_{\mathrm{MF}}^{(\mathrm{ss})}$ obtained from $h_{\mathrm{MF}} = (A_e I/2)s_0^{(I)}(\Delta_{\mathrm{MF}})$ may have multiple solutions since $s_0^{(I)}(\Delta_{\mathrm{MF}})$ is a highly nonlinear function of Δ_{MF} and hence h_{MF} (see Fig. 2). The condition for a given solution $h_{\mathrm{MF}}^{(\mathrm{ss})}$ to be stable is $(d\dot{h}_{\mathrm{MF}}/dh_{\mathrm{MF}})_{h_{\mathrm{MF}}^{(\mathrm{ss})}} < 0$.

As an example, we consider a typical QD containing $N = 10^4$ identical nuclear spin-9/2's. The *intrinsic* steady-state Overhauser shift $h_{\mathrm{MF}}^{(\mathrm{ss})}$ is shown in Figs. 3(a) and 3(d). For a given detuning Δ, there are at most three possible Overhauser shifts $h_{\mathrm{MF}}^{(\mathrm{ss})}$, with two being stable (black lines) and one being unstable (gray lines). For $\omega_N < 0$ [Fig. 3(a)], when sweeping the laser frequency from large (blue or red) detuning towards resonance, $h_{\mathrm{MF}}^{(\mathrm{ss})}$ starts from the weak polarization phase (curve I) and gets trapped into the strong polarization phase (curve II) when the detuning becomes smaller than a critical value. In the strong polarization phase, $h_{\mathrm{MF}}^{(\mathrm{ss})}$ always tends to compensates the "bare" detuning Δ and locks the effective detuning $\Delta_{\mathrm{MF}} = \Delta - h_{\mathrm{MF}}^{(\mathrm{ss})}$ onto the resonance condition for both $\Delta < 0$ and $\Delta > 0$. By contrast, the electron-induced DNP is weakly dependent on Δ and locks the effective detuning onto the resonance for $\Delta > 0$ (or $\Delta < 0$) only [6, 19, 20]. As a result of the hole-induced DNP, the sharp Lorentzian optical absorption peak is broadened symmetrically into a round top with abrupt edges, where bistable Overhauser shift manifesting as hysteretic loops [3(b)]. By identifying $|0\rangle$ as the vacuum and $|1\rangle$ as the spin-up neutral exciton, the hole-induced DNP qualitatively explains the puzzling observation of hysteretic bidirectional locking of the blue neutral exciton absorption peak [12]. Taking $\gamma_1 = \gamma_2 = \Omega_R = 1\ \mathrm{ns}^{-1}$, $a_h = 0.2a_e = 2\ \mu\mathrm{s}^{-1}$, and a typical hole mixing coefficient $\eta = 0.1$ for self-assembled QDs [21, 22], the DNP buildup time $\tau_p \equiv 1/\Gamma_p \approx 5$ s agrees reasonably with the observed value $\tau_p^{\exp} \approx 1$ s.

For $\omega_N > 0$, $h_{\mathrm{MF}}^{(\mathrm{ss})}$ in Fig. 3(d) always tends to repel the effective detuning $\Delta_{\rm MF}$ away from the resonance condition for both blue detuning and red detuning. As a result, the symmetric Lorentzian absorption peak is shifted hysteretically to finite detunings by the bistable Overhauser shift [Fig. 3(e)].

Locking of the Overhauser shift $\hat{h}$ onto the resonance condition $\Delta_{\rm MF} = 0$ suppresses the nuclear fluctuation [19]. For identical nuclei, the rate equation Eq. (8) leads to the Fokker-Planck equation [6, 19] for the probability distribution $p(s,t) \equiv {\rm Tr}\,\hat{\rho}_N(t)\delta(s-\hat{s})$ of the average (degree of) nuclear polarization $\hat{s} \equiv (1/N)\sum_j \hat{I}_j^z/I$ (N is the number of QD nuclei):

$$\frac{d}{dt}p(s,t) = \frac{\partial}{\partial s}\left[D(s)\frac{\partial}{\partial s}p(s,t) - v(s)p(s,t)\right] \qquad (11)$$

with the drift coefficient $v(s) = a[G_+(s) - G_-(s)]$ and the diffusion coefficient $D(s) = (a^2/2)[G_+(s) + G_-(s)]$, where $a = 1/(NI)$ is the change of $\hat{s}$ by each nuclear spin flip, $G_\pm(s) \equiv N_\mp(s)W_\pm(\Delta - A_e I s/2)$, and $N_\pm(s) \equiv NI[2(I+1)/3 \pm s]$. The steady-state solution to Eq. (11) is $p^{(\rm ss)}(s) = p^{(\rm ss)}(s^*)\exp[\int_{s^*}^{s} v(s')/D(s')ds']$, with the most probable nuclear polarization s^* determined by $v(s^*) = 0$, which is equivalent to the mean-field approximation in Eq. (10). The stability condition $(\partial v(s)/\partial s)_{s=s^*} < 0$ for s^* is also equivalent to $(d\dot{h}_{\rm MF}/dh_{\rm MF})_{h^{(\rm ss)}_{\rm MF}} < 0$. First-order Taylor expansion around s^* shows that $p^{(\rm ss)}(s)$ assumes a Gaussian form centered at s^*, with the standard deviation

$$\sigma_s^{\rm DNP}(s^*) = \sqrt{\left|\frac{D(s^*)}{[\partial v(s)/\partial s]_{s=s^*}}\right|} \sim \frac{1}{N}\sqrt{\frac{\gamma_2^2}{a_e I\,|\omega_N|}}. \qquad (12)$$

By contrast, in the absence of optical pumping, the nuclear spins are in the unpolarized state with standard deviation $\sigma_s^{\rm eq} = [(I+1)/(3NI)]^{1/2}$.

For a typical QD (e.g., InAs QD) with $I = 9/2$ and (units: ns^{-1}) $\gamma_2 = 1, A_e = 100$, and $|\omega_N| = 0.2$ (corresponding to $|B| \sim$ 2 T), we plot the ratio $\sigma_s^{\rm DNP}(s^*)/\sigma_s^{\rm eq}$ in Figs. 3(c) and 3(f). Corresponding to the two stable phases (curves I and II) of the nuclear polarizations in Figs. 3(a) and 3(d), there are two possible nuclear fluctuations $\sigma_s^{\rm DNP}$(I) (curve I) and $\sigma_s^{\rm DNP}$(II) (curve II) in Figs. 3(c) and 3(f). For $\omega_N < 0$ [Fig. 3(c)], once the nuclear spins are trapped into the strong polarization phase [curve II in Fig. 3(a)] by frequency sweeping of the laser, the nuclear fluctuation is suppressed below its thermal equilibrium value by a factor $\sim$ 10, in reasonable agreement with the one pump experiment in Ref. [11]. Interestingly, the strongest ($\sim$ 15-fold) suppression of the nuclear fluctuation occurs at the resonance condition $\Delta = \Delta_{\rm MF} = 0$, where the nuclear polarization vanishes. This corresponds to a $\sim$ 15-fold enhancement of the electron spin coherence time. For $\omega_N > 0$ [Fig. 3(f)], the resonance condition $\Delta_{\rm MF} = 0$ and hence maximal suppression appears at finite detunings.

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