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The Nuclear Dark State under Dynamical Nuclear Polarization *

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We analyze aspects of nuclear dark states. The formation of the dark states prevents the further establishment of nuclear polarizations, while inhomogeneous nuclear spin precessions can result in leakage from these states. An optimal efficiency for pumping nuclear polarization is achieved when the dynamical nuclear polarization (DNP) cycling rate is comparable to the dark-state leakage rate. When the DNP rate is much larger, the nuclear spin bath can be locked on the dark states. We propose schemes where the inhomogeneous precessions can be suppressed for the realization of large-scale dark states. As the dark states correspond to low transverse nuclear field fluctuations, they can be used to suppress the decoherence of the electron induced by the nuclear spins.

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A single electron spin localized in a semiconductor quantum dot is a promising candidate as a building block for a solid-state quantum computer.[1] Quantum controls of single electron spins by electrical and optical means have been experimentally demonstrated in various types of quantum-dot systems, most of which are hosted by Ⅲ–V semiconductor compounds.[2−14] In Ⅲ–V compounds, the single electron spin is coupled to many lattice nuclear spins through the hyperfine interaction. This is recognized as the main cause of electron spin decoherence at low temperatures (∼0.1–1 K) when phonon-induced dephasing becomes suppressed. The above temperature range is still much larger compared with the nuclear Zeeman energy, which is of the order ∼mK in a field of 1 Tesla. Thus, the nuclear spin bath at thermal equilibrium has large statistical fluctuations. As a result, the electron Zeeman frequency has large inhomogeneous broadening. Typically, the ensemble dephasing time of the electron spin is T_2^*∼ns.[15−18] Nuclear spin interactions can also result in dynamic fluctuations of the hyperfine field, which can cause electron spin decoherence in a timescale of T_2∼μs, when the inhomogeneous broadening is eliminated.[19−21] This leaves much room to improve the electron spin coherence time if the inhomogeneous broadening of the nuclear spin bath can be suppressed.

Various approaches have been proposed and implemented to suppress inhomogeneous broadening. Spin echo types can rephase inhomogeneous evolutions and transiently recover the coherence in certain time windows.[14,22−25] An alternative approach is to prepare the nuclear spin bath prior to quantum control such that the statistical fluctuations in the nuclear field are suppressed.[26−32] Once prepared, nuclear spins can remain on such configurations for a sufficiently long time for the subsequent quantum controls, as their dephasing and relaxation processes are extremely slow. An intuitive approach is to polarize the nuclear spins to suppress the longitudinal fluctuations.[35,36] Nuclear spins can be polarized through dynamical nuclear polarization (DNP) processes, in which the electron passes its spin polarization to the nuclei through electron-nuclear flip-flop.[37] However, experimentally achieved nuclear spin polarization has been limited, even though the DNP cycling rate is controlled so it is much faster than the nuclear spin relaxation time. In small quantum dots, polarizations of ∼10–70% were reported in various setups,[18,38−42] and in large quantum dots the polarization is even lower at ∼1%.[13,32] With the experimental parameters, a high nuclear spin polarization close to 1 is expected if the nuclear spin bath is assumed to be semiclassical, i.e., with the quantum coherence between nuclear spins neglected. One possible explanation for the low nuclear polarization could be the formation of nuclear dark states due to the mutual coherence established by the electron-nuclear flip-flop.[35,43] In addition, more sophisticated approaches utilizing quantum feedback mechanisms were also proposed and experimentally implemented.[26−34]

In this Letter, we analyze the general aspects of nuclear dark states in DNP processes, defined as those states annihilated by the collective nuclear flip operator in electron-nuclear flip-flop coupling. We show that the fluctuations of the transverse nuclear field can be substantially suppressed in such dark states. The conclusion is obvious when nuclear spins have homogeneous coupling strength with the electron: the transverse fluctuation is $\sqrt{J}$ in dark states as compared to its value of $\sim J$ in thermal equilibrium, where J is the total spin of the nuclei. When the number of nuclear spins N is large, the most probable value is $J \sim \sqrt{N}$. Our numerical calculations show that the inhomogeneous dark states have also suppressed the transverse fluctuations, which is similar to the homogeneous example. This shows another possibility to prepare the nuclear spin bath for a better electron

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spin qubit: by aligning one of the transverse directions with the external magnetic field through a global rotation of nuclear spins, the electron spin coherence time can be enhanced. We further discuss the effects of inhomogeneous nuclear precessions by longitudinal hyperfine coupling. The inhomogeneous precessions can result in leakage out of the dark states, and the leakage rate is proportional to the inhomogeneity in the hyperfine coupling coefficients. We find that the effective rate for pumping nuclear polarization has a non-monotonic dependence on the DNP cycling rate. An optimal pumping efficiency is achieved when the DNP rate is comparable to the dark-state leakage rate. When the DNP rate is much larger, the nuclear spin bath can be locked on the dark states.

The essential physics can be illustrated by considering an electron spin coupled to a bath of many nuclear spins with homogeneous hyperfine coupling strength. Neglecting the weak noncollinear (e.g., $\hat{I}_x^n \hat{S}_z$) terms,[44–46] the Hamiltonian reads

$$\hat{H} = \omega_{\rm e}\hat{S}_z + \sum_n [\omega_N \hat{I}_z^n + \frac{a_t}{2}(\hat{I}_+^n \hat{S}_- + \hat{I}_-^n \hat{S}_+) + a_l \hat{I}_z^n \hat{S}_z], \tag{1}$$

where $\hat{\boldsymbol{S}}$ denotes the spin of the single electron and $\hat{\boldsymbol{I}}^n$ denotes the spin of the nth nuclei; $\omega_{\rm e}$ and ω_N are the electron and nuclear Zeeman energy, respectively. If we define the collective spin operator of the nuclear spin bath: $\hat{\boldsymbol{J}} \equiv \sum_n \hat{\boldsymbol{I}}^n$, the hyperfine interaction simply becomes: $a_t(\hat{J}_+\hat{S}_- + \hat{J}_-\hat{S}_+)/2 + a_l\hat{J}_z\hat{S}_z$. We use $|J, m, \beta\rangle$ to denote the common eigenstates of the collective spin operators $\hat{J}^2$ and $\hat{J}_z$ with eigenvalues $J(J+1)$ and m, respectively; β denotes the additional set of quantum numbers for resolving the degeneracy.

The experimentally accessible temperature for quantum-dot systems varies from ∼0.1–1 K, which can be considered as a high limit for the nuclear spin. Thus, for a nuclear spin bath at thermal equilibrium, the density matrix can be written as $\rho_T = \frac{1}{2^N}\sum_{J,m,\beta}|J, m, \beta\rangle\langle J, m, \beta|$, i.e., all states are equally populated. When N is large, the distribution of the total magnetic quantum number is a normal distribution: $P_m(m) \approx \frac{1}{\sqrt{2\pi}\sigma}\exp(-\frac{m^2}{2\sigma^2})$, where $\sigma \sim \sqrt{N}$. We find the distribution of the total spin quantum number $P_J(J) \propto J\exp(-\frac{J^2}{2\sigma^2})$, and the most probable value $J \sim \sqrt{N}$. The electron spin decoherence is determined by the fluctuations of the collective spin operator: $\Delta J_\nu \equiv (\langle \hat{J}_\nu^2\rangle - \langle \hat{J}_\nu\rangle^2)^{1/2}$, where ν denotes the three orthogonal directions. ρ_T is invariant under rotation, thus $\langle \hat{J}_\nu\rangle = 0$ and the fluctuation $\Delta J_\nu \sim \sqrt{N}$.

In a DNP process, the flip-flop action on the nuclear spin bath is described by the collective spin flip operator $\hat{J}^-$. The repeated actions of $\hat{J}^-$ bring every possible initial state $|J, m, \beta\rangle$ to the dark state $|J, m = -J, \beta\rangle$. The dark states have two remarkable features. First, because of the mutual coherence established between the nuclear spins, the nuclear polarization cannot be further increased by the collective spin flip since $\hat{J}^-|J, -J, \beta\rangle$=0. Second, in dark states the transverse fluctuation of the nuclear hyperfine field is minimized evidently from the equality $\langle J, m, \beta|\hat{J}_x^2 + \hat{J}_y^2|J, m, \beta\rangle = J^2 + J - m^2$ (see Fig. 1(c)).

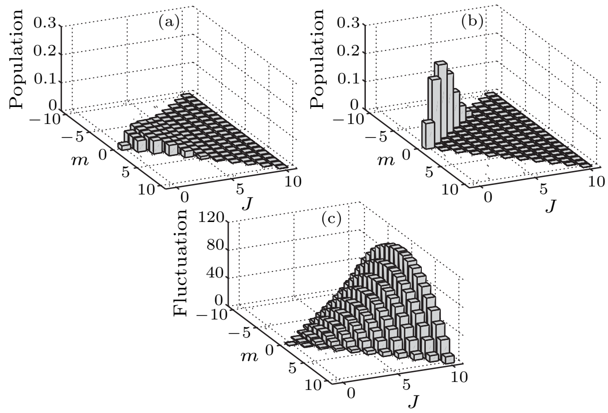

Fig. 1. (a) Population distribution $P(J, m)$ for a bath of 20 spins in the thermal equilibrium at high temperature limit. (b) Population distribution when the bath is fully pumped to the dark state from the thermal equilibrium with the total spin quantum number J conserved. (c) Fluctuation of the transverse field for total spin eigenstates $|J, m, \beta\rangle$ for various values of J and m.

If we consider a nuclear spin bath initially in the state ρ_T, an efficient DNP process will saturate it to the dark state $\rho_D = \sum_{J,\beta}\frac{2J+1}{2^N}|J, -J, \beta\rangle\langle J, -J, \beta|$ (see Fig. 1(b)). In this state, the nuclear polarization $\frac{\langle \hat{J}_z\rangle}{N} \sim \frac{1}{\sqrt{N}}$ is negligibly small for large N. The longitudinal fluctuation is $\Delta J_z \sim \sqrt{N}$, which is comparable to that of the thermal distribution. Remarkably, the transverse fluctuations $\Delta J_{x,y} \sim N^{1/4}$ are substantially reduced. Thus, suppression of the nuclear hyperfine field along two orthogonal directions can be achieved by pumping the nuclear spin bath to the dark states.

The above model with homogeneous hyperfine coupling coefficients for different nuclear spins is an idealized situation. In the more realistic case, the hyperfine coupling is $\sum_n[\frac{a_{n,t}}{2}(\hat{I}_+^n\hat{S}_- + \hat{I}_-^n\hat{S}_+) + a_{n,l}\hat{I}_z^n\hat{S}_z]$, i.e., the coupling strength a_n varies for different nuclear spins; a_n is proportional to the electron density at the given nuclear site. For a confined electron, the density distribution is always inhomogeneous, hence the a_n for different sites are always inhomogeneous.

The nuclear spins can be grouped into shells according to the hyperfine coupling strength. For nuclear spins in the same shell, the coupling a_n are the same, and thus only their collective spin couples to the electron. Taking the phosphorus donor in silicon for example, the first several shells are (A,6.0,6), (B,4.5,12), (C,3.3,4), (D,2.2,12), and (F,1.7,12), where the letter is the shell label by convention, the second number is the hyperfine coupling strength in units of MHz, and the last is the number of equivalent sites on the shell.[47,48] When the electron wavefunction is more localized, the number of shells is smaller and the inhomogeneity of a_n for different shells is stronger. For deep centers such as NV centers in diamond, the electron is strongly coupled with a few shells with a strong inhomogeneity: the first two shells have a hyperfine coupling strength of $\sim$130 MHz (three sites) and $\sim$70 MHz (nine sites), respectively, while for the rest of the shells the hyperfine coupling strength is less than 10 MHz.[51]

The electron-nuclear flip-flop has an energy cost $\bar{\omega}_{\rm e} - \omega_N + a_{n,l}$, where $\bar{\omega}_{\rm e}$ is the electron Zeeman energy in the total magnetic field (the external field plus the nuclear field). When the difference in a_n is sufficiently large, it is possible to pump each shell independently by tuning the resonances in the DNP controls.[48–50] In such a case, different shells can be independently prepared to dark states. This reduces to the homogeneous situation discussed previously.

In what follows, we focus on nuclear spin baths where the inhomogeneity in a_n is small and insufficient for resolving the different shells in the DNP controls. Similar to the homogeneous case, we can define the nuclear hyperfine field operators: $\hat{A}_{\pm} \equiv \sum_n \frac{a_{n,t}}{\bar{a}_t} \hat{I}^n_{\pm}$ and $\hat{A}_z \equiv \sum_n \frac{a_{n,l}}{\bar{a}_l} \hat{I}^n_z$, where $\bar{a}$ denotes an average of a_n. The hyperfine coupling can then be written as $\bar{a}_t(\hat{A}_+\hat{S}_- + \hat{A}_-\hat{S}_+)/2 + \bar{a}_l \hat{A}_z \hat{S}_z$. In the DNP process, nuclear spins are collectively flipped by $\hat{A}_-$. Similar dark states exist in this inhomogeneous case.[43]

In the homogeneous case, the $2J+1$ states $|J,m,\beta\rangle$ with the same J and β quantum number form a multiplet, i.e., an irreducible invariant subspace of the total spin. These collective spin eigenstates can be expanded in the product basis of individual spins: $|J,m,\beta\rangle = \sum_{\{m_n\}} c_{J,\beta}(\{m_n\}) \prod_n |I_n, m_n\rangle$, where $c_{J,\beta}$ are the Clebsch–Gordan coefficients. The collective operator $\hat{J}_-$ couples states in the same multiplet: $\hat{J}_-|J,m,\beta\rangle = \sqrt{(J+m)(J-m+1)}|J,m-1,\beta\rangle$, like lowering a ladder. The inhomogeneous case has an isomorphic structure. One can find a complete while nonorthogonal basis $|\mathcal{L}_{J,m,\beta}\rangle = \mathcal{N}_{J,m,\beta} \sum_{\{m_n\}} \mathfrak{c}_{J,\beta} \prod_n |I_n, m_n\rangle$, such that $\hat{A}_-|\mathcal{L}_{J,m,\beta}\rangle = \frac{\mathcal{N}_{J,m-1,\beta}}{\mathcal{N}_{J,m,\beta}} \sqrt{(J+m)(J-m+1)}$ $|\mathcal{L}_{J,m-1,\beta}\rangle$, where $\mathfrak{c}_{J,\beta} = c_{J,\beta} \prod_n a_n^{I_n - m_n}$, and $\mathcal{N}_{J,m,\beta} \sim \bar{a}^{-(J-m)}$ is the normalization factor. $|\mathcal{L}_{J,m,\beta}\rangle$ are known as the generalized eigenvectors of $\hat{A}_-$. The dimensions of the multiplet for the homogeneous and inhomogeneous case are identical. Three differences from the homogenous case are noted. First, $|\mathcal{L}_{J,m,\beta}\rangle$ are not eigenstates of $\hat{A}_z$. Second, $|\mathcal{L}_{J,m,\beta}\rangle$ with the same m while different J or β are not orthogonal in general. Third, $\hat{A}_-$ and its Hermitian conjugate $\hat{A}_+$ do not have the same set of generalized eigenvectors as $\hat{J}_-$ and $\hat{J}_+$. $\hat{A}_+$ can couple generalized eigenvectors of $\hat{A}_-$ from different multiplets (see Fig. 2(b)). It can also be proved that $\langle \mathcal{L}_{J',m',\beta'}|\hat{A}_+|\mathcal{L}_{J,m,\beta}\rangle = 0$ for $|J-J'|>1$ or $m' \neq m+1$.

At the bottom of each multiplet ($m=-J$), we have the inhomogeneous dark states with $\hat{A}_-|\mathcal{L}_{J,-J,\beta}\rangle = 0$.[35,43] Thus, efficient DNP pumping with $\hat{A}_-$ will saturate the nuclear spin bath from the arbitrary initial state towards the inhomogeneous dark states $\rho_{ID} = \sum_{J,m,\beta} p'(J,m,\beta) |\mathcal{L}_{J,m,\beta}\rangle\langle\mathcal{L}_{J,m,\beta}|$. Obviously $\langle \mathcal{L}_{J,-J,\beta}|\hat{A}_{x,y}|\mathcal{L}_{J,-J,\beta}\rangle = 0$, i.e., the expectation value of the transverse nuclear field is zero. In the following, we show numerically that, in the inhomogeneous dark states, the transverse fluctuations are well suppressed, just like in the homogeneous case.

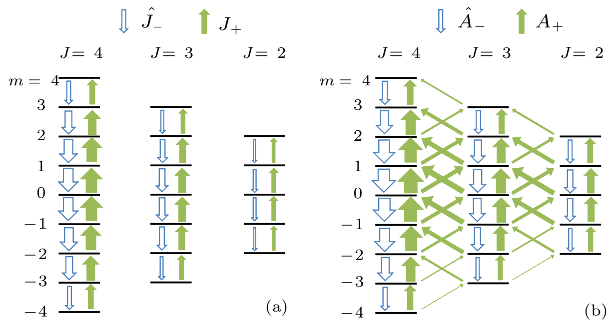

Fig. 2. (Color online) The isomorphism between generalized eigenstates of the homogeneous lowering operator $\hat{J}_-$ and the inhomogeneous $\hat{A}_-$, illustrated for an ensemble of two spins with $I_1 = 1/2$ and $I_2 = 3/2$, respectively. $\hat{J}_- = \hat{I}_{1,-} + \hat{I}_{2,-}$, and $\hat{A}_- = 1.2\hat{I}_{1,-} + 0.8\hat{I}_{2,-}$. In (a), the horizontal lines denote $|J,m,\beta\rangle$. The hollow (solid) arrows show the coupling by $\hat{J}_-$ ($\hat{J}_+$), with the coupling strength indicated by the thickness of the arrow. In (b), the horizontal lines denote $|\mathcal{L}_{J,m,\beta}\rangle$, the generalized eigenstates of $\hat{A}_-$. Hollow (solid) arrows show the coupling by $\hat{A}_-$ ($\hat{A}_+$), with the strength indicated by the arrow thickness.

In our calculation, we evaluate the inhomogeneous dark states of 1600 nuclear spins distributed on eight shells. Each shell has $n_i = 200$ nuclear spins coupled to the single electron with hyperfine coupling strength a_i, where i denotes the shell index. $\{a_i\}$ is set as an arithmetic sequence. Other choices of n_i and a_i give similar values on the transverse fluctuation when the average $\bar{a} \equiv \frac{\sum_i n_i a_i}{\sum_i n_i}$ and standard deviation $\Delta a \equiv \sqrt{\frac{\sum_i n_i a_i^2}{\sum_i n_i} - \bar{a}^2}$ take comparable values. We consider the nuclear spin bath saturated in dark states by pumping with $\hat{A}_-$ from the initial state ρ_T. We evaluate $\langle \mathcal{L}_{J,-J,\{j_i\}}|\hat{A}_x^2 + \hat{A}_y^2|\mathcal{L}_{J,-J,\{j_i\}}\rangle$, with J and $\{j_i\}$ randomly selected according to the thermal distribution. Table 1 summarizes this fluctuation with different $\Delta a/\bar{a}$.

Clearly for dark states in the weak inhomogeneous case, the transverse fluctuations are still in the order of $\sim N^{1/4}$, comparable to the homogeneous case. This has a practical and important use for the spin quantum bit where the statistical fluctuations from the nuclear spin bath are the main source of decoherence. Simply through saturation towards the dark states by efficient DNP pumping, the transverse fluctuations of the nuclear hyperfine field get suppressed from the initial thermal value of $\sim N^{1/2}$ to the dark state value of $\sim N^{1/4}$. One can then rotate the quantization axis of the electron spin to one of the transverse directions in subsequent operations, and the qubit coherence time can be enhanced by a factor of $\sim N^{1/4}$. In nuclear spin baths with large N (e.g., for a gate-defined dot where $N \sim 10^7$), the enhancement could be significant.

Table 1. Fluctuation of the transverse field of the dark states against the standard deviation of a_i. Here we take $N = 1600$ qubits for the nuclear bath, and thus the typical value of J in the homogenous case is $J_{\rm typ} = 20$. We set $n_i = 200$, and thus the contribution to the total fluctuation of each group is comparable.

$\Delta a/\bar{a}$	0	0.1	0.2	0.3	0.4
$\langle \hat{A}_x^2 + \hat{A}_y^2 \rangle$	20	22.26	26.84	31.71	39.27

In the above discussions, we have considered the effects of the collective pumping by $\hat{A}_-$. The nuclear spins also precess with inhomogeneous frequencies because of the longitudinal hyperfine coupling $\hat{S}_z \hat{A}_z$. The inhomogeneous precessions cause transitions between the generalized eigenstates of $\hat{A}_-$ since they are not the eigenstates of $\hat{A}_z$. A straightforward calculation shows that the matrix element $\langle \mathcal{L}_{J',m',\beta'} | \hat{A}_z | \mathcal{L}_{J,m,\beta} \rangle$ is non-zero only if $J' = J, J \pm 1$ and $m' = m$.[48,49,52] Thus, for the dark state, we have $\hat{A}_z | \mathcal{L}_{J,-J,\beta} \rangle = \kappa_0 | \mathcal{L}_{J,-J,\beta"} \rangle + \kappa_+ | \mathcal{L}_{J+1,-J,\beta'} \rangle$. $| \mathcal{L}_{J+1,-J,\beta'} \rangle$ is a state next to the dark state in a multiplet of a larger dimension (see Fig. 3). Such states can be further pumped with $\hat{A}_-$ to the new dark states $| \mathcal{L}_{J+1,-J-1,\beta'} \rangle$, which correspond to a larger nuclear polarization. The weak noncollinear terms such as $\hat{I}_x^n \hat{S}_z$ that we have ignored provide an additional leakage mechanism from the dark states, while as long as their strength is weak, the main leakage mechanism should be the $\hat{S}_z \hat{A}_z$ term.

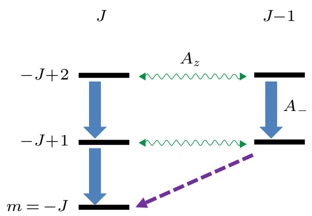

Fig. 3. (Color online) Effective population leakage due to inhomogeneous precession. Each horizontal line denotes $| \mathcal{L}_{J,m,\beta} \rangle$, with the given values of J and m. The blue solid arrows and green wavy lines indicate $\hat{A}_-$ pumping and $\hat{A}_z$ precession, respectively. The purple dashed line shows the effective population leakage.

DNP in the presence and absence of inhomogeneous nuclear spin precessions are qualitatively different. When pumping alone by $\hat{A}_-$, the nuclear spin bath becomes saturated at the dark states and nuclear polarization is limited to small values. This is the manifestation of the mutual coherence between nuclear spins established by the collective spin flips. The inhomogeneous precessions can dephase the nuclear spins and allow the nuclear polarization to be further increased. When nuclear inhomogeneous precessions and collective spin flips are both taking effect, the steady state is the fully polarized state.

We can estimate the leakage rate by calculating the fidelity $F(t) \equiv \langle \mathcal{L}_{J,m,\beta} | e^{-i\hat{A}_z t} | \mathcal{L}_{J,m,\beta} \rangle$. A dark state is a linear superposition of the product states $\prod_n |I_n, m_n\rangle$, which are eigenstates of $\hat{A}_z$ as $\hat{A}_z \prod_n |I_n, m_n\rangle = E_{\{m_n\}} \prod_n |I_n, m_n\rangle$ with $E_{\{m_n\}} = \sum_n a_n m_n$. Thus $F(t) = \sum |\mathfrak{c}_{J,\beta}|^2 e^{-itE_{\{m_n\}}} \simeq e^{-\kappa^2 t^2/2}$, and we have the leakage rate $\kappa = \sqrt{\sum_{\{m_n\}} |\mathfrak{c}_{J,\beta}|^2 E_{\{m_n\}}^2 - (\sum_{\{m_n\}} |\mathfrak{c}_{J,\beta}|^2 E_{\{m_n\}})^2}$. For $J \sim \sqrt{N}$, we find $\kappa \sim \Delta a \sqrt{N}$. Thus, the leakage rate is proportional to the inhomogeneity of a_n.

The above picture provides an explanation on the inefficiency of DNP in large quantum-dot systems.[32] In each DNP cycle, a collective spin flip by $\hat{A}_-$ is applied on the nuclear spin bath. Because of the existence of the dark states, the effective rate η for polarizing nuclear spins is not determined by the DNP cycling rate $\mathit{\Gamma}$, rather by the dark state leakage rate κ for large $\mathit{\Gamma}$. In smaller quantum dots, such as the self-assembled quantum dot,[18,38] a larger nuclear polarization is achieved. This is consistent with the above picture since the inhomogeneity Δa is much larger in these small quantum dots.

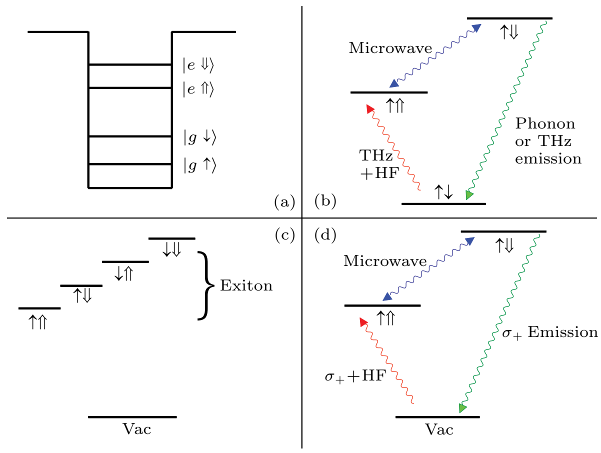

Fig. 4. Two pumping schemes to reach the dark states, (a) and (b) the double spin method, (c) and (d) the exciton method; (a) and (c) the energy levels, and (b) and (d) the pumping cycles of the electron (exciton) spins.

The above picture also leads to an interesting conclusion that the DNP is most efficient when $\mathit{\Gamma} \sim \kappa$. In a simplified picture, we can consider the dynamics in the subspace formed by the three states: $| \mathcal{L}_{J,-J,\beta} \rangle$, $| \mathcal{L}_{J+1,-J,\beta'} \rangle$, and $| \mathcal{L}_{J+1,-J-1,\beta'} \rangle$. $\hat{A}_z$ couples $| \mathcal{L}_{J,-J,\beta} \rangle$ and $| \mathcal{L}_{J+1,-J,\beta'} \rangle$ with a strength $\kappa_+ \sim \kappa \sim \Delta a \sqrt{N}$, and $\hat{A}_-$ pumps the population incoherently from $| \mathcal{L}_{J+1,-J,\beta'} \rangle$ to $| \mathcal{L}_{J+1,-J-1,\beta'} \rangle$ with the cycling rate $\mathit{\Gamma}$. For $\mathit{\Gamma} \le \kappa_+$, the effective pumping rate $\eta = \mathit{\Gamma}$. In this limit, the DNP of nuclear spins behaves as a semiclassical process, i.e., the mutual nuclear coherence can be neglected due to the fast inhomogeneous precession. For $\mathit{\Gamma} > \kappa_+$, the effective pumping rate is $\eta = \mathit{\Gamma} - \sqrt{\mathit{\Gamma}^2 - \kappa_+^2}$. In the limit $\mathit{\Gamma} \gg \kappa_+$, the inhomogeneous nuclear precession is inhibited by the strong incoherent pumping ($\eta \sim \kappa_+^2/2\mathit{\Gamma}$). This is a manifestation of the quantum Zeno effect. DNP control with a large cycling rate can not only pump the nuclear spin bath to dark states, but also lock it on such states. The most efficient DNP is achieved when $\mathit{\Gamma} \sim \kappa_+$.

For the purpose of suppression, the nuclear field fluctuation and enhancement of the electron spin coherence time, it is desirable to lock the nuclear spin bath on some dark states with J taking the typical value $\sim\sqrt{N}$. From the above discussions, this can be achieved by pumping from the thermal distribution with the DNP cycling rate $\Gamma \gg \kappa \sim \Delta a\sqrt{N}$. This could be possible in a system with very weak inhomogeneity. On the other hand, if one can suppress the inhomogeneous precessions from $\hat{A}_z$ while performing DNP, then the desired dark states can be easier to access. In the following, we propose two schemes for such purposes.

In the first scheme, we consider a quantum dot charged with two electrons (see Fig. 4). The four lowest energy single electron states are: orbital ground state with spin up ($|g\uparrow\rangle$) and spin down ($|g\downarrow\rangle$), and first-excited state with spin up ($|e\Uparrow\rangle$) and spin down ($|e\Downarrow\rangle$). The two-electron ground state is the singlet state $|g\uparrow, g\downarrow\rangle$, where the hyperfine interaction with nuclear spins vanishes. For a typical quantum dot, the orbital ground state and excited state have an energy difference of several meV and can be coupled by a terahertz laser.[53,54] The optical coupling does not change the spin. The DNP control makes use of the optical excitation of a spin-forbidden transition from $|g\uparrow, g\downarrow\rangle$ to $|g\uparrow, e\Uparrow\rangle$ (see Figs. 4(a) and 4(b)). By tuning the laser frequency to be in resonance with this transition in an external magnetic field, all spin-allowed transitions are suppressed by the detuning, while the desired spin-forbidden transition can be optically excited through the assistance by the electron-nuclear flip-flop.[55] The transition is accompanied by an action of $\hat{A}^-$ on the nuclear spin bath. A microwave field then pumps the population from $|g\uparrow, e\Uparrow\rangle$ to $|g\uparrow, e\Downarrow\rangle$, which can relax back to the ground state either by emitting a phonon or a terahertz photon. This completes one cycle of the DNP process. With a strong microwave field and an efficient relaxation channel back to the ground state, the steady-state population on the two intermediate states $|g\uparrow, e\Uparrow\rangle$ and $|g\uparrow, e\Downarrow\rangle$ can be small. Since the ground state has no hyperfine coupling with nuclear spins, the inhomogeneous nuclear precessions by $\hat{A}_z$ are suppressed, while $\hat{A}_-$ is repeatedly applied.

The second scheme is similar, except that it makes use of the excitonic transitions in a neutral quantum dot. There are four exciton ground states, $|\uparrow\Uparrow\rangle$, $|\uparrow\Downarrow\rangle$, $|\downarrow\Uparrow\rangle$ and $|\downarrow\Downarrow\rangle$, where $\uparrow$ ($\downarrow$) denotes a spin up (down) electron and $\Uparrow$ ($\Downarrow$) denotes a spin up (down) hole. The state with the absence of the electron and hole is denoted as vac. This scheme also makes use of the spin optical transition selection rules in the III–V semiconductor: an optical field with σ^+ (σ^-) circular polarization couples vac to the bright exciton state $|\uparrow\Downarrow\rangle$ ($|\downarrow\Uparrow\rangle$) only. The transition from vac to the other two exciton states $|\uparrow\Uparrow\rangle$ and $|\downarrow\Downarrow\rangle$ are spin-forbidden and known as the dark excitons. In an external magnetic field, the four exciton states are split in energy (see Fig. 4(c)). A σ^+ polarized laser with a frequency in resonance with the dark exciton $|\uparrow\Uparrow\rangle$ can drive this spin-forbidden excitonic transition with the assistance of the electron-nuclear flip-flop.[56] A microwave field can couple this dark exciton to the bright exciton $|\uparrow\Downarrow\rangle$, where the electron and hole recombine, by emitting a σ^+ photon. This DNP cycle pumps the nuclear spin by operation $\hat{A}_-$, during which the effects of $\hat{A}_z$ are suppressed because most of the time the quantum dot stays in the vac state with the absence of the electron and hole.

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