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Many-body singlets by dynamic spin polarization

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We show that dynamic spin polarization by collective raising and lowering operators can drive a spin ensemble from arbitrary initial state to many-body singlets, the zero-collective-spin states with large-scale entanglement. For an ensemble of N arbitrary spins, both the variance of the collective spin and the number of unentangled spins can be reduced to $O(1)$ [versus the typical value of $O(N)$], and in the steady state many-body singlets are occupied with a population of ∼20% independent of the ensemble size. We discuss a potential implementation in a mesoscopic ensemble of nuclear spins using dynamic nuclear spin polarization by an electron. The result is of twofold significance for spin quantum technology: (1) a cleaner surrounding and less quantum noise for the electron spin and (2) a resource of entanglement for nuclear-spin-based quantum information processing. The scheme can also be applied to other spin systems where collective raising and lowering operations are available.

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Many-body singlets (MBSs) are the zero-collective-spin states of a spin ensemble with large-scale quantum entanglement and zero spin uncertainties. They appear in a variety of contexts in quantum physics and in condensed matter physics, e.g., as horizon states of the quantum black hole,[1] and as ground states of quantum antiferromagnetic models.[2] Their special characteristics place them at the center of attention for quantum applications. First, MBSs are invariant under a simultaneous unitary rotation on all spins. This makes MBSs suitable for spanning a decoherence-free subspace,[3] for quantum communications without a shared reference frame,[4] and for metrology of the spatial gradient or fluctuations of external fields.[5] Second, MBSs are an extreme example for the squeezing of spin uncertainties.[6–9] The collective spin has zero variance in all directions and thus a source of quantum noise is removed, e.g., in the context of a quantum object affected by a spin bath. Third, MBSs contain large-scale quantum entanglement: every spin is entangled with the rest of the ensemble. An example of pure MBSs is the product of 2-qubit singlets (Bell pairs). In the maximally mixed state of all MBSs, the distillable bipartite entanglement is logarithmic in the ensemble size.[1]

Despite the successful generation of a photonic analog of 4-qubit singlets by parametric down conversion,[3,10] realization of MBSs in a general spin ensemble is an outstanding goal awaiting technically feasible approaches. Theoretical study shows that spin squeezing based on quantum nondemolition measurement can reduce the total collective spin variance of an atomic ensemble by a factor of 5 in the lossless case.[5] However, in such squeezed state the weighting of MBSs is small and vanishes in large N limits. Other relevant studies include the proposal of a physical model for projective measurement of MBSs by spin-dependent scattering.[11]

Here we introduce an approach for squeezing of collective spin uncertainties and generation of large-scale entanglement. The approach uses collective spin raising and lowering operations only and is applicable to an ensemble of N arbitrary spins initially in arbitrary state. The state after squeezing is significant in figures of merit: in the low-loss limit, MBSs are occupied with an N-independent population of ∼20% in the steady state, and both the variance of the total collective spin and the number of spins unentangled with the rest are $O(1)$ [versus the typical values of $O(N)$]. We discuss potential implementation in a mesoscopic ensemble of nuclear spins, a spin system of extensive interest either as a noise source or as a superior information storage in quantum technology. Our scheme uses only generic features of dynamic nuclear spin polarization processes by an electron. MBSs can be a valuable resource of quantum entanglement for nuclear-spin quantum information processing.[12–14] In electron-spin-based quantum computation, reduction in the collective spin fluctuations of the peripheral nuclei means less quantum noise for the electron spin. The approach can be applied to other spin (pseudospin) systems where collective raising/lowering operations are available.

We refer to the definition of spin squeezing in the generalized sense,[5,9] where the degree of squeezing is quantified by $\langle \hat{\boldsymbol{J}}^2\rangle$, with $\hat{\boldsymbol{J}} \equiv \sum_{n=1}^{N} \hat{\boldsymbol{I}}_n$ being the total collective spin for an ensemble of N particles with equal or different spins. $\langle \hat{\boldsymbol{J}}^2\rangle = 0$ indicates perfect squeezing where the N spins are in the MBSs. $\langle \hat{\boldsymbol{J}}^2\rangle(\bar{s})^{-1}$ gives an upper bound on the number of spins unentangled with others, where $\bar{s}$ is the average spin per particle.[5,9] States of the spin ensemble can be grouped into multiplets, i.e., irreducible invariant subspaces of the total spin. A multiplet $\{|J,M,\alpha_J^k\rangle, M = -J,\ldots,J\}$ is denoted in short as $\{J,\alpha_J^k\}$, where α_J^k is a general index for distinguishing the set of orthogonal $(2J+1)$-dimensional multiplets. The aim is to transfer population from all multiplets to those singlets with $J = 0$.

Our scheme is motivated by the discovery of an identity

$$\frac{\langle J+1,-J+1,\alpha_{J+1}|(\hat{j}_A^+ - \hat{j}_B^+)|J,-J,\alpha_J\rangle}{\langle J,-J,\alpha_J|(\hat{j}_A^+ - \hat{j}_B^+)|J+1,-J-1,\alpha_{J+1}\rangle^*} = -[(J+1)(2J+1)]^{-\frac{1}{2}}, \tag{1}$$

where $\hat{\boldsymbol{j}}_A$ is the collective spin of an arbitrary subset of the ensemble and $\hat{\boldsymbol{j}}_B \equiv \hat{\boldsymbol{J}} - \hat{\boldsymbol{j}}_A$ [Fig. 1(a)]. Moreover, $\langle J',M',\alpha_{J'}|\hat{j}_A^+ - \hat{j}_B^+|J,-J,\alpha_J\rangle = 0$ for $|J'-J| > 1$ or $M' \neq -J+1$. Under the condition that each multiplet is initialized on the spin coherent state $|J,-J,\alpha_J\rangle$, application of the $\hat{j}_A^+ - \hat{j}_B^+$ operator tends to transfer populations from multiplets of larger dimension to multiplets of smaller dimension [Fig. 1(c)].

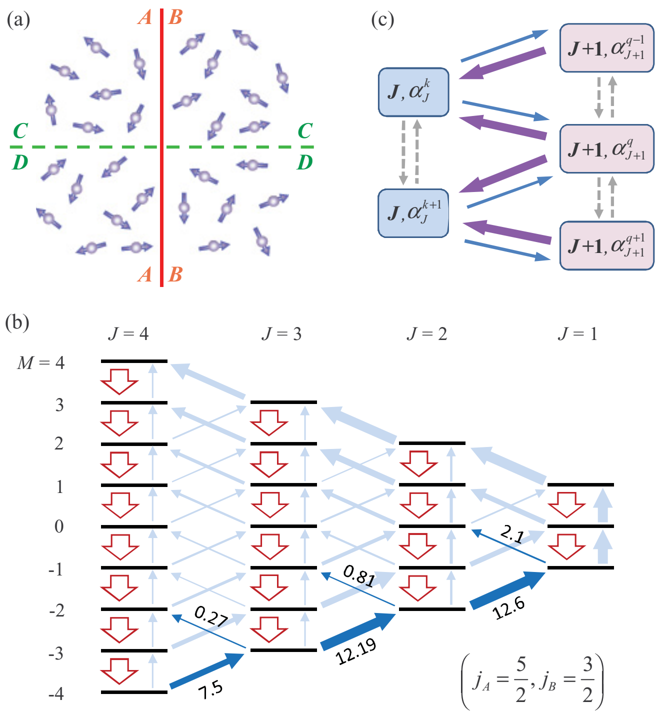

FIG. 1. (Color online) (a) A spin ensemble partitioned into subsets A and B (C and D) by the solid (dashed) line. (b) An example of how the operator $\hat{j}_A^+ - \hat{j}_B^+$ couples various basis states $|J,M,j_A,j_B\rangle$ (solid arrows). The absolute value squared of the transition matrix elements are indicated with the thickness of the arrows. The transitions related to the spin coherent states $|J,-J\rangle$ are highlighted. Hollow vertical arrows show the coupling by $\hat{J}^-$. (c) Schematics of the population transfer rates between multiplets under the condition that each multiplet is initialized on the spin coherent state $|J,-J\rangle$ when the $\hat{j}_A^+ - \hat{j}_B^+$ operator is applied. The transfer rate of $\{J,\alpha_J^k\} \leftarrow \{J,\alpha_J^{k'}\}$ is identical to the rate of the backward transfer $\{J,\alpha_J^{k'}\} \rightarrow \{J,\alpha_J^k\}$. The rate of $\{J+1,\alpha_{J+1}^q\} \rightarrow \{J,\alpha_J^k\}$ is by a factor of $(J+1)(2J+1)$ larger than that of the backward transfer $\{J,\alpha_J^k\} \rightarrow \{J+1,\alpha_{J+1}^q\}$.

Squeezing of collective spin uncertainties can thus be realized by dynamic spin polarization with the lowering operator $\hat{J}^-$ and raising operators of the form $\hat{j}_A^+ - \hat{j}_B^+$. Consider the use of two such operators $\hat{j}_A^+ - \hat{j}_B^+$ and $\hat{j}_C^+ - \hat{j}_D^+$, where C and D constitute a different bipartition of the ensemble [Fig. 1(a)]. The Hilbert space can be divided into independent subspaces according to the quantum numbers $\{j_{A\cap D}, j_{B\cap D}, j_{B\cap C}, j_{A\cap C}\}$, conserved by the raising/lowering operations, and their values determine the number of $(2J+1)$-dimensional multiplets $n(J)$. If $\hat{J}^-$ is applied more frequently, such that the system is in spin coherent states every time $\hat{j}_A^+ - \hat{j}_B^+$ or $\hat{j}_C^+ - \hat{j}_D^+$ is applied, we find the steady state in each subspace: $\rho = \sum_J f(J) \sum_{k=1}^{n(J)} |J,-J,\alpha_J^k\rangle\langle J,-J,\alpha_J^k|$, where $f(J) = (J+1)(2J+1)f(J+1)$. Most subspaces contain at least one MBS,[15] and $n(J) \leqslant n(0)(2J+1)$. Thus, in the steady state, MBSs are occupied with a population $n(0)f(0) \geqslant [\sum_J g(J)]^{-1} = 0.20$, and the variance $\langle \hat{\boldsymbol{J}}^2\rangle \leqslant [\sum_J g(J)]^{-1}\sum_J J(J+1)g(J) = 2.44$, where $g(J) \equiv (2J+1)[\prod_{i=0}^{J-1}(i+1)(2i+1)]^{-1}$.

Dynamic spin polarization by the collective raising and lowering operators can be described by Lindblad terms in the master equation $\dot{\rho} = -\frac{1}{2}\sum_{m=1}^{3}(\hat{L}_m^\dagger \hat{L}_m \rho + \rho \hat{L}_m^\dagger \hat{L}_m - 2\hat{L}_m \rho \hat{L}_m^\dagger)$, where $\hat{L}_1 \equiv \sqrt{\Lambda_h}\hat{J}^-$, $\hat{L}_2 \equiv \sqrt{\Lambda_o}(\hat{j}_A^+ - \hat{j}_B^+)$, and $\hat{L}_3 \equiv \sqrt{\Lambda_o}(\hat{j}_C^+ - \hat{j}_D^+)$. An example of the simulated squeezing dynamics is given in Fig. 2. The initial density matrix is the completely mixed one in the subspace with $\{j_{A\cap D} = 7/2,\ j_{B\cap D} = 7/2,\ j_{B\cap C} = 7/2,\ j_{A\cap C} = 7/2\}$, which corresponds to $\langle \hat{\boldsymbol{J}}^2\rangle = 63$. The polarization rates used are $\Lambda_h = 2000\Lambda_o$. After a pumping time of $t = 0.5\Lambda_o^{-1}$, MBSs are occupied with a population of 0.21 and $\langle \hat{\boldsymbol{J}}^2\rangle$ is substantially reduced to 2.44.

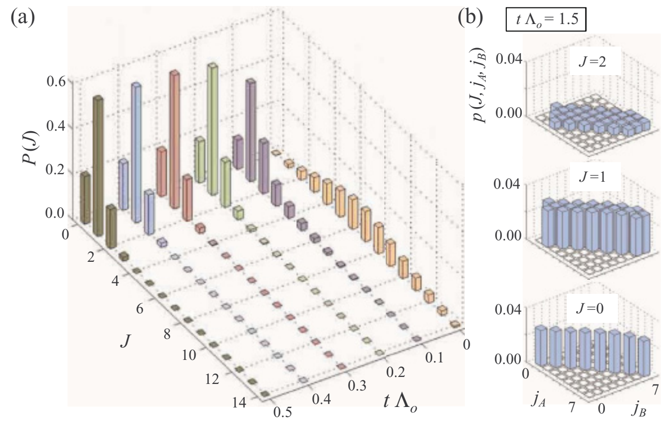

FIG. 2. (Color online) Simulation of squeezing control with the operators $\hat{J}^-$, $\hat{j}_A^+ - \hat{j}_B^+$, and $\hat{j}_C^+ - \hat{j}_D^+$ (see text). The system is initially in the completely mixed state in the subspace with $\{j_{A\cap D} = 7/2,\ j_{B\cap D} = 7/2,\ j_{B\cap C} = 7/2,\ j_{A\cap C} = 7/2\}$. The polarization rates are $\Lambda_h = 2000\Lambda_o$. (a) $P(J)$ gives the integrated probability of finding the system with total collective spin J at various time. At $t = 0.5\Lambda_o^{-1}$, MBSs are occupied with the population $P(0) = 0.21$, and $\langle \hat{\boldsymbol{J}}^2\rangle = 2.44$. (b) Population distribution on various MBSs, triplets and quintets at $t = 1.5\Lambda_o^{-1}$. The sum of populations on all other multiplets is less than 2%.

Conditions for efficient squeezing – The $\hat{J}^-$ and $\hat{j}_A^+ - \hat{j}_B^+$ operators are applied with the rates $\Lambda_h|\langle\psi_f|\hat{J}^-|\psi_i\rangle|^2$ and $\Lambda_o|\langle\psi_f|\hat{j}_A^+ - \hat{j}_B^+|\psi_i\rangle|^2$, respectively, and the scheme requires that the former rate shall always be larger. We note that $\langle J,M,j_A,j_B|(\hat{j}_A^- - \hat{j}_B^-)(\hat{j}_A^+ - \hat{j}_B^+)|J,M,j_A,j_B\rangle$ increases with the decrease of J and reaches the maximal value of $\sim(j_A + j_B)^2$ for small J, while $\langle J,M|\hat{J}^+\hat{J}^-|J,M\rangle \sim J^2$. Thus we find the requirement $\Lambda_h/\Lambda_o > (j_{A\cap D} + j_{B\cap D} + j_{B\cap C} + j_{A\cap C})^2$, the latter quantity $\sim 4Ns^2$ in an ensemble of N spin s particles.

Spin decoherence can compete with the squeezing, since it causes the population decay of MBSs with a rate $\sim N\gamma_n$, γ_n being the single-spin decoherence rate. When $\Lambda_h/\Lambda_o > 4Ns^2$ is satisfied, the inhomogeneous raising operator pumps population into MBSs with a rate $\Lambda_o|\langle\psi_f|\hat{j}_A^+ - \hat{j}_B^+|\psi_i\rangle|^2 \sim N\Lambda_o$. We thus anticipate that decoherence will have negligible effects on the squeezing under the low-loss condition $\frac{1}{4Ns^2}\Lambda_h > \Lambda_o \gg \gamma_n$. This is indeed confirmed by numerical simulations where single-spin decoherence terms are included in the master equation [Fig. 3(a)]. The effects of spin decoherence are almost invisible for $\gamma_n = 0.1\Lambda_o$.

Using inhomogeneous operators of a general form – A general collective operator $\sum_n c_n \hat{I}_n^+$ with inhomogeneous c_n can be expanded as $\sum_A \beta_A(\hat{J}^+ - 2\hat{j}_A^+)$, with A being various

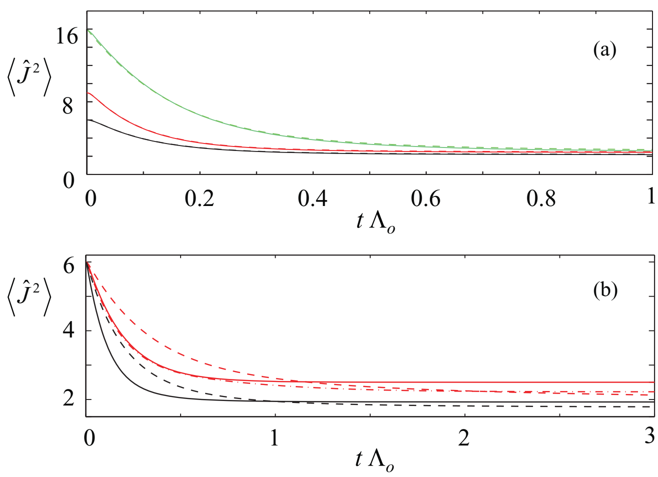

FIG. 3. (Color online) (a) Effects of spin decoherence. Black curves: 8 spin-$\frac{1}{2}$, $\Lambda_h/\Lambda_o = 40$; red (dark gray) curves: 12 spin-$\frac{1}{2}$, $\Lambda_h/\Lambda_o = 60$; green (light gray) curves: 8 spin-1, $\Lambda_h/\Lambda_o = 200$. $\gamma_n = 0$ for solid curves, and $\gamma_n = 0.1\Lambda_o$ for dashed curves. The spins are initially in the maximally mixed states. (b) Squeezing of 8 spin-$\frac{1}{2}$ particles using different collective operators. Red (dark gray) curves are squeezing by $\hat{J}^-$ and only one inhomogeneous operator: $\hat{j}_A^+ - \hat{j}_B^+$ (solid); $0.7(\hat{j}_A^+ - \hat{j}_B^+) + 0.3(\hat{j}_C^+ - \hat{j}_D^+)$ (dashed); $\sum_n \exp(i\frac{n}{4}\pi)\hat{I}_n^+$ (dash-dotted). $\Lambda_h/\Lambda_o = 50$. Black curves are squeezing by $\hat{J}^-$ and two inhomogeneous operators: $\hat{j}_A^+ - \hat{j}_B^+$ and $\hat{j}_C^+ - \hat{j}_D^+$ (solid); $0.7(\hat{j}_A^+ - \hat{j}_B^+) + 0.3(\hat{j}_C^+ - \hat{j}_D^+)$ and $0.7(\hat{j}_E^+ - \hat{j}_F^+) + 0.3(\hat{j}_G^+ - \hat{j}_H^+)$ (dashed). $\Lambda_h/\Lambda_o = 100$. $\{A|B\} = \{1234|5678\}$, $\{C|D\} = \{1278|3456\}$, $\{E|F\} = \{1357|2468\}$, and $\{G|H\} = \{1458|2376\}$ give different bipartition of the eight spins.

subsets of the ensemble. Since Eq. (1) holds for $-\hat{j}_A^+ + \hat{j}_B^+ \equiv \hat{J}^+ - 2\hat{j}_A^+$ with an arbitrary choice of A, we have this same identity for such a general linear combination. Squeezing to MBSs can thus be achieved using $\hat{J}^-$ and inhomogeneous collective raising operators of the general form. This allows great flexibility for implementation in various physical systems. The numerical simulations in Fig. 3(b) compare the squeezing efficiency by operators of various inhomogeneous form. Even better efficiency is achieved using operators with more inhomogeneity than $\hat{J}^+ - 2\hat{j}_A^+$.

Implementation in the nuclear spin bath of an electron – The homogeneous and inhomogeneous collective raising/lowering operations of nuclear spins are realized in the process of dynamic nuclear spin polarization (DNSP), a major tool for manipulation of nuclear spins.[16–29] We consider the hyperfine interaction $\hat{H}_0 = \sum_n |\psi(\boldsymbol{r}_n)|^2 \hat{\boldsymbol{I}}_n \cdot \overleftrightarrow{\boldsymbol{A}} \cdot \hat{\boldsymbol{S}}$, coupling the electron spin $\hat{\boldsymbol{S}}$ to peripheral lattice nuclear spins $\hat{\boldsymbol{I}}_n$. $\overleftrightarrow{\boldsymbol{A}}$ is the hyperfine coupling constant in tensor form, and the position dependence of coupling enters through the envelope function $\psi(\boldsymbol{r})$ of the electron only. $\hat{H}_0$ generally describes the hyperfine interaction of an electron or hole system in quantum dots or shallow donors formed in group IV or III-V materials.[30,31] In most DNSP schemes, $\hat{H}_0$ induces the electron-nuclear flip-flop in passing electron spin polarization to the nuclei and the energy cost is compensated by emission/absorption of phonons or photons.[19–23] These DNSP schemes are termed as the dc type hereafter. Alternatively, DNSP can also

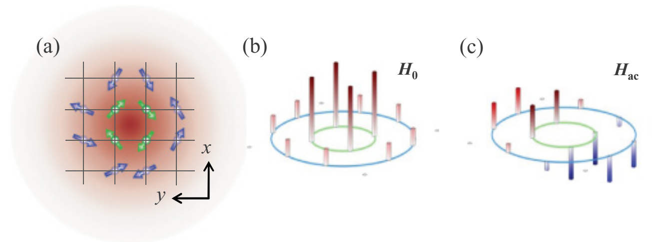

FIG. 4. (Color online) (a) Schematics of an electron in a nuclear spin bath. The first coordination shell has four nuclear spins in green (light gray) color and the second shell has eight nuclear spins in blue (dark gray) color. (b) dc hyperfine coupling coefficients at the various lattice sites. (c) ac hyperfine coupling coefficients with ac electric field in the x direction. The heights of the bar give the magnitude.

utilize the ac correction to the hyperfine coupling, $\hat{H}_{ac} = \sum_n (\boldsymbol{d}_\omega \cdot \nabla |\psi(\boldsymbol{r}_n)|^2) \cos(\omega t) \hat{\boldsymbol{I}}_n \cdot \overleftrightarrow{\boldsymbol{A}} \cdot \hat{\boldsymbol{S}}$, when an ac electric field induces an electron displacement $\boldsymbol{d}_\omega \cos(\omega t)$, with energy cost for electron-nuclear flip-flop directly supplied by the ac field.[27–29] Such a DNSP process is termed hereafter as the ac type.

For nuclear spins on the periphery of an electron, MBSs can be realized by combining dc and ac DNSP processes which polarize nuclear spins in opposite directions with the operators $\sum_n |\psi(\boldsymbol{r}_n)|^2 \hat{I}_n^-$ and $\sum_n \frac{\partial}{\partial\mu}|\psi(\boldsymbol{r}_n)|^2 \hat{I}_n^+$, respectively. Here μ is the direction of ac electric field. The lattice sites with equal electron density $|\psi(\boldsymbol{r})|^2$ are grouped into coordination shells. On each shell, $\sum_n |\psi(\boldsymbol{r}_n)|^2 \hat{I}_n^-$ is the homogeneous collective lowering operator while $\sum_n \frac{\partial}{\partial\mu}|\psi(\boldsymbol{r}_n)|^2 \hat{I}_n^+$ is of the inhomogeneous form. Figure 4(a) shows the schematic of an electron with a two-dimensional (2D) Gaussian envelope function. The 12 lattice nuclear spins form two coordination shells according to the dc hyperfine coupling strength. For the green shell with four lattice nuclear spins, $\hat{H}_{ac} = \tilde{a}\hat{S}^-(\hat{j}_A^+ - \hat{j}_B^+)e^{i\omega t}E_x + \tilde{a}\hat{S}^-(\hat{j}_C^+ - \hat{j}_D^+)e^{i\omega t}E_y + c.c.$, where $E_{x(y)}$ is the ac electric field in the x (y) direction. For the blue shell, the ac DNSP operators are of a more general inhomogeneous form, and numerical simulations in Fig. 3(b) have demonstrated efficient squeezing using one such operator (dashed red) or using two such operators, assuming an electric field alternating between the x and y directions (dashed black).

Under the influence of incoherent electron spin flips in the DNSP process, the large shell-to-shell difference in the dc hyperfine coupling strength causes loss of intershell coherence in a time scale much faster than the squeezing. Thus different coordination shells are independently squeezed toward MBSs. For a shell of N spin-s nuclei, the collective spin variance can be reduced by a factor of $\sim Ns^2$ under the low-loss condition. The improvement on the electron spin coherence thus depends on the constitution of the spin bath. Moreover, the steady-state population is on the extremal state $|J,-J\rangle$ for all multiplets, which corresponds to a different nuclear magnetic field $\langle a \sum_n \hat{I}_n^z \rangle$. Distillation of MBSs is thus possible on a shell with large a through projective measurement of the electron spin resonance.

Interaction between neighboring nuclear spins causes spin diffusion and spin dephasing, which can result in loss of MBSs.

The dipolar interaction between neighboring lattice sites is of the strength ~0.1 kHz. Nuclear spin diffusion by the $\hat{I}_n^+ \hat{I}_m^-$ coupling terms is efficiently suppressed when the shell-to-shell inhomogeneity in the hyperfine coupling is large. By the $\hat{I}_n^z \hat{I}_m^z$ term, the nuclear spins are subject to a dipolar magnetic field dependent on the configuration of their neighbors, which leads to dephasing with a rate of $\gamma_n \sim 1$ kHz. For nuclei with spin larger than 1/2, quadrupolar interaction by inhomogeneous electric field can also contribute to γ_n. To realize efficient squeezing, fast DNSP mechanisms are desired.

For optically controllable electron spin, e.g., in quantum dot or impurity in III-V semiconductors, fast dc DNSP can be realized by the hyperfine-mediated optical excitation of spin-forbidden excitonic transitions.[21,32] Assuming the electron Zeeman splitting $\omega_e \sim 0.2$ GHz, the intrinsic broadening of a charged exciton $\gamma_t \sim 0.2$ GHz, and an optical Rabi frequency $\Omega \sim 3$ GHz for the excitonic transition, we estimate the DNSP rate $\Lambda_h = \frac{a^2\Omega^2}{\omega_e^2\gamma_t} \sim 10$ MHz on a coordination shell with hyperfine coupling $a = 3$ MHz. For other electron-nuclear spin systems, fast dc DNSP may be realized through the bath-assisted electron-nuclear flip-flop in the presence of an efficient energy dissipation channel, e.g., an electron Fermi sea in nearby leads.[20]

ac DNSP is of the rate $\Lambda_o = \frac{\tilde{a}^2}{\gamma_s}$, where γ_s is the broadening of the electron spin resonance.[28] The magnitude of the ac hyperfine interaction $\tilde{a}$ depends on the strength of the ac electric field and the inhomogeneity of the electron envelop function. Giving the phosphorus donor in silicon as an example, we have the first several shells: $(A, 6.0, 6)$, $(B, 4.5, 12)$, $(C, 3.3, 4)$, $(D, 2.2, 12)$, and $(F, 1.7, 12)$, where the first letter is the label of the shell by convention, the second number is the hyperfine coupling strength in units of megahertz, and the third is the number of equivalent sites on the shell.[33] The distance between neighboring shells is in the order of ~0.1 nm. Thus, we estimate $\tilde{a} \sim$ MHz by a moderate displacement of the electron $d_\omega \sim 0.1$ nm. Assuming $\gamma_s \sim 0.1$ GHz, Λ_o can be of ~10 kHz on these shells. Thus we conclude that the low-loss condition can indeed be satisfied for nuclear spins on the periphery of a strongly confined electron.

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