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<td>Yao, W</td>
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Many-body singlets by dynamic spin polarization

Wang Yao
Department of Physics and Center of Theoretical and Computational Physics, The University of Hong Kong, Hong Kong, China
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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 $\sim 20\%$ 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.

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 in a mesoscopic ensemble of nuclear spins using dynamic nuclear spin polarization by an electron. The result is a spin system of extensive interest either as a noise source or as a superior information storage in quantum technology. MBSs can be a valuable resource of quantum entanglement for nuclear-spin quantum information processing.10–12 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,5,6 where the degree of squeezing is quantified by $\langle \hat{J}_z^2 \rangle = 0$ indicates perfect squeezing where the $N$ spins are in the MBSs. $\langle \hat{J}_z^2 \rangle$ 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 $\{|J,M,\alpha J\rangle, M = -J,\ldots,J\}$ is denoted in short as $[J,\alpha J]$, where $\alpha J$ 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

$$
\langle J + 1, -J + 1, \alpha J + 1 | (\hat{j}_A^+ - \hat{j}_B^+) | J, -J, \alpha J \rangle
$$

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

where $\hat{j}_A$ is the collective spin of an arbitrary subset of the ensemble and $\hat{j}_B = \hat{J}_A$ [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 $M' = -J + 1$. 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)].

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(c) Schematics of the population transfer rates between multiplets

Schematics of the population transfer rates between multiplets

(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 \( \hat{J}_z^A - \hat{J}_z^B \) 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 \( \hat{J}^- \).

Schematics of the population transfer rates between multiplets

Consider the use of two such operators \( \hat{J}_z^A - \hat{J}_z^B \) and \( \hat{J}^- \), 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 \( |J\rangle, |M\rangle, j_A, j_B\rangle \), conserved by the raising/lowering operations, and their values determine the number of \( (2J+1)\)-dimensional multiplets \( n(J) \). If \( J^- \) is applied more frequently, such that the system is in spin coherent states every time \( \hat{J}_z^A - \hat{J}_z^B \) or \( \hat{J}^- \) is applied, we find the steady state in each subspace: \( \rho = \sum_j \sum_{m=1}^{\Delta m} f(J) g(j) \langle J, -J, j_A, j_B\rangle \langle J, -J, j_A, j_B\rangle^\dagger \), where \( f(J) = (J + 1)/2(J + 1) \). Most subspaces contain at least one MBS, and \( n(J) \leq n(0)(2J + 1) \). Thus, in the steady state, MBSs are occupied with a population \( n(0) f(0) \geq \sum_j g(j) \sum_{m=1}^{\Delta m} f(J) g(j) \langle J, -J, j_A, j_B\rangle \langle J, -J, j_A, j_B\rangle^\dagger = 0.20 \), and the variance \( \langle \hat{J}^- \rangle^2 \leq \sum_j g(j) \sum_{m=1}^{\Delta m} f(J) g(j) \langle J, -J, j_A, j_B\rangle \langle J, -J, j_A, j_B\rangle^\dagger = 2.44 \), where \( g(j) \equiv (2J + 1)/2(J + 1) \).

Dynamic spin polarization by the collective raising and lowering operators can be described by Lindblad terms in the master equation

\[
\dot{\rho} = -\frac{i}{\hbar} [\hat{H}, \rho] + \sum_{m=1}^{\Delta m} \gamma_m (\hat{J}_m^+ \rho \hat{J}_m^- - \hat{J}_m^- \rho \hat{J}_m^+) + \frac{1}{2} (\hat{J}_m^- \rho \hat{J}_m^+ + \hat{J}_m^+ \rho \hat{J}_m^- - \hat{J}_m^- \rho \hat{J}_m^+ - \hat{J}_m^+ \rho \hat{J}_m^-).
\]

With \( \gamma_m = \Delta_m \gamma \), the average population on each MBS is given by

\[
\langle \hat{J}_m^- \rangle = \frac{1}{\Delta m} \sum_{m=1}^{\Delta m} \gamma_m.
\]

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 \( \hat{J}_z^A - \hat{J}_z^B \) 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 \( \hat{J}^- \).

FIG. 2. (Color online) Simulation of squeezing control with the operators \( \hat{J}^-, \hat{J}_z^A - \hat{J}_z^B \), and \( \hat{J}_z^C - \hat{J}_z^D \) (see text). The system is initially in the completely mixed state in the subspace with \( |J_{A/D} = 7/2, J_{B/C} = 7/2, J_{A/C} = 7/2\rangle \). The polarization rates are \( \Lambda_a = 2000\Lambda_o \). (a) \( P(J) \) gives the integrated probability of finding the system with total collective spin \( J \) at various times. At \( t = 0.5\Lambda_o^{-1}, \) MBSs are occupied with the population \( P(0) = 0.21 \), and \( \langle J^2 \rangle = 2.44 \). (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 \( \hat{J}^-, \hat{J}_z^A - \hat{J}_z^B \), and \( \hat{J}_z^C - \hat{J}_z^D \) operators are applied with the rates \( \Lambda_a (|\langle J/\sqrt{J} \rangle |^2 \right)^2 \) and \( \Lambda_a (|\langle J/\sqrt{J} \rangle |^2 \right)^2 \), respectively, and the scheme requires that the former rate shall always be larger. We note that \( \langle J, M, j_A, j_B | (\hat{J}_z^A - \hat{J}_z^B) J, M, j_A, j_B \rangle \) increases with the decrease of \( J \) and reaches the maximal value of \( \sim (J_a + j_B)^2 \) for small \( J \), while \( \langle J, M | \hat{J}_z^C - \hat{J}_z^D | J, M \rangle \sim J^2 \). Thus, we find the requirement \( \Lambda_a/\Lambda_o > (J_{A/D} + j_{B/C} + j_{A/C})^2 \), the latter quantity \( \sim N \gamma a^2 \) in an ensemble of \( N \) spin particles.

Spin decoherence can compete with the squeezing, since it causes the population decay of MBSs with a rate \( \sim \gamma_a \), \( \gamma_a \) being the single-spin decoherence rate. When \( \Lambda_a/\Lambda_o > 4N \gamma a^2 \) is satisfied, the inhomogeneous raising operator pumps population into MBSs with a rate \( \Lambda_a (|\langle J/\sqrt{J} \rangle |^2 \right)^2 \sim N \gamma a \). We thus anticipate that decoherence will have negligible effects on the squeezing under the low-loss condition \( 1/N \gamma a \approx \Lambda_a \gg \gamma a \). 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 \( \gamma a = 0.1 \Lambda_o \).

Using inhomogeneous operators of a general form – A general collective operator \( \sum_n c_n \hat{J}_m^\dagger \) with inhomogeneous \( c_n \) can be expanded as \( \sum_A \beta_A (\hat{J} - \hat{J}_A) \), with \( A \) being various.
FIG. 3. (Color online) (a) Effects of spin decoherence. Black curves: 8 spin-\(\frac{1}{2}\), \(\Lambda_0/\Lambda_n = 40\); red (dark gray) curves: 12 spin-\(\frac{1}{2}\), \(\Lambda_0/\Lambda_n = 60\); green (light gray) curves: 8 spin-1, \(\Lambda_0/\Lambda_n = 200\). \(\gamma_c = 0\) for solid curves, and \(\gamma_c = 0.1\Lambda_n\) 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); \(\sum_n \exp(i\pi n)\hat{J}_n^+\) (dash-dotted). \(\Lambda_0/\Lambda_n = 50\). Black curves are squeezing by \(\hat{J}_-\) and two inhomogeneous operators: \(\hat{J}_{n1}^+ - \hat{J}_{n2}^+\) and \(\hat{J}_{n3}^+ - \hat{J}_{n4}^+\) (solid); \(0.7(\hat{J}_{n1}^+ - \hat{J}_{n2}^+) + 0.3(\hat{J}_{n3}^+ - \hat{J}_{n4}^+)\) and \(0.7(\hat{J}_{n1}^+ - \hat{J}_{n2}^+) + 0.3(\hat{J}_{n3}^+ - \hat{J}_{n4}^+)\) (dashed). \(\Lambda_0/\Lambda_n = 100\). \(|A|B| = 1234[5678], |C|D| = 1278[3456], |E|F| = 1357[2468], |G|H| = 1458[2376]\) give different bipartition of the eight spins.

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.

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 MBSSs 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 (DNPS), a major tool for manipulation of nuclear spins.\(^{16-29}\) We consider the hyperfine interaction \(\hat{H}_0 = \sum_n \psi(r_n)\hat{I}_n \cdot \mathbf{A} \cdot \hat{S}\), coupling the electron spin \(\hat{S}\) to peripheral lattice nuclear spins \(\hat{I}_n, \mathbf{A}\) is the hyperfine coupling constant in tensor form, and the position dependence of coupling enters through the envelope function \(\psi(r)\) 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 DNPS 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 DNPS schemes are termed as the dc type hereafter. Alternatively, DNPS can also utilize the ac correction to the hyperfine coupling, \(\hat{H}_{ac} = \sum_n (d_{n,x} \partial_x - \psi(r_n)^2) \cos(\omega t) \hat{I}_n \cdot \mathbf{A} \cdot \hat{S}\), when an ac electric field induces an electron displacement \(d_{n,x} \cos(\omega t)\), with energy cost for electron-nuclear flip-flop directly supplied by the ac field.\(^{31-28}\) Such a DNPS process is termed hereafter as the ac type.

For nuclear spins on the periphery of an electron, MBSSs can be realized by combining dc and ac DNPS processes which polarize nuclear spins in opposite directions with the operators \(\sum_n \psi(r_n)^2 \hat{I}_n^-\) and \(\sum_n \frac{1}{\gamma_c} \psi(r_n)^2 \hat{I}_n^+\), respectively. Here \(\mu\) is the direction of ac electric field. The lattice sites with equal electron density \(\psi(r)^2\) are grouped into coordination shells. On each shell, \(\sum_n \psi(r_n)^2 \hat{I}_n^-\) is the homogeneous collective lowering operator while \(\sum_n \frac{1}{\gamma_c} \psi(r_n)^2 \hat{I}_n^+\) 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, \(\hat{H}_{ac} = \alpha \hat{S} \cdot (\hat{J}_A^+ - \hat{J}_B^+) \cos(\omega t) + \beta \hat{S} \cdot (\hat{J}_C^+ - \hat{J}_D^+) \cos(\omega t) + c.c.,\) where \(E_{\text{dc}}(y)\) is the ac electric field in the \(y\) direction. For the blue shell, the ac DNPS 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 DNPS 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 MBSSs. For a shell of \(N\) spin-\(\frac{1}{2}\) nuclei, the collective spin variance can be reduced by a factor of \(\sim N^2\) under the low-loss condition. The improvement on the electron spin coherence thus depends on the condition 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 \(\langle \mathbf{A} \rangle\). Distillation of MBSSs is thus possible on a shell with large \(\alpha\) 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 MBSSs.
The dipolar interaction between neighboring lattice sites is of the strength $\sim 0.1$ kHz. Nuclear spin diffusion by the $I^+_n I^-_n$ coupling terms is efficiently suppressed when the shell-to-shell inhomogeneity in the hyperfine coupling is large. By the $I^+_n I^-_n$ 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 $\gamma_n$. 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. 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 $\gamma_{dn} = \frac{2\Omega^2}{\gamma_t^2} \sim 10$ 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.  

15The states unconnected with MBS are $\sim 1\%$ of the entire Hilbert space if two raising operators of the form $\hat{I}^+ - \hat{I}^-$ are used, and can be reduced to $\sim 0.01\%$ if three such operators are used in the dynamic spin polarization.