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We present a scheme for achieving coherent spin squeezing of nuclear spin states in semiconductor quantum dots. The nuclear-polarization-dependence of the electron spin resonance generates a unitary evolution that drives nuclear spins into a collective entangled state. The polarization dependence of the resonance generates an area-preserving, "twisting" dynamics that squeezes and stretches the nuclear spin Wigner distribution without the need for nuclear spin flips. Our estimates of squeezing times indicate that the entanglement threshold can be reached in current experiments.

Entanglement generation and detection are two of the most sought-after goals in the field of quantum control. Besides offering a means to probe some of the most peculiar and fundamental aspects of quantum mechanics, entanglement in many-body systems can be used as a tool to reduce fluctuations below the standard quantum limit[1]. Recently, squeezing of the collective spin state of many atoms[2] was achieved using atom-light or atom-atom interactions[3–6], allowing unprecedented precision of measurements in atomic ensembles[7]. Similarly, future progress in spin-based information processing hinges on our ability to find ways of precisely controlling the dynamics of nuclear spins in nanoscale solid-state devices[8, 9]. In particular, electron spin coherence times[10, 11] can be improved by driving the nuclear spin bath into reduced-entropy "narrowed" states[12–17], as seen in experiments[18]. Furthermore, with quantum control, a nuclear spin bath can be turned into a resource, serving as a long-lived quantum memory[19–21], or a medium for high-precision magnetic field sensing[22].

Here we describe a coherent spin squeezing mechanism for gate-defined quantum dots[23], see Fig. 1a. With suitable modification, our approach can also be applied to other systems which can be approximately described by a central-spin model. We consider a single electron in a quantum dot, in contact with a large group of nuclear spins, $\{\hat{\mathbf{I}}_n\}$. The electron and nuclear spins are coupled by the hyperfine interaction $H_{\mathrm{HF}} = \sum_n A_n \hat{\mathbf{S}} \cdot \hat{\mathbf{I}}_n$, where $\hat{\mathbf{S}}$ is the electron spin, and each coupling constant A_n is proportional to the local electron density at the position of nucleus n. The electron spin is driven by an applied RF field with frequency close to the electron spin resonance (ESR) in the presence of an externally applied magnetic field. Because the electron spin evolves rapidly on the timescale of nuclear spin dynamics, the nuclear spins are subjected to an effective hyperfine field (the "Knight field") produced by the time-averaged electron spin polarization. Nuclear spin squeezing results from the dependence of the electronic hyperfine field on the detuning from the ESR condition, which in turn depends on the nuclear polarization, see Fig. 1b.

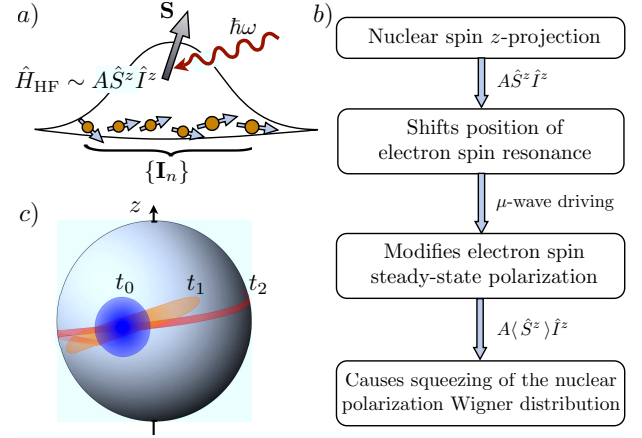In a system comprised of many spins $\{\hat{\mathbf{I}}_n\}$, such as a

FIG. 1: Nuclear spin squeezing in a quantum dot. a) An electron in a quantum dot, with the electron spin $\mathbf{S}$ coupled to a large group of nuclear spins $\{\mathbf{I}_n\}$. Electron spin resonance is excited by microwave radiation applied in the presence of an external magnetic field. b) Flowchart describing the squeezing mechanism. c) Schematic depiction of twisting dynamics on the Bloch sphere, shown in a rotating frame where the mean polarization is stationary. We focus on dynamics between the initial time t_0 and an intermediate time t_1, during which the phase space (Wigner) distribution is squeezed within a small, flat region of the Bloch sphere, see Eqs.(7) and (8). At longer times, indicated by t_2, the distribution extends around the Bloch sphere.

quantum dot or an atomic ensemble, the collective total spin $\hat{\mathbf{I}} = \sum_n \hat{\mathbf{I}}_n$ is a quantum mechanical angular-momentum variable. Because different vector components of $\hat{\mathbf{I}}$ do not commute, they are subject to the Heisenberg uncertainty relations

$$\Delta I^y \Delta I^z \geq \frac{\hbar}{2} |\langle \hat{I}^x \rangle|, \qquad (1)$$

and its cyclic permutations[2, 24] (without loss of generality, we focus on the spin 1/2 case). Squeezing is achieved by reducing fluctuations in one spin component below the "standard quantum limit," $\Delta I^\alpha = \sqrt{\frac{1}{2}\hbar |\langle \hat{I}^x \rangle|}$. As we discuss below, depending on the application, a variety of criteria can be used for identifying "useful" levels of squeezing (see Refs.[2, 7, 24]).

Typically, the inequality in Eq.(1) is far from saturated

in quantum dots under ambient conditions. In equilibrium, the typical nuclear polarization and the uncertainties ΔI^α are all of order $\sqrt{N}$ (see e.g. Ref.[25]), where $N \sim 10^6$ is the number of nuclear spins in the quantum dot. Here we consider an initial state prepared by polarizing nuclear spins to a fraction p of the maximal polarization, and then rotating this polarization into the equatorial plane of the Bloch sphere such that the mean spin points along x, $\langle \hat{I}^x \rangle = pN\hbar/2$. Experimentally, nuclear spin polarizations of up to 40% have been reported for electrically-driven systems[26], and up to 60% in optically-pumped systems[27].

To quantify the degree of squeezing for arbitrary polarization, Wineland et al.[7] introduced the parameter $\xi = \sqrt{N}\Delta I^z/|\langle \hat{I}^x \rangle| \approx 2\Delta I^z/(p\hbar\sqrt{N})$, which characterizes the angular resolution of the squeezed state relative to that of an uncorrelated product state. Different physical effects are described by three different conditions:

$$1)\ \xi < 1/p, \quad 2)\ \xi < 1/\sqrt{p}, \quad 3)\ \xi < 1. \qquad (2)$$

Condition 1 is sufficient to achieve ESR narrowing in a quantum dot in a large magnetic field, where the electron Zeeman energy is sensitive to the Overhauser shift, proportional to ΔI^z. Condition 2 indicates that the standard quantum limit has been surpassed. Finally, the most stringent condition ($\xi < 1$) is sufficient to imply entanglement of the constituent spin-1/2 particles (c.f. Ref. [24]) and enhanced resolution for atomic clocks.

Below we demonstrate that with realistic values of p, all three conditions (2) can be met. Compared with the ideal case $p = 1$, we find that incomplete initial polarization, $p < 1$, and fluctuations in the prepared value of p should not hamper efforts to obtain useful squeezing (all three conditions are close for p of order 1).

In Ref.[28], Fernholz et al. achieved squeezing of the internal spin variables of individual composite particles, Cesium atoms with total spin $F = 4$. In contrast, here we describe a mechanism for squeezing the collective spin state of a large ensemble of spatially-distributed spins which can in principle be selectively addressed.

To describe the coupled electron-nuclear spin dynamics, we model the system with the microscopic Hamiltonian (below we set $\hbar = 1$)

$$H = \omega_Z \hat{S}^z + \omega_0 \hat{I}^z + A\hat{I}^z\hat{S}^z + \frac{A}{2}(\hat{I}^+\hat{S}^- + \hat{I}^-\hat{S}^+) + H_{\text{el}}, \ (3)$$

where ω_Z is the electron Zeeman energy in the magnetic field, ω_0 is the nuclear Larmor frequency, and H_{el} describes the driving of the electron spin and its coupling to an environment, which leads to fast dephasing and relaxation. For simplicity, here we consider a single species of nuclear spin, and take all hyperfine coupling constants to be equal, $A_n = A$. The latter condition amounts to the assumption that electron density is approximately constant inside the dot, and zero outside. In this case,

the electron spin couples directly to the *total* nuclear spin $\hat{\mathbf{I}} = \sum_n \hat{\mathbf{I}}_n$, with the square of the total nuclear spin, $\hat{I}^2$, conserved by the dynamics. The effects of non-uniform couplings will be discussed at the end.

We begin by writing the Heisenberg equation of motion for the total nuclear spin operator $\hat{\mathbf{I}}$, $d\hat{\mathbf{I}}/dt = i[\hat{\mathbf{I}}, H]$:

$$\frac{d\hat{\mathbf{I}}}{dt} = \mathbf{b} \times \hat{\mathbf{I}}, \quad \mathbf{b} = \omega_0\mathbf{z} + A\hat{\mathbf{S}}. \qquad (4)$$

In the motional-narrowing regime where electron dynamics are fast compared to the nuclear spin evolution, we use Eq. (3) to adiabatically eliminate the electron spin from the right hand side of Eq. (4). Due to the large mismatch between the electron and nuclear Zeeman energies, $\omega_Z/\omega_0 \gg 1$, averaging over fast oscillations of the electron allows us to replace $\hat{\mathbf{S}}$ by an operator-valued semiclassical mean polarization $S^z(\hat{I}^z)$ which depends on the nuclear polarization $\hat{I}^z$ through the Overhauser shift of the ESR frequency, c.f. Ref.[29]:

$$S^z = \frac{1}{2}\frac{(\delta\omega - A\hat{I}^z)^2 + \gamma^2}{(\delta\omega - A\hat{I}^z)^2 + \tilde{\gamma}^2}, \quad \tilde{\gamma}^2 = \gamma^2 + \frac{\gamma}{\Gamma_1}\Omega^2, \quad (5)$$

where $\delta\omega$ is the detuning between the driving frequency and ω_Z, Ω is the driving strength, $\gamma \equiv 1/T_2$ is the electron spin dephasing rate, Γ_1 is the electron spin relaxation rate. Linearizing Eq. (5) in $A\hat{I}^z$ around the optimal detuning $\delta\omega_* = \tilde{\gamma}/\sqrt{3}$ where S^z is most sensitive to nuclear-polarization-dependent frequency shifts, see Fig.2, and substituting into Eq. (4), we obtain an effective Hamiltonian for the collective nuclear spin:

$$H \approx \omega_0\hat{I}^z + \frac{1}{2}\lambda(\hat{I}^z)^2, \quad \lambda = A \left.\frac{\partial S^z}{\partial \hat{I}^z}\right|_{\substack{\hat{I}^z=0 \\ \delta\omega=\delta\omega_*}}, \qquad (6)$$

with $\hat{I}^2 = I(I+1)$, $I \le N/2$, conserved by the dynamics. Note that here we have absorbed a constant shift into the nuclear Larmor frequency ω_0. The Hamiltonian in Eq.(6) is of the canonical squeezing Hamiltonian form[2]. It is interesting to note that while the end results, Eqs.(5) and (6), have a similar form to those obtained for spin squeezing in atomic systems[4–6], the underlying microscopic mechanisms are quite different.

To illustrate the squeezing induced by the Hamiltonian in Eq. (6), and to facillitate further investigation of the effects of classical fluctuations in the initial state, as well as the effect of time-dependent electron spin fluctuations around the steady state S^z, we now analyze the evolution of the nuclear spin Wigner distribution. For a large initial polarization p, where $I = pN/2$, and for short to intermediate times $t_0 \lesssim t \lesssim t_1$ (see Fig.1c), the "uncertainty region" associated with the nuclear state is small on the scale of the total spin and we can consider evolution in a locally flat patch of the Bloch sphere. Here the operators $\hat{I}^y$ and $\hat{I}^z$ approximately obey canonical commutation relations, and the initial nuclear spin state
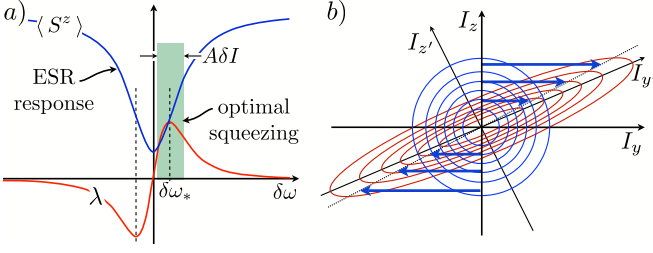
FIG. 2: a) Time-averaged electron spin polarization S^z, Eq.(5), and squeezing strength λ, Eq.(6), versus RF detuning $\delta\omega$ from the ESR frequency. The average electron spin polarization depends on $\hat{I}^z$ through the dependence of the detuning on the Overhauser shift, as indicated by the shaded region. b) Contour plot representation of the Wigner distribution of a large collective spin on a locally-flat patch of the Bloch sphere, in the rotating frame where $\omega_0 = 0$. The mean spin points along x. Before squeezing, the Wigner distribution is isotropic (blue). After squeezing, the Wigner distribution, Eq.(7), is squeezed along an axis z', and stretched along an orthogonal axis y' (red).

(polarized along x) is described by an isotropic 2D Gaussian Wigner distribution with width set by the initial transverse fluctuations, $\Delta I \equiv \Delta I_0^{y,z}$.

Semiclassically, Eq.(6) induces precession of the total spin vector about the z axis with a *polarization-dependent* Larmor frequency $\eta = \partial H/\partial I^z = \omega_0 + \lambda I^z$. Correspondingly, the Gaussian Wigner distribution evolves as

$$f_t(I^y, I^z) = \mathcal{A}\exp\left(-\frac{(I^z)^2 + (I^y + I\lambda t I^z)^2}{2\Delta I^2}\right), \quad (7)$$

where without loss of generality we set $\omega_0 = 0$. The initial (isotropic) and evolved (squeezed) distributions are shown in Fig.2b.

The quadratic form in the exponential in Eq.(7) is diagonalized in a suitably chosen orthonormal basis y', z' [35]. As shown in Fig.2b, stretching in one direction (y') is accompanied by squeezing in the perpendicular direction (z'), such that the phase space volume of the Wigner distribution is exactly preserved if fluctuations of the electron spin are ignored. For times $t \gtrsim t_S = (|\lambda| I)^{-1}$, the uncertainty $\widetilde{\Delta I}$ of the squeezed component decreases as

$$\widetilde{\Delta I}(t) \approx \Delta I \frac{t_S}{t}, \quad t_S \approx \frac{16\Gamma_1 \tilde{\gamma}^3}{3\sqrt{3}IA^2\gamma\Omega^2}. \quad (8)$$

Squeezing proceeds until long times when the phase space distribution begins to extend around the Bloch sphere, see Fig.1c. The curvature of the Bloch sphere imposes a limit on the maximum achievable squeezing[2].

For an order-of-magnitude estimate of the squeezing time, we set $\Omega = \Gamma_1 = \frac{1}{5}\gamma$. This choice selects the regime of moderately strong electron spin dephasing where the resonance is broader than the minimum value $\gamma = \frac{1}{2}\Gamma_1$. In this practically relevant regime, the motional-averaging approximation can be safely applied. Taking the 'intrinsic' width of the resonance to

be twice larger than the typical Overhauser field fluctuations, $\gamma \approx A\sqrt{N}$, we obtain

$$t_{S,\text{min}} \approx 20\frac{\sqrt{N}}{IA}. \quad (9)$$

Using a typical value of the hyperfine coupling for GaAs, $A \approx 0.1\,\mu\text{s}^{-1}$, we obtain $t_{S,\text{min}} \approx 200\,\mu s(\sqrt{N}/I)$. The estimate for $t_{S,\text{min}}$ can be improved slightly by optimizing the expression for t_S in Eq. (8) with respect to driving power Ω. The fast relaxation rate $\Gamma_1 \sim A\sqrt{N}$ can be achieved by working in a regime of efficient electron spin exchange with the reservoirs in the leads. We see that the squeezing time is inversely proportional to the initial length of the nuclear spin vector, i.e. the degree of nuclear polarization before squeezing.

To derive the squeezing time t_S in Eq.(9), a coherent nuclear spin state with $\Delta I = \sqrt{I/2}$ was used. As discussed above, however, when classical uncertainty in the nuclear spin state is included, the initial width of the Wigner distribution is given by $\Delta I = \sqrt{N}/2$. Given that the width $\widetilde{\Delta I}$ of the squeezed component decays as $1/t$, see Eq.(8), the effect of the classical transverse fluctuations is simply to increase the time required to reach a desired level of fluctuations by an order-one factor $\sqrt{N/2I} = \sqrt{1/p}$.

Besides fluctuations in the transverse components of the initial polarization, the DNP process used to prepare the initial nuclear spin state will also leave behind uncertainty in the length I of the net spin (typically with a scale much smaller than I itself). However, because the rate of angular precession depends only on the z-component of the total spin, Eq.(7), sections of the phase space distribution with constant I^z but varying Bloch sphere radii will rigidly precess. Therefore fluctuations in the initial polarization I do not pose a significant threat to squeezing.

In addition to uncertainty in the initial nuclear spin state, we must also consider the effect of time-dependent fluctuations of the electron spin about its mean-field value S^z, Eq.(5). The mean-field approximation to Eq.(4) applies in the motion-averaged limit when the electron spin evolves quickly on the time scale of the nuclear spin dynamics, and hence the contribution of time-dependent electron spin fluctuations is small. The residual effect of such fluctuations is to add a diffusive component to the nuclear-polarization-dependent precession induced by the time-averaged electron spin. It can be shown that the diffusivity κ associated with this phase diffusion approximately goes as $\kappa \sim 1/\Gamma$, where $\Gamma \sim W, \Gamma_1$ is the characteristic rate of electron spin dynamics[35]. Thus phase diffusion is indeed suppressed by motional-averaging. At long times, the competition between coherent twisting dynamics, which squeezes fluctuations as $1/t$, and phase diffusion, which tends to increase fluctuations as $t^{1/2}$, slows down squeezing to $\widetilde{\Delta I} \sim t^{-1/2}$, but does not prevent it.

The results above are based on a semiclassical mean-field treatment of Eq.(4) supplemented by electron-spin-fluctuation-driven phase diffusion. This intuitive approach is quantitatively supported by a lengthier calculation based on the full density matrix of the combined electron-nuclear system, to be presented elsewhere. The more powerful density-matrix approach can also be used to study squeezing in the coherent driving regime of electron spin dynamics where large correlations can build up between the electron and nuclear spins.

Is the approximation of uniform hyperfine coupling justified? The hyperfine interaction in a quantum dot is strong near the center, where electron density is high, and weak at the edges. Notably, the atomic systems[4] display a similar level of spatial inhomogeneity, since there is a full modulation of coupling between zero and maximum coupling in a standing wave of light. The observation of robust squeezing in atomic clouds of size comparable to the wavelength of light indicates that spatial variation of the coupling does not compromise the effect.

For $p \approx 20\%$, squeezing sets in after $t_S \sim 2$ μs, and fluctuations are suppressed by a factor of 10 within approximately 20 μs (neglecting phase diffusion). Due to classical fluctuations in the initial state, the first $\sqrt{1/p}$-fold ($1/p$-fold) squeezing goes toward reaching the standard quantum limit (entanglement threshold). Taking into account phase diffusion, we arrive at timescales that are at least 10 times shorter than typical nuclear decoherence times (recently measured to be ~ 1 ms in vertical double quantum dots[31]). It should thus be possible to squeeze the nuclear spin state faster than it decoheres due to dipole-dipole interactions, etc.

All elements required for achieving and demonstrating squeezing, i.e. dynamical nuclear polarization[26, 27], controlled rotations using NMR pulses[30, 31], and coherent control of single electron spins[23, 32, 33], have been realized. In particular, we note that in Ref. [23] electron spin resonance was achieved by excitation using microwave magnetic fields, with driving amplitudes comparable to the random nuclear field acting on the electron spin, $A\delta I$. The corresponding transition rates are of the order of 10 MHz. In order to reach the motional averaging regime, the electron spin relaxation rate, Γ_1, must be comparable to the transition rate W, which can be easily accomplished by allowing cotunneling to the electron reservoirs next to the dot. The degree of squeezing ξ, see Eq. (2), can be ascertained by the combination of two separate measurements on the final state: (1) an NMR pulse[30, 31] which rotates the minimum uncertainty axis (z') into the z-axis followed by an electron spin dephasing measurement[34] of ΔI^z and (2) an NMR pulse which rotates the net polarization $\langle I^x \rangle$ into to the z-axis, followed by a measurement of the average nuclear field along z.

In summary, squeezed and entangled states of nuclear spins in quantum dots driven near the ESR are generated by unitary evolution which does not involve incoherent spin flips. Our estimates of the timescales for various effects that compete with squeezing indicate that squeezing is feasible and can be realized with current capabilities. Such schemes may potentially open the door to unprecedented levels of quantum control over collective degrees of freedom in nanoscale systems with mesoscopic numbers ($N \sim 10^4$ to 10^6) of nuclear spins.

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[35] see Supplementary Material for the details of the analysis