Spin Squeezing in a Quadrupolar Nuclei NMR System

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We have produced and characterized spin-squeezed states at a temperature of 26 °C in a nuclear magnetic resonance quadrupolar system. The experiment was carried out on ^{133}Cs nuclei of spin $I = 7/2$ in a sample of lyotropic liquid crystal. The source of spin squeezing was identified as the interaction between the quadrupole moment of the nuclei and the electric field gradients present within the molecules. We use the spin angular momentum representation to describe formally the nonlinear operators that produce the spin squeezing on a Hilbert space of dimension $2I + 1 = 8$. The quantitative and qualitative characterization of this spin-squeezing phenomenon is expressed by a squeezing parameter and squeezing angle developed for the two-mode Bose-Einstein condensate system, as well as by the Wigner quasiprobability distribution function. The generality of the present experimental scheme points to potential applications in solid-state physics.

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The role of nonclassical states in atomic physics and optics has been extensively investigated in the last two decades [1–3]. A great deal of this interest is due to the collective behavior of atoms in the so-called spin-squeezed state. Quantum states of this kind were first studied in relation to the production of atom-atom entanglement by means of a nonlinear spin-spin interaction [4,5]—the one-axis twisting (OAT) model. Since then, many theoretical developments [[6–8], [9–17]] have led to various applications in the domain of quantum information processing, such as proposals for spin squeezing to be exploited in quantum entanglement [8,18–20], in quantum metrology, where the idea of a fundamental noise limit set by quantum-mechanical laws is explored [15,21], and in atom chip based investigations [22].

In parallel, several experimental observations [21–36] of spin-squeezed states have been achieved mainly in many-body atom-light interaction scenarios involving collective spin in an ensemble of N atoms. Particularly, important experiments of this type were performed with a cesium atom ensemble [24,27]. In these, the hyperfine ground states $F = 3$ and $F = 4$ were exploited in Hilbert spaces of dimension $(2F + 1) = 7$ and 9. Such an approach matches a low dimensionality regime, which may be extended to nuclear quadrupolar systems and qubits in solid-state physics [17,35]. On this behalf, the present experimental development is a way to bring atomic and solid state physics closer to the general purpose of producing, in the near future, a quantum computer. Particularly, solid state systems could be used efficiently on a quantum computer as data storage devices. To achieve this, the formalism and procedures presented here are of general applicability to any physical system that explores the concepts of quadrupolar nuclei in solid state NMR.

The route followed in this Letter to achieve spin squeezing is to investigate a quadrupolar NMR system, with nuclear spin $7/2$, and characterize the squeezed states through a squeezing parameter and squeezing angle, as well as by the Wigner quasiprobability distribution function. In our system, it is the physical nature of the quadrupolar nuclei that leads to the spin-squeezed states. Essentially, the inherent electric field gradients in a quadrupolar system evolving in the NMR setup produce the signal that indicates spin squeezing. This is basically different, for example, from the liquid state NMR simulation in [37], where spin $1/2$ constituents with a scalar coupling are refocused by appropriate pulses. Here, we use a liquid-crystal platform that, within the NMR framework, allows control of the quantum-level couplings of nuclei and the collective behavior of molecules, so as to compensate for thermal vibrations in order to execute an efficient experimental implementation. Our experimental tasks can be carried out by invoking the concept of coherent states, one- and two-mode BEC-like systems, protocols of classical bifurcation developed in [38–40] and protocols of quantum information processing [41–46].

To be more specific, part of the theoretical framework that we use comes from studies of the two-mode BEC system [11–14]. In these, in order to reach a spin-squeezed state, an OAT model is implemented. The associated Hamiltonian is also known as the two-site Bose-Hubbard model and the particle description can be mapped on to an angular momentum description, by using the Schwinger representation. More specifically, it is possible to map a Hamiltonian described in terms of creation $\hat{a}_i^\dagger$ and annihilation $\hat{a}_i$ operators, satisfying the commutation relations $[\hat{a}_i, \hat{a}_j^\dagger] = \delta_{ij}$ $(i, j = 1, 2)$, on to an angular momentum algebraic realization [1,3,6,11,13,14,47], with operators

that obey the commutation relations $[\hat{I}_i, \hat{I}_j] = i\hbar\epsilon_{ijk}\hat{I}_k$ $(i, j, k = x, y, z)$. Here we must make the following distinction: in the NMR framework, the operator $\hat{I}$ refers to the internal nuclear spin angular momentum. A related discussion about squeezing has been carried out in a quantum dots system, where it arises from the nuclear-electron interaction [16]. The common formalism used in these investigations is the collective spin representation. We are interested in the internal nuclei spin, and the relevance of those results to our case is that it is possible to explore an equivalent formalism in the context of NMR quadrupolar nuclei. Here , the OAT model comes from an analogous term given by a quadratic nuclear spin angular momentum operator along the z axis in the NMR quadrupolar Hamiltonian. In this light, we report here the experimental observation of the dynamics of spin squeezing in a NMR quadrupolar system in a lyotropic liquid crystal sample held at a temperature of 26 °C in order to keep the liquid crystalline phase stable.

A spin-squeezed state can be reached by means of an interaction that depends nonlinearly on Cartesian orbital angular momentum components, for which $\hat{I}_{\mathbf{n}} = \mathbf{n}\cdot(\hat{I}_x, \hat{I}_y, \hat{I}_z)$, perpendicular to the mean spin $\langle\hat{I}\rangle$. This procedure is used with the OAT model [5,24] or the two-axis countertwisting model [5,27]. The OAT model, as we approach it here, is applied to the description of internal nuclei spin [17,27], and is characterized by a quadratic term in the z component of the orbital angular momentum $\kappa\hat{I}_z^2$, where κ is the strength of this interaction. We start from a coherent spin state $|i, i\rangle_x$, which corresponds to a symmetric quasiprobability distribution on the spherical phase space around the x axis, and then, after a transformation, it appears squeezed in the y-z plane in a rotated y'-z' basis [27].

To quantify the degree of squeezing, we adopt the criteria of Ref. [11–13], developed from Ref. [5], so that the parameter of squeezing is defined by $\xi = (\Delta\hat{I}_{\mathbf{n}})_{\min}/\sqrt{I/2}$, where $(\Delta\hat{I}_{\mathbf{n}})_{\min}$ represents the smallest variance of a spin component $\hat{I}_{\mathbf{n}}$ normal to the mean spin $\langle\hat{I}\rangle$; specifically,

$$\xi = \frac{\sqrt{\frac{1}{2}C - \frac{1}{2}\sqrt{A^2 + B^2}}}{\sqrt{I/2}} < 1, \quad \text{and} \tag{1}$$

$$\alpha_\xi = \frac{1}{2}\arctan(B/A), \tag{2}$$

where I is the nuclear spin ($I = 7/2$ for ^{133}Cs), $A = \langle\hat{I}_z^2 - \hat{I}_y^2\rangle$, $B = \langle\hat{I}_z\hat{I}_y + \hat{I}_y\hat{I}_z\rangle$, and $C = \langle\hat{I}_z^2 + \hat{I}_y^2\rangle$ are appropriate combinations of spin components $\hat{I}_z$ and $\hat{I}_y$, chosen with respect to the orientation of $\mathbf{n}$, in this case along x. α_ξ is the *squeezing* angle, which is a geometrical property that characterizes the orientation of the squeezing [12,13]. The expression in Eq. (1) differs from a more general expression [see Eq. (2) in [18]]. Whereas Eq. (1) is capable of detecting squeezing, it fails to detect entanglement, as Eq. (2) in [18] does. The issue of entanglement and other quantum correlations in NMR experiments have been addressed in various publications [48,49], but this is not the purpose of the present Letter, since our system is composed of noninteracting quadrupole nuclei in a liquid crystal (see below).

The NMR formalism for any quadrupolar system is based on a nuclear spin $I > 1/2$ and $m = I, I-1, \ldots, -I$ as its quantization rule. The laboratory frame representation is used to set up the Hamiltonian, with basically four kinds of contribution: the first is the Zeeman term, due to the interaction of the nuclear magnetic moment $-\hbar\gamma(\hat{I}_x, \hat{I}_y, \hat{I}_z)$ with a strong static magnetic field B_0 aligned in the z direction. This first contribution is expressed by $-\hbar\gamma B_0\hat{I}_z$, where γ is the gyromagnetic ratio of the nuclear species and $\hbar$ is the reduced Planck's constant. The second is the effective quadrupolar term [50], which arises from the interaction of the quadrupole moment (Q) of the nuclei with the electric field gradient internally present in the sample ($V_{\alpha,\beta}$). It is expressed as $eQ/(4I(2I-1))(V_{zz}(3\hat{I}_z^2 - \hat{I}^2) + (V_{xx} - V_{yy})(\hat{I}_x^2 - \hat{I}_y^2))$, and the electric field gradient satisfies the Laplace's equation $\sum_\alpha V_{\alpha\alpha} = 0$. In an ordered nuclear system with axial symmetry the condition $|V_{xx}| \approx |V_{yy}| \ll |V_{zz}|$ is satisfied. This allows us to simplify the second contribution to the form, $eQV_{zz}/(4I(2I-1))(3\hat{I}_z^2 - \hat{I}^2)$. This term will be the generator of the nuclear spin-squeezed state. The third is the radio-frequency (rf) term, due to the interaction of the nuclear magnetic moment with a time-dependent external magnetic field perturbation $\mathbf{B}_1(t) = B_1(\cos(\omega_{\rm rf}t + \phi), \sin(\omega_{\rm rf}t + \phi), 0)$ perpendicular to the strong static magnetic field B_0. Finally, the fourth term is due to contributions from the environment ($\mathcal{H}_{\rm env}$) and represents effective weak interactions with other nuclear species, electrons, field fluctuations, and so on [49]. In a rotating frame representation, the total NMR Hamiltonian is thus described by

$$\mathcal{H}_{\rm NMR} = -\hbar(\omega_L - \omega_{\rm rf})\hat{I}_z + \hbar\frac{\omega_Q}{6}(3\hat{I}_z^2 - \hat{I}^2) + \hbar\omega_1(\hat{I}_x\cos\phi + \hat{I}_y\sin\phi) + \mathcal{H}'_{\rm env}, \tag{3}$$

where $\omega_Q = 3eQV_{zz}/(2I(2I-1)\hbar)$ is the quadrupolar coupling, $\omega_1 = \gamma B_1$ the rf strength, and $\omega_L = \gamma B_0$ the Larmor frequency of the nuclear species. The coupling parameters of our physical quadrupolar system satisfy the inequality $|\omega_Q| \ll |\omega_L|$.

Let us set $\omega_{\rm rf} = \omega_L$ and $\phi = 0$ to transform the NMR Hamiltonian into Hamiltonian (1) of Ref. [13], which corresponds to the one-axis twisting model (when $\omega_1 = 0$) of spin squeezing, after dropping the constant term $-(\hbar\omega_Q/6)\hat{I}^2$. The Hamiltonian for the experimental setup is

$$\mathcal{H}^s_{\rm NMR} = \frac{\hbar\omega_Q}{2}\hat{I}_z^2. \tag{4}$$

Before going into some of the details, we offer the following intuitive account of what happens in this

experiment: Our sample is subject to a strong magnetic field such that the nuclear spins are polarized. By an appropriate choice of radio frequency fields we then reach an initial state, which will be transformed by Hamiltonian (4); owing to these changes, the nuclear spin precesses and produces a characteristic signal, from which we can, by using quantum state tomography, obtain the associated density matrix. The free evolution of the state under (4) produces the squeezing; an extended experimental description can be found in the Supplemental Material [51].

The NMR experimental setup employs cesium nuclei (^{133}Cs) with quadrupolar spin system $I = 7/2$, making the dimension of the Hilbert space $d = 2I + 1 = 8$. A lyotropic liquid crystal sample was prepared with 42.5 wt% cesium-pentadecafluorooctanoate (Cs-PFO) and 57.5 wt% deuterated water (D_2O). The experiment was carried out in a Varian 500 MHz spectrometer with a 5 mm probe for liquids. The Larmor and quadrupolar frequencies of ^{133}Cs nuclei are, respectively, $\omega_L/2\pi = 65.598$ MHz and $\omega_Q/2\pi = 7.58$ kHz. The length of the π pulse was calibrated at 26 μs. The transverse and longitudinal relaxation times are $T_2 \approx 30$ ms and $T_1 \approx 650$ ms, respectively. The recycle delay time is 3.5 s.

To describe a quantum state in a NMR system, we use the density operator at thermal equilibrium, in which populations are represented by the Boltzman-Gibbs distribution. The density operator is denoted by $\rho = (1/\mathcal{Z})\hat{1} + \epsilon\rho_0$, where $\epsilon = \omega_L\hbar/k_BT\mathcal{Z}$ is the polarization value ($\sim 10^{-6}$), k_B is Boltzmann's constant, T the room temperature (in our case 26 °C), $\mathcal{Z}$ the partition function, and $\rho_0 = \hat{I}_z$ the deviation density matrix [49]. The deviation density matrix is transformed by a method adapted from the strongly modulating pulse technique, in order to achieve a nuclear spin coherent state NSCS, the equivalent of the so-called pseudopure state [39,49] in a NMR qubit system $|\zeta(\theta,\varphi)\rangle = \sum_{m=-I}^{I} \binom{2I}{I+m}^{1/2} \cos(\theta/2)^{I-m} \sin(\theta/2)^{I+m} e^{-i(I+m)\varphi} |I, m\rangle$, where $|I, m\rangle$ are eigenstates of $\hat{I}_z$ with eigenvalue m [39,40]. The density operator changes to $\rho = ((1-\epsilon)/\mathcal{Z})\hat{1} + \epsilon\Delta\rho$, such that $\Delta\rho \equiv |\zeta(\theta,\varphi)\rangle\langle\zeta(\theta,\varphi)|$ is the deviation density operator, for any $0 \le \theta \le \pi$ and $0 \le \varphi \le 2\pi$ [39,51]. Specifically, by choosing $\theta_0 = \pi/2$ and $\varphi_0 = \pi$, we implement the initial quantum state, denoted by $|\zeta(\pi/2,\pi)\rangle$, such that it is suitable to implement the spin-squeezing protocol [11–13]. Following the nuclear spin-squeezing protocol, the $|\zeta(\pi/2,\pi)\rangle$ evolves by application of the operator defined by $\exp[-i\mathcal{H}^s_{\mathrm{NMR}}\tau_k/\hbar]$, in 44 time steps of $\tau_{k+1} - \tau_k = 3$ μs, with $k = 0, 1, \ldots, 44$, so that the discrete $\tau_k \in [0, 132\ \mu\mathrm{s}]$. The readout at each time step of the evolved initial quantum state is performed by quantum state tomography [51].

The efficiency of the implementation of $|\zeta(\pi/2,\pi)\rangle$ and its time evolution can be estimated by means of the Wigner quasiprobability distribution function, which is applied to the experimental deviation density matrix of the tomographed quantum state $|\zeta(\theta_k,\varphi_k)\rangle$. Thus, by definition, we have [67–69]

$$W(\theta,\varphi) = \sqrt{\frac{2I+1}{4\pi}} \sum_{K=0}^{2I} \sum_{Q=-K}^{K} \varrho_{KQ}(\theta,\varphi;\theta_k,\varphi_k) Y_{KQ}(\theta,\varphi), \tag{5}$$

for $\theta \in [0,\pi]$ and $\varphi \in [0,2\pi]$, where $\varrho_{KQ}(\theta,\varphi;\theta_k,\varphi_k) = \mathrm{Tr}_{\theta,\varphi}\{|\zeta(\theta_k,\varphi_k)\rangle\langle\zeta(\theta_k,\varphi_k)|\hat{T}^{\dagger}_{KQ}\}$, $\hat{T}_{KQ}$ being the spherical tensor operators (or irreducible tensor operators [67–69]) and $Y_{KQ}(\theta,\varphi)$ the spherical harmonics.

Next, we analyze the 4th tomographed quantum state with the Wigner formalism for the theoretical prediction [top of Fig. 1(a)] and experimental results [top of Fig. 1(b)]. We can observe the qualitative signature of the spin-squeezing phenomenon, depicting the compression of the probability distribution in the direction denoted by the arrows contained in the y-z plane. Analogously, we show the same phase space at the 17th time step, where the squeezing effect is attenuated, but at the 40th time step the squeezing effect is recovered. Finally, at the 44th time step, we can observe from the shape of the probability distribution that the squeezing cycle has been completed and a new one has started. From that set of figures, we observe a correspondence between a description following the theoretical development of the matrix operators and a free evolution of the nuclear spins, monitored by quantum state tomography.

We also investigate the dynamics of the squeezing parameter (ξ), computed by Eq. (1), finding that the system evolves under spin squeezing as shown in Fig. 1(c), where a periodic behavior is seen, which may also arise in collapse and revival phenomena [70–72]. Theoretical results (black solid line) are generated by transforming a theoretical initial NSCS $|\zeta(\pi/2,\pi)\rangle$ under the evolution operator that depends on the Hamiltonian $\mathcal{H}^s_{\mathrm{NMR}}$, using numerical calculations for any $\tau \in [0, 132\ \mu\mathrm{s}]$. Experimental results (dark green dots) are computed at 44 steps from the tomographed deviation density matrix, such that $\tau_k \in [0, 132\ \mu\mathrm{s}]$. Note that the evolution of the spin squeezing has a periodic behavior, which depends on the inverse of the quadrupolar frequency $\nu_Q^{-1} = 132\ \mu s$, and that it matches the choice of time window used to monitor the spin system. The error bars for each experimental dot represents an error of $\sim 10\%$ (see [51] for a detailed discussion). Similarly, we analyze the evolution of the squeezing angle, Eq. (2), which is plotted over the same time window in Fig. 1(d). The theoretical prediction (black solid line) starts at a value of $\pi/4$ and falls monotonically to $-\pi/4$, coinciding with the end of the periodical behavior of the spin system, which then starts a new cycle. Precisely at this time there is a discontinuity in the squeezing angle, which switches from the negative value back to the positive value. Experimental data (dark green dots) follow closely the solid line computed by the theoretical procedure.

To conclude, we have accomplished, in a liquid-crystal NMR quadrupolar nuclear spin system, an experimental characterization of a spin-squeezing process by adopting a one-axis twistinglike model [5,24] in a regime of low dimensionality. The theoretical framework established in

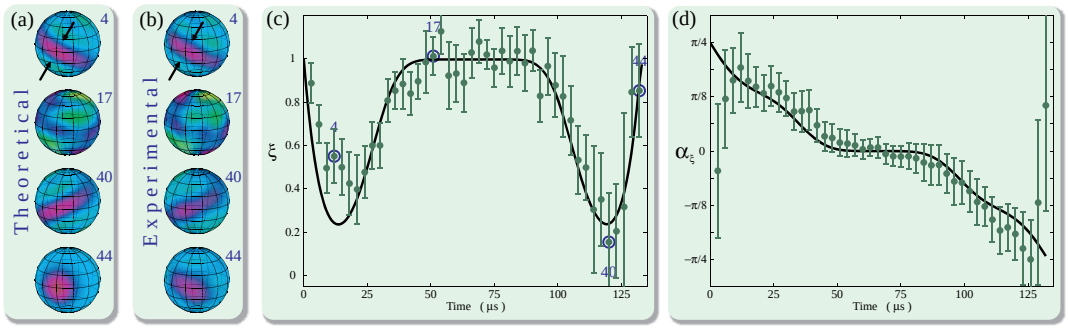

FIG. 1 (color online). Experimental results of spin squeezing under the one-axis twisting model. The NSCS $|\zeta(\pi/2,\pi)\rangle$ was evolved under the Hamiltonian $\mathcal{H}^{s}_{\mathrm{NMR}}$. The dynamics of spins was monitored in time steps of $\delta\tau = 3\ \mu$s over the time interval $[0, \nu_Q^{-1}]$. (a) Theoretical Wigner quasiprobability distribution function computed from a density matrix at time steps τ_k, where $k = 4, 17, 40$, and 44. (b) Experimental Wigner quasiprobability distribution function calculated from the tomographed density matrix. (c) Dynamics of the squeezing parameter (ξ) from theoretical prediction (black solid line) and experimental results (dark green dots). The error bars are discussed in [51]. Blue open circles correspond to analysis of the Wigner quasiprobability distribution function of (a) and (b). (d) Dynamics of the squeezing angle (α_ξ).

the atomic physics formalism for symmetrical traps of two-mode BECs is applied to the case of nuclear spin systems, with respect to their algebraic structure and commutation rules. The spin-squeezing process is generated in a natural way in NMR experiments by quadrupolar nuclei. We observed this process by monitoring the free evolution of a nuclear quadrupolar spin system, exploiting inherent physical properties of the nuclei, such as the quadrupole moment and electric field gradient interaction, and not by the interaction between spins. We use an ensemble of Cs-PFO molecules in which single quadrupolar nuclei are used to attain our objective, so that just one nucleus "mimics" the effect of seven particles in a two-mode BEC. This is one of the strong points of our investigation, because in most cases it is challenging to manipulate few particles coherently. Also, we conjecture that this low effective number of particles is what allows us to see explicitly the collapse and revival features appearing in Fig. 1(c). Although our experimental setup runs at room temperature, the pure part of the density matrix, which is proportional to ϵ, holds the quantum behavior of the nuclear spin system and indicates this squeezing phenomenon. In our view, the main challenge is to keep many particles correlated efficiently for a long time as it happens in cold atoms. From this point of view, we are showing that there are some molecular systems that share features similar to those of ultracold atoms and, as a benchmark, we present a study on squeezed states. Also, there is a growing interest in the physics of the two-site Bose-Hubbard model, which is, in terms of angular momentum, formally identical to the system investigated here, so that we expect our approach to be useful for the discussion of such phenomena as tunneling, Bose-Einstein condensates in periodic potentials, the Josephson effect, and topological excitations.

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