Entanglement and Extreme Spin Squeezing

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For any mean value of a Cartesian component of a spin vector we identify the smallest possible uncertainty in any of the orthogonal components. The corresponding states are optimal for spectroscopy and atomic clocks. We show that the results for different spin J can be used to identify entanglement and to quantify the depth of entanglement in systems with many particles. With the procedure developed in this Letter, collective spin measurements on an ensemble of particles can be used as an experimental proof of multiparticle entanglement.

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The commutator relation for angular momentum operators leads to Heisenberg's uncertainty relation for the Cartesian components

$$\Delta J_x \cdot \Delta J_y \ge |\langle J_z \rangle|/2. \tag{1}$$

Without violating Heisenberg's uncertainty relation, it is possible to redistribute the uncertainty unevenly between J_x and J_y , so that a measurement of either J_x or J_y becomes more precise than the standard quantum limit $\sqrt{|\langle J_z \rangle|/2}$. States with this property are called spin squeezed states in analogy with the squeezed states of a harmonic oscillator.

A two-level atom can be represented as a spin- $\frac{1}{2}$ system, and, in experiments on a large number of atoms N which all start out in the same initial state and which are all subject to the same Hamiltonian, one can conveniently express the collective observables of the gas by means of an effective spin J = N/2, so that, e.g., the difference in the number of atoms populating the two internal states is given by the J_z component. The state with all atoms in the "spin up" internal state is equivalent to the $|J_z = J\rangle$ eigenstate of the macroscopic spin. If one measures the x component of the spin of a single atom, it is projected onto the internal superposition states $(|\downarrow\rangle \pm |\uparrow\rangle)/\sqrt{2}$ with equal probability. The value of the total J_x is given by the difference in the number of atoms in the two states, and it fluctuates binomially with a variance J/2, which matches precisely the equality sign in (1) with $\Delta J_x = \Delta J_y$.

The states and the amount of spin squeezing produced by applying Hamiltonians J_x^2 and $J_x^2 - J_y^2$ to an initial $|J_z = J\rangle$ state were studied [1], and the squeezed states, which satisfy the equality sign in (1), the so-called minimumuncertainty-product states, have been identified [2–4]. Interaction of atoms with broadband squeezed light [5,6] is an experimentally verified means to produce spin squeezing [7]. Spin squeezing based on ideas from quantum computing was recently suggested [8], and recent ideas for neutral atom spin squeezing based on quantum nondemolition detection of the atomic spin state [9] and on the collisional interactions between atoms [10,11] give reason to believe that sizable spin squeezing. In Ramsey-type spectroscopy on a collection of twolevel atoms, a signal proportional to the length of the mean collective spin pointing, say, along the z axis is recorded and the noise is given by the uncertainty of one of the orthogonal components. Wineland *et al.* have shown [12] that the frequency resolution in spectroscopy on N twolevel atoms contains the factor

$$\xi = \frac{\sqrt{2J}\,\Delta J_x}{|\langle J_z \rangle|},\tag{2}$$

which is reduced by spin squeezing. In this way, spin squeezing becomes an important ingredient in high precision spectroscopy and in atomic clocks, which are at present limited precisely by the fundamental spin noise [13]. Furthermore, spin squeezing is an important ingredient in quantum information, because the ensuing quantum entanglement leads to possibilities, e.g., for atomic teleportation [14].

In the derivation of Eq. (2) it is assumed that no other sources of noise are present. The states which minimize the quantity ξ are obtained in the limit $\langle J_z \rangle$, $\Delta J_x \to 0$, where any other source of noise will, however, deteriorate the spectroscopic resolution. In practice, the ideal states for spectroscopy are therefore states which minimize the noise ΔJ_x for a given (not too small) value of $\langle J_z \rangle$. In this Letter we identify this minimum, i.e., we optimize the signal-to-noise ratio by identifying the states with minimum quantum noise for a given value of the signal. For photons a similar analysis has been performed in Ref. [15]. Having identified the minimum of ΔJ_x we use this information to derive an experimental criterion for entanglement. A measurement of two collective operators for a collection of atoms produces a sufficient criterion for entanglement which can even quantify the "depth" of entanglement, i.e., the minimum number of particles forming multiparticle entangled states in the sample.

To get a lower limit on ΔJ_x as a function of $\langle J_z \rangle$, one can use the inequality $\langle J_x^2 \rangle + \langle J_y^2 \rangle + \langle J_z \rangle^2 \leq J(J+1)$ which, together with Heisenberg's uncertainty relation (1), yields the limit

$$(\Delta J_x)^2 \geq \frac{1}{2} \left[J(J+1) - \langle J_z \rangle^2 - \sqrt{[J(J+1) - \langle J_z \rangle^2] - \langle J_z \rangle^2} \right].$$
(3)

This does not present a tight minimum for $\operatorname{Var}(J_x)$, but for large J and $\langle J_z \rangle \approx J$ it is close to the actual minimum found by the numerical approaches discussed below. For low values of $\langle J_z \rangle$ it differs by a factor of 2. The precise analysis of the minimum becomes quite different for integer spins and for half-integer spins, and we shall deal with them separately.

For integer spins our calculations show that the state which minimizes $\operatorname{Var}(J_x)$ for a given $\langle J_z \rangle$ has vanishing $\langle J_x \rangle$ and $\langle J_y \rangle$, so that it is also a minimum of $\langle J_x^2 \rangle$. Accordingly, these states can be found by minimizing $\mu \langle J_z \rangle + \langle J_x^2 \rangle$, where μ is a Lagrange multiplier, ensuring the value of $\langle J_z \rangle$. For J values up to several hundred, it is straightforward to numerically determine the minimum, by determining the smallest eigenvalue of the operator $\mu J_z + J_x^2$ for a wide range of values of μ . By determining $\langle J_z \rangle$ and $\langle J_x^2 \rangle$ in the corresponding eigenstates, one finds exactly the minimum value of $\operatorname{Var}(J_x) = \langle J_x^2 \rangle$ for the particular value of $\langle J_z \rangle$. The results for different values of J are shown in Fig. 1. For J = 1 it is possible to diagonalize $\mu J_z + J_x^2$ analytically, and we get $\operatorname{Var}(J_x)_{\min} = (1 - \sqrt{1 - \langle J_z \rangle^2})/2$. For half-integer spins, it is not true that the state

For half-integer spins, it is not true that the state minimizing $\langle J_x^2 \rangle$ also minimizes $\operatorname{Var}(J_x)$ at a given value of $\langle J_z \rangle$. The reason is that, for half-integer spins, the operator J_x^2 has eigenvalues $\frac{1}{4}, \frac{9}{4}, \ldots$, and its expectation



FIG. 1. Maximal squeezing for different values of J. The curves starting at the origin represent the minimum obtainable variance as a function of the mean spin. Starting from above, the curves represent J = 1/2, 1, 3/2, 2, 3, 4, 5, and 10. The dotted curve for J = 1/2 is the limit identified in Ref. [11]. The solid curves are obtained by diagonalization of the operator $\mu J_z + J_x^2$. The dashed curve represents the position of a bifurcation in the solution for half-integer spins. To the right of this curve the diagonalization may be applied. To the left of the curve the minimum is found by a variational calculation (dash-dotted curve for J = 3/2).

value will thus always exceed $\frac{1}{4}$. The variance of J_x , however, can come arbitrarily close to zero, if the system approaches a J_x eigenstate. Consider, for instance, a $J = \frac{1}{2}$ particle, where all (pure) states can be obtained as a simple rotation of the spin-up state. In this case the components perpendicular to the mean spin are never squeezed; their variances are both $\frac{1}{4}$. But if we compute the variance of J_x and the mean value of J_z , one finds that they obey the relation, $Var(J_x)_{min} = \langle J_z \rangle^2$, where both sides approach zero when the state approaches either of the two $J_x = \pm \frac{1}{2}$ eigenstates. In that case, of course, the mean spin also has a component along the x direction. The state is spin squeezed in the sense of the relation (1), but not in the sense where one deals explicitly with a spin component perpendicular to the mean spin vector.

For large half-integer J it is more difficult to find the most squeezed states. The reason is that the problem cannot be formulated as a linear quantum mechanicals problem like the diagonalization of an operator containing a Lagrange multiplier term, which we used for integer spins. To compute a variance, we have to determine the square of a mean value which is an expression to fourth order in wave function amplitudes. It is easy, however, to implement a Monte Carlo variational calculation which minimizes $\mu \langle J_z \rangle + \operatorname{Var}(J_x)$, by randomly modifying the amplitudes of a state vector as long as the variational functional is reduced. Like above, the Lagrange multiplier term is used to adjust the mean value of J_z , so that for each value of μ the identified state vector minimizes Var (J_x) for the given value of $\langle J_z \rangle$. When applied to larger halfinteger values of J this method shows that large values of $\langle J_z \rangle$ are accompanied by vanishing mean values of J_x and J_{y} , and the solution thus coincides with the one found by the diagonalization method. But, for a critical value of $\langle J_z \rangle$, the solution bifurcates, and two states with opposite nonvanishing mean values of J_x have the smallest variance (see Fig. 2). These states approach the two $J_x = \pm \frac{1}{2}$ states in the limit of small $\langle J_z \rangle$. Because of the noise in the



FIG. 2. Bifurcation in the solution for J = 3/2. The points represent the mean value of the spin in the maximally squeezed states. The points are obtained by a Monte Carlo variational calculation which minimizes $\mu \langle J_z \rangle + \text{Var}(J_x)$. Above $\langle J_z \rangle \approx 1.32$ there is a unique solution with $\langle J_x \rangle = 0$. Below $\langle J_z \rangle \approx 1.32$ the solution bifurcates, and $\langle J_x \rangle$ approaches $\pm 1/2$.

simulation, the Monte Carlo method is not efficient for a precise determination of the critical point of the bifurcation for large values of J. Before the bifurcation the state is the eigenstate corresponding to the lowest eigenvalue of the operator $\mu J_z + J_x^2$, and after the bifurcation the state is a superposition of the different eigenvectors with amplitudes on states with higher eigenvalues. We can therefore determine the position of the bifurcation from the properties of the eigenvectors, and, for different values of J < 100, we find that the bifurcation happens in the interval

$$0.83 < \frac{\langle J_z \rangle}{J} < 0.88\,,\tag{4}$$

except for the special case J = 1/2 where $\langle J_z \rangle = J$ at the bifurcation. If we do not break the $\pm J_x$ symmetry the variance from this point flattens out to the value $\frac{1}{4}$, but, in either of the states with the broken symmetry, the variance decreases towards zero when smaller values of $\langle J_z \rangle$ and nonvanishing values of $\langle J_x \rangle$ are considered. The position of the bifurcation is plotted in Fig. 1. To the right of the dashed line the minimum may be found by diagonalization. To the left of the curve the variational approach has to be applied for half-integer spins.

It is the nonlinearity of the problem that leads to the bifurcation and symmetry breaking of the solution. Classical approximations to many-body quantum problems often show such bifurcations associated with phase transitions in the problem, e.g., lasing. It is interesting that a similar phenomenon appears here, in the study of a single quantum system in a (very) low-dimensional Hilbert space. We emphazise that we are not discussing an extension of quantum theory to include nonlinear terms, we are simply identifying the quantum states that minimize a variance, and this is a nonlinear problem.

Since we have identified the maximally squeezed states as eigenstates of the operator $\mu J_z + J_x^2$, even without having explicit expressions for these eigenstates, we can devise a method to produce them. This method works only for integer spin, and for half-integer spin which are squeezed to values of the mean spin exceeding the value at the bifurcation (4). The system is first prepared in the $|J_z = J\rangle$ eigenstate, and one switches on a Hamiltonian $H(t) = \omega J_z + \chi(t) J_x^2$, where $\chi(t)$ increases very slowly from the value zero and where $\omega < 0$. If the state of the spin follows this Hamiltonian adiabatically, it evolves through the minimum energy eigenstates of the instantaneous H(t), which is precisely the family of states identified by the above diagonalization procedure. The adiabatic process may be difficult to perform in physical systems of interest, and for practical purposes it is relevant to point out that the straightforward application of a Hamiltonian $H = J_x^2$ also leads to spin squeezing [1] and, in the regime with large $|\langle J_z \rangle|$, the squeezing resulting from this Hamiltonian is actually close to the optimum. The Hamiltonian $H = J_r^2 - J_y^2$, also discussed in Ref. [1], leads to similar squeezing for large $|\langle J_z \rangle|$, and it follows the optimum for a larger range of parameters.

We have identified the minimum variance of J_x given the value of $\langle J_z \rangle$. Any measurement of these two quantities can be plotted as a point in Fig. 1, and this point must lie on or above the curve for the relevant J. We note that the curves depend on J, and in the chosen units, large spins can be more squeezed than small spins. This implies that the collective spin variables $\vec{J} = \sum_i \vec{J}_i$ for several spin-J particles can be more squeezed than the individual spins themselves. We will now show that this requires the state of the spins to be an entangled state. It is already known [11] that, for spin-1/2 particles, reduction of the parameter ξ below unity for the collective spin implies entanglement of the spins. We generalize this property to arbitrary spins.

A separable (nonentangled) state of N spin-J particles is defined as a weighted sum of products of density matrices with positive weights p_k [16,17]:

$$\rho = \sum_{k} p_k \rho_1^{(k)} \otimes \rho_2^{(k)} \dots \otimes \rho_N^{(k)}, \qquad (5)$$

where $\rho_i^{(k)}$ is the density matrix of the *i*th particle in the k^{th} term of the weighted sum. The variance of J_x in such a state obeys the inequality

$$\operatorname{Var}(J_{x}) \geq \sum_{k} p_{k} \sum_{i=1}^{N} (\Delta J_{x}^{2})_{i}^{(k)}$$
$$\geq \sum_{k} p_{k} \sum_{i=1}^{N} JF_{J}(\langle J_{z} \rangle_{i}^{(k)} / J), \qquad (6)$$

where the function $F_J(\cdot)$ is the minimum variance of J_x divided by J for the spin-J particle, i.e., the curves plotted in Fig. 1, and $\langle J_z \rangle_i^{(k)}$ is the mean value of J_z of the i^{th} particle in the k^{th} realization in the sum (5).

As it appears from Fig. 1, all the curves $F_J(\cdot)$ are convex. We can prove this property for integer spins, and for half-integer spins in the range of large $|\langle J_z \rangle|$, by considering the production of the states by adiabatic passage from the $|J_z = J\rangle$ eigenstate. The positive factor in front of the J_x^2 component in the Hamiltonian $\omega J_z + \chi(t)J_x^2$ is gradually increased, and the rate of change of $\langle J_x^2 \rangle$ and $\langle J_z \rangle$ at any given time are given by Ehrenfest's theorem:

$$\frac{d}{dt} \langle J_x^2 \rangle = \frac{1}{i\hbar} \langle [J_x^2, \omega J_z] \rangle,$$

$$\frac{d}{dt} \langle J_z \rangle = \frac{1}{i\hbar} \langle [J_z, \chi(t) J_x^2] \rangle.$$
(7)

The mean values on the right-hand side should be evaluated in the maximally spin squeezed state, i.e., they are not known explicitly. But, we observe that they contain the same operator, and the ratio between the two rates of changes is therefore simply $-\omega/\chi(t)$. This implies that, along the family of maximally squeezed states, the relative change of $\langle J_x^2 \rangle$ and $\langle J_z \rangle$, i.e., the slope of the curve $F_J(\cdot)$, is monotonically increasing [since $\chi(t)$ is an increasing function of time and $\omega < 0$]. It follows that the second derivative of the function $F_J(\cdot)$ is positive, i.e., the function is convex.

From the convexity, it follows that the functions $F_J(\cdot)$ obey Jensen's inequality, which states that any linear combination of $F_J(a_i)$'s with positive coefficients is larger than or equal to the function F_J evaluated on the linear combination of the arguments. It therefore follows that, in any separable state,

$$\operatorname{Var}(J_{x}) \geq \sum_{k} p_{k} NJF_{J} \left(\frac{1}{NJ} \sum_{i=1}^{N} \langle J_{z} \rangle_{i}^{(k)} \right)$$
$$\geq NJF_{J} \left(\sum_{k} p_{k} \frac{1}{NJ} \sum_{i=1}^{N} \langle J_{z} \rangle_{i}^{(k)} \right)$$
$$= NJF_{J} \left(\frac{\langle J_{z} \rangle}{NJ} \right). \tag{8}$$

This relation is the main result of this paper. In an experiment with a collection of N spin-J particles, it is possible to measure the collective J_z and J_x , and to determine their mean value and variance. If the data point $[\langle J_z \rangle/NJ, \operatorname{Var}(J_x)/NJ]$ lies below the relevant curve in Fig. 1, the systems are not in a separable state, i.e., they are experimentally proven to be in an entangled state.

The extent to which the measured data point falls below the curve in the plot is a measure of the degree of entanglement. A quantitative measure of entanglement in a multiparticle system is the number of elements that must at least have gone together in entangled states. We define a k-particle entangled state to be a state of Nparticles which *cannot* be decomposed into a convex sum of products of density matrices with all density matrices involving less than k particles: at least one of the terms is a k particle entangled density matrix. If, for example, N spin $-\frac{1}{2}$ particles form N/2 entangled pairs, the degree of macroscopic spin squeezing of the system is limited by the inequality (9) with J = 1 and N replaced by N/2. If the measured macroscopic $\langle J_z \rangle$ and $Var(J_x)$ also lie below the corresponding J = 1 curve, the measurement unambiguously implies that the systems are entangled in larger than binary ensembles. The size of these ensembles is a measure of the depth of entanglement, which can be determined experimentally. This criterion may be compared to the one used in [18], where the fidelity of production of maximally entangled N-particle states is used as proof of N-particle entanglement.

As a final point we demonstrate how our procedure can be applied to identify substantial multiparticle entanglement in recent theoretical proposals for spin squeezing. In Ref. [11] it is predicted that appreciable spin squeezing of atoms can be obtained in a two-component Bose-Einstein condensate. In the calculation, a reduction of Var(J_x) by a factor of 1000 is found for a reduction of $\langle J_z \rangle$ of only 1% with 10⁵ atoms in the condensate. By using Eq. (3), these numbers imply a depth of entanglement of $\sim 2 \times 10^4$. In ion traps it has been shown that it is possible to implement a Hamiltonian J_x^2 by applying bichromatic light to all ions in the trap [8]. This Hamiltonian can be used to create a maximally entangled state of all the ions. If the decoherence in the trap is such that one cannot produce a maximally entangled state, a different strategy could be to apply the light for a short time so that squeezing is produced. For small times the squeezing obtained from the Hamiltonian J_x^2 is close to the optimal curves in Fig. 1, and our theory identifies a depth of entanglement close to the total number of ions in the trap. In this way, one could produce and verify the production of an entangled state of many ions.

We have considered squeezing and entanglement related to collective vector operators J_z and J_x . We emphazise that the collective spin components of a multiparticle atomic system are readily available by standard spectroscopic methods, which require no access to the individual components. Given the large interest in spin squeezing, a criterion of entanglement based on this property is an important tool. Recall, however, that systems may well be entangled without being spin squeezed: The spin squeezing measurement provides a sufficient criterion for the depth of entanglement, not a necessary one.

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