

## Spin Squeezing Inequalities for Arbitrary Spin

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We determine the complete set of generalized spin squeezing inequalities, given in terms of the collective angular momentum components, for particles with an arbitrary spin. They can be used for the experimental detection of entanglement in an ensemble in which the particles cannot be individually addressed. We also present a large set of criteria involving collective observables different from the angular momentum coordinates. We show that some of the inequalities can be used to detect  $k$ -particle entanglement and bound entanglement.

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With an interest towards fundamental questions in quantum physics, as well as applications, larger and larger entangled quantum systems have been realized with photons, trapped ions, and cold atoms [1]. Quantum entanglement can be used as a resource for certain quantum information processing tasks [1], and it is also necessary for a wide range of interferometric schemes to achieve the maximum sensitivity in metrology [2]. Hence, the verification of the presence of entanglement is a crucial but exceedingly challenging task, especially in an ensemble of many, say  $10^6$ – $10^{12}$ , particles. In such systems, typically the particles are not accessible individually and only collective operators can be measured. A ubiquitous entanglement criterion in this context is the spin squeezing inequality [3]

$$\frac{(\Delta J_x)^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2} \geq \frac{1}{N}, \quad (1)$$

where  $N$  is the number of spin- $\frac{1}{2}$  particles,  $J_l := \sum_{n=1}^N j_l^{(n)}$  for  $l = x, y, z$  are the collective angular momentum components and  $j_l^{(n)}$  are the single spin angular momentum components acting on the  $n$ th particle. If a state violates Eq. (1), then it is entangled (i.e., not fully separable [4]). Such spin squeezed states [5] have been created in numerous experiments with cold atoms and trapped ions [1,6], and can be used, for instance, in atomic clocks to achieve a precision higher than the shot noise limit [5].

Recently, after several generalized spin squeezing inequalities (SSIs) for the detection of entanglement appeared in the literature [7–9] and were used experimentally [10], a complete set of such entanglement conditions has been presented in Ref. [11]. However, all of the above mentioned conditions are for spin-1/2 particles (qubits), and so far the literature on systems of particles with  $j > \frac{1}{2}$  is limited to a small number of conditions, specialized for certain quantum states or particles with a low dimension [7,12,13]. At this point the question arises: Could one

obtain a complete set of inequalities for  $j > \frac{1}{2}$ ? Such conditions would be very relevant from the practical point of view since in most of the experiments the physical spin of the particles is larger than  $\frac{1}{2}$  and the spin- $\frac{1}{2}$  subsystems are created artificially. Thus, knowing the full set of entanglement criteria for  $j > \frac{1}{2}$ , many experiments for realizing large scale entanglement could be technologically less demanding, and fundamentally new experiments could also be carried out. The solution is not simple: Known methods for detecting entanglement for spin- $\frac{1}{2}$  particles by spin squeezing cannot straightforwardly be generalized to higher spins. For example, for  $j > \frac{1}{2}$ , Eq. (1) can also be violated without entanglement between the spin- $j$  particles [14].

In this Letter, we present the complete set of optimal spin squeezing inequalities for the collective angular momentum coordinates for a system of  $N$  particles with spin  $j$ . We also show how existing entanglement conditions for spin- $\frac{1}{2}$  particles can be transformed into entanglement conditions for spin- $j$  particles with  $j > \frac{1}{2}$  (i.e., qudits with a dimension  $d = 2j + 1$ ). Finally, we present a large set of entanglement conditions for qudit systems that involve operators different from the angular momentum coordinates, and investigate in detail one of the conditions.

*Definitions.*—The basic idea for the qudit case is that besides  $j_l$ , other single-qudit quantities can also be measured. Let us consider particles with  $d$  internal states.  $a_k$  for  $k = 1, 2, \dots, M$  will denote single-particle operators with the property  $\text{Tr}(a_k a_l) = C \delta_{kl}$ , where  $C$  is a constant. As we will show later, the  $a_k$  operators can be, for instance, the  $\text{SU}(d)$  generators for a  $d$  dimensional system. Moreover, for obtaining our generalized spin squeezing inequalities, we will need the upper bound  $K$  for the inequality  $\sum_{k=1}^M \langle a_k^{(n)} \rangle^2 \leq K$ .

The  $N$ -qudit collective operators used in our criteria will be denoted by  $A_k = \sum_n a_k^{(n)}$ . In the qubit case, the SSIs were developed based on the first and second moments and

variances of the such collective operators [11]. For  $j > 1/2$ , we define the modified second moment

$$\langle \tilde{A}_k^2 \rangle := \langle A_k^2 \rangle - \left\langle \sum_n (a_k^{(n)})^2 \right\rangle = \sum_{m \neq n} \langle a_k^{(n)} a_k^{(m)} \rangle \quad (2)$$

and the modified variance

$$(\tilde{\Delta A}_k)^2 := (\Delta A_k)^2 - \left\langle \sum_n (a_k^{(n)})^2 \right\rangle. \quad (3)$$

In the following, the quantities Eqs. (2) and (3) will be used instead of second moments and variances because otherwise it is not possible to obtain tight inequalities for separable states [13].

*SSIs for qudits.*—First, we present a general inequality from which the entanglement conditions for the different operator sets can be obtained.

*Observation 1.*—For separable states, i.e., for states that can be written as a mixture of product states [4],

$$(N-1) \sum_{k \in I} (\tilde{\Delta A}_k)^2 - \sum_{k \notin I} \langle \tilde{A}_k^2 \rangle \geq -N(N-1)K \quad (4)$$

holds, where each index set  $I \subseteq \{1, 2, \dots, M\}$  defines one of the  $2^M$  inequalities. Note that  $I = \emptyset$  and  $I = \{1, 2, \dots, M\}$  are among the possibilities. The proof can be found in the Appendix. It is remarkable that the bound on the right-hand side of Eq. (4) is tight, independent of  $I$ , and independent of the particular choice of the  $a_k$  operators except for the value of  $K$ .

Equation (4) is the basis for the entanglement conditions we present in Observations 2 and 4.

*Observation 2.*—Optimal spin squeezing inequalities for qudits. For fully separable states of spin- $j$  particles, all the following inequalities are fulfilled

$$\langle J_x^2 \rangle + \langle J_y^2 \rangle + \langle J_z^2 \rangle \leq Nj(Nj+1), \quad (5a)$$

$$(\Delta J_x)^2 + (\Delta J_y)^2 + (\Delta J_z)^2 \geq Nj, \quad (5b)$$

$$\langle \tilde{J}_k^2 \rangle + \langle \tilde{J}_l^2 \rangle - N(N-1)j^2 \leq (N-1)(\tilde{\Delta J}_m)^2, \quad (5c)$$

$$(N-1)[(\tilde{\Delta J}_k)^2 + (\tilde{\Delta J}_l)^2] \geq \langle \tilde{J}_m^2 \rangle - N(N-1)j^2, \quad (5d)$$

where  $k, l, m$  take all possible permutations of  $x, y, z$ . Violation of any of the inequalities (5) implies entanglement. The inequalities (5) are a full set for large  $N$  in the sense that it is not possible to add a new entanglement condition detecting other states based on  $\langle J_k \rangle$  and  $\langle \tilde{J}_k^2 \rangle$ .

*Proof.*—We applied Observation 1 with  $\{a_k\} = \{J_x, J_y, J_z\}$ ,  $K = j^2$  and used  $j_x^2 + j_y^2 + j_z^2 = j(j+1)1$  [15,16]. For  $j = \frac{1}{2}$ , the inequalities (5) are identical to the optimal SSIs for qubits [11]. For this case, the completeness has already been shown [11]. That is, for all values of  $\langle J_k \rangle$  and  $\langle \tilde{J}_k^2 \rangle$  that fulfill Eqs. (5) there is a corresponding separable state in the large  $N$  limit. Direct calculation shows that if a separable quantum state  $\varrho_{\text{sep}, \frac{1}{2}} = \sum_m p_m \rho_m^{(1)} \otimes \rho_m^{(2)} \otimes \dots \otimes \rho_m^{(N)}$ , where  $\rho_m^{(n)}$  are single-qubit pure states, saturates one of the inequalities Eqs. (5) for

$j = \frac{1}{2}$ , then the state  $\varrho_{\text{sep}, j} = \sum_m p_m \omega_m^{(1)} \otimes \omega_m^{(2)} \otimes \dots \otimes \omega_m^{(N)}$ , saturates the same inequality of Eqs. (5) for spin- $j$  particles. Here,  $\omega_m^{(n)}$  are single-qudit pure-state density matrices such that  $\text{Tr}(\rho_m^{(n)} \sigma_i) j = \text{Tr}(\omega_m^{(n)} j_i)$ . For instance, if the first state is  $|+\frac{1}{2}\rangle_x$ , then the second one is  $|+j\rangle_x$ . Thus the proof of completeness of Ref. [11] can be extended to prove the completeness of the criteria Eqs. (5). ■

Equation (5a) is valid for all quantum states. States maximally violating Eq. (5b) are angular momentum singlets, while for Eq. (5c), for even  $N$ , they are symmetric Dicke states of the form  $\binom{N/2}{N}^{-1/2} \sum_k \mathcal{P}_k (|+j\rangle^{\otimes N/2} \otimes |-j\rangle^{\otimes N/2})$ , where  $\mathcal{P}_k$  denotes all different permutations [17].

It is also possible to obtain entanglement conditions for spin- $j$  particles from criteria for qubit systems.

*Observation 3.*—Let us consider an inequality valid for  $N$ -qubit separable states of the form

$$f(\{\langle J_l \rangle\}_{l=x,y,z}, \{\langle \tilde{J}_l^2 \rangle\}_{l=x,y,z}) \geq \text{const}, \quad (6)$$

where  $f$  is a concave function of its variables. All of the generalized SSIs in the literature have this form. Then, the entanglement condition Eq. (6) can be transformed to a criterion for a system of  $N$  spin- $j$  particles by the substitution

$$\langle J_l \rangle \rightarrow \frac{1}{2j} \langle J_l \rangle, \quad \langle \tilde{J}_l^2 \rangle \rightarrow \frac{1}{4j^2} \langle \tilde{J}_l^2 \rangle. \quad (7)$$

*Proof.*—Let us consider product states of  $N$  spin- $j$  particles of the form  $\varrho_j = \otimes_n \varrho_j^{(n)}$ , and define the quantities  $r_l^{(n)} = \langle J_l^{(n)} \rangle / j$ . Then, the first and second moments can be rewritten as  $\langle J_l \rangle = j \sum_n r_l^{(n)}$  and  $\langle \tilde{J}_l^2 \rangle = j^2 \sum_{m \neq n} r_l^{(n)} r_l^{(m)}$ . The only constraint for the physically allowed values for  $r_l^{(n)}$  is  $|r_l^{(n)}| \leq 1$  for all  $j$ . Hence, for an arbitrary function  $f$ ,

$$\begin{aligned} & \min_{\varrho_j} f(\{\frac{1}{2j} \langle J_l \rangle_{\varrho_j}\}_{l=x,y,z}, \{\frac{1}{4j^2} \langle \tilde{J}_l^2 \rangle_{\varrho_j}\}_{l=x,y,z}) \\ &= \min_{\varrho_{1/2}} f(\{\langle J_l \rangle_{\varrho_{1/2}}\}_{l=x,y,z}, \{\langle \tilde{J}_l^2 \rangle_{\varrho_{1/2}}\}_{l=x,y,z}). \end{aligned}$$

If  $f$  is a concave function of its variables then we have the same minimum for separable states. ■

Using Observation 3, for instance, the standard spin squeezing inequality Eq. (1) from Ref. [3] becomes

$$\frac{(\Delta J_x)^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2} + \frac{\sum_n (j^2 - \langle (j_x^{(n)})^2 \rangle)}{\langle J_y \rangle^2 + \langle J_z \rangle^2} \geq \frac{1}{N}. \quad (8)$$

Equation (8) is violated only if there is entanglement between the spin- $j$  particles. Because of the second, non-negative term on the left-hand side of Eq. (8), for  $j > \frac{1}{2}$  there are states that violate Eq. (1), but do not violate Eq. (8). Remarkably, it can be proven that Eq. (5c) is strictly stronger than Eq. (8) [17].

The last application of Observation 1 is the following.

*Observation 4.*—For a system of  $d$ -dimensional particles, we can define collective operators based on the  $\text{SU}(d)$  generators  $\{g_k\}_{k=1}^M$  with  $M = d^2 - 1$  as  $G_k = \sum_{n=1}^N g_k^{(n)}$ . The SSIs for  $G_k$  have the general form

$$(N-1) \sum_{k \in I} (\tilde{\Delta} G_k)^2 - \sum_{k \notin I} \langle \tilde{G}_k^2 \rangle \geq -2N(N-1) \frac{(d-1)}{d}. \quad (9)$$

For instance, for the  $d = 3$  case, the  $SU(d)$  generators can be the Gell-Mann matrices [18].

*Proof.*—We used Observation 1 with  $C = 2$  and  $K = 2(1 - \frac{1}{d})$  [15,19]. ■

Observation 4 presents an abundance of inequalities. Here, we will analyze in detail Eq. (9) for  $I = \{1, 2, \dots, M\}$ . Using  $\sum_k g_k^2 = 2(d+1)(1 - \frac{1}{d}) \mathbb{1}$  [15], Eq. (9) for this case can be rewritten as

$$\sum_{k=1}^{d^2-1} (\Delta G_k)^2 \geq 2N(d-1). \quad (10)$$

Equation (10) is maximally violated by many-body  $SU(d)$  singlets. Such states appear often in statistical physics of spin systems and condensed matter physics [20]. They are invariant under operations of the type  $U^{\otimes N}$  [4], which can be exploited in differential magnetometry [21], encoding quantum information in decoherence free subspaces and sending information independent from the reference frame direction [22].

*Noise tolerance of Eq. (10).*—First, we will ask how efficiently Eq. (10) can be used for entanglement detection. Let us consider  $SU(d)$  singlet states (i.e., states with  $\langle G_k^2 \rangle = 0$ ) mixed with white noise as  $\rho_{\text{noisy}} = (1 - p_{\text{noise}}) \rho_{\text{singlet}} + p_{\text{noise}} \frac{1}{d^N} \mathbb{1}$ . Direct calculation shows that such a state is detected as entangled if  $p_{\text{noise}} < \frac{d}{d+1}$ . Thus, the noise tolerance in detecting  $SU(d)$  singlets is increasing with  $d$ . Note that Eq. (5b) detects a noisy state as entangled for an analogous situation if  $p_{\text{noise}} < \frac{2}{d+1}$ .

*Equation (10) detects  $k$ -particle entanglement.*—The criteria presented so far detect any type of nonseparability. It would be important to find similar criteria that detect higher forms of entanglement, that is,  $k$  entanglement. This type of strong entanglement, rather than simple nonseparability, is needed, for instance, to achieve maximal precision in many interferometric tasks [23]. A pure state is said to possess  $k$  entanglement if it cannot be written as a tensor product  $\otimes_n |\psi_n\rangle$  such that each  $|\psi_n\rangle$  is a state of at most  $k-1$  qubits. A mixed state is  $k$  entangled if it cannot be obtained mixing states that are at most  $k-1$  entangled [24]. Otherwise the state is called  $(k-1)$  producible.

While Eq. (10) can be maximally violated by two-producible states for  $j = \frac{1}{2}$  [21], it is not the case for  $j > \frac{1}{2}$ . For the  $SU(d)$  case, a  $d$ -particle entangled state is needed to violate Eq. (10) maximally [15]. Thus, the amount of violation of Eq. (10) can be used to detect  $k$  entanglement.

*Observation 5.*—For two-producible states the following bound holds

$$\sum_{k=1}^{d^2-1} (\Delta G_k)^2 \geq \begin{cases} 2N(d-2) & \text{for even } N, \\ 2N(d-2) + 2 & \text{for odd } N. \end{cases} \quad (11)$$

The violation of Eq. (11) signals 3-particle entanglement. Note that for large  $d$  the bound in Eq. (11) is very close to the bound for separable states in Eq. (10). The proof can be found in the Appendix.

*Equation (10) detects bound entanglement.*—In Ref. [11], it has already been shown the optimal SSIs for the  $j = \frac{1}{2}$  case can detect bound entanglement [25], i.e., entangled states with a positive partial transpose (PPT, [26]), in the thermal states of common spin models. We find numerically that the criterion Eq. (10) detects bound entanglement in the thermal state of several Hamiltonians, such as for example  $H = \sum_k G_k^2$ , even for  $j > \frac{1}{2}$  [17].

*Symmetric states.*—Next, it is important to ask how our entanglement criteria behave for symmetric states, as such states naturally appear in many systems such as Bose-Einstein condensates of two-state atoms.

*Observation 6.*—(i) Symmetric states can violate Eq. (4) for some  $I$  only if  $\rho_{\text{av}2}^{T1} \neq 0$ , where  $T1$  denotes the partial transposition [26] and the average two-qudit density matrix is defined as  $\rho_{\text{av}2} = \frac{1}{N(N-1)} \sum_{m \neq n} \rho_{mn}$ . (ii) For symmetric states, if  $a_k$  are the  $SU(d)$  generators  $g_k$ , Eq. (4) is equivalent to

$$\sum_{k \in I} N(\tilde{\Delta} G_k)^2 + \langle G_k \rangle^2 \geq 0. \quad (12)$$

For this case, Eq. (12) is violated for at least one  $I$  and some choice of the collective operators if and only if  $\rho_{\text{av}2}^{T1} \neq 0$ . For the proof, see the Appendix.

*Implementation.*—The angular momentum coordinates  $J_k$  and their variances can be measured in cold atoms by coupling the atomic spin to a light field, and then measuring the light [6]. The collective spin can be rotated by magnetic fields. Measuring the operators  $\sum_n (j_k^{(n)})^2$  can be realized by rotating the spin by a magnetic field, and then measuring the populations of the  $j_z$  eigenstates. In some cold atomic systems, such operators might also be measured directly, as in such systems in the Hamiltonian a  $s_z(j_k^{(n)})^2$  term appears, where  $\vec{s}$  is the photonic pseudospin [27]. For the  $SU(d)$  generators, the  $G_k$  operators can be measured in a similar manner, however,  $SU(2)$  rotations realized with a magnetic field are not sufficient. For larger spins, it is advantageous to choose the  $g_k$  operators to be  $(|k\rangle\langle l| + |l\rangle\langle k|)/\sqrt{2}$ ,  $i(|k\rangle\langle l| - |l\rangle\langle k|)/\sqrt{2}$  and  $|k\rangle\langle k|$  [28]. The corresponding collective operators can all be measured based an  $SU(2)$  rotation within a two-dimensional subspace and a population measurement of at most two quantum states.

In summary, we have presented a complete set of generalized SSIs for detecting entanglement in an ensemble of qudits based on knowing only  $\langle J_k \rangle$  and  $\langle \tilde{J}_k^2 \rangle$  for  $k = x, y, z$ . We extended our approach to collective observables based on the  $SU(d)$  generators. We showed that some of the inequalities can be used to detect  $k$  entanglement and bound entanglement. Finally, we discussed the experimental implementation of the criteria.

*Appendix: Proof of Observation 1.*—We consider product states of the form  $|\Phi\rangle = \otimes_n |\phi_n\rangle$ . For such states, we have  $\langle \tilde{A}_k^2 \rangle_\Phi = \langle A_k \rangle^2 - \sum_n \langle a_k^{(n)} \rangle^2$ . Hence, the left-hand side of Eq. (4) equals  $-\sum_n (N-1) \sum_{k \in I} \langle a_k^{(n)} \rangle^2 - \sum_{k \notin I} (\langle A_k \rangle^2 - \sum_n \langle a_k^{(n)} \rangle^2) \geq -\sum_n (N-1) \sum_{k=1}^M \langle a_k^{(n)} \rangle^2 \geq -N(N-1)K$ . We used that  $\langle A_k \rangle^2 \leq N \sum_n \langle a_k^{(n)} \rangle^2$  [11]. ■

*Proof of Observation 5.*—We will find a lower bound on the left-hand side of Eq. (11) for  $N = 2$ . Let us consider first antisymmetric states. We will use that  $\sum_k \langle G_k^2 \rangle = \sum_k \langle g_k^2 \otimes \mathbb{1} \rangle + \sum_k \langle \mathbb{1} \otimes g_k^2 \rangle + 2 \sum_k \langle g_k \otimes g_k \rangle$ . Then, we need that  $\sum_k g_k \otimes g_k = 2F - \frac{2}{d} \mathbb{1}$  where  $F$  is the flip operator [15,19]. Hence,  $\sum_k \langle G_k^2 \rangle = 4(d+1)(1 - \frac{2}{d})$ . For the nonlinear part, we have that  $\sum_k \langle g_k \rangle_{\rho_{\text{red}}}^2 = 2\text{Tr}(\rho_{\text{red}}^2) - \frac{2}{d}$  [15,19], and using the Cauchy-Schwarz inequality for  $\sum_k \langle g_k \otimes \mathbb{1} \rangle \langle \mathbb{1} \otimes g_k \rangle$ , we obtain a bound  $\sum_k \langle G_k \rangle^2 \leq 4 - \frac{8}{d}$ . Here we used that for antisymmetric states, for the reduced single-qudit state  $\text{Tr}(\rho_{\text{red}}^2) \leq \frac{1}{2}$  [29]. This leads to Eq. (11) for antisymmetric states. For symmetric states the bound on the left-hand side of Eq. (11) can be obtained similarly and it is larger. Finally, since the equation is invariant under the permutation of qudits, the variances give the same value for  $\rho$  as for  $\frac{1}{2}(\rho + F\rho F) \equiv P_a \rho P_a + P_s \rho P_s$ , where  $P_s$  and  $P_a$  are the projectors to the symmetric and antisymmetric subspaces, respectively. Thus, it is sufficient to consider mixtures of symmetric and antisymmetric states. The bound for the product of such two-qudit states and of single-qudit states for the left-hand side of Eq. (11) can be obtained using  $[\Delta(a \otimes \mathbb{1} + \mathbb{1} \otimes a)]_{\psi_1 \otimes \psi_2}^2 = (\Delta a)_{\psi_1}^2 + (\Delta a)_{\psi_2}^2$ . Because of the concavity of the variance, the bound is the same for mixed 2-producible states. ■

*Proof of Observation 6.*—Equation (4) can be rewritten as  $\sum_{k \in I} N \langle \tilde{A}_k \rangle^2 + \langle A_k \rangle^2 \geq \sum_{k=1}^M \langle \tilde{A}_k^2 \rangle - N(N-1)K$ , which can be reexpressed as  $\sum_{k \in I} N \langle \langle a_k \otimes a_k \rangle_{\rho_{\text{av2}}} - \langle a_k \otimes \mathbb{1} \rangle_{\rho_{\text{av2}}}^2 \rangle \geq \sum_k \langle a_k \otimes a_k \rangle_{\rho_{\text{av2}}} - K$ . From Eq. (4) for  $I = \emptyset$  it follows that  $\sum_k \langle a_k \otimes a_k \rangle_{\rho_{\text{av2}}} = \frac{1}{N(N-1)} \sum_k \langle \tilde{A}_k^2 \rangle \leq K$ , while the equality holds for symmetric states for  $SU(d)$  generators  $g_k$  [15]. We also need that a density matrix of a two-qudit symmetric state has a positive partial transpose if and only if  $\langle O \otimes O \rangle - \langle O \otimes \mathbb{1} \rangle^2 \geq 0$  for every  $O$  [30]. Hence the statement of Observation 6 follows. For qubits, we obtain the results of Ref. [8]. ■

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[28] For these  $d^2$  operators we have  $K = 1$  [15].  
[29] This is because all pure two-qudit antisymmetric states can be written in some basis as  $\alpha_{12} |\Psi_{12}^- \rangle + \alpha_{34} |\Psi_{34}^- \rangle + \alpha_{56} |\Psi_{56}^- \rangle + \dots$ , where  $\alpha_{nm}$  are constants and  $|\Psi_{mn}^- \rangle = (|mn\rangle - |nm\rangle)/\sqrt{2}$ . See J. Schliemann *et al.*, *Phys. Rev. A* **64**, 022303 (2001).  
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