

Spin squeezing and entanglement for arbitrary spin

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A complete set of generalized spin-squeezing inequalities is derived for an ensemble of particles with an arbitrary spin. Our conditions are formulated with the first and second moments of the collective angular momentum coordinates. A method for mapping the spin-squeezing inequalities for spin- $\frac{1}{2}$ particles to entanglement conditions for spin- j particles is also presented. We apply our mapping to obtain a generalization of the original spin-squeezing inequality to higher spins. We show that, for large particle numbers, a spin-squeezing parameter for entanglement detection based on one of our inequalities is strictly stronger than the original spin-squeezing parameter defined in [A. Sørensen *et al.*, Nature **409**, 63 (2001)]. We present a coordinate system independent form of our inequalities that contains, besides the correlation and covariance tensors of the collective angular momentum operators, the nematic tensor appearing in the theory of spin nematics. Finally, we discuss how to measure the quantities appearing in our inequalities in experiments.

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I. INTRODUCTION

One of the most rapidly developing areas in quantum physics is creating larger and larger entangled quantum systems with photons, trapped ions, and cold neutral atoms [1–12]. Entangled states can be used for metrology in order to obtain a sensitivity higher than the shot-noise limit [13–15] and can also be used as a resource for certain quantum information processing tasks [16–19]. Moreover, experiments realizing macroscopic quantum effects might give answers to fundamental questions in quantum physics [20, 21].

Spin squeezing is one of the most successful approaches for creating large-scale quantum entanglement [13, 22–37]. It is used in systems of very many particles in which only collective quantities can be measured. For an ensemble of N particles with a spin j , the most relevant collective quantities are the collective spin operators defined as

$$J_l := \sum_{n=1}^N j_l^{(n)} \quad (1)$$

for $l = x, y, z$, where $j_l^{(n)}$ are the components of the angular momentum operator for the n^{th} spin.

Spin-squeezed states are typically almost fully polarized states for which the angular momentum variance is small in a direction orthogonal to the mean spin [22]. They can be used to achieve a high accuracy in certain very general metrological tasks [14, 15]. On the other hand, in spin- $\frac{1}{2}$ systems spin squeezing is closely connected to multipartite entanglement. A ubiquitous cri-

terion for detecting the entanglement of spin-squeezed states is [13]

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

Any fully separable state of N qubits, that is, a state that can be written as [38]

$$\varrho = \sum_k p_k \varrho_k^{(1)} \otimes \varrho_k^{(2)} \otimes \dots \otimes \varrho_k^{(N)}, \quad \sum_k p_k = 1, \quad p_k > 0, \quad (3)$$

satisfies Eq. (2). Any state violating Eq. (2) is not fully separable and is therefore entangled.

Apart from the original inequality Eq. (2), several other generalized spin-squeezing entanglement conditions have been presented [39–54] and even the complete set of such criteria for multi-qubit systems has been found in Ref. [55]. While most of the conditions are for a fixed particle number, conditions for the case of nonzero particle number variance have also been derived [56, 57].

So far most of the attention has been focused on ensembles of spin- $\frac{1}{2}$ particles. The literature on systems of particles with $j > \frac{1}{2}$ has been limited to a small number of conditions, specialized to certain sets of quantum states or particles with a low spin [48–54]. The reason is that 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. (2) can also be violated without entanglement between the spin- j particles, as we will discuss later [33].

In spite of the difficulties in deriving entanglement conditions for particles with a higher spin, they are very much needed in quantum experiments nowadays. As most of such experiments are done with atoms with $j > \frac{1}{2}$, such conditions can make the complexity of experiments much smaller: The artificially created spin- $\frac{1}{2}$ subsystems must be manipulated by lasers, while the physi-

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cal spin- j particles can directly be manipulated by magnetic fields. Moreover higher spin systems could make it possible to perform quantum information processing tasks different from the ones possible with spin- $\frac{1}{2}$ particles or to create different kind of entangled states [58–64].

In this paper, we will start from the complete set presented for spin- $\frac{1}{2}$ particles in Ref. [55]. All spin-squeezing entanglement criteria of this set are based on the first and second moments of collective angular momentum coordinates. It has been possible to obtain a full set of tight inequalities by analytical means only due to certain advantageous properties of the spin- $\frac{1}{2}$ case. For the case of particles with $j > \frac{1}{2}$, the inequalities presented in the literature are either based on numerical optimization [48] or are analytical but not tight [51]. The reason for this is that for $j > \frac{1}{2}$, the second moments of the collective observables are not only connected to the two-body correlations, as in the spin- $\frac{1}{2}$ case, but also to the local second moments.

In order to solve this problem, we define modified second moments and the corresponding variances as follows

$$\begin{aligned} \langle \tilde{J}_l^2 \rangle &:= \langle J_l^2 \rangle - \langle \sum_n (j_l^{(n)})^2 \rangle = \sum_{n \neq m} \langle j_l^{(n)} j_l^{(m)} \rangle, \\ (\tilde{\Delta} J_l)^2 &:= \langle \tilde{J}_l^2 \rangle - \langle J_l \rangle^2, \end{aligned} \quad (4)$$

where $l = x, y, z$. The modified quantities do not contain anymore the local second moments. We will show that by using the first moments and the modified second moments of the collective operators, it is possible to write down tight entanglement conditions analytically also for the $j > \frac{1}{2}$ case [65]. We will also discuss that the local second moments are related to single-particle spin squeezing (see Sec. VIA).

The main results of our paper are as follows.

(i) We will find the complete set of conditions for the $j > \frac{1}{2}$ case, which we will call *optimal spin-squeezing inequalities for spin- j particles*. They are a complete set since, for large N , they detect all entangled states that can be detected knowing only the first moments and the modified second moments. For instance, they can be used to verify the entanglement of singlet states, symmetric Dicke states and planar squeezed states [52].

(ii) We also present a generalization of the original spin squeezing parameter ξ_s^2 defined in Eq. (2) that can be used for entanglement detection even for particles with $j > \frac{1}{2}$

$$\xi_{s,j}^2 := N \frac{(\tilde{\Delta} J_x)^2 + N j^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2}. \quad (5)$$

If $\xi_{s,j}^2 < 1$ then the state is entangled. For spin- $\frac{1}{2}$ particles, the definitions of Eqs. (2) and (5) are the same.

(iii) Finally, we will show that, in the large particle number limit, the entanglement condition based on the following entanglement parameter

$$\xi_{os}^2 := (N-1) \frac{(\tilde{\Delta} J_x)^2 + N j^2}{\langle \tilde{J}_y \rangle + \langle \tilde{J}_z \rangle} \quad (6)$$

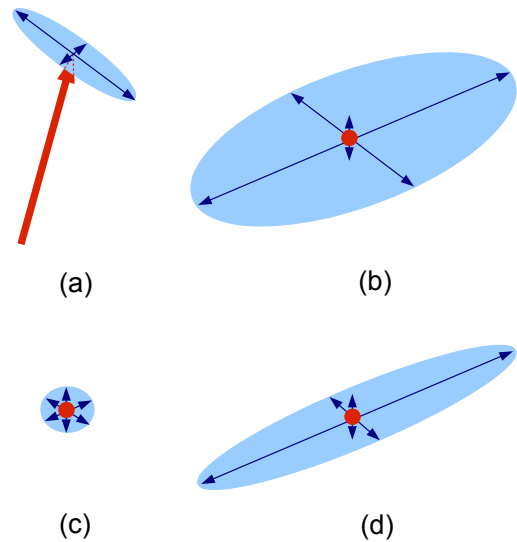


FIG. 1: (Color online) Different types of spin-squeezed states. (a) *Almost fully polarized spin-squeezed states* detected by $\xi_{s,j}^2$, given in Eq. (5) and also by the new parameter, ξ_{os}^2 , defined in Eq. (6). (b) States close to *symmetric Dicke states* with $\langle J_z \rangle = 0$ with a small variance for one of the angular momentum components and large variances in the two orthogonal directions. Such states can be detected by ξ_{os}^2 but are not detected by $\xi_{s,j}^2$. (c) States close to *many-body singlets* with a small variance for all the three angular momentum components. Such states are detected by the criterion (9b). (d) *Planar squeezed states* with a small variance for two of the angular momentum components and a large variance in the orthogonal direction. Such states are detected by the criterion (9d).

is strictly stronger than the condition based on $\xi_{s,j}^2$. Note that ξ_{os}^2 is defined only for $\langle \tilde{J}_y \rangle + \langle \tilde{J}_z \rangle > 0$. In this way ξ_{os}^2 will always be non-negative. In Eq. (6), the subscript “os” refers to the optimal spin-squeezing inequalities since we obtain ξ_{os}^2 , essentially, by dividing the left-hand side of one of the inequalities by the right-hand side. For clarity, we give Eq. (6) explicitly for the $j = \frac{1}{2}$ case

$$\xi_{os}^2 = (N-1) \frac{(\Delta J_x)^2}{\langle J_y \rangle + \langle J_z \rangle - \frac{N}{2}}. \quad (7)$$

If $\xi_{os}^2 < 1$ then the state is entangled. The parameter (5) is appropriate only for spin-squeezed states with a large total spin depicted in Fig. 1(a), while the parameter (6) detects also states that have zero total spin, as shown in Fig. 1(b). Moreover, we will also show that for large particle numbers, if $\xi_{s,j}^2 < 1$ then we also have

$$\xi_{os}^2 < \xi_{s,j}^2. \quad (8)$$

Thus, ξ_{os}^2 is a better indicator of entanglement than $\xi_{s,j}^2$.

The paper is organized as follows. In Ref. [66], we have already presented a generalization of the complete set of spin-squeezing inequalities valid for systems of spin- j particles with $j > \frac{1}{2}$. In this paper, we extend the results of Ref. [66] in several directions. In Sec. II, we present the optimal spin-squeezing inequalities for spin- j particles and discuss some of their fundamental properties. In Sec. III, we study states that violate the inequalities maximally. In Sec. IV, we show a method for mapping existing entanglement conditions for spin- $\frac{1}{2}$ particles to analogous conditions for spin- j particles with $j > \frac{1}{2}$. Using the mapping, we derive the spin-squeezing parameter $\xi_{s,j}^2$. In Sec. V, we present the spin-squeezing parameter ξ_{os}^2 and examine its properties. In Sec. VI, we consider various issues concerning the efficient application of our spin-squeezing inequalities.

II. COMPLETE SET OF SPIN-SQUEEZING INEQUALITIES FOR SPIN- j PARTICLES.

In this section, we present our spin-squeezing inequalities for particles with an arbitrary spin j and we also examine the connection of these inequalities to the entanglement of the reduced two-particle state, and to the criterion based on the positivity of the partial transpose.

A. The optimal spin-squeezing inequalities for qudits

Observation 1.—The following generalized spin-squeezing inequalities are valid for separable states given by Eq. (3) for an ensemble of spin- j particles even with $j > \frac{1}{2}$

$$\langle J_x^2 \rangle + \langle J_y^2 \rangle + \langle J_z^2 \rangle \leq Nj(Nj+1), \quad (9a)$$

$$(\Delta J_x)^2 + (\Delta J_y)^2 + (\Delta J_z)^2 \geq Nj, \quad (9b)$$

$$\langle \tilde{J}_l^2 \rangle + \langle \tilde{J}_m^2 \rangle - N(N-1)j^2 \leq (N-1)(\tilde{\Delta} J_k)^2, \quad (9c)$$

$$(N-1) \left[(\tilde{\Delta} J_k)^2 + (\tilde{\Delta} J_l)^2 \right] \geq \langle \tilde{J}_m^2 \rangle - N(N-1)j^2. \quad (9d)$$

Here k, l, m may take all the possible permutations of x, y, z . If a quantum state violates one of the inequalities (9), then it is entangled.

Proof. We will prove that for separable states the following inequality holds

$$(N-1) \sum_{l \in I} (\tilde{\Delta} J_l)^2 - \sum_{l \notin I} \langle \tilde{J}_l^2 \rangle \geq -N(N-1)j^2, \quad (10)$$

where I is a subset of indices including the two extremal cases $I = \emptyset$ and $I = \{x, y, z\}$. We consider first pure product states of the form $|\Phi\rangle = \otimes_n |\phi_n\rangle$. For such states, the modified variances and the modified second moments

can be obtained as

$$\begin{aligned} (\Delta \tilde{J}_l)_\Phi^2 &= - \sum_n \langle j_l^{(n)} \rangle^2, \\ \langle \tilde{J}_l^2 \rangle_\Phi &= \langle J_l \rangle^2 - \sum_n \langle j_l^{(n)} \rangle^2 = \sum_{n \neq m} \langle j_l^{(n)} \rangle \langle j_l^{(m)} \rangle. \end{aligned} \quad (11)$$

Substituting Eq. (11) into the left-hand side of Eq. (10), we obtain

$$\begin{aligned} & - \sum_n (N-1) \sum_{l \in I} \langle j_l^{(n)} \rangle^2 - \sum_{l \notin I} \left(\langle J_l \rangle^2 - \sum_n \langle j_l^{(n)} \rangle^2 \right) \\ & \geq - \sum_n (N-1) \sum_{l=x,y,z} \langle j_l^{(n)} \rangle^2 \geq -N(N-1)j^2. \end{aligned} \quad (12)$$

The two inequalities in Eq. (12) follow from the inequality [55]

$$\langle J_l \rangle^2 \leq N \sum_n \langle j_l^{(n)} \rangle^2, \quad (13)$$

and from the well-known bound for an angular momentum component $\langle j_l \rangle \leq j$. Hence we proved that Eq. (10) is valid for pure product states. Due to the left-hand side of Eq. (10) being concave in the state, it is also valid for separable states.

From Eq. (10) we can obtain all inequalities of Eq. (9a)–(9d), knowing that

$$\langle J_x^2 \rangle + \langle J_y^2 \rangle + \langle J_z^2 \rangle = \langle \tilde{J}_x^2 \rangle + \langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle + Nj(j+1), \quad (14)$$

which is a consequence of the identity [67]

$$j_x^2 + j_y^2 + j_z^2 = j(j+1)\mathbf{1}. \quad (15)$$

Hence, we proved that Eq. (9) is valid for separable states. ■

In order to evaluate Eq. (9), six operator expectation values are needed. These are the vector of the expectation values of the three collective angular momentum components

$$\vec{J} := (\langle J_x \rangle, \langle J_y \rangle, \langle J_z \rangle), \quad (16)$$

and the vector of the modified second moments

$$\vec{\tilde{K}} := (\langle \tilde{J}_x^2 \rangle, \langle \tilde{J}_y^2 \rangle, \langle \tilde{J}_z^2 \rangle). \quad (17)$$

For the spin- $\frac{1}{2}$ case, the modified second moments can be obtained from the true second moments since $\langle \tilde{J}_x^2 \rangle = \langle J_x^2 \rangle - \frac{N}{4}$. For spin- j particles with $j > \frac{1}{2}$, the elements of $\vec{\tilde{K}}$ typically cannot be measured directly. Instead, we measure the true second moments

$$\vec{K} := (\langle J_x^2 \rangle, \langle J_y^2 \rangle, \langle J_z^2 \rangle) \quad (18)$$

and the sum of the squares of the local second moments

$$\vec{M} := \left(\langle \sum_n (j_x^{(n)})^2 \rangle, \langle \sum_n (j_y^{(n)})^2 \rangle, \langle \sum_n (j_z^{(n)})^2 \rangle \right). \quad (19)$$

Then, \vec{K} can be obtained as the difference between the true second moments and the sum of local second moments as

$$\vec{K} = \vec{K} - \vec{M}. \quad (20)$$

In Sec. VIC, we discuss how to measure \vec{K} based on the measurement of \vec{K} and \vec{M} .

For any value of the mean spin \vec{J} , Eq. (9) defines a polytope in the $(\langle \tilde{J}_x^2 \rangle, \langle \tilde{J}_y^2 \rangle, \langle \tilde{J}_z^2 \rangle)$ -space. The polytope is depicted in Figs. 2(a) and 2(b) for different values for \vec{J} . It is completely characterized by its extremal points. Direct calculation shows that the coordinates of the extreme points in the $(\langle \tilde{J}_x^2 \rangle, \langle \tilde{J}_y^2 \rangle, \langle \tilde{J}_z^2 \rangle)$ -space are

$$\begin{aligned} A_x &:= [N(N-1)j^2 - \kappa(\langle J_y \rangle^2 + \langle J_z \rangle^2), \kappa \langle J_y \rangle^2, \kappa \langle J_z \rangle^2], \\ B_x &:= \left[\langle J_x \rangle^2 + \frac{\langle J_y \rangle^2 + \langle J_z \rangle^2}{N} - Nj^2, \kappa \langle J_y \rangle^2, \kappa \langle J_z \rangle^2 \right], \end{aligned} \quad (21)$$

where $\kappa := \frac{N-1}{N}$. The points $A_{y/z}$ and $B_{y/z}$ can be obtained in an analogous way. Note that the coordinates of the points A_l and B_l depend nonlinearly on $\langle J_l \rangle$.

Let us see briefly the connection between the inequalities and the facets of the polytope. The inequality with three second moments, Eq. (9a), corresponds to the facet $A_x - A_y - A_z$ in Fig. 2(a). The inequality with three variances, Eq. (9b), corresponds to the facet $B_x - B_y - B_z$. The inequality with one variance, Eq. (9c) corresponds to the facets $B_x - A_y - A_z$, $B_y - A_z - A_x$, and $B_z - A_x - A_y$. The inequality with two variances, Eq. (9d), corresponds to the facets $B_x - B_y - A_z$, $B_y - B_z - A_x$ and $B_z - B_x - A_y$.

B. Completeness of Eq. (9)

In this section, we will show that, in the large N limit, all points inside the polytope correspond to separable states. This implies that the criteria of Observation 1 are complete, that is, if the inequalities are not violated then it is not possible to prove the presence of entanglement based only on the first and the modified second moments. In other words, it is not possible to find criteria detecting more entangled states based on these moments. To prove this, first we can observe that if some quantum states satisfy Eq. (9) then their mixture also satisfies it. Thus, it is enough to investigate the states corresponding to the extremal points of the polytope. We will give a straightforward generalization of the proof for the spin- $\frac{1}{2}$ case presented in Ref. [55].

Observation 2.—(i) For any value of \vec{J} there are separable states corresponding to A_k for $k \in \{x, y, z\}$.
(ii) Let us define $J := Nj$,

$$c_x := \sqrt{1 - \frac{\langle J_y \rangle^2 + \langle J_z \rangle^2}{J^2}}, \quad (22)$$

and $p := \frac{1}{2}[1 + \frac{\langle J_x \rangle}{Jc_x}]$. If Np is an integer then there exists also a separable state corresponding to B_x . Similar statements hold for B_y and B_z . Note that this condition is always fulfilled, if $\vec{J} = 0$ and N is even.

(iii) There are always separable states corresponding to points B'_k such that their distance from B_k is smaller than j^2 . In the limit $N \rightarrow \infty$ for a fixed normalized angular momentum $\frac{\vec{J}}{N}$, the points B_k and the B'_k cannot be distinguished by measurement, for that a precision j^2 or better would be needed when measuring $\langle \tilde{J}_x^2 \rangle$, which is unrealistic. Hence in the macroscopic limit the characterization is complete.

Proof. A separable state corresponding to A_x is

$$\rho_{A_x} := p(|\psi_+\rangle\langle\psi_+|)^{\otimes N} + (1-p)(|\psi_-\rangle\langle\psi_-|)^{\otimes N}. \quad (23)$$

Here $|\psi_{\pm}\rangle$ are the single-particle states with $(\langle j_x \rangle, \langle j_y \rangle, \langle j_z \rangle) = j(\pm c_x, \frac{\langle J_y \rangle}{J}, \frac{\langle J_z \rangle}{J})$.

If $M := Np$ is an integer, we can also define the state corresponding to the point B_x as

$$|\phi_{B_x}\rangle := |\psi_+\rangle^{\otimes M} \otimes |\psi_-\rangle^{\otimes (N-M)}. \quad (24)$$

Since there is a separable state for each extreme point of the polytope, for any internal point a corresponding separable state can be obtained by mixing the states corresponding to the extreme points.

If M is not an integer, we can approximate B_x by taking $m := M - \varepsilon$ as the largest integer smaller than M , defining the state

$$\begin{aligned} \rho' &:= (1-\varepsilon)(|\psi_+\rangle\langle\psi_+|)^{\otimes m} \otimes (|\psi_-\rangle\langle\psi_-|)^{\otimes (N-m)} \\ &+ \varepsilon(|\psi_+\rangle\langle\psi_+|)^{\otimes (m+1)} \otimes (|\psi_-\rangle\langle\psi_-|)^{\otimes (N-m-1)}. \end{aligned} \quad (25)$$

It has the same coordinates as B_x , except for the value of $\langle \tilde{J}_x^2 \rangle$, where the difference is $4j^2 c_x^2 \varepsilon (1-\varepsilon) \leq j^2$. ■

The extremal states that correspond to the vertices of the polytope defined by the optimal spin-squeezing inequalities are, in a certain sense, generalizations of the coherent spin states defined as [39, 68]

$$|\Psi_{\text{CSS}}\rangle = |\Psi\rangle^{\otimes N}, \quad (26)$$

where $|\Psi\rangle$ is a state with maximal $\langle j_x \rangle^2 + \langle j_y \rangle^2 + \langle j_z \rangle^2$. All states of the form (26) saturate all the inequalities, as can be seen by direct substitution into Eq. (9). Further extremal states can be obtained as tensor products or mixtures of coherent spin states. Note that they exist for all the possible values of the mean spin \vec{J} , while spin coherent states Eq. (26) were fully polarized.

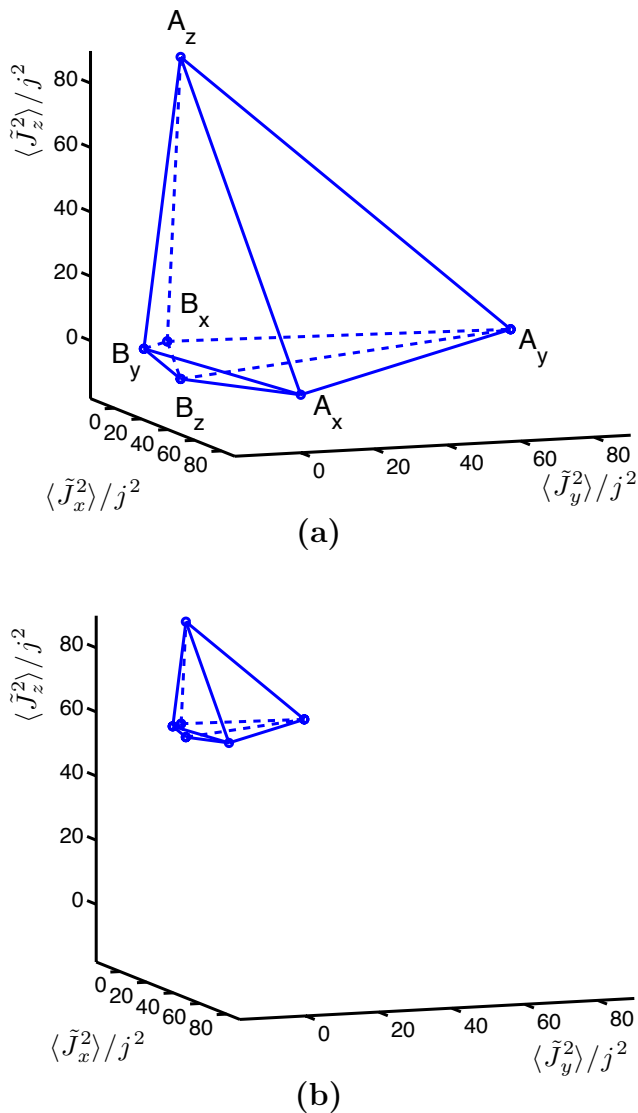


FIG. 2: (Color online) (a) The polytope of separable states corresponding to Eqs. (9) for $N = 10$ spin- j particles and for $\vec{J} = 0$. The completely mixed state defined in Eq. (39) corresponds to the origin of the coordinate axis, i.e., the point $(0, 0, 0)$ and it is inside the polytope. (b) The same polytope for $\vec{J} = (0, 0, 8)j$. Note that this polytope is a subset of the polytope in (a). For the coordinates of the points A_l and B_l see Eq. (21).

C. Relation of Eq. (9) to two-particle entanglement

Since the optimal spin-squeezing inequalities (9) contain only first moments and modified second moments of the angular momentum components, they can be reformulated with the average two-body correlations. For that, we define the average two-particle density matrix as

$$\rho_{\text{av}2} := \frac{1}{N(N-1)} \sum_{m \neq n} \rho_{mn}, \quad (27)$$

where ρ_{mn} is the two-particle reduced density matrix for the m^{th} and n^{th} particles.

Next, we formulate our entanglement conditions with the density matrix $\rho_{\text{av}2}$.

Observation 3.—The optimal spin-squeezing inequalities Eq. (9) for arbitrary spin can be given in terms of the average two-body density matrix as

$$N \sum_{l \in I} (\langle j_l \otimes j_l \rangle_{\text{av}2} - \langle j_l \otimes \mathbb{1} \rangle_{\text{av}2}^2) \geq \Sigma - j^2, \quad (28)$$

where we have defined the expression Σ as the sum of all the two-particle correlations of the local spin operators

$$\Sigma := \sum_{l=x,y,z} \langle j_l \otimes j_l \rangle_{\text{av}2}. \quad (29)$$

The right-hand side of Eq. (28) is nonpositive. For the $j = \frac{1}{2}$ case, the right-hand side of Eq. (28) is zero for all symmetric states, while for $j > \frac{1}{2}$ it is zero only for some symmetric states.

Proof. Equation (10) can be transformed into

$$N \sum_{l \in I} (\tilde{\Delta} J_l)^2 + \sum_{l \in I} \langle J_l \rangle^2 \geq \sum_l \langle \tilde{J}_l^2 \rangle - N(N-1)j^2. \quad (30)$$

Next, let us see how Eq. (30) behaves for symmetric states. We know from angular momentum theory that Eq. (9a) of the optimal spin-squeezing inequalities is saturated only when the state is symmetric. For the $j = \frac{1}{2}$ case, all symmetric states saturate Eq. (9a), while for $j > \frac{1}{2}$ only some of the symmetric states saturate it. Based on these and Eq. (14), we know that, for spin- $\frac{1}{2}$ particles in a symmetric state the right-hand side of Eq. (30) is zero. On the other hand, for spin- j particles with $j > \frac{1}{2}$ in a symmetric state, the right-hand side can also be negative.

Let us now turn to the reformulation of Eq. (30) in terms of the two-body reduced density matrix. The modified second moments and variances can be expressed with the average two-particle density matrix as

$$\begin{aligned} \langle \tilde{J}_l^2 \rangle &= \sum_{m \neq n} \langle j_l^{(n)} j_l^{(m)} \rangle = N(N-1) \langle j_l \otimes j_l \rangle_{\text{av}2}, \\ (\tilde{\Delta} J_l)^2 &= -N^2 \langle j_l \otimes \mathbb{1} \rangle_{\text{av}2}^2 + N(N-1) \langle j_l \otimes j_l \rangle_{\text{av}2}. \end{aligned} \quad (31)$$

Substituting Eq. (31) into Eq. (30), we obtain Eq. (28). As in the case of Eq. (30), the right-hand side of Eq. (28) is zero for symmetric states of spin- $\frac{1}{2}$ particles. ■

Not that, as in the spin- $\frac{1}{2}$ case, there are states detected as entangled that have a separable two-particle density matrix [55]. Such states are, for example, permutationally invariant states with certain symmetries for which the reduced single-particle density matrix is completely mixed. For large N , due to permutational invariance and the symmetries mentioned above, the two-particle density matrices are very close to the a com-

pletely mixed matrix as well and hence they are separable. Still, some of such states can be detected as entangled by the optimal spin-squeezing inequalities. Examples of such states are the permutationally invariant singlet states discussed later in Sec. III B.

D. Relation of Eq. (9) to the criterion based on the positivity of the partial transpose

Our inequalities are entanglement conditions. Thus, it is important to compare them to the most useful entanglement condition known so far, the condition based on the positivity of the partial transpose (PPT) [69].

In Ref. [55], it has been shown for the spin- $\frac{1}{2}$ case that the optimal spin-squeezing inequalities can detect the thermal states of some spin models that have a positive partial transpose for all bi-partitions of the system. Such states are extreme forms of bound entangled states: they are non-distillable even if the qubits of the two partitions are allowed to unite with each other. We found that for the $j > \frac{1}{2}$ case, the inequality (9b) also detects such bound entangled states in the thermal states of spin models. An example of such a state for $j = 1$ and $N = 3$ is

$$\varrho_{\text{BES}} \propto e^{-\frac{J_x^2 + J_y^2 + J_z^2}{T}}. \quad (32)$$

The state (32) is detected by our criterion below the temperature bound $T_s \approx 3.66$ while it is detected by the PPT criterion below the bound $T_{\text{PPT}} \approx 3.57$.

Finally, we will consider the special case of symmetric states. In this case, the PPT condition applied to the reduced two-body density matrix detects all states detected by the spin-squeezing inequalities.

Observation 4.—The PPT criterion for the average two-particle density matrix defined in Eq. (27) detects all symmetric entangled states that the optimal spin-squeezing inequalities detect for $j > \frac{1}{2}$. The two conditions are equivalent for symmetric states of particles with $j = \frac{1}{2}$.

Proof. We will connect the violation of Eq. (28) to the violation of the PPT criterion by the reduced two-particle density matrix $\varrho_{\text{av}2}$. If a quantum state is symmetric, its reduced state $\varrho_{\text{av}2}$ is also symmetric. For such states, the PPT condition is equivalent to [70]

$$\langle A \otimes A \rangle_{\text{av}2} - \langle A \otimes \mathbb{1} \rangle_{\text{av}2}^2 \geq 0 \quad (33)$$

holding for all Hermitian operators A . Based on Observation 3, it can be seen by straightforward comparison of Eqs. (28) and (33) that, for $j = \frac{1}{2}$, Eq. (28) holds for all possible choices of I and for all possible choices of coordinate axes, i.e., all possible j_l , if and only if Eq. (33) holds for all Hermitian operators A . For $j > \frac{1}{2}$ there is no equivalence between the two statements. Only from the latter follows the former. ■

III. STATES THAT VIOLATE THE OPTIMAL SPIN-SQUEEZING INEQUALITIES FOR SPIN j

In this section we will study, what kind of states violate maximally our spin-squeezing inequalities. We will also examine, how much noise can be mixed with these states such that they are still detected as entangled by our inequalities.

A. The inequality with three second moments, Eq. (9a)

The first two equations of Eqs. (9) are invariant under the exchange of coordinate axes x, y , and z . As a consequence of basic angular momentum theory, Eq. (9a), the inequality with three second moments is valid for all quantum states, thus it cannot be violated. As discussed in the proof of Observation 3, for the $j = \frac{1}{2}$ case, all symmetric states saturate Eq. (9a), while for $j > \frac{1}{2}$ only some of the symmetric states saturate it. In both cases, states of the form (26) are a subset of the saturating states.

B. The inequality with three variances, Eq. (9b)

The states maximally violating Eq. (9b) are the many-body singlet states. The characteristic values of the collective operators for many-body singlets are shown in Table I. States violating Eq. (9b) have a small variance for all the components of the angular momentum as shown in Fig. 1(c).

Let us see now some examples of many-body singlets states. For $j = \frac{1}{2}$, a pure singlet state can be constructed, for example, as a tensor product of two-particle singlets of the form

$$|\Psi^-\rangle = \frac{1}{\sqrt{2}} (|+\frac{1}{2}, -\frac{1}{2}\rangle_z - |-\frac{1}{2}, +\frac{1}{2}\rangle_z). \quad (34)$$

Any permutation of such a state is a singlet as well. The mixture of all such permutations is a permutationally invariant singlet defined as

$$\rho_{\text{s,PI}} = \frac{1}{N!} \sum_{k=1}^{N!} \Pi_k (|\Psi^-\rangle \langle \Psi^-| \otimes \cdots \otimes |\Psi^-\rangle \langle \Psi^-|) \Pi_k^\dagger, \quad (35)$$

where Π_k are all the possible permutations of the qubits. It can be shown that for even N , Eq. (35) equals the $T = 0$ thermal ground state of the Hamiltonian [58, 59]

$$H_s = J_x^2 + J_y^2 + J_z^2. \quad (36)$$

For even N and $j = \frac{1}{2}$, the state $\rho_{\text{s,PI}}$ is the only permutationally invariant singlet state. For $j = \frac{1}{2}$, all singlets are outside of the symmetric subspace.

In the case of spin-1 particles, the following two-particle symmetric state

$$|\phi_{\text{s}1}\rangle = \frac{1}{\sqrt{3}} (|1, -1\rangle + |0, 0\rangle + |-1, 1\rangle), \quad (37)$$

is also a singlet. It is very important from the point of view of experimental realizations with Bose-Einstein condensates that for $j > \frac{1}{2}$ there are singlet states in the symmetric subspace.

Next, we mix the spin- j singlet state with white noise and examine up to how much noise it is still violating Eq. (9b). The noisy singlet state is the following

$$\varrho_{\text{s,noisy}}(p_n) = (1 - p_n)\varrho_{\text{s}} + p_n\varrho_{\text{cm}}, \quad (38)$$

where ϱ_{s} is a singlet state maximally violating Eq. (9b), and p_n is the amount of noise and we defined the completely mixed state as

$$\rho_{\text{cm}} = \frac{1}{d^N} \mathbb{1}, \quad (39)$$

where the dimension of the qudit is $d = 2j + 1$. The vectors of the collective quantities ($\vec{J}_{\text{cm}}, \vec{K}_{\text{cm}}, \vec{M}_{\text{cm}}$) are shown in Table I for the completely mixed state. Based on these, simple calculations show that the state (38) is detected as entangled by Eq. (9b) if

$$p_n < \frac{1}{j+1} = \frac{2}{d+1}. \quad (40)$$

Hence, the white-noise tolerance decreases with d .

Finally note that for any j the modified second moments of the collective angular momentum components are zero for the completely mixed state, i.e.,

$$\vec{K}_{\text{cm}} = (0, 0, 0). \quad (41)$$

Thus, the completely mixed state belongs to a point at the origin of the coordinate system of the modified second moments for $\vec{J} = 0$. In contrast, in the space of true second moments the singlet state is at the origin, since for the singlet we have $\langle J_l^2 \rangle = \langle J_l \rangle = 0$ for $l = x, y, z$.

Eq. (9b) has been proposed to detect entanglement in optical lattices of cold atoms [49]. A related inequality was presented for entanglement detection in condensed matter systems by susceptibility measurements [50]. Experimentally, it has been used for entanglement detection in photonic systems [12] and in fermionic cold atoms [11]. An ensemble of d -state fermions naturally fills up the energy levels of a harmonic oscillator such that all levels have d fermions in a multipartite $SU(d)$ singlet state. Such a state is also a singlet, maximally violating the optimal spin-squeezing inequality with three variances, Eq. (9b). Singlets can also be obtained through spin squeezing in cold atomic ensembles [58, 59]. Finally, the ground state of the system Hamiltonian for certain spinor Bose-Einstein condensates is a singlet state [62].

C. The inequality with only one variance, Eq. (9c)

Next, we will consider the optimal spin-squeezing inequality with one variance Eq. (9c). This entanglement criterion is very useful to detect the almost fully polarized spin-squeezed states shown in Fig. 1(a). It can also

be used to detect symmetric Dicke states with a maximal $\langle J_x^2 + J_y^2 + J_z^2 \rangle$ and $\langle J_z \rangle = 0$. States close to such symmetric Dicke states have a small variance for one component of the angular momentum while they have a large variance in two orthogonal directions as shown in Fig. 1(b).

Dicke states $|\lambda, \lambda_z, \alpha\rangle$ are quantum states obeying the eigenequations

$$\begin{aligned} (J_x^2 + J_y^2 + J_z^2)|\lambda, \lambda_z, \alpha\rangle &= \lambda(\lambda + 1)|\lambda, \lambda_z, \alpha\rangle, \\ J_z|\lambda, \lambda_z, \alpha\rangle &= \lambda_z|\lambda, \lambda_z, \alpha\rangle, \end{aligned} \quad (42)$$

where α is a label used to distinguish the different eigenstates corresponding to the same eigenvalues λ and λ_z . In particular, we will show that Eq. (9c) is very useful to detect entanglement close to the symmetric Dicke state

$$|D_{N,j}\rangle := |Nj, 0\rangle, \quad (43)$$

where N must be even for half integer j 's. In this case, the α label is not needed, as the two eigenvalues determine the state uniquely. The state (43) for $j = \frac{1}{2}$ has already been known to have intriguing entanglement properties [43] and it is optimal for certain very general quantum metrological tasks [15].

We will now show that the state (43) maximally violates Eq. (9c) for $j = \frac{1}{2}$ and is close to violating it maximally for $j > \frac{1}{2}$. In order to show this, we rewrite Eq. (9c) for $(k, l, m) = (z, x, y)$ as

$$\begin{aligned} \langle J_x^2 + J_y^2 + J_z^2 \rangle - N(\Delta J_z)^2 - \langle J_z \rangle^2 + N \sum_n \langle (j_z^{(n)})^2 \rangle \\ \leq Nj(Nj + 1). \end{aligned} \quad (44)$$

The state (43) maximally violates Eq. (9c) for $j = \frac{1}{2}$ since it maximizes all terms with a positive coefficient and minimizes all terms with a negative one on the left-hand side of Eq. (44). This statement is almost true also for the case $j > \frac{1}{2}$, except for the term with the local second moments which has a value

$$\sum_n \langle (j_z^{(n)})^2 \rangle = \frac{N(N-1)j^2}{2jN-1}. \quad (45)$$

The proof of Eq. (45) is given in the appendix. Based on these, our symmetric Dicke state is detected as entangled for any j .

In a practical situation, it is also important to know how much additional noise is tolerated such that the noisy state is still detected as entangled. Next, we look at the noise tolerance of the inequality (44) for our case. We mix the symmetric Dicke state (43) with white noise as

$$\varrho_{\text{D,noisy}}(p_n) = (1 - p_n)|D_{N,j}\rangle\langle D_{N,j}| + p_n\varrho_{\text{cm}}. \quad (46)$$

The expectation values and the relevant moments of the collective angular momentum components for the Dicke state (43) are given in Table I. Based on these, a noisy Dicke state is detected as entangled if

$$p_{\text{noise}} < \frac{N}{N(2j+1)-1}. \quad (47)$$

TABLE I: Expectation values of collective quantities appearing in the optimal spin-squeezing inequalities (9) for various quantum states. \vec{J} , \vec{K} , and \vec{M} are defined in Eqs. (16), (18), and (19), respectively.

Singlet state discussed in Sec. III B	$\vec{J}_s = (0, 0, 0)$ $\vec{K}_s = (0, 0, 0)$ $\vec{M}_s = \left(\frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N\right)$
Completely mixed state defined in Eq. (39)	$\vec{J}_{\text{cm}} = (0, 0, 0)$ $\vec{K}_{\text{cm}} = \left(\frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N\right)$ $\vec{M}_{\text{cm}} = \left(\frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N, \frac{j(j+1)}{3}N\right)$
Symmetric Dicke state, $ D_{N,j}\rangle$, discussed in Sec. III C	$\vec{J}_D = (0, 0, 0)$ $\vec{K}_D = \left(\frac{Nj(Nj+1)}{2}, \frac{Nj(Nj+1)}{2}, 0\right)$ $\vec{M}_D = \left(\frac{Nj(j+1)}{2} - \frac{N(N-1)j^2}{4jN-2}, \frac{Nj(j+1)}{2} - \frac{N(N-1)j^2}{4jN-2}, \frac{N(N-1)j^2}{2jN-1}\right)$

For large N , the bound on the noise is $\frac{1}{2j+1}$.

Entangled states close to Dicke states have been observed in photonic experiments with a condition similar to the optimal spin-squeezing inequality with one variance, Eq. (9c) [2–4]. Symmetric Dicke states can be created dynamically in Bose-Einstein condensate [61, 62]. Cold trapped ions also seem to be ideal to create symmetric Dicke states, thus the use of our inequalities is expected even in these systems [6, 42, 71].

D. The inequality with two variances, Eq. (9d)

As the last case let us consider the optimal spin-squeezing inequality (9d). Typical states strongly violating Eq. (9d) have a small variance for two components of the angular momentum while having a large variance in the orthogonal direction, see Fig. 1(d). As we will see, for certain values for j , singlet states [Fig. 1(c)] also violate Eq. (9d).

Now it is hard to compute the maximally violating state, because an independent optimization for the different terms does not seem to lead to a state maximizing the whole expression even for $j = \frac{1}{2}$. Thus, we will consider examples of important states violating the inequality and compare it to other similar conditions.

Let us consider the multi-particle spin singlet states. Based on \vec{J}_s , \vec{K}_s , and \vec{M}_s given in Table I, we find that the optimal spin-squeezing inequality (9d) is violated whenever

$$j < \frac{2N-3}{N}. \quad (48)$$

Thus, for $N \geq 7$, the singlet state is violating this inequality for $j = \frac{1}{2}, 1$, and $\frac{3}{2}$.

An alternative of the entanglement condition with two variances (9d), the planar squeezing entanglement condition [52, 72], is of the form

$$(\Delta J_x)^2 + (\Delta J_y)^2 \geq NC_j, \quad (49)$$

where the constant C_j is $\frac{1}{4}$ for $j = \frac{1}{2}$ and $\frac{7}{16}$ for $j = 1$, respectively. For larger j , the constant C_j is determined numerically. For even N , the criterion (49) is maximally violated by the many-particle singlet state for any j .

Let us compare the entanglement condition (9d) to the planar squeezing entanglement condition (49). Using Eq. (15), Eq. (9d) can be rewritten for $(k, l, m) = (x, y, z)$ as

$$(\Delta J_x)^2 + (\Delta J_y)^2 \geq Nj + \frac{1}{N-1}\langle J_z^2 \rangle - \frac{N}{N-1}M_z. \quad (50)$$

For $j = \frac{1}{2}$ and for large N , it can be seen that the right-hand side of Eq. (50) equals $\frac{N}{4} + \frac{1}{N-1}\langle J_z^2 \rangle$. A comparison with Eq. (49) shows that our condition (50) is strictly stronger in this case. For $j > \frac{1}{2}$, Eq. (50) is not strictly stronger any more, but still is more effective in detecting quantum states with a large $\langle J_z^2 \rangle$.

This seems to be the advantage of our inequality compared to Eq. (49): It has information not only about the variances in the x and y directions, but also about the second moment in the third direction.

IV. SPIN- $\frac{1}{2}$ ENTANGLEMENT CRITERIA TRANSFORMED TO HIGHER SPINS

In this section, we present a method to map spin- $\frac{1}{2}$ entanglement criteria to criteria for higher spins. We use it to transform the original spin-squeezing parameter Eq. (2) to a spin-squeezing parameter for higher spins. We show that two of the optimal spin-squeezing inequalities are strictly stronger than the transformed original spin-squeezing criterion. We also convert some other spin- $\frac{1}{2}$ entanglement criteria to criteria for higher spins.

A. The original spin-squeezing parameter for higher spins

Next, we present a mapping that can transform every spin-squeezing inequality for an ensemble of spin- $\frac{1}{2}$ particles written in terms of the first and the modified second moments of the collective spin operators to an entanglement condition for spin- j particles, also given in terms of the first and the modified second moments.

Observation 5.—Let us consider an entanglement condition (i.e., a necessary condition for separability) for spin- $\frac{1}{2}$ particles of the form

$$f(\{\langle J_l \rangle\}, \{\langle \tilde{J}_l^2 \rangle\}) \geq \text{const.}, \quad (51)$$

where f is a six-dimensional function. Then, the inequality obtained from Eq. (51) by the substitution

$$\langle J_l \rangle \longrightarrow \frac{1}{2j} \langle J_l \rangle, \quad \langle \tilde{J}_l^2 \rangle \longrightarrow \frac{1}{4j^2} \langle \tilde{J}_l^2 \rangle. \quad (52)$$

is an entanglement condition for spin- j particles. Any quantum state that violates it is entangled.

Proof. Let us consider a product state of N spin- j particles

$$\rho_j = \bigotimes_n \rho_j^{(n)} \quad (53)$$

and define the quantities $r_l^{(n)} = \frac{1}{j} \langle j_l^{(n)} \rangle$. Then the first and modified second moments of the collective spin can be rewritten in terms of those quantities as

$$\frac{\langle J_l \rangle}{2j} = \frac{1}{2} \sum_n r_l^{(n)}, \quad \frac{\langle \tilde{J}_l^2 \rangle}{4j^2} = \frac{1}{4} \sum_{n \neq m} r_l^{(n)} r_l^{(m)}. \quad (54)$$

For the length of the single-particle Bloch vectors we have the constraints

$$0 \leq \sum_l (r_l^{(n)})^2 \leq 1. \quad (55)$$

Both the lower and the upper bound are sharp, and these are the only constraints for physical states for every j [73]. Thus, the set of allowed values for $\{\frac{1}{2j} \langle J_l \rangle\}_{l=x,y,z}$ and $\{\frac{1}{4j^2} \langle \tilde{J}_l^2 \rangle\}_{l=x,y,z}$ for product states of the form Eq. (53) are independent from j . This is also true for separable states since separable states are mixtures of product states. Let us now consider the range of

$$f(\{\frac{1}{2j} \langle J_l \rangle\}, \{\frac{1}{4j^2} \langle \tilde{J}_l^2 \rangle\}) \quad (56)$$

for separable states. We have seen that the set of allowed values for the arguments of the function in Eq. (56) for separable states is independent of j . Thus, the range of Eq. (56) for separable states is also independent of j . Hence the statement of Observation 5 follows [74]. ■

Note that the complete set of optimal spin-squeezing inequalities (9) for $j > \frac{1}{2}$ can be obtained from the complete set for the spin- $\frac{1}{2}$ case presented in Ref. [55] using Observation 5.

Next, we will transform the spin-squeezing parameter $\xi_{s,j}$ to higher spins.

Observation 6.—Based on Observation 5, the original spin-squeezing parameter defined in Eq. (2) for spin- $\frac{1}{2}$ particles is transformed into the spin-squeezing parameter Eq. (5) for spin- j particles.

Proof. Let us first write down the entanglement condition for spin- $\frac{1}{2}$ particles based on the spin-squeezing parameter (2) in terms of the modified variance as

$$\xi_s^2 \equiv N \frac{(\tilde{\Delta} J_x)^2 + \frac{N}{4}}{\langle J_y \rangle^2 + \langle J_z \rangle^2} \geq 1. \quad (57)$$

Then, we use Observation 5 to obtain

$$\xi_{s,j}^2 \equiv N \frac{(\tilde{\Delta} J_x)^2 + N j^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2} \geq 1. \quad (58)$$

■

It is instructive to rewrite Eq. (58) as

$$\xi_{s,j}^2 \equiv N \frac{(\Delta J_x)^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2} + N \frac{\sum_n [j^2 - \langle (j_x^{(n)})^2 \rangle]}{\langle J_y \rangle^2 + \langle J_z \rangle^2} \geq 1. \quad (59)$$

Equation (59) can be further reformulated such that the second term depends only on the average single-particle density matrix, $\rho_{\text{av}1}$, as

$$\xi_{s,j}^2 = N \frac{(\Delta J_x)^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2} + \frac{j^2 - \langle j_x^2 \rangle_{\text{av}1}}{\langle j_y \rangle_{\text{av}1}^2 + \langle j_z \rangle_{\text{av}1}^2}, \quad (60)$$

where

$$\rho_{\text{av}1} := \frac{1}{N} \sum_n \rho_n \equiv \text{Tr}_2(\rho_{\text{av}2}), \quad (61)$$

and ρ_n is the single-particle reduced density matrix for the n^{th} particle. Thus, in Eq. (60) we wrote down the new spin-squeezing parameter $\xi_{s,j}^2$ as the sum of the original parameter ξ_s^2 given in Eq. (2) and a second term that depends only on single particle observables and is related to single particle spin squeezing. For $j = \frac{1}{2}$, this second term in Eq. (60) is zero. For $j > \frac{1}{2}$, it is nonnegative. Hence, for $j > \frac{1}{2}$ there are states that violate Eq. (2), but do not violate $\xi_{s,j}^2 \geq 1$. This is shown in a simple example with qutrits.

Example 1.—Let us consider a multi-particle state of the form

$$|\Psi(\alpha)\rangle = (\sqrt{\alpha}|1\rangle + \sqrt{1-\alpha}|0\rangle)^{\otimes N} \quad (62)$$

for $j = 1$. For $\alpha > 0.5$ and for any $N \geq 1$, the original spin-squeezing inequality (2) is violated by the state (62). On the other hand, no separable state can violate $\xi_{s,j}^2 \geq 1$, thus, it is the correct formulation of the original spin-squeezing inequality for $j > \frac{1}{2}$.

There is another interpretation on how to use the original spin-squeezing inequality (2) for the $j > \frac{1}{2}$ case. Equation (2) is inherently for ensembles of spin- $\frac{1}{2}$ particles. When used for higher spins, N should be the number of spin- $\frac{1}{2}$ constituents rather than the number of spin- j particles. Then, Eq. (2) detects entanglement between the spin- $\frac{1}{2}$ constituents of the particles, and cannot distinguish between entanglement among the spin- j particles and entanglement within the spin- j particles [33].

Observation 7.—The optimal spin-squeezing inequality with three variances, Eq. (9b), and the one with one variance, Eq. (9c), for $(k, l, m) = (x, y, z)$ are strictly stronger than the spin-squeezing inequality $\xi_{s,j}^2 \geq 1$ [$\xi_{s,j}$ is defined in Eq. (5)], since they detect strictly more states.

Proof. To see this, let us rewrite Eq. (9c) for the particular choice of coordinate axes as

$$(N-1) \left[(\tilde{\Delta}J_x)^2 + Nj^2 \right] \geq \langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle. \quad (63)$$

Then, from Eqs. (9b) and (14) follows

$$\langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle \geq -Nj^2 + \langle J_y \rangle^2 + \langle J_z \rangle^2 - (\tilde{\Delta}J_x)^2. \quad (64)$$

Clearly, the left-hand-side of Eq. (63) is not smaller than the right-hand side of Eq. (64). Hence, the condition $\xi_{s,j}^2 \geq 1$ can be obtained.

So far we have shown that all quantum states detected by the criterion $\xi_{s,j}^2 \geq 1$ are also detected by Eq. (9b) or by Eq. (9c) for $(k, l, m) = (x, y, z)$. We have now to present a quantum state that is detected by Eq. (9b) or by Eq. (9c) but not detected by the condition $\xi_{s,j}^2 \geq 1$. Such states are the many-body singlet states or the symmetric Dicke states (43). ■

Finally, note that the original spin-squeezing parameter ξ_s^2 can also be generalized to higher spins without introducing the modified quantities, however, in this case the bounds must be obtained numerically [48].

B. Other spin- $\frac{1}{2}$ criteria transformed to higher spins

In this section, we transform two generalized spin-squeezing criteria for spin- $\frac{1}{2}$ particles found in the literature to criteria for higher spins.

First, let us consider the criterion of Refs. [41, 42], which is valid for multiqubit systems. It can be rewritten in terms of the expectation values and the modified second moments as

$$\sqrt{\left(\langle \tilde{J}_l^2 \rangle + \langle \tilde{J}_k^2 \rangle \right)^2 + (N-1)^2 \langle J_n \rangle^2 - \langle \tilde{J}_n^2 \rangle} \leq \frac{N(N-1)}{4}. \quad (65)$$

Equation (65) is violated for some choice of the coordinate axes if the average reduced two-particle state is entangled [75].

Observation 8.—Using Observation 5, Eq. (65) can be transformed to a system of spin- j particles as

$$\sqrt{\left(\langle \tilde{J}_l^2 \rangle + \langle \tilde{J}_k^2 \rangle \right)^2 + 4(N-1)^2 j^2 \langle J_n \rangle^2 - \langle \tilde{J}_n^2 \rangle} \leq N(N-1)j^2. \quad (66)$$

As a second example, let us consider now the entanglement condition based on the planar squeezing inequality [52] for $j = \frac{1}{2}$ [i.e., Eq. (49) with $C_j = \frac{1}{4}$].

Observation 9.—Using Observation 5, the planar squeezing criterion can be transformed to particles with $j > \frac{1}{2}$ as

$$(\tilde{\Delta}J_x)^2 + (\tilde{\Delta}J_y)^2 \geq -Nj^2. \quad (67)$$

It is instructive to compare Eq. (67) to the planar spin-squeezing inequality Eq. (49). Note again that Eq. (67) is analytical for any j , while Eq. (49) is based on numerics.

V. A STRONGER ALTERNATIVE OF THE ORIGINAL SPIN-SQUEEZING PARAMETER

In this section, we show that the spin-squeezing parameter ξ_{os}^2 given in Eq. (6), based on the optimal spin-squeezing inequality (9c), is stronger than $\xi_{s,j}^2$ [Eq. (5)]. In particular, it not only detects almost completely polarized spin-squeezed quantum states, but also quantum states for which $\langle J_l \rangle = 0$ for $l = x, y, z$, e.g., Dicke states.

How can one obtain a spin-squeezing parameter based on an entanglement condition given as an inequality? We will use the most straightforward way and divide the right-hand side of the inequality by the left-hand side, after some rearrangement of the terms. After completing our calculations, we became aware that the parameter (7) has appeared in Ref. [45]. It was obtained in the way described above from one of the optimal spin-squeezing inequalities for the spin- $\frac{1}{2}$ case [i.e., Eq. (9c) with $j = \frac{1}{2}$] given in Ref. [55]. It was used to study the entanglement dynamics in the modified Lipkin-Meshkov-Glick model and its time evolution was found to be similar to the time evolution of ξ_s^2 . Reference [45] also describes a phase space method for the efficient calculation of the spin-squeezing parameters for large systems [76].

Next, we show explicitly the relation between the spin-squeezing parameter Eq. (6) and the corresponding optimal spin-squeezing inequality (9c). Then, we prove important properties of the parameter.

Observation 10.—A spin-squeezing parameter ξ_{os}^2 based on the optimal spin-squeezing inequality with one variance, Eq. (9c), can be defined as given in Eq. (6). Equation (9c) for $(k, l, m) = (x, y, z)$ is violated if and only if $\xi_{os}^2 < 1$.

Proof. Equation (9c) can be rewritten as

$$\langle \tilde{J}_l^2 \rangle + \langle \tilde{J}_m^2 \rangle \leq (N-1)[(\tilde{\Delta}J_k)^2 + Nj^2]. \quad (68)$$

The spin-squeezing parameter Eq. (6) can be obtained after dividing the right-hand side of Eq. (68) by its left-hand side. Such a derivation is valid only if the left-hand side of Eq. (68) is positive. Straightforward calculations show that if the left-hand side of Eq. (68) is nonpositive then Eq. (9c) cannot be violated for $(k, l, m) = (x, y, z)$. ■

We will now show that ξ_{os}^2 is comparable to the original spin-squeezing parameter ξ_{s}^2 .

Observation 11.—For large N and $\xi_{\text{s},j}^2 < 1$, the spin-squeezing parameter ξ_{os}^2 is smaller than $\xi_{\text{s},j}^2$, i.e., Eq. (8) holds. Thus all states detected by $\xi_{\text{s},j}^2$ are also detected by ξ_{os}^2 and the squeezing parameter ξ_{os}^2 is even more sensitive.

Proof. The basic idea of the proof is that for large N the parameter ξ_{os}^2 defined in Eq. (6) can be obtained from $\xi_{\text{s},j}^2$ given in Eq. (5) by replacing $\langle J_l \rangle^2$ with $\langle \tilde{J}_l^2 \rangle$ for $l = y, z$. Knowing that

$$\langle \tilde{J}_l^2 \rangle \approx \langle J_l^2 \rangle \geq \langle J_l \rangle^2 \quad (69)$$

proves the claim.

We will now present a formal derivation. Straightforward algebra leads from Eq. (5) to

$$\xi_{\text{s},j}^2 = N \frac{[(\tilde{\Delta}J_x)^2 + Nj^2] - j(j+1)\xi_{\text{s},j}^2}{\langle J_y \rangle^2 + \langle J_z \rangle^2 - Nj(j+1)}. \quad (70)$$

Let us consider first the case when the denominator of Eq. (70) is positive. Then, we need the relation between the expectation values and the second moments

$$\langle J_l^2 \rangle \geq \langle J_l \rangle^2, \quad (71)$$

and the relation between the modified second moments and the true second moments

$$\langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle \geq \langle J_y^2 \rangle + \langle J_z^2 \rangle - Nj(j+1). \quad (72)$$

Equation (72) can be easily derived from Eq. (14). Based on Eqs. (71) and (72), we obtain an inequality for the usual spin-squeezing parameter

$$\xi_{\text{s},j}^2 \geq (N-1) \frac{[(\tilde{\Delta}J_x)^2 + Nj^2] - j(j+1)\xi_{\text{s},j}^2}{\langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle}. \quad (73)$$

Let us compare Eq. (73) with the fraction in Eq. (6). One can see that the only difference is the $j(j+1)\xi_{\text{s},j}^2$ term in the numerator of Eq. (73). If $\xi_{\text{s},j}^2 < 1$ then for large N the first term in the numerator in Eq. (73) is much larger than the second one

$$[(\tilde{\Delta}J_x)^2 + Nj^2] \gg j(j+1)\xi_{\text{s},j}^2. \quad (74)$$

This can be seen noting that $(\tilde{\Delta}J_x)^2 + Nj^2 \geq (\Delta J_x)^2$ holds and for large particle numbers the variance of an angular momentum component is, in practice, much

larger than ~ 1 . Thus, for large particle numbers the right-hand side of Eq. (73) equals ξ_{os}^2 .

Finally, note that if the denominator of Eq. (70) is nonpositive then the condition $\xi_{\text{s},j}^2 < 1$ can be satisfied only if $(\tilde{\Delta}J_x)^2 + Nj^2 \leq j(j+1)$ which would be possible if $(\tilde{\Delta}J_x)^2 \sim 1$ and hence is not realistic for large particle numbers. ■

Observation 11 is valid only for large particle numbers. For small particles numbers, there are quantum states that are detected by the original spin-squeezing parameter generalized for arbitrary spin, Eq. (5), but not detected by the spin-squeezing parameter ξ_{os}^2 defined in Eq. (6). For instance, such a state is a ground state of the five-qubit Hamiltonian

$$H_5 = J_x^2 + \frac{1}{4}J_z^2 + \frac{3}{4}J_z. \quad (75)$$

The Hamiltonian (75) has a four dimensional subspace of ground states. Any state in this subspace has $\xi_{\text{s}}^2 = 0.97$ while $\xi_{\text{os}}^2 = 1.29$. Due to Observation 7, these states must violate the optimal spin-squeezing inequality with three variances, Eq. (9b), which can be verified by direct calculation.

It is instructive to see, how the spin-squeezing parameter ξ_{os}^2 behaves for an ensemble of particles almost fully polarized in the z direction. For a fully polarized ensemble, the first and second moments of the angular momentum components are

$$\begin{aligned} \langle J_x^2 \rangle &= \langle J_y^2 \rangle = \frac{1}{2}Nj, & \langle J_z^2 \rangle &= N^2j^2, \\ \langle J_x \rangle &= \langle J_y \rangle = 0, & \langle J_z \rangle &= Nj. \end{aligned} \quad (76)$$

Based on these, we obtain the following formulas, which are approximately valid for almost fully polarized ensembles

$$\xi_{\text{os}}^2 \approx \frac{(\tilde{\Delta}J_x)^2 + Nj^2}{Nj^2 + \frac{1}{(N-1)}[\langle J_y^2 \rangle + \sum_n \langle (j_x^{(n)})^2 \rangle - Nj]}, \quad (77a)$$

$$\xi_{\text{s},j}^2 \approx \frac{(\tilde{\Delta}J_x)^2 + Nj^2}{Nj^2}. \quad (77b)$$

In Eq. (77), we substituted the value for completely polarized states for $\langle J_z \rangle$ and $\langle J_z^2 \rangle$. We also used Eq. (15) to eliminate j_y and j_z from Eq. (77a). The second term in the denominator of Eq. (77a) is negligible compared to the first term which is $\propto N$. Hence, the two spin-squeezing parameters are approximately equal

$$\xi_{\text{os}}^2 \approx \xi_{\text{s},j}^2. \quad (78)$$

Thus, the spin-squeezing parameter ξ_{os}^2 detects the fully polarized entangled states detected by $\xi_{\text{s},j}^2$.

In practical situations, the almost completely polarized state is mixed with noise. Next, we will discuss noisy spin-squeezed states.

Observation 12.—The spin-squeezing parameter ξ_{os}^2 is much more efficient than $\xi_{\text{s},j}^2$ in detecting almost completely polarized spin-squeezed states mixed with white noise.

Proof. Let us consider a state ϱ that is almost completely polarized in the z direction and spin-squeezed in the x direction. After mixing ϱ with white noise, we obtain

$$\varrho_{\text{noisy}}(p_n) = (1 - p_n)\varrho + p_n\rho_{\text{cm}}, \quad (79)$$

where p_n is the ratio of noise and ρ_{cm} is defined in Eq. (39). Then, using that we have $\langle J_x \rangle = 0$, straightforward calculations show that the original spin-squeezing parameter increases more

$$\xi_{s,j,\text{noisy}}^2 = \frac{1}{(1 - p_n)}\xi_{s,j}^2 + \frac{p_n}{(1 - p_n)^2} \frac{N^2 j^2}{(\langle J_y \rangle^2 + \langle J_z \rangle^2)}, \quad (80)$$

than our alternative spin-squeezing parameter

$$\xi_{\text{os},\text{noisy}}^2 = \xi_{\text{os}}^2 + \frac{p_n}{1 - p_n} \frac{N(N - 1)j^2}{(\langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle)}. \quad (81)$$

Since Eq. (78) and $\langle J_y \rangle^2 + \langle J_z \rangle^2 \approx \langle \tilde{J}_y^2 \rangle + \langle \tilde{J}_z^2 \rangle$ hold for almost fully polarized spin-squeezed states and for large particle numbers, we obtain

$$\xi_{\text{os},\text{noisy}}^2 \approx \xi_{s,j,\text{noisy}}^2 (1 - p_n). \quad (82)$$

This proves our claim. \blacksquare

Besides almost completely polarized states, our spin-squeezing parameter ξ_{os}^2 can also detect the entanglement of unpolarized states. This is due to the fact that it is defined in Eq. (6) based on the spin-squeezing inequality (9c), which can be used to detect the symmetric Dicke state $|D_{N,j}\rangle$, given in Eq. (43). Such states have $\langle J_l \rangle = 0$ for $l = x, y, z$, and thus they are not detected by $\xi_{s,j}^2$ [77]. We will now analyze how it is possible that Eq. (6) can be used to detect both usual spin-squeezed states with a large polarization $|\vec{J}|$ and states with $\vec{J} = 0$.

For that, let us rewrite Eq. (6) such that the denominator contains both variances of the spin components and their expectation values

$$\xi_{\text{os}}^2 = (N - 1) \frac{(\tilde{\Delta}J_x)^2 + Nj^2}{(\tilde{\Delta}J_y)^2 + (\tilde{\Delta}J_z)^2 + \langle J_y \rangle^2 + \langle J_z \rangle^2}. \quad (83)$$

Thus, the states detected by $\xi_{\text{os}}^2 < 1$ have to have a small variance of a spin component in some direction. Then, in the orthogonal directions either they have to have a large spin component or a large variance of one of those spin components.

Let us see now an application of the ideas above.

Observation 13.—Consider a set of quantum states ϱ_k such that (i) ϱ_k are all detected as entangled by the spin-squeezing parameter ξ_{os}^2 and (ii)

$$(\tilde{\Delta}J_x)_{\varrho_k}^2 = (\tilde{\Delta}J_x)_{\varrho}^2, \quad (84)$$

for all k . Then, their mixture

$$\varrho = \sum_k p_k \varrho_k \quad (85)$$

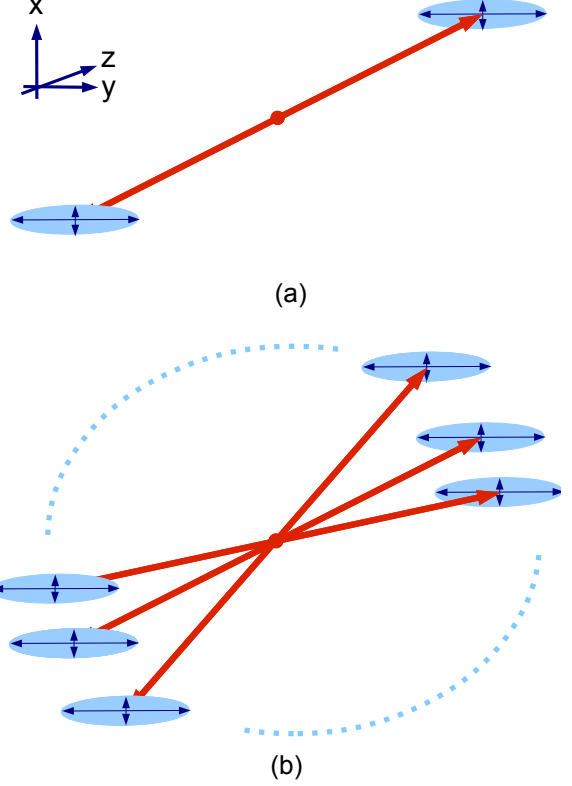


FIG. 3: (Color online) Different types of spin-squeezed states violating the criterion based on the spin-squeezing parameter ξ_{os}^2 [Eq. (6)] having $\vec{J} = 0$. (a) Mixture of two almost completely polarized spin-squeezed states pointing into opposite directions. (b) Mixture of several almost completely polarized spin-squeezed states. The original spin-squeezing inequality based on the spin-squeezing parameter and its generalization for arbitrary spin j , $\xi_{s,j}^2$ [Eq. (5)] cannot detect these states since for these states the mean spin is zero.

for $p_k > 0$ and $\sum_k p_k = 1$ is always detected as entangled by ξ_{os}^2 . This is not the case for the spin-squeezing parameter $\xi_{s,j}^2$, defined in Eq. (5). For an illustration, see Fig. 3.

Proof. The observation can be proved by straightforward substitution of Eqs. (84) and (85) into Eq. (6). \blacksquare

Following Observation 13, let us consider a spin-squeezed state ϱ_{ss} of many particles that is almost completely polarized in the z direction and spin squeezed along the x direction. Such a state is detected by the spin-squeezing parameter ξ_{os}^2 defined in Eq. (5) and also by the parameter $\xi_{s,j}^2$. Due to Observation 13, the following state is also detected by ξ_{os}^2

$$\varrho_{\text{ss,rot}} = \frac{1}{2\pi} \int_0^{2\pi} d\phi e^{-iJ_x\phi} \varrho_{\text{ss}} e^{+iJ_x\phi}. \quad (86)$$

The quantum state (86) has $\langle J_l \rangle = 0$ for $l = x, y, z$, a

large value for $(\tilde{\Delta}J_y)^2 + (\tilde{\Delta}J_z)^2$ and a small value for $(\tilde{\Delta}J_x)^2$. From the point of view of collective observables, the state (86) is similar to the symmetric Dicke state given in Eq. (43). Such a state is clearly not detected as entangled by the parameter $\xi_{s,j}^2$. The state $\rho_{ss,rot}$ is depicted in Fig. 3(b).

Finally, note that spin-squeezing parameters can be defined based on the optimal spin squeezing inequality with three variances Eq. (9b) as [58]

$$\xi_{\text{singlet}}^2 = \frac{\sum_l (\Delta J_l)^2}{Nj}. \quad (87)$$

For a pure state, the quantity $N\xi_{\text{singlet}}^2$ gives an upper bound on the number of particles not entangled with other particles [58, 78]. ξ_{singlet}^2 can also be interpreted through connections to robustness measures [47].

It is also possible to define a spin squeezing parameter based on the inequality with two variances Eq. (9d) as

$$\xi_{\text{planar squeezing}}^2 = (N-1) \frac{(\tilde{\Delta}J_x)^2 + (\tilde{\Delta}J_y)^2 + Nj^2}{\langle \tilde{J}_z^2 \rangle}. \quad (88)$$

If the parameter (88) is smaller than 1, and the denominator is positive then the state is entangled. Equation (88) can be used to characterize planar squeezing.

VI. FURTHER CONSIDERATIONS

Next, we will discuss several issues connected mostly to practical aspects of using the spin squeezing inequalities for entanglement detection.

A. The nematic tensor and single-particle spin squeezing

In this section we discuss that single-particle spin squeezing becomes possible for particles with $j > \frac{1}{2}$, and it is characterised by the local second moments.

As mentioned in the introduction, for spin- $\frac{1}{2}$ particles the local second moments $\langle \sum_n (j_l^{(n)})^2 \rangle$ are constants. For $j > \frac{1}{2}$, the local second moments are not constants any more. In order to characterize the collective local second moments in any direction, we introduce the following matrix

$$Q_{kl} := \frac{1}{N} \sum_n \left(\frac{1}{2} \langle j_k^{(n)} j_l^{(n)} + j_l^{(n)} j_k^{(n)} \rangle - Q_0 \delta_{kl} \right), \quad (89)$$

where for convenience we define

$$Q_0 := \frac{j(j+1)}{3}. \quad (90)$$

The traceless Q matrix is the rank-2 quadrupole or nematic tensor [62, 79–84]. It depends only on the average single particle density matrix thus it can be rewritten as

$$Q_{kl} = \left(\frac{1}{2} \langle j_k j_l + j_l j_k \rangle_{\text{av1}} - Q_0 \delta_{kl} \right), \quad (91)$$

where the average single-body density matrix ρ_{av1} is defined in Eq. (61). The second moment of any angular momentum component can be obtained as

$$\langle j_{\vec{n}}^2 \rangle_{\text{av1}} = \vec{n}^T (Q + Q_0 \mathbf{1}) \vec{n}, \quad (92)$$

where the unit vector \vec{n} describes the direction of the component.

The matrix Q , together with the average single particle spin

$$\langle \vec{j} \rangle = \frac{1}{N} (\langle J_x \rangle, \langle J_y \rangle, \langle J_z \rangle), \quad (93)$$

contains all the information to calculate the single-particle average spin squeezing parameter

$$\xi_{s,j,\text{av1}}^2 = 2j \frac{(\Delta j_{\vec{n}})_{\text{av1}}^2}{\langle j_{\vec{n}\perp 1} \rangle_{\text{av1}}^2 + \langle j_{\vec{n}\perp 2} \rangle_{\text{av1}}^2}, \quad (94)$$

where \vec{n} is some direction, and $j_{\vec{n}\perp k}$ are two directions perpendicular to \vec{n} and to each other. If $\xi_{s,j,\text{av1}}^2 < 1$ then there is entanglement between the $2j$ spin- $\frac{1}{2}$ constituents within the average single-particle state [85]. For $j > \frac{1}{2}$, it is possible to obtain spin squeezing within the particles, which can lead to improvement in metrological applications, but does not involve interparticle entanglement [33, 86].

In Eq. (4), we defined the modified second moments and modified variances that do not contain the local second moments. Thus, our inequalities for the spin- j particles can be interpreted as entanglement conditions that separate the entanglement between the spin- $\frac{1}{2}$ constituents of the spin- j particles and entanglement between the spin- j particles. Our inequalities detect only spin squeezing due to interparticle entanglement.

B. Coordinate system independent form of the spin squeezing inequalities

In this section, we show how to write down the optimal spin squeezing inequalities for a general j in a form that is independent from the choice of the coordinate axes. Such a form of our inequalities is very useful, as one does not have to look for the optimal choice of the coordinate axes for the spin squeezing inequalities to detect a given quantum state as entangled.

First, we define the quantities that are necessary to characterize the second moments and covariances of collective angular momentum components [87]

$$\begin{aligned} C_{kl} &:= \frac{1}{2} \langle J_k J_l + J_l J_k \rangle, \\ \gamma_{kl} &:= C_{kl} - \langle J_k \rangle \langle J_l \rangle. \end{aligned} \quad (95)$$

The matrices C and γ have already been defined for the optimal spin squeezing inequalities for $j = \frac{1}{2}$ [55, 88]. For the $j > \frac{1}{2}$ case, we also need the nematic matrix Q given in Eq. (89) to characterize the local second moments of the angular momentum coordinates.

Based on these, we define the matrix that will play a central role in our entanglement conditions

$$\mathfrak{X} := (N-1)\gamma + C - N^2Q. \quad (96)$$

The matrix \mathfrak{X} has also been introduced for spin- $\frac{1}{2}$ particles in Ref. [55]. For such systems $Q = 0 \cdot \mathbb{1}$ and $\mathfrak{X} = (N-1)\gamma + C$, which agrees with the definition in Ref. [55].

We can now present our coordinate system independent entanglement criteria.

Observation 14.—The coordinate system independent form of the optimal spin squeezing inequalities for spin- j particles is

$$\text{Tr}(C) \leq Nj(Nj+1), \quad (97a)$$

$$\text{Tr}(\gamma) \geq Nj, \quad (97b)$$

$$\lambda_{\min}(\mathfrak{X}) \geq \text{Tr}(C) - Nj(Nj+1) + N^2Q_0, \quad (97c)$$

$$\lambda_{\max}(\mathfrak{X}) \leq (N-1)\text{Tr}(\gamma) - N(N-1)j + N^2Q_0, \quad (97d)$$

where $\lambda_{\min}(A)$ and $\lambda_{\max}(A)$ are the smallest and largest eigenvalues of the matrix A , respectively.

Proof. Equation (97a) can be obtained straightforwardly by replacing the sum of the three second moments by $\text{Tr}(C)$ on the left-hand sides of Eq. (9a). Similarly, Eq. (97b) can be obtained by replacing the sum of the three variances by $\text{Tr}(\gamma)$ on the left-hand side of Eq. (9b).

In order to obtain Eq. (97c) from Eq. (9c), we need to add $\langle \tilde{J}_k^2 \rangle$ to both sides of Eq. (9c)

$$\langle \tilde{J}_k^2 \rangle + \langle \tilde{J}_l^2 \rangle + \langle \tilde{J}_m^2 \rangle - N(N-1)j^2 \leq (N-1)(\tilde{\Delta}J_k)^2 + \langle \tilde{J}_k^2 \rangle. \quad (98)$$

Then, we need to write down explicitly a diagonal element of the matrix defined in Eq. (96) with the modified second moments and variances as

$$\mathfrak{X}_{kk} = (N-1)(\tilde{\Delta}J_k)^2 + \langle \tilde{J}_k^2 \rangle + N^2Q_0, \quad (99)$$

where $k \in \{x, y, z\}$. Using Eqs. (14) and (99), the optimal spin squeezing inequality with a single variance, Eq. (98), can be rewritten as

$$\mathfrak{X}_{kk} - N^2Q_0 \geq \text{Tr}(C) - Nj(Nj+1). \quad (100)$$

\mathfrak{X}_{kk} is the only quantity in Eq. (100) that depends on the choice of coordinate axes. Equation (100) is violated for some choice of the coordinate axes, if and only if Eq. (97c) is violated. A similar derivation leads from Eq. (9d) to Eq. (97d). ■

C. Measuring the second moments of local operators

In this section, we will discuss the additional complexity arising from the need to measure the modified second moments of the collective angular momentum components, given in Eq. (4), rather than the true second

moments, for $j > \frac{1}{2}$. We will show that for each inequality it is sufficient to measure at most only one of the quantities M_l defined in Eq. (19).

Let us now take the four inequalities in Eq. (9) and examine whether they need the measurement of the modified second moments. Two of the inequalities, namely Eqs. (9a) and (9b), are already written in terms of the true variances and second moments. In Eq. (9c), all the three expectation values, M_k , M_l , and M_m , appear. Based on Eq. (15), from Eq. (9c) we obtain

$$(N-1)(\Delta J_k)^2 - NM_k \geq -Nj(Nj+1) + [\langle J_l^2 \rangle + \langle J_m^2 \rangle]. \quad (101)$$

In an analogous way, we can transform Eq. (9d) to

$$(N-1)[(\Delta J_k)^2 + (\Delta J_l)^2] \geq N(N-1)j + \langle J_m^2 \rangle - NM_m. \quad (102)$$

Note that a similar equation has already been used in Eq. (50) to describe planar squeezing. It can be seen explicitly that both for Eqs. (101) and (102) only the measurement of one of the second moments of the local operators is needed.

Measuring the expectation value of the operator $\sum_n (j_l^{(n)})^2$ can be realized in two different ways: (i) by rotating the spin by a magnetic field, and then measuring the populations of the j_z eigenstates. Let us denote the eigenvalues of j_z by χ_z . The sum of the local second moments can be obtained with the populations of the j_z eigenstates, N_{χ_z} , as

$$M_z = \sum_{\chi_z = -j, -j+1, \dots, j} N_{\chi_z} \chi_z^2. \quad (103)$$

For spin-1 systems, $M_z = N_{-1} + N_{+1} = N - N_0$.

(ii) In some cold atomic systems, such operators might also be measured directly, as in such systems in the Hamiltonian a $(j_l^{(n)})^2$ term coupled to the pseudo spin of the light appears [30, 89, 90].

One might try to eliminate the need for measuring quantities of the type $\langle M_m \rangle$ in Eq. (101) by looking for the minimum of $(\Delta J_k)^2$ for a given $[\langle J_l^2 \rangle + \langle J_m^2 \rangle]$. This problem is very complex and possibly can only be solved numerically for large spins. Analogously, a condition similar to Eq. (102), but without the need for measuring $\langle M_m \rangle$ can be obtained by looking for the minimum of $[(\Delta J_k)^2 + (\Delta J_l)^2]$ for a given $\langle J_m^2 \rangle$.

VII. SUMMARY

In summary, we have presented a complete set of generalized spin squeezing inequalities for detecting entanglement in an ensemble of spin- j particles with $j > \frac{1}{2}$ based on knowing only $\langle J_l \rangle$ and the modified second moments $\langle \tilde{J}_l^2 \rangle$ for $l = x, y, z$. We have called the inequalities

optimal spin squeezing inequalities for spin- j particles. We have also presented a mapping from spin-squeezing inequalities valid in qubit systems to spin-squeezing inequalities valid in qudit systems. We have shown how to transform the original spin squeezing parameter to an ensemble of particles with a spin larger than $\frac{1}{2}$. We have shown that a new spin squeezing parameter based on the optimal spin squeezing inequality with a single variance is, for large particle numbers, strictly stronger than the original spin squeezing parameter and its version mapped to higher spins. We have also examined the entanglement properties of the states detected by our inequalities and computed the noise tolerances of our inequalities for these states. We have also discussed how to measure the modified second moments in experiments.

In the future, it would be interesting to extend our research to entanglement conditions based on collective observables different from angular momentum operators, with collective operators based on the $SU(d)$ generators [66, 91]. Moreover, it would also be interesting to find entanglement conditions with the true second moments, without the need for measuring the modified second moments even if this involves numerical calculations rather than analytical ones.

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Appendix A: Proof of Eq. (45)

In this appendix, we present a proof of the formula Eq. (45) for symmetric Dicke states with a maximal $\langle J_x^2 + J_y^2 + J_z^2 \rangle$ and $\langle J_z \rangle = 0$. For completeness, we will consider a more general case, namely, states for which $\langle J_z \rangle \neq 0$ is also allowed. For carrying out our calculations, we need to map states of N spin- j particles to states of $2Nj$ spin- $\frac{1}{2}$ particles.

Observation 15.—Let us consider symmetric Dicke states of N spin- j particles

$$|Nj, \lambda_z\rangle_j, \quad (\text{A1})$$

which fulfill the eigenequations

$$\begin{aligned} (J_x^2 + J_y^2 + J_z^2)|Nj, \lambda_z\rangle_j &= Nj(Nj+1)|Nj, \lambda_z\rangle_j, \\ J_z|Nj, \lambda_z\rangle_j &= \lambda_z|Nj, \lambda_z\rangle_j, \end{aligned} \quad (\text{A2})$$

and the subscript j indicates that they are states of spin- j particles. For such states,

$$\sum_n \langle (j_z^{(n)})^2 \rangle = \frac{N(N-1)j^2}{2jN-1} + \frac{2j-1}{(2Nj-1)} \lambda_z^2. \quad (\text{A3})$$

Proof. First note for the quantum state (A1) $\langle J_x^2 + J_y^2 + J_z^2 \rangle$ is maximal. All such states are uniquely characterised by the two eigenvalues in the eigenequations Eq. (A2). Thus, a third parameter to distinguish states with degenerate eigenvalues is not needed.

Analogously, a symmetric Dicke state of $2Nj$ spin- $\frac{1}{2}$ particles satisfying also the property that $\langle J_x^2 + J_y^2 + J_z^2 \rangle$ is maximal can be denoted as

$$|Nj, \lambda_z\rangle_{\frac{1}{2}}, \quad (\text{A4})$$

where quantum state (A4) also fulfils the eigenequations Eq. (A2).

The symmetric Dicke state of spin- j particles, Eq. (A1), can be mapped to the Dicke state of spin- $\frac{1}{2}$ particles, Eq. (A4)

$$|Nj, \lambda_z\rangle_j \rightarrow |Nj, \lambda_z\rangle_{\frac{1}{2}}. \quad (\text{A5})$$

The moments of the collective angular momentum components, $\langle J_l^m \rangle$, are the same for the states $|Nj, \lambda_z\rangle_j$ and $|Nj, \lambda_z\rangle_{\frac{1}{2}}$. We can imagine that we represent the spin- j particle as $2j$ spin- $\frac{1}{2}$ particles in a symmetric state. For example, the spin-1 state $|0\rangle$ is mapped to a symmetric two-qubit state

$$|1, 0\rangle_1 \equiv |0\rangle \rightarrow |1, 0\rangle_{\frac{1}{2}} \equiv \frac{1}{\sqrt{2}}(|+\frac{1}{2}, -\frac{1}{2}\rangle + |-\frac{1}{2}, +\frac{1}{2}\rangle). \quad (\text{A6})$$

Operators can be mapped in an analogous way. The expectation value of the square of the single-particle operator for spin- j particles can be expressed with operators acting on the spin- $\frac{1}{2}$ state as

$$\langle (j_z^{(n)})^2 \rangle_j = \left\langle \left(\sum_{k=1}^{2j} j_z^{(n,k)} \right)^2 \right\rangle_{\frac{1}{2}}. \quad (\text{A7})$$

The left-hand side is an expectation value evaluated on $|Nj, \lambda_z\rangle_j$, while the right-hand side is an expectation value evaluated on $|Nj, \lambda_z\rangle_{\frac{1}{2}}$. The superscript (n, k) denotes the k^{th} spin- $\frac{1}{2}$ constituent of the to the n^{th} qudit.

In the following, we will refer only to the state with spin- $\frac{1}{2}$ particles and hence will omit the $\frac{1}{2}$ subscript. Due to symmetry we can express the right-hand side of Eq. (A7) with single-particle and two-particle expectation values as

$$\left\langle \left(\sum_{k=1}^{2j} j_z^{(n,k)} \right)^2 \right\rangle = 2j \langle (j_z^{(n,1)})^2 \rangle + 2j(2j-1) \langle j_z^{(n,1)} j_z^{(n,2)} \rangle. \quad (\text{A8})$$

The single-particle second moment is

$$\langle (j_z^{(n,1)})^2 \rangle = \frac{1}{4}. \quad (\text{A9})$$

Moreover, the two-body correlations can be calculated from $\langle J_z^2 \rangle = \lambda_z^2$ as

$$\langle j_z^{(n,1)} j_z^{(n,2)} \rangle = -\frac{1}{4(2Nj-1)} + \frac{1}{2Nj(2Nj-1)} \lambda_z^2. \quad (\text{A10})$$

Substituting Eqs. (A9) and (A10) into Eq. (A8), Eq. (A3) follows. ■

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