Fisher information and multiparticle entanglement

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The Fisher information F gives a limit to the ultimate precision achievable in a phase estimation protocol. It has been shown recently that the Fisher information for a linear two-mode interferometer cannot exceed the number of particles if the input state is separable. As a direct consequence, with such input states the shot-noise limit is the ultimate limit of precision. In this work, we go a step further by deducing bounds on F for several multiparticle entanglement classes. These bounds imply that genuine multiparticle entanglement is needed for reaching the highest sensitivities in quantum interferometry. We further compute similar bounds on the average Fisher information $\overline{F}$ for collective spin operators, where the average is performed over all possible spin directions. We show that these criteria detect different sets of states and illustrate their strengths by considering several examples, also using experimental data. In particular, the criterion based on $\overline{F}$ is able to detect certain bound entangled states.

I. INTRODUCTION

Entanglement is a distinguishing feature of quantum theory and will play a key role in the development of future technologies. Indeed, by using many-particle entangled states it is possible to perform several tasks better than feasible with any classical means [1]. A valuable example is the estimation of a phase shift θ as done in quantum interferometry [2–4]. In this case, by using a probe state of N classically correlated particles it is possible to reach, at maximum, a phase uncertainty which scales as $\Delta\theta \sim 1/\sqrt{N}$. This bound, generally indicated as the shot noise limit, is not fundamental and can be surpassed by preparing the N particles in a proper entangled state. It is therefore important to have a precise classification of entangled states and study their usefulness for specific applications.

While the structure of the set of entangled bipartite quantum states is understood quite well, less is known about the classification and quantification of the entanglement of multipartite quantum states [5–8]. Commonly applied criteria to distinguish between different entanglement classes include entanglement witnesses [9–12], criteria inspired by or derived from Bell inequalities [13–21], and spin-squeezing inequalities [22–25]. Recently, other approaches have led to criteria which can be evaluated directly from elements of the density matrix [26,27]. Further recent work on the detection of multiparticle entanglement can be found in Refs. [28–31] and in the recent review Ref. [8].

In this paper, we introduce criteria which can distinguish between different multipartite entanglement classes and which

are deeply connected to phase estimation. This extends previous works [22,32–36] on the interplay between entanglement and phase sensitivity. Our criteria are based on the quantum Fisher information (QFI) for linear two-mode transformations and can be easily computed for any density matrix ρ of an arbitrary number of particles. The first set of criteria is obtained by optimizing the QFI for different multipartite entanglement classes. We discuss bounds on the QFI that can be beaten only by increasing the number of entangled particles in the probe state. Our classification distinguishes quantum phase estimation in the sense that genuine multiparticle entanglement is necessary to accomplish this quantum task in the best possible way. The second set of criteria is based on the QFI for linear collective spin operators, averaged over all spin directions in the Bloch sphere. The sets of states detected by the two criteria are different and not contained in each other. We consider several examples in order to assess the strength of the criteria. In particular, using experimental data we apply our criteria for several states of $N = 4$ photons.

The article is organized as follows. We start by introducing the basic concepts related to general phase estimation protocols, linear two-mode interferometers, and the classification of multiparticle entanglement in Sec. II. In Sec. III we derive and compare the entanglement criteria based on the QFI and on the average QFI. In Sec. IV, we apply the criteria to several families of entangled states, including experimental data. We conclude in Sec. V.

II. BASIC CONCEPTS

A. Phase estimation and entanglement

In a general phase estimation scenario, a probe state ρ is transformed into $\rho(\theta) = e^{-i\theta\hat{H}} \rho \, e^{+i\theta\hat{H}}$, depending on the (unknown) phase shift θ and the operator $\hat{H}$. The phase shift

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is inferred as the value assumed by an estimator, $\theta_{est}(\{\mu_i\}_m)$, depending on the results $\{\mu_i\}_m = \{\mu_1, \ldots, \mu_m\}$ of m independent repeated measurements of a positive operator valued measurement (POVM) with elements $\{\hat{E}_\mu\}_\mu$. We indicate with $\langle\theta_{est}\rangle$ and $(\Delta\theta_{est})^2 = \langle\theta_{est}^2\rangle - \langle\theta_{est}\rangle^2$ the mean value and variance of the estimator, respectively, calculated over all possible sequences $\{\mu_i\}_m$. If the estimator is unbiased, that is, its mean value coincides with the true value of the phase shift, $\langle\theta_{est}\rangle = \theta$, then its minimal standard deviation is limited by the bounds [37,38]

$$\Delta\theta_{est} \geqslant \frac{1}{\sqrt{mF}} \geqslant \frac{1}{\sqrt{mF_Q}}. \tag{1}$$

The quantity F in the first inequality is the Fisher information, defined as

$$F = \sum_\mu \frac{1}{P(\mu|\theta)}[\partial_\theta P(\mu|\theta)]^2, \tag{2}$$

where $P(\mu|\theta) = \text{Tr}[\rho(\theta)\hat{E}_\mu]$ are conditional probabilities. The maximum likelihood estimator is an example of an estimator which is unbiased and saturates $\Delta\theta_{est} = 1/\sqrt{mF}$ in the central limit, for a sufficiently large m [39]. According to Eq. (1), F thus quantifies the asymptotic usefulness of a quantum state for phase estimation, given the operator $\hat{H}$ and the chosen final measurement. Maximizing F over all possible POVMs leads to the so-called QFI F_Q, and thus to the second inequality in Eq. (1). For a mixed input state $\rho = \sum_l \lambda_l |l\rangle\langle l|$ (with $\lambda_l > 0$, $\sum_l \lambda_l = 1$) the QFI is given by [40]

$$F_Q[\rho; \hat{H}] = 2\sum_{l,l'} \frac{(\lambda_l - \lambda_{l'})^2}{\lambda_l + \lambda_{l'}}|\langle l|\hat{H}|l'\rangle|^2, \tag{3}$$

where the sum runs over indices such that $\lambda_l + \lambda_{l'} > 0$. For pure input states this reduces to $F_Q = 4(\Delta\hat{H})^2$, where $(\Delta\hat{H})^2 = \langle\hat{H}^2\rangle - \langle\hat{H}\rangle^2$ is the variance of the generator of the phase shift, $\hat{H}$ [41].

In this paper we focus on linear two-mode interferometers and input states of N particles. In this case,

$$\hat{H}_{lin} = \frac{1}{2}\sum_{l=1}^N \hat{\sigma}_{\vec{n}_l}^{(l)}, \tag{4}$$

where $\hat{\sigma}_{\vec{n}_l}^{(l)} = \vec{n}_l \cdot \vec{\hat{\sigma}}^{(l)} = \alpha_l\hat{\sigma}_x^{(l)} + \beta_l\hat{\sigma}_y^{(l)} + \gamma_l\hat{\sigma}_z^{(l)}$ is an operator decomposed as the sum of Pauli matrices acting on the particle l, and $\vec{n}_l \equiv (\alpha_l, \beta_l, \gamma_l)$ is a vector on the Bloch sphere ($\alpha_l^2 + \beta_l^2 + \gamma_l^2 = 1$). If all local directions are the same, $\vec{n}_l = \vec{n}$, then $\hat{H}_{lin} \equiv \hat{J}_{\vec{n}} = \vec{n} \cdot \vec{\hat{J}}$, where $\vec{\hat{J}} \equiv \frac{1}{2}\sum_{l=1}^N \vec{\hat{\sigma}}^{(l)}$ is a collective spin operator. The operators $\hat{J}_x$, $\hat{J}_y$, and $\hat{J}_z$ fulfill the commutation relations of angular momentum operators. As an example for a linear, collective, two-mode interferometer, we mention the Mach-Zehnder interferometer, whose generator is $\hat{H}_{lin} = \hat{J}_y$ [42].

For linear phase shift generators $\hat{H}_{lin}$ as in Eq. (4), the QFI provides a direct connection between entanglement and phase uncertainty. We remind the reader that a state of N particles is entangled if it cannot be written as a separable state $\rho_{sep} = \sum_\alpha p_\alpha \bigotimes_{l=1}^N |\psi_\alpha^{(l)}\rangle\langle\psi_\alpha^{(l)}|$, where $\{p_\alpha\}$ forms a probability distribution [43]. It has been recently shown that the QFI for separable states and linear generators is [33,34]

$$F_Q[\rho_{sep}; \hat{H}_{lin}] \leqslant N. \tag{5}$$

Taking into account Eqs. (1) and (5) and the definition of QFI, $F_Q \geqslant F$, we conclude that the phase uncertainty attainable with separable states is $\Delta\theta_{est} \geqslant \Delta\theta_{SN}$, where

$$\Delta\theta_{SN} = \frac{1}{\sqrt{mN}}. \tag{6}$$

This bound holds for any linear interferometer and any final measurement and is generally called the shot-noise limit. It is not fundamental and can be surpassed by using proper entangled states. For general probe states of N particles, we have [33,34]

$$F_Q[\rho; \hat{H}_{lin}] \leqslant N^2, \tag{7}$$

where the equality can only be saturated by certain maximally entangles states. From the maximum value of the QFI we obtain the optimal bound for the phase uncertainty, called the Heisenberg limit,

$$\Delta\theta_{HL} = \frac{1}{\sqrt{m}\,N}. \tag{8}$$

We thus expect that, in order to increase the QFI and the sensitivity of a linear interferometer, it is necessary to increase the number of entangled particles in the probe state. The purpose of this paper is to quantitatively investigate this effect and to derive bounds on the QFI for multiparticle entanglement classes.

B. Multiparticle entanglement

We consider the following classification of multiparticle entanglement from Ref. [16,44,45] (see also [22]; alternative classifications can be found in Refs. [46,47]). A pure state of N particles is k-producible if it can be written as $|\psi_{k\text{-prod}}\rangle = \otimes_{l=1}^M |\psi_l\rangle$, where $|\psi_l\rangle$ is a state of $N_l \leqslant k$ particles (such that $\sum_{l=1}^M N_l = N$). A state is k-particle entangled if it is k-producible but not $(k-1)$-producible. Therefore, a k-particle entangled state can be written as a product $|\psi_{k\text{-ent}}\rangle = \otimes_{l=1}^M |\psi_l\rangle$ which contains at least one state $|\psi_l\rangle$ of $N_l = k$ particles which does not factorize. A mixed state is k-producible if it can be written as a mixture of $(k_l \leqslant k)$-producible pure states, that is, $\rho_{k\text{-prod}} = \sum_l p_l |\psi_{k\text{-prod}}\rangle\langle\psi_{k\text{-prod}}|$, where $k_l \leqslant k$ for all l. Again, it is k-particle entangled if it is k-producible but not $(k-1)$ producible. We denote the set of k-producible states by S_k. We later use that S_k is convex for any k. Note that, formally, a fully separable state is 1-producible and that a decomposition of a $k < N$-particle entangled state of N particles may contain states where different sets of particles are entangled.

Let us illustrate the classification by considering states of $N = 3$ particles. A state $|\psi_{1\text{-prod}}\rangle = |\phi\rangle_1 \otimes |\varphi\rangle_2 \otimes |\chi\rangle_3$ is fully separable. A state $|\psi_{2\text{-ent}}\rangle = |\phi\rangle_{12} \otimes |\chi\rangle_3$ which cannot be written as $|\psi_{1\text{-prod}}\rangle$ (i.e., $|\phi\rangle_{12}$ does not factorize, $|\phi\rangle_{12} \neq |\phi\rangle_1 \otimes |\varphi\rangle_2$) is two-particle entangled. A state $|\psi_{3\text{-ent}}\rangle$ which does not factorize is three-particle entangled.

III. CRITERIA FOR MULTIPARTICLE ENTANGLEMENT FROM THE QUANTUM FISHER INFORMATION

Now we are in a position to derive the desired bounds. We start by computing the maximum of the QFI $F_Q[\rho_{k\text{-prod}}; \hat{H}_{\text{lin}}]$ for k-producible states and linear Hamiltonians $\hat{H}_{\text{lin}}$, including the case of collective spin operators $\hat{H}_{\text{lin}} = \hat{J}_{\vec{n}}$. Then we derive similar bounds for the QFI for a generator $\hat{J}_{\vec{n}}$, now averaged over all directions $\vec{n}$. At the end of this section, we investigate the question of whether or not the criteria are different by comparing the sets of states they detect.

A. Entanglement criterion derived from F_Q

Observation 1 (F_Q^{k+1} criterion). For k-producible states and an arbitrary linear two-mode interferometer $\hat{H}_{\text{lin}}$ defined in Eq. (4), the QFI is bounded by

$$F_Q[\rho_{k\text{-prod}}; \hat{H}_{\text{lin}}] \leqslant sk^2 + r^2, \qquad (9)$$

where $s = \lfloor \frac{N}{k} \rfloor$ is the largest integer smaller than or equal to $\frac{N}{k}$ and $r = N - sk$. Hence, a violation of the bound (9) proves $(k+1)$-particle entanglement. The bounds are uniquely saturated by a product of s GHZ states of k particles and another GHZ states of r particles, where [48]

$$|\text{GHZ}_\nu\rangle = \frac{1}{\sqrt{2}}(|0\rangle^{\otimes \nu} + |1\rangle^{\otimes \nu}), \qquad (10)$$

known as the Greenberger-Horne-Zeilinger (GHZ) [49] or NOON [50] state of ν particles.

Proof. The basic ingredients of the derivations are the following. (i) The sets $\mathcal{S}_k$ of k-producible states are convex. (ii) The Fisher information is convex in the states; that is, for any fixed phase transformation and any fixed output measurement the relation $F[p\rho_1 + (1-p)\rho_2] \leqslant pF[\rho_1] + (1-p)F[\rho_2]$ holds for $p \in [0,1]$ [51]. Since the QFI is equal to the Fisher information for a particular measurement, this holds also for F_Q. (iii) It is easy to see that for a product state $|\phi\rangle_A \otimes |\chi\rangle_B$, $(\Delta \hat{H}_{\text{lin}}^{(AB)})^2_{|\phi\rangle_A \otimes |\chi\rangle_B} = (\Delta \hat{H}_{\text{lin}}^{(A)})^2_{|\phi\rangle_A} + (\Delta \hat{H}_{\text{lin}}^{(B)})^2_{|\chi\rangle_B}$. Here $\hat{H}_{\text{lin}}^{(AB)}$ acts on all the particles while $\hat{H}_{\text{lin}}^{(A)}$ acts on the particles of $|\psi\rangle_A$ only and in analogy for $\hat{H}_{\text{lin}}^{(B)}$. (iv) For a state with N particles, $4(\Delta \hat{H}_{\text{lin}})^2 \leqslant N^2$ holds [33]. The inequality is saturated uniquely by the GHZ state.

It follows from (i) and (ii) that the maximum of F_Q for a fixed Hamiltonian $\hat{H}_{\text{lin}}$ and k-producible mixed states is reached on pure k-producible states $|\psi_{k\text{-prod}}\rangle$ [52]. Therefore, our task is to maximize $F_Q[|\psi_{k\text{-prod}}\rangle; \hat{H}_{\text{lin}}] = 4(\Delta \hat{H}_{\text{lin}})^2_{|\psi_{k\text{-prod}}\rangle}$ with respect to the probe state $|\psi_{k\text{-prod}}\rangle$ and linear operator $\hat{H}_{\text{lin}}$. Since the local directions of $\hat{H}_{\text{lin}}$ [Eq. (4)] can be changed by local unitary operations [36], which do not change the entanglement properties of the state, we can, without loss of generality, fix $\hat{H}_{\text{lin}} = \hat{J}_z$. Due to (iii) and (iv), we obtain $\max_{|\psi_{k\text{-prod}}\rangle}(\Delta \hat{J}_z)^2_{|\psi_{k\text{-prod}}\rangle} = \max_{|\psi_{k\text{-prod}}\rangle} \sum_{l=1}^M (\Delta \hat{J}_z^{(l)})^2_{|\psi_l\rangle} = \max_{\{N_l\}} \frac{1}{4} \sum_{l=1}^M N_l^2$. Since $(N_1 + 1)^2 + (N_2 - 1)^2 \geqslant N_1^2 + N_2^2$ if $N_1 \geqslant N_2$, the QFI is increased by making the N_l as large as possible. Hence, the maximum is reached by the product of $s = \lfloor \frac{N}{k} \rfloor$ GHZ state of $N_l = k$ particles and one GHZ state of
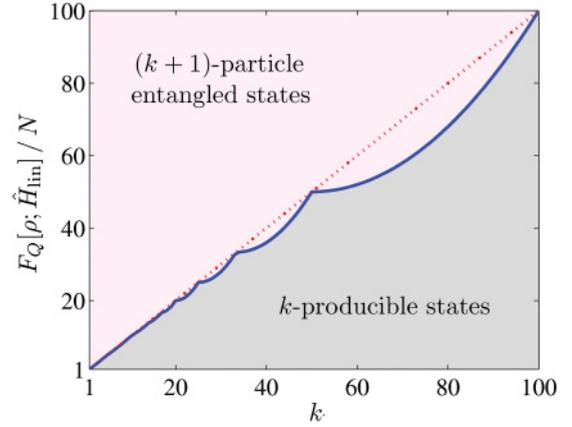

FIG. 1. (Color online) F_Q^{k+1} criterion. The solid line is the bound $F_Q[\rho; \hat{H}_{\text{lin}}] = sk^2 + r^2$ which separates k-producible states (below the line) from $(k+1)$-particle entangled states (above the line). For comparison, the function $F_Q[\rho; \hat{H}_{\text{lin}}] = Nk$ is plotted (dotted line). Here $N = 100$.

$r = N - sk$ particles. Therefore, for k-producible states, the QFI is bounded by Eq. (9). ∎

Given the operator $\hat{H}_{\text{lin}}$ and the probe state ρ, the criterion (9) has a clear operational meaning. If the bound is surpassed, then the probe state contains useful $(k+1)$-particle entanglement: When used as input state of the interferometer defined by the transformation $e^{-i\theta \hat{H}_{\text{lin}}}$, ρ enables a phase sensitivity better than any k-producible state. A plot of the bound Eq. (9) is presented in Fig. 1 as a function of k and for $N = 100$. Since the bound increases monotonically with k, the maximum achievable phase sensitivity increases with the number of entangled particles. For $k = 1$ we recover the bound (5) for separable states. For $k = N - 1$, the bound is $F_Q[\rho_{(N-1)\text{-prod}}; \hat{H}_{\text{lin}}] \leqslant (N-1)^2 + 1$ and a QFI larger than this value signals that the state is fully N-particle entangled. The maximum value of the bound is obtained for $k = N$ (thus, $s = 1$ and $r = 0$), when $F_Q[\rho_{N\text{-ent}}; \hat{H}_{\text{lin}}] = N^2$, saturating the equality sign in Eq. (7).

Given the probe state ρ, the F_Q^{k+1} criterion can be used to detect $(k+1)$-particle entanglement. In order to maximize $F_Q[\rho; \hat{H}_{\text{lin}}]$, it is advantageous to optimize the local directions $\vec{n}_l$ in $\hat{H}_{\text{lin}}$ [36] [see Eq. (4)]. While the general problem needs to be solved numerically, a simple analytic solution can be obtained if we restrict ourselves to collective spin operators $\hat{H}_{\text{lin}} = \hat{J}_{\vec{n}}$. In this case we have [36]

$$F_Q[\rho; \hat{J}_{\vec{n}}] = \vec{n}^T \Gamma_C \vec{n}. \qquad (11)$$

The matrix Γ_C is real and symmetric and has the entries

$$[\Gamma_C]_{ij} = 2 \sum_{l,l'} \frac{(\lambda_l - \lambda_{l'})^2}{\lambda_l + \lambda_{l'}} \mathcal{R}[\langle l|\hat{J}_i|l'\rangle\langle l'|\hat{J}_j|l\rangle], \qquad (12)$$

where the states $|l\rangle$ and the variables λ_l are defined by the eigenvalue decomposition of the input state, $\rho = \sum_l \lambda_l |l\rangle\langle l|$, and $\mathcal{R}(z)$ is the real part of z. The sum runs over indices where

$\lambda_l + \lambda_{l'} > 0$. Maximizing Eq. (11) with respect to $\vec{n}$ leads to

$$F_Q^{\max}[\rho] \equiv \max_{\vec{n}} F_Q[\rho; \hat{J}_{\vec{n}}] = \lambda_{\max}(\Gamma_C), \quad (13)$$

where $\lambda_{\max}(\Gamma_C)$ is the maximal eigenvalue of Γ_C. In the case of collective operations, the criteria Eq. (9) can be substituted by

$$\lambda_{\max}(\Gamma_C) \leqslant sk^2 + r^2. \quad (14)$$

For any pure symmetric input state this is the optimal value of F also if arbitrary local unitary operations can be used [36]. Note that while this optimization increases F_Q, it might happen that for a fixed output measurement the Fisher information F is actually reduced by this optimization because the measurement would have to be adapted as well [40].

Finally, note that the result Eq. (9) can be obtained directly by using the Wigner-Yanase information I [53]. The bound (9) has been derived previously for I in Ref. [44] and directly applies to the QFI since I is convex in the states and agrees with the Fisher information on pure states, $F[|\psi\rangle; \hat{H}] = 4I(|\psi\rangle, \hat{H})$. See Ref. [54] for a more general discussion of convex quantities which are equal to the Fisher information on the pure states. Note also that a bound similar to Eq. (9) has been discussed for the class of so-called spin-squeezed states [22].

B. Entanglement criterion derived from $\overline{F}_Q$

Let us now consider the estimation of a fixed (unknown) phase shift θ with an interferometer that, in each run of the experiment, is given by $\exp[-i\hat{J}_{\vec{v}}\theta]$ with a random direction $\vec{v}$ of probability $P(\vec{v})$. For $m \gg 1$ independent repetitions of the phase measurement, the phase estimation uncertainty approaches

$$\Delta\theta_{\text{est}} \geqslant \frac{1}{\sqrt{mF^P[\rho]}}, \quad (15)$$

where

$$F^P[\rho] = \int_{|\vec{v}|^2=1} d^3\vec{v}\, P(\vec{v})\, F[\rho; \hat{J}_{\vec{v}}; \{\hat{E}_\mu\}], \quad (16)$$

and $P(\vec{v})$ is normalized to one. The direction-averaged Fisher information [Eq. (16)] is bounded by

$$F_Q^P[\rho] = \int_{|\vec{v}|^2=1} d^3\vec{v}\, P(\vec{v})\, F_Q[\rho; \hat{J}_{\vec{v}}]. \quad (17)$$

The latter quantity can be used to introduce an infinite set of multiparticle entanglement criteria, depending on the function $P(\vec{v})$. If $P(\vec{v}) = \delta_{\vec{v},\vec{n}}$, then we recover the standard situation of a fixed collective spin direction and the criteria Eq. (13). We here consider the opposite case, $P(\vec{v}) = 1/4\pi$, where all directions $\vec{v}$ on the Bloch sphere appear with equal probability. We indicate the corresponding average of the QFI as $\overline{F}_Q[\rho]$. It can be written as $\overline{F}_Q[\rho] = \frac{1}{4\pi} \sum_{ij} [\Gamma_C]_{ij} \int_{|\vec{v}|^2=1} d^3\vec{v}\, v_i v_j$ [see Eq. (11)] and evaluating the integrals leads to

$$\overline{F}_Q[\rho] = \frac{\text{Tr}[\Gamma_C]}{3} = \frac{F_Q[\rho; \hat{J}_x] + F_Q[\rho; \hat{J}_y] + F_Q[\rho; \hat{J}_z]}{3}. \quad (18)$$

The sum of three Fisher informations for the phase generators $\hat{J}_x$, $\hat{J}_y$, and $\hat{J}_z$ on the right-hand side appeared already in Refs. [55,56] as a criterion for entanglement. We would like to determine bounds on $\overline{F}_Q$ for k-producible states in analogy to the bounds that we found for F_Q. We directly state the results and derive them afterwards.

Observation 2 ($\overline{F}_Q^{k+1}$ criterion). For k-producible states, the average QFI defined in Eq. (18) is bounded by

$$\overline{F}_Q[\rho_{k\text{-prod}}] \leqslant \tfrac{1}{3}[s(k^2 + 2k - \delta_{k,1}) + r^2 + 2r - \delta_{r,1}], \quad (19)$$

where $s = \lfloor \frac{N}{k} \rfloor$, $r = N - sk$, and δ is the Kronecker δ. Hence, a violation of the bound (19) proves $(k+1)$-particle entanglement. For separable states, corresponding to $k = 1$, the bound becomes

$$\overline{F}_Q[\rho_{\text{sep}}] \leqslant \tfrac{2}{3}N. \quad (20)$$

The maximal value for any quantum state is given by

$$\overline{F}_Q \leqslant \tfrac{1}{3}[N^2 + 2N]. \quad (21)$$

Proof. Let us first prove Eq. (21). Since $\overline{F}_Q$ can be written as the sum of three QFIs, it is also convex in the states. Therefore, the maximum is again reached for pure states. Hence, $\overline{F}_Q \leqslant \frac{4}{3} \max_{|\psi\rangle}[\langle \vec{\hat{J}}^2 \rangle_{|\psi\rangle} - |\langle \vec{\hat{J}} \rangle_{|\psi\rangle}|^2] \leqslant \frac{4}{3}j(j+1)$, where $\langle \vec{\hat{J}}^2 \rangle \equiv \langle \vec{\hat{J}} \cdot \vec{\hat{J}} \rangle = \langle \hat{J}_x^2 \rangle + \langle \hat{J}_y^2 \rangle + \langle \hat{J}_z^2 \rangle$ and $|\langle \vec{\hat{J}} \rangle|^2 = \langle \hat{J}_x \rangle^2 + \langle \hat{J}_y \rangle^2 + \langle \hat{J}_z \rangle^2$. This leads to Eq. (21) because $|\langle \vec{\hat{J}} \rangle_{|\psi\rangle}|^2 \geqslant 0$ and

$$\langle \vec{\hat{J}}^2 \rangle_{|\psi\rangle} \leqslant j(j+1) \quad (22)$$

holds in general, while equality is reached by the symmetric states of N particles [57].

For the k-producible pure state $|\psi_{k\text{-prod}}\rangle = \bigotimes_{l=1}^M |\psi_l\rangle$, the average QFI is given by $\overline{F}_Q[|\psi_{k\text{-prod}}\rangle] = \frac{4}{3} \sum_{l=1}^M [\langle \vec{\hat{J}}_l^2 \rangle_{|\psi_l\rangle} - |\langle \vec{\hat{J}}_l \rangle_{|\psi_l\rangle}|^2] \leqslant \frac{1}{3} \sum_{l=1}^M [N_l^2 + 2N_l - 4|\langle \vec{\hat{J}}_l \rangle_{|\psi_l\rangle}|^2]$, where $\vec{\hat{J}}_l$ is the vector of collective spin operators acting on the particles contained in state $|\psi_l\rangle$. The inequality is due to Eq. (22). In the same way as it was for F_Q, in order to maximize the bound it is advantageous to increase the N_l as much as possible. This is true even though if $N_l = 1$ then $\overline{F}_Q$ is reduced by $\frac{1}{3}$ since $|\langle \vec{\hat{J}} \rangle_{|\psi_l\rangle}|^2 = \frac{1}{4}$ in this case. For $k \in [1, N]$, we obtain the bound (19), where $s = \lfloor \frac{N}{k} \rfloor$ and $r = N - sk$ as above, and we obtain Eq. (20) for $k = 1$. ∎

The bound in Eq. (19) is shown in Fig. 2 as a function of k. Let us note that the bound for $k = N - 1$ is

$$\overline{F}_Q[\rho_{(N-1)-\text{prod}}] \leqslant \tfrac{1}{3}[N^2 + 1]. \quad (23)$$

Again, the bounds for a given k are saturated by using s GHZ states of k particles and one GHZ state of r particles. However, as we discuss presently, these states are not uniquely saturating the bounds, in contrast to what happens in the case of the F_Q^{k+1} criterion.

C. F_Q^{k+1} criterion vs $\overline{F}_Q^{k+1}$ criterion

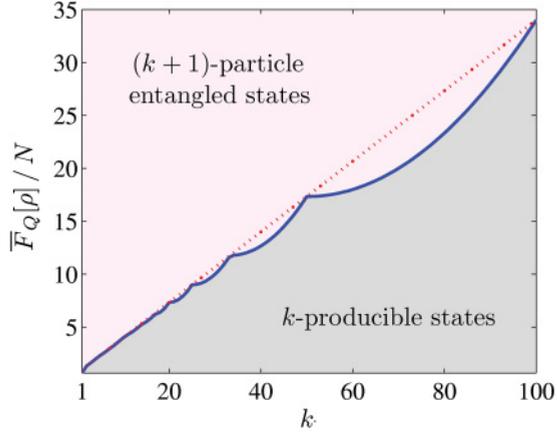To start the comparison, let us first discuss states with extremal values for the criteria. In particular, we consider

FIG. 2. (Color online) $\overline{F}_Q^{k+1}$ criterion. The solid line shows the bound in Eq. (19) [$\overline{F}_Q^{k+1}$-criterion] as a function of k. For comparison, the function $N(k+2)/3$ is plotted (dotted line). Here $N = 100$.

the cases $k = 1$, where the criteria detect any kind of entanglement, and $k = N - 1$, where the criteria detect genuine multiparticle entanglement. The states we use to illustrate the criteria are the N-particle GHZ state from Eq. (10), the fully separable state $|1\rangle^{\otimes N}$, and the Dicke state with $N/2$ excitations [57],

$$\left|D_N^{(N/2)}\right\rangle = \mathcal{S}(|0\rangle^{\otimes N/2} \otimes |1\rangle^{\otimes N/2}), \tag{24}$$

known as twin-Fock state for indistinguishable particles [60]. In Table I, we list the Γ_C matrices for these states for all N. Since all states are symmetric under the exchange of any two particles, we can directly read off the optimal values of F_Q and $\overline{F}_Q$ from these matrices [36]. For the pure separable state, $F_Q[|1\rangle^{\otimes N}; \hat{J}_{\vec{n}}] = N$ for any direction $\vec{n}$ in the x-y plane because $|1\rangle$ is an eigenstate of $\hat{\sigma}_z$. Hence, this state saturates the bound for separable states both for F_Q and for $\overline{F}_Q$. As noted before, the GHZ state maximizes both F_Q and $\overline{F}_Q$. The Dicke state $|D_N^{(N/2)}\rangle$ has a N^2 scaling in F_Q as the GHZ state with a prefactor $\frac{1}{2}$; therefore, it does not saturate the maximum value $F_Q = N^2$. However, it saturates the maximal value of $\overline{F}_Q$ from Eq. (21). In fact, the criterion F_Q^N detects $|D_N^{(N/2)}\rangle$ as N-particle entangled if $N \leqslant 5$ only, while the criterion $\overline{F}_Q^N$ detects the state as N-particle entangled for any value of N. Hence, $\overline{F}_Q$ is not uniquely saturated by the GHZ state as F_Q. We use this fact in the proof of the following Observation which shows that the two criteria in general detect strictly different sets of states.

Observation 3. (a) For all pairs (k, N) with $k < N$, the F_Q^{k+1} criterion detects the entanglement of some states for which the

TABLE I. Comparison of the maximal values of F_Q and $\overline{F}_Q$ for three different input states.

State	Γ_C	$F_Q^{\max}$	$\overline{F}_Q$
$|1\rangle^{\otimes N}$	$\mathrm{diag}(N,N,0)$	N	$\frac{2}{3}N$
$|\mathrm{GHZ}_N\rangle$	$\mathrm{diag}(N,N,N^2)$	N^2	$\frac{1}{3}(N^2+2N)$
$|D_N^{(N/2)}\rangle$	$\frac{1}{2}(N^2+2N)\mathrm{diag}(1,1,0)$	$\frac{1}{2}(N^2+2N)$	$\frac{1}{3}(N^2+2N)$

$\overline{F}_Q^{k+1}$ does not detect entanglement. (b) For all pairs (k, N) with $2 < k < N$, the $\overline{F}_Q^{k+1}$ criterion detects the entanglement of some states for which the F_Q^{k+1} criterion does not detect entanglement.

The proof can be found in the Appendix. Part (b) of Observation 3 can be extended also to cases where $k = 1 < N$ and $k = 2 < N$, as shown in Secs. IV D and IV E below.

IV. EXAMPLES

We now turn to illustrate the strength of the described criteria for their utilization in entanglement detection and in quantum metrology applications. To this end, we evaluate the criteria for different sets of states. We first consider an actual experimental setting of different types of entangled four-qubit states. Second, we will consider various three-qubit entangled states including bound entangled states. We will compare different means to detect their entanglement by computing the amount of detected states. Finally, we construct an example extending Observation 3 before we examine two families of bound entangled states.

A. Experimental GHZ and Dicke states

We start by applying the above criteria to entangled states of $N = 4$ photonic qubits produced experimentally by parametric downconversion from the Refs. [61,62]. The qubits are encoded in the polarization with $|0\rangle \equiv |H\rangle$ and $|1\rangle \equiv |V\rangle$, where H stands for horizontal and V for vertical polarization. In Ref. [61], a large family of entangled states of $N = 4$ qubits has been produced. We investigate the data of the state $\frac{1}{\sqrt{2}}(|0011\rangle + |1100\rangle)$, which can be converted to a GHZ state [cf. Eq. (10)] by flipping the state of the last two qubits, and the state $|\psi^+\rangle \otimes |\psi^+\rangle$, where $|\psi^+\rangle = \frac{1}{\sqrt{2}}(|01\rangle + |10\rangle) = |D_2^{(1)}\rangle$. Hence, this state is a product of two-particle Dicke states [57]. Note that by flipping the state of the second and of the fourth qubit, this state can be transformed into $|\mathrm{GHZ}_2\rangle \otimes |\mathrm{GHZ}_2\rangle$. Finally, we also use the data of Ref. [62], where the Dicke state $|D_4^{(2)}\rangle$ has been produced. The states were observed with fidelities $\mathcal{F}_{\mathrm{GHZ}_4} = 0.8303 \pm 0.0080$, $\mathcal{F}_{(D_2^{(1)})^{\otimes 2}} = 0.9255 \pm 0.0091$ [61], and $\mathcal{F}_{D_4^{(2)}} = 0.8872 \pm 0.0055$ [62]. For comparison, the data of the separable state $|+\rangle^{\otimes 4}$ measured in Ref. [62] is used, which was observed with a fidelity $\mathcal{F}_{|+\rangle^{\otimes 4}} = 0.9859 \pm 0.0062$. Here $|+\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$.

The optimized Fisher information $F_Q^{\max}$ and $\overline{F}_Q$ for the different states are calculated from the measured density matrices. We compare the experimental results with the ideal cases and with the bounds on k-producible states from Observations 1 and 2 for $N = 4$. In order to do so, we apply the bit flips mentioned above to the experimental data where necessary. The results are shown in Fig. 3. For the $N = 4$ GHZ and Dicke states, four-particle entanglement is proven with a high statistical significance by $\overline{F}_Q$. In particular, for the Dicke state, the statistical significance for the proof of four-particle entanglement from $F_Q^{\max}$ is much lower. This is a consequence of the fact that the ideal Dicke states reaches the maximal value of $\overline{F}_Q$ for any N, while the deviation of $F_Q^{\max}$ from the maximal value increases with increasing N [cf. Table I].
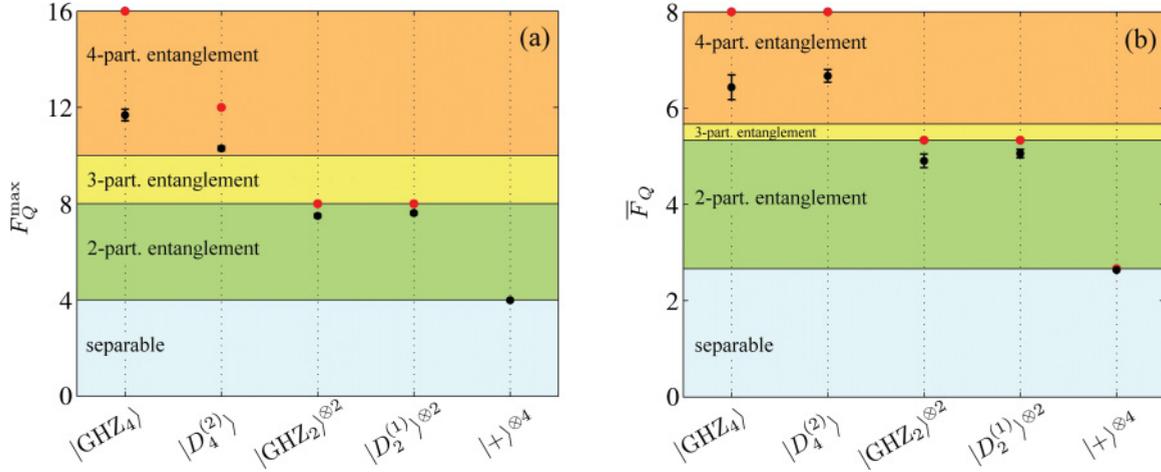
FIG. 3. (Color online) Black dots are the values of $F_Q^{\max}$ [in panel (a)] and of $\overline{F}_Q$ [in panel (b)] for states generated experimentally, calculated from the experimental density matrices. Error bars are calculated with a parametric bootstrap method [63] (see also the supplementary material of Ref. [62]). The red dots are ideal values. More explicitly, we have $F_Q^{\max} = 11.681 \pm 0.238$ and $3\overline{F}_Q = 19.296 \pm 0.256$ for the $|GHZ_4\rangle$ state; $F_Q^{\max} = 10.291 \pm 0.094$ and $3\overline{F}_Q = 20.004 \pm 0.131$ for the $|D_4^{(2)}\rangle$ state; $F_Q^{\max} = 7.495 \pm 0.070$ and $3\overline{F}_Q = 14.713 \pm 0.141$ for the $|GHZ_2\rangle^{\otimes 2}$ state; $F_Q^{\max} = 7.612 \pm 0.058$ and $3\overline{F}_Q = 15.174 \pm 0.089$ for the $|D_2^{(1)}\rangle^{\otimes 2}$ state; $F_Q^{\max} = 4.002 \pm 0.025$ and $3\overline{F}_Q = 7.902 \pm 0.015$ for the separable state $|+\rangle^{\otimes 4}$. In panels (a) and (b), the vertical lines are bounds for the F_Q^{k+1} and $\overline{F}_Q^{k+1}$ criteria, respectively.

The very high fidelity of the experimental product of two $N = 2$ Dicke states is reflected in the fact that $F_Q^{\max}$ and $\overline{F}_Q$ nearly reach the optimal values for the states $|D_2^{(1)}\rangle^{\otimes 2}$ and $|GHZ_2\rangle^{\otimes 2}$, and entanglement is clearly proven, while the bounds for two-particle entangled states are not violated.

As a final remark, we would like to point out that the multiparticle entanglement of the states could be proved with less experimental effort and a generally larger statistical significance by witness operators [61]. However, in this case this does not give any direct information about the usefulness for a given task, in particular for phase estimation (see Ref. [62] for a detailed comparison of the F_Q criteria with a witness operator for the state $|D_4^{(2)}\rangle$).

B. Pure states of three particles

In order to get an impression of the strength of the criteria, we randomly choose a three-qubit state $|\psi\rangle$ and analyze it using various criteria. First, we evaluate the criteria F_Q^2 and $\overline{F}_Q^2$ which detect entanglement. Further, we compare several criteria detecting multiparticle entanglement: (i) the entanglement witness $\mathcal{W} = \frac{1}{2}\mathbb{1} - |GHZ\rangle\langle GHZ|$, which has a positive expectation value for all two-particle entangled states [9], (ii) the density matrix element (DME) condition which states that

$$|\rho_{18}| \leqslant \sqrt{\rho_{22}\rho_{77}} + \sqrt{\rho_{33}\rho_{66}} + \sqrt{\rho_{44}\rho_{55}} \quad (25)$$

for all two-entangled states (ρ_{ij} denote coefficients of a given density matrix $\rho = |\psi\rangle\langle\psi|$) [26], and (iii) the multiparticle criteria F_Q^3 and $\overline{F}_Q^3$.

To generate a random pure state [64], we take a vector of a random unitary matrix distributed according to the Haar measure on U(8):

$$|\psi\rangle = (\cos\alpha_7, \cos\alpha_6 \sin\alpha_7 e^{i\phi_7}, \cos\alpha_5 \sin\alpha_6 \sin\alpha_7 e^{i\phi_6},$$
$$\dots, \sin\alpha_1 \cdots \sin\alpha_7 e^{i\phi_1}), \quad (26)$$

where $\alpha_i \in [0, \pi/2]$ and $\phi_k = [0, 2\pi)$. The parameters are drawn with the probability densities $P(\alpha_i) = i \sin(2\alpha_i)(\sin\alpha_i)^{2i-2}$ and $P(\phi_i) = 1/2\pi$. The calculations were performed for a set of 10^6 states. The results are presented in Table II. The averaged criteria seem to detect more states in general. It is surprising that the witness condition detects nearly as many states as the criteria F_Q^3 and $\overline{F}_Q^3$. This may be an artifact of the small N we chose.

TABLE II. Percentage of detected two-particle and three-particle entangled pure three-qubit states. See text for details. DME′ denotes the whole family of DME conditions, which is obtained by permuting the qubits of the state.

Criterion	Detected two-particle entangled (%)
F_Q^2	94.32
$\overline{F}_Q^2$	98.38
	Detected three-particle entangled (%)
$\mathcal{W}$	18.99
DME	80.63
DME′	82.61
F_Q^3	22.93
$\overline{F}_Q^3$	27.99

TABLE III. Percentage of GHZ-diagonal three-particle entangled states which are detected by the entanglement witness, the criterion F_Q^3 and the criterion $\overline{F}_Q^3$. In the middle column, only states violating the DME condition (25) have been generated, while in the last column, also states violating any of the other DME conditions obtained by permutations of the particles have been generated.

Criterion	Detected DME (%)	Detected DME′ (%)
$\mathcal{W}$	50.56	12.27
F_Q^3	19.45	4.77
$\overline{F}_Q^3$	13.14	3.25

C. GHZ-diagonal states

The DME criterion (25) and the criteria obtained thereof by permutations of the qubits completely characterize the GHZ-diagonal states of three qubits [26], which can be written as

$$\frac{1}{\mathcal{N}}\begin{pmatrix} \lambda_1 & 0 & 0 & 0 & 0 & 0 & 0 & \mu_1 \\ 0 & \lambda_2 & 0 & 0 & 0 & 0 & \mu_2 & 0 \\ 0 & 0 & \lambda_3 & 0 & 0 & \mu_3 & 0 & 0 \\ 0 & 0 & 0 & \lambda_4 & \mu_4 & 0 & 0 & 0 \\ 0 & 0 & 0 & \mu_4 & \lambda_5 & 0 & 0 & 0 \\ 0 & 0 & \mu_3 & 0 & 0 & \lambda_6 & 0 & 0 \\ 0 & \mu_2 & 0 & 0 & 0 & 0 & \lambda_7 & 0 \\ \mu_1 & 0 & 0 & 0 & 0 & 0 & 0 & \lambda_8 \end{pmatrix}, \quad (27)$$

with real coefficients λ_i and μ_i, where $\mathcal{N}$ is a normalization factor. If $\lambda_i = \lambda_{9-i}$ for $i = 5,6,7,8$, then these states are diagonal in the GHZ basis $|\psi_{l_1 l_2}^\pm\rangle = \frac{1}{\sqrt{2}}(|0l_1 l_2\rangle \pm |1\bar{l}_1\bar{l}_2\rangle)$, where l_1 and l_2 are equal to 0 or 1, and $\bar{1} = 0$ and $\bar{0} = 1$. We generated 10^6 random states of this form violating Eq. (25) directly, which states $|\mu_1| \leqslant \lambda_2 + \lambda_3 + \lambda_4$ in this case. The results are shown in Table III in the middle column. Then we generated again 10^6 states violating Eq. (25) or its other forms obtained by permuting the qubits. The results are shown in the right column of Table III. The witness criterion detects significantly more states than the criteria based on the Fisher information. Contrary to the case of pure states, the F_Q^3 criterion detects more states than $\overline{F}_Q^3$ in this case. Note that the percentage of detected states reduces significantly for all criteria in the DME′ case. The reason is that all criteria work best for the symmetric GHZ state $\frac{1}{\sqrt{2}}(|000\rangle + |111\rangle)$, which has the highest weight in the state if only condition (25) is used [26].

The family of states (27) also comprises bound entangled states if $\lambda_1 = \lambda_8 = \mu_1 = 1$ and $\lambda_7 = 1/\lambda_2$, $\lambda_6 = 1/\lambda_3$, $\lambda_5 = 1/\lambda_4$, and $\mu_2 = \mu_3 = \mu_4 = 0$, as long as $\lambda_2\lambda_3 \neq \lambda_4$. Then the states have a positive partial transpose (PPT) [65] for any bipartition of the three particles while still being entangled [47]. It follows that the state cannot be distilled to a GHZ state [46,66]. We generated again 10^6 random states of this class and applied F_Q^2 and $\overline{F}_Q^2$, but neither criterion detected any of these states. However, we will see presently that $\overline{F}_Q$ is, in fact, able to detect bound entanglement.

D. Extension of the observation 3 for $N = 4$

Observation 3(b) can be extended to pairs (k, N), where $1 \leqslant k < N$. We now construct an explicit example for the cases $N = 4$ and $k = 1, 2$. The basic idea is to use states with the property $\Gamma_C = c_N \mathbb{1}$ which are extremal in the sense that they saturate the inequality $\max_{\vec{n}} F_Q[\rho; \hat{J}_{\vec{n}}] \geqslant \overline{F}_Q[\rho]$. Hence, they provide the minimal $F_Q^{\max}$ compared to $\overline{F}_Q$. One way of constructing such states is by considering a symmetric state $|\psi_S\rangle = \sum_\mu \gamma_\mu |j,\mu\rangle$ [57], and by choosing the γ_μ such that $\langle \vec{\hat{J}} \rangle = 0$ and $\langle \hat{J}_x^2 \rangle = \langle \hat{J}_y^2 \rangle = \langle \hat{J}_z^2 \rangle$. If $\gamma_\mu \neq 0$ and $\gamma_{\mu'} \neq 0$ only if $|\mu - \mu'| > 2$ then $\langle \hat{J}_x \rangle = \langle \hat{J}_y \rangle = \langle \hat{J}_i \hat{J}_j \rangle = 0$ for $(i,j) = (x,y)$, $(i,j) = (x,z)$, and $(i,j) = (y,z)$. For $N = 4$, all the conditions above are fulfilled by the states

$$|\psi_S^4 \pm\rangle = \sqrt{\frac{1}{3}}|2, \pm 2\rangle + \sqrt{\frac{2}{3}}|2, \mp 1\rangle, \quad (28)$$

leading to $\Gamma_C = 8\mathbb{1}$, and hence $F_Q^{\max} = \overline{F}_Q = 8$. With this state, $\overline{F}_Q$ reaches the maximal value possible for $N = 4$ [cf. Eq. (21)], while $F_Q^{\max}$ saturates the bound of Eq. (9) for $k = 2$. This provides the example for Observation 3(b) for $(N,k) = (4,2)$. If we mix $|\psi_S^4\pm\rangle$ with the identity, then using Eq. (A2) from the Appendix it can be shown that for $p^* = \frac{7}{32}(1 + \sqrt{113/7})$ we obtain $\Gamma_C[\rho(p^*)] = 4\mathbb{1}$. Hence, $\rho(p^*)$ saturates the F_Q^{k+1} criterion but violates the $\overline{F}_Q^{k+1}$ criterion for $k = 1$. This provides the example for Observation 3(b) for $(N,k) = (4,1)$.

Note that the state $|\psi_S^4-\rangle$ has appeared also in other contexts [8]. For instance, it is the most nonclassical state for total spin $j = 2$ [67], and it is a maximally entangled state of four qubits for multipartite entanglement measures based on antilinear operators and combs [68]. Finally, symmetric states with $\Gamma_C \propto \mathbb{1}$ have the highest sensitivity to small misalignments of Cartesian reference frames [69]. The quantity to be optimized in the derivations is $3\overline{F}_Q$. For $N = 4$, again the state $|\psi_S^4-\rangle$ is optimal, and several other examples of symmetric states with $\Gamma_C \propto \mathbb{1}$ for even N have been found in Ref. [69].

The bound entangled Dür and Smolin states considered in Sec. IV E below provide further examples, for $k = 1$ and any N.

E. Detecting bound entangled states

We consider two families of states where the state has a PPT with respect to some bipartitions, but not with respect to others. Due to the PPT bipartitions it is not possible to distill these states to a GHZ state nonetheless [46]. Both families of states provide examples for situations where the $\overline{F}_Q^{k+1}$ criterion detects states which the F_Q^{k+1} does not detect for $k = 1$ and for any value of N. This extends the results summarized in Observation 3 from Sec. III C.

1. Dür states

Interestingly, the $\overline{F}_Q^2$ criterion (20) can reveal entanglement of a bound entangled state introduced by Dür [70]:

$$\rho_{\text{Dür}}^{(N)} = \frac{1}{N+1}\left(|\text{GHZ}_\varphi\rangle\langle\text{GHZ}_\varphi| + \frac{1}{2}\sum_{l=1}^N (P_l + \bar{P}_l)\right), \quad (29)$$

TABLE IV. Nonvanishing factors contributing to Γ_C for the Dür states [Eq. (29)]. The multiplicity is the number of occurrences.

Factor	Value	Multiplicity
$\langle \mathrm{GHZ}_0 | \hat{J}_x | 1_l \rangle$	$\frac{1}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_0 | \hat{J}_x | 0_l \rangle$	$\frac{1}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_\pi | \hat{J}_x | 1_l \rangle$	$\frac{1}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_\pi | \hat{J}_x | 0_l \rangle$	$-\frac{1}{\sqrt{8}}$	N
$\langle 1_l | \hat{J}_x | (N-2)_{\mathcal{P}} \rangle$	$\frac{1}{2}$	$N(N-1)$
$\langle 0_l | \hat{J}_x | 2_{\mathcal{P}} \rangle$	$\frac{1}{2}$	$N(N-1)$
$\langle \mathrm{GHZ}_0 | \hat{J}_y | 1_l \rangle$	$-\frac{i}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_0 | \hat{J}_y | 0_l \rangle$	$\frac{i}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_\pi | \hat{J}_y | 1_l \rangle$	$-\frac{i}{\sqrt{8}}$	N
$\langle \mathrm{GHZ}_\pi | \hat{J}_y | 0_l \rangle$	$-\frac{i}{\sqrt{8}}$	N
$\langle 1_l | \hat{J}_y | (N-2)_{\mathcal{P}} \rangle$	$-\frac{i}{8}$	$N(N-1)$
$\langle 0_l | \hat{J}_y | 2_{\mathcal{P}} \rangle$	$-\frac{i}{2}$	$N(N-1)$
$\langle \mathrm{GHZ}_\pi | \hat{J}_z | \mathrm{GHZ}_0 \rangle$	$\frac{N}{2}$	1

with $|\mathrm{GHZ}_\varphi\rangle = \frac{1}{\sqrt{2}}[|0\rangle^{\otimes N} + e^{i\varphi}|1\rangle^{\otimes N}]$, where φ is an arbitrary phase. We will consider $\varphi = 0$ in the following. Further, P_l is the projector on the state $|0\rangle^{\otimes l-1} \otimes |1\rangle \otimes |0\rangle^{\otimes N-l} \equiv |1_l\rangle$ and $\bar{P}_l$ is obtained from P_l by exchanging $0 \leftrightarrow 1$.

We can directly state the eigenstates and eigenvalues. The state $|\mathrm{GHZ}_0\rangle$ is an eigenstate with eigenvalue $\frac{1}{N+1}$ and the states $|1_l\rangle$ and $|0_l\rangle$ are eigenstates with eigenvalue $\frac{1}{2(N+1)}$. The kernel is spanned by the state $|\mathrm{GHZ}_\pi\rangle$ and by all states of the form $|n_{\mathcal{P}}\rangle \equiv \mathcal{P}(|0\rangle^{\otimes n} \otimes |1\rangle^{\otimes N-n})$, where $\mathcal{P}$ is a permutation of the qubits and $n = 2, 3, \ldots, N-2$. Now we can compute the elements of the correlation matrix Γ_C using Eq. (12). The nonvanishing factors $\langle l|\hat{J}_i|l'\rangle$ are given in Table IV. We obtain

$$\Gamma_C = N \,\mathrm{diag}\left(\frac{3N-1}{3N+3}, \frac{3N-1}{3N+3}, \frac{N}{N+1} \right). \quad (30)$$

The matrix Γ_C is diagonal because the factors $\langle l|\hat{J}_{x,z}|l'\rangle$ are real while the factors $\langle l|\hat{J}_y|l'\rangle$ are imaginary and since $\langle l|\hat{J}_x|l'\rangle$ vanishes for the eigenstates where $\langle l|\hat{J}_z|l'\rangle \neq 0$ and vice versa. We observe that $F_Q^{\max} < N$ for all N while

$$\overline{F}_Q = \frac{9N-2}{9N+9}N > \frac{2}{3}N \quad (31)$$

for all N.

Hence, the F_Q^2 criterion does not detect the entanglement in any of these cases [cf. Eq. (5)]. Therefore, these states represent an example of Observation 3(b) for $k = 1$ and any N. In conclusion, the states are not useful for sub-shot-noise interferometry for any direction $\vec{n}$, even though they are more useful than separable states when averaging over all directions.

2. Generalized Smolin states

As a second example, we consider the generalized $N = 2n$-qubit Smolin state [71]

$$\rho_{\mathrm{Smolin}}^{(N)} = \frac{1}{2^N}\left(\mathbb{1} + (-1)^n \sum_{i=1}^3 \sigma_i^{\otimes N} \right), \quad (32)$$

TABLE V. Nonvanishing factors contributing to Γ_C for the Smolin states [Eq. (32)]. The multiplicity is the number of occurrences.

Factor	Value	Multiplicity
$\langle \mathrm{GHZ}_0^{i_1,\ldots,i_N} | \hat{J}_z | \mathrm{GHZ}_\pi^{i_1,\ldots,i_N} \rangle$	$\frac{(N-2N_1)}{2}$	$\binom{N}{N_1}$
$\langle \mathrm{GHZ}_0^{i_1,\ldots,i_r,\ldots,i_N} | \hat{J}_x | \mathrm{GHZ}_0^{i_1,\ldots,\bar{i}_r,\ldots,i_N} \rangle$	$\frac{1}{2}$	$N2^{N-2}$
$\langle \mathrm{GHZ}_0^{i_1,\ldots,i_r,\ldots,i_N} | \hat{J}_y | \mathrm{GHZ}_\pi^{i_1,\ldots,\bar{i}_r,\ldots,i_N} \rangle$	$(-1)^{\bar{i}_r}\frac{i}{2}$	$N2^{N-2}$

which can be written as a mixture of $2n$-qubit GHZ-type states,

$$\rho_{\mathrm{Smolin}}^{(N)} = \frac{1}{2^{N-2}} \sum_{\sum_j i_j \text{ even/odd}} |\mathrm{GHZ}_0^{i_1,\ldots,i_N}\rangle\langle \mathrm{GHZ}_0^{i_1,\ldots,i_N}|, \quad (33)$$

where $|\mathrm{GHZ}_\varphi^{i_1,\ldots,i_N}\rangle = \frac{1}{\sqrt{2}}[|i_1,i_2,\ldots,i_N\rangle + e^{i\varphi}|\bar{i}_1,\bar{i}_2,\ldots,\bar{i}_N\rangle]$. The index i_j can take the values 0 and 1, and if $i_j = 0$ then $\bar{i}_j = 1$ and vice versa. For even n, then sum $\sum_{j=1}^N i_j \equiv N_1$ can take even values $\{0, 2, \ldots, n\}$, while if n is odd, then the sum can take odd values $\{1, 3, \ldots, n\}$. The kernel of ρ_{Smolin}^N is spanned by the states $|\mathrm{GHZ}_\pi^{i_1,\ldots,i_N}\rangle$ for any set $\{i_j\}$ such that $N_1 = 0, 1, \ldots, n$, and the states $|\mathrm{GHZ}_0^{i_1\ldots i_N}\rangle$ with $N_1 = 1, 3, \ldots, n-1$ if n is even and $N_1 = 0, 2, \ldots, n-1$ if n is odd.

Now we can compute the elements of the correlation matrix Γ_C using Eq. (12). The nonvanishing factors $\langle l|\hat{J}_i|l'\rangle$ are given in Table V. We obtain

$$\Gamma_C = N \cdot \mathbb{1} \quad (34)$$

for any even N. The matrix Γ_C is diagonal for the same reasons as in the previous case. We observe that $F_Q^{\max} = N$ for all N while

$$\overline{F}_Q = N > \frac{2}{3}N \quad (35)$$

for all N. Therefore, these states represent an example of Observation 3(b) for $k = 1$ and any even N.

Hence, similarly as previously, the F_Q^2 criterion does not detect the entanglement in any of these cases [cf. Eq. (5)], so the states are also not useful for sub-shot-noise interferometry for any direction $\vec{n}$, even though they are more useful than separable states when averaging over all directions.

V. CONCLUSIONS AND OUTLOOK

We have introduced two criteria based on the quantum Fisher information (QFI) for the detection of entangled states of different multiparticle entanglement classes, and consequently of their usefulness for sub-shot-noise phase estimation. Our first criterion is obtained from $F_Q[\rho, \hat{H}_{\mathrm{lin}}]$, for general linear operators of N qubits. Our second criterion is related to QFI for collective spin operators, averaged over all directions on the Bloch sphere. Both sets of criteria can be easily evaluated for a given state ρ of an arbitrary number of particles, even if the state is mixed. We considered several examples, showing in particular that the average QFI can be used to detect bound entangled states. It remains an interesting

open question whether or not there exist bound entangled states which are detected by the QFI, since this would imply that such states could be used for sub-shot-noise interferometry.

Note added in proof. Independently from our work, an article on the relationship between multiparticle entanglement and the Fisher information has appeared [72].

APPENDIX: PROOF OF OBSERVATION 3

We consider states of the form

$$\rho(p) = p|\psi\rangle\langle\psi| + (1-p)\frac{\mathbb{1}}{2^N}, \qquad (A1)$$

mixtures of a pure state and the totally mixed state. It can be shown directly from Eq. (12) that

$$\Gamma_C[\rho(p)] = \gamma_{p,N}\Gamma_C[|\psi\rangle], \quad \gamma_{p,N} = \frac{p^2 2^{N-1}}{p(2^{N-1}-1)+1} \quad (A2)$$

holds. The criteria (9) and (19) can be rewritten as $\gamma_{p,N} \leqslant \alpha_{N,k}$ and $\gamma_{p,N} \leqslant \bar{\alpha}_{N,k}$, respectively, where

$$\alpha_{N,k} = \frac{sk^2 + r^2}{F_Q[|\psi\rangle]} \qquad (A3)$$

and

$$\bar{\alpha}_{N,k} = \frac{s(k^2 + 2k - \delta_{k,1}) + r^2 - 2r - \delta_{r,1}}{4\mathrm{Tr}(\Gamma_C[|\psi\rangle])}. \qquad (A4)$$

In order to violate the criteria,

$$p > x \cdot \frac{1 - 2^{1-N}}{2}\left[1 + \sqrt{1 + \frac{1}{x}\frac{2^{3-N}}{(1-2^{1-N})^2}}\right]$$

has to hold, where $x = \alpha_{N,k}$ or $x = \bar{\alpha}_{N,k}$. The right-hand side is strictly monotonic increasing with x. If, for instance, $\alpha_{N,k} < \bar{\alpha}_{N,k}$, then the F_Q^N criterion detects the states as multiparticle entangled already for a smaller value of p than the $\overline{F}_Q^N$ criterion. Therefore, we can prove the claim by comparing the α coefficients for different states $|\psi\rangle$. However, the minimal x has to be such that at least one criterion detects the state for $p \leqslant 1$.

For $|\psi\rangle$ we employ the GHZ states from Eq. (10) and the Dicke states from Eq. (24). The results summarized in Table I ensure the following: (i) There will always be a $p \in (0,1]$ such that F_Q^{k+1} and $\overline{F}_Q^{k+1}$ detect $\rho(p)$ when $|\psi\rangle = |\mathrm{GHZ}_N\rangle$, and (ii) there will always be a $p \in (0,1]$ such that $\overline{F}_Q^{k+1}$ detects $\rho(p)$ when $|\psi\rangle = |D_N^{(N/2)}\rangle$.

Let us start with the GHZ states. We check whether $\alpha_{N,k} < \bar{\alpha}_{N,k}$ is fulfilled. This condition is equivalent to

$$2sk(N-k) - Ns\,\delta_{k,1} + 2r(N-r) - N\delta_{r,1} > 0. \qquad (A5)$$

Checking explicitly the cases (i) $1 < r < k$, (ii) $1 = r < k$, (iii) $r = 0, k > 1$, and (iv) $k = 1$ it can be shown that Eq. (A5) is always fulfilled. Hence, for the family of states $\rho(p)$ from Eq. (A1) with $|\psi\rangle = |\mathrm{GHZ}_N\rangle$, for every N and $1 \leqslant k < N$ the F_Q^{k+1} criterion detects always states in addition to the states that the $\overline{F}_Q^{k+1}$ criterion detects. This proves part (a) of Observation 3.

Let us now consider the Dicke states and check whether $\alpha_{N,k} > \bar{\alpha}_{N,k}$ is always fulfilled in this case. The condition is equivalent to

$$(sk^2 + n\delta_{k,1} - 2nk) + (r^2 + \delta_{r,1} - 2r) > 0. \qquad (A6)$$

Again checking all the cases, it can be seen that this is fulfilled for $k > 2$ and any $r < k$. Hence, in these cases the criterion $\overline{F}_Q^{k+1}$ detects states in addition to those that F_Q^{k+1} detects. In fact, the criterion F_Q^{k+1} may not even detect any of the states of this family. ∎

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