Activating hidden metrological usefulness

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We consider bipartite entangled states that cannot outperform separable states in any linear interferometer. Then, we show that these states can still be more useful metrologically than separable states if several copies of the state are provided or an ancilla is added to the quantum system. We present a general method to find the local Hamiltonian for which a given quantum state performs the best compared to separable states. We obtain analytically the optimal Hamiltonian for some quantum states with a high symmetry. We show that all bipartite entangled pure states outperform separable states in metrology. Some potential applications of the results are also suggested.

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Entanglement lies at the heart of quantum mechanics and plays an important role in quantum information theory [1]. Recently, it has been realized that entanglement can be a useful resource in very general metrological tasks. By using entangled states it is possible to overcome the shot-noise limit, corresponding to classical interferometers, in the precision of parameter estimation [2–7]. On the other hand, separable states, i.e., states without entanglement cannot overcome the classical limit. It has even been shown that quantum states with a very weak form of entanglement, called bound entanglement [8–10], can also be metrologically useful in this sense [11, 12]. However, there are highly entangled states that are not useful for metrology [13].

In what sense is metrological usefulness the property of the quantum state? It is clear that, starting from many entangled quantum states that are not useful for metrology, with local operations and classical communication (LOCC) it is possible to distill singlets, which are metrologically useful. This finding is almost trivial, as metrological "uselessness" is not conserved by LOCC operations. On the other hand, in quantum metrology experiments most LOCC operations are typically not possible. Here, we investigate how metrological usefulness can change in the two simplest cases very relevant in practice: We consider adding an ancilla to a single copy of the bipartite quantum state. We also consider providing two copies of the state [14]. These two scenarios follow the spirit in which the activation of bound entanglement and nonlocality has been studied [10, 15–17] (see Fig. 1).

In this Letter, we show that some bipartite entangled quantum states that are not useful in linear interferome-

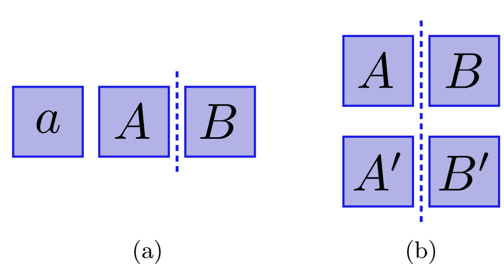

FIG. 1. (a) An ancilla ("a") is added to bipartite state ϱ_{AB}. (b) An additional copy or a different state is added to the state. In both cases, a new bipartite state is obtained, where the two parties are separated by a dashed line.

ters become useful in the cases mentioned above. These findings are quite surprising: including uncorrelated ancilla qubits can make a state metrologically useful. To support our claims, we present a general method to find the *local* Hamiltonian for which a given bipartite quantum state provides the largest gain compared to separable states. Note that this task is different, and in a sense more complex, than maximizing the quantum Fisher information. The reason is that by changing the Hamiltonian, the sensitivity achievable by separable states can also change.

Quantum Fisher information.—Before discussing our main results, we review some of the fundamental relations of quantum metrology. A basic metrological task in a *linear* interferometer is estimating the small angle θ for a unitary dynamics $U_\theta = \exp(-i\mathcal{H}\theta)$, where the Hamiltonian is the sum of *local* terms. That is, all local terms act within the subsystem and there are no interactions between the subsystems. In particular, for bipartite systems it is

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2, \tag{1}$$

where $\mathcal{H}_n$ are single-subsystem operators. The precision is limited by the Cramér-Rao bound as [18–21]

$$(\Delta\theta)^2 \geq \frac{1}{m\mathcal{F}_Q[\varrho, \mathcal{H}]}, \tag{2}$$

where m is the number of indepedendent repetitions, and the quantum Fisher information, a central quantity in quantum metrology is defined by the formula [18]

$$\mathcal{F}_Q[\varrho, \mathcal{H}] = 2\sum_{k,l} \frac{(\lambda_k - \lambda_l)^2}{\lambda_k + \lambda_l} |\langle k|\mathcal{H}|l\rangle|^2. \tag{3}$$

Here, λ_k and $|k\rangle$ are the eigenvalues and eigenvectors, respectively, of the density matrix ϱ, which is used as a probe state for estimating θ.

Metrological usefulness of a quantum state.—We call a quantum state metrologically useful, if it can outperform separable states in some metrological task, i.e., if

$$\mathcal{F}_Q[\varrho, \mathcal{H}] > \max_{\varrho_{\rm sep}} \mathcal{F}_Q[\varrho_{\rm sep}, \mathcal{H}] =: \mathcal{F}_Q^{(\rm sep)}(\mathcal{H}). \tag{4}$$

It is an intriguing task to find the operator $\mathcal{H}$, for which a given state performs the best compared to separable states. For that we define the metrological gain compared to separable states by

$$g_{\mathcal{H}}(\varrho) = \mathcal{F}_Q[\varrho, \mathcal{H}]/\mathcal{F}_Q^{(\rm sep)}(\mathcal{H}). \tag{5}$$

We are interested in the quantity

$$g(\varrho) = \max_{{\rm local}\mathcal{H}} g_{\mathcal{H}}(\varrho), \tag{6}$$

where a local Hamiltonian is just the sum of single system Hamiltonians as in Eq. (1). The maximization task looks challenging since we have to maximize a fraction, where both the numerator and the denominator depend on the Hamiltonian. (See the Supplemental Material for basic properties of the metrological gain [22].)

Maximally entangled state.—As we have mentioned, it is a difficult task to obtain $g(\varrho)$ and the optimal local Hamiltonian for any ϱ. As a first step, we consider the $d \times d$ maximally entangled state, which is defined as

$$|\Psi^{(\rm me)}\rangle = \frac{1}{\sqrt{d}} \sum_{k=1}^{d} |k\rangle|k\rangle. \tag{7}$$

Due to the symmetry of the state, the optimal Hamiltonian can straightforwardly be obtained as

$$\mathcal{H}^{(\rm me)} = D \otimes \mathbb{1} + \mathbb{1} \otimes D, \tag{8}$$

where the diagonal matrix D is given as

$$D = {\rm diag}(+1, -1, +1, -1, ...). \tag{9}$$

The details are given in the Supplemental Material [22]. For the 3×3-case, we consider the noisy quantum state

$$\varrho_{AB}^{(p)} = (1-p)|\Psi^{(\rm me)}\rangle\langle\Psi^{(\rm me)}| + p\mathbb{1}/d^2, \tag{10}$$

which is useful if [22]

$$p < \frac{25 - \sqrt{177}}{32} \approx 0.3655. \tag{11}$$

(See the Supplemental Material for the definition of the related notion of robustness of metrological usefulness [22].)

Activation by an ancilla qubit.—Now we consider the previous state, after a pure ancilla qubit is added

$$\varrho^{(\rm anc)} = |0\rangle\langle 0|_a \otimes \varrho_{AB}^{(p)}. \tag{12}$$

The setup is depicted in Fig. 1(a). Then, with the operator

$$\mathcal{H}^{(\rm anc)} = 1.2 C_{aA} \otimes \mathbb{1}_B + \mathbb{1}_{aA} \otimes D_B, \tag{13}$$

where an operator acting on the ancilla and A is

$$C_{aA} = \frac{9}{20}(2\sigma_x + \sigma_z)_a \otimes |0\rangle\langle 0|_a + \mathbb{1}_a \otimes (|2\rangle\langle 2|_a - |1\rangle\langle 1|_a), \tag{14}$$

we have $g_{\mathcal{H}^{(\rm anc)}}(\varrho^{(\rm anc)}) > 1$ if $p < 0.3752$ [c.f. Eq. (11)]. Hence larger part of the noisy maximally entangled states are useful in the case with the ancilla.

Activation by adding extra copies.—We consider now two copies of the noisy 3×3 maximally entangled state

$$\varrho^{(\rm tc)} = \varrho_{AB}^{(p)} \otimes \varrho_{A'B'}^{(p)}. \tag{15}$$

The setup is shown in Fig. 1(b). Then, with the two-copy operator

$$\mathcal{H}^{(\rm tc)} = D_a \otimes D_{A'} \otimes \mathbb{1}_{BB'} + \mathbb{1}_{AA'} \otimes D_B \otimes D_{B'}, \tag{16}$$

we have $g_{\mathcal{H}^{(\rm tc)}}(\varrho^{(\rm tc)}) > 1$ if $p < 0.4164$ [c.f. Eq. (11)]. Hence larger part of the noisy maximally entangled states are useful in the two-copy case, than with a single copy. So far we have studied the 3×3 case. For the 2×2-case, see the Supplemental Material [22].

Observation 1.—In summary, we have just shown that there are bipartite states with the following properties. (i) They are not more useful than separable states considering any local Hamiltonian. (ii) By adding an ancilla or two copies, they are more useful than separable states for some local Hamiltonian. For the case of an added ancilla, the new subsystems are now aA and B, and the Hamiltonian contains interactions between the ancilla a and A. In the two-copy case, the new subsystems are AA' and BB', and the Hamiltonian contains interactions between A and A', and between B and B'. Note that in both cases, the extra interactions increase the metrological capabilities of separable states. Still, simple algebra shows that in both cases the metrological gain can stay the same or can increase, but cannot decrease [22].

So far, we exploited the symmetries of quantum states to obtain the Hamiltonian leading to the largest metrological gain. We now present a general method to compute $g(\varrho)$ numerically.

Method for finding the optimal Hamiltonian.—We need to maximize $\mathcal{F}_Q[\varrho,\mathcal{H}]$ over $\mathcal{H}$ for a given ϱ. However, since it is convex in $\mathcal{H}$, maximizing it over $\mathcal{H}$ is a difficult task. Instead of the quantum Fisher information, let us consider the error propagation formula

$$(\Delta\theta)^2_M = \frac{(\Delta M)^2}{\langle i[M,\mathcal{H}]\rangle^2}, \tag{17}$$

which provides a bound on the quantum Fisher information [22, 36–38]

$$\mathcal{F}_Q[\varrho,\mathcal{H}] \geq 1/(\Delta\theta)^2_M. \tag{18}$$

We will now minimize Eq. (17).

Observation 2.—The error propagation formula given in Eq. (17) can be minimized over $\mathcal{H}$ for a given M and ϱ as follows.

Proof. Simple algebra yields

$$\langle i[M,\mathcal{H}]\rangle = \mathrm{Tr}(A_1\mathcal{H}_1) + \mathrm{Tr}(A_2\mathcal{H}_2), \tag{19}$$

where $A_n = \mathrm{Tr}_{\{1,2\}\backslash n}(i[\varrho, M])$ are operators acting on a single subsytem. Hence, we have to maximize Eq. (19) over $\mathcal{H}_1$ and $\mathcal{H}_2$. We choose the constraints

$$c_n \mathbb{1} \pm \mathcal{H}_n \geq 0, \tag{20}$$

where $n = 1,2$ and $c_n > 0$ is some constant. This way we make sure that $\sigma_{\min}(\mathcal{H}_n) \geq -c_n$, and $\sigma_{\max}(\mathcal{H}_n) \leq +c_n$, for $n = 1,2$, where $\sigma_{\min}(X)$ and $\sigma_{\max}(X)$ denote the smallest and largest eigenvalues of X. The optimal $\mathcal{H}_n$ is the one that maximizes $\mathrm{Tr}(A_n\mathcal{H}_n)$ under these constraints. It can straightforwardly be obtained as

$$\mathcal{H}_n^{(\rm opt)} = U_n \tilde{D}_n U_n^\dagger, \tag{21}$$

where the eigendecompisition of A is given as $A_n = U_n D_n U_n^\dagger$ and $(\tilde{D}_n)_{k,k} = c_n s((D_n)_{k,k})$, where $s(x) = 1$ if $x \geq 0$, and -1 otherwise. Clearly, $\mathcal{H}_n^{(\rm opt)}$ has the same eigenvectors as A_n and has only eigenvalues $+c_n$ and $-c_n$. ■

We already know how to optimize $\mathcal{H}$ for a given M. However, how do we find the optimal M? This can be done with the well-known formula for the symmetric logarithmic derivative [21]

$$M_{\rm opt} = 2i\sum_{k,l}\frac{\lambda_k - \lambda_l}{\lambda_k + \lambda_l}|k\rangle\langle l|\langle k|\mathcal{H}|l\rangle. \tag{22}$$

For a given $\mathcal{H}$, the error propagation formula given in Eq. (17) is minimized for $M = M_{\rm opt}$ [22, 37].

Iterative method.—We can now construct the following procedure for minimizing Eq. (17). First we choose a random M. Then, repeat the following two steps.

(Step 1) Determine the optimal $\mathcal{H}$ for a given M using Observation 2.

(Step 2) Determine the optimal M for a given $\mathcal{H}$ using Eq. (22).

A see-saw procedure similar in spirit has been used to make the optimization of the metrological performance over density matrices in Refs. [12, 39, 40].

After several iterations of the two steps above, we obtain the maximal quantum Fisher information over a certain set of Hamiltonians. Based on that, we can calculate the quantity

$$g_{c_1,c_2}(\varrho) = \max_{\mathcal{H}_1,\mathcal{H}_2}\frac{\mathcal{F}_Q(\varrho,\mathcal{H}_1\otimes\mathbb{1}+\mathbb{1}\otimes\mathcal{H}_2)}{\mathcal{F}_Q^{(\rm sep)}(c_1,c_2)}, \tag{23}$$

where we assumed that $\mathcal{H}_n$ are constrained with Eq. (20). The separable limit for Hamiltonians of the form (1) is [12, 41]

$$\mathcal{F}_Q^{(\rm sep)}(\mathcal{H}) = \sum_{n=1,2}[\sigma_{\max}(\mathcal{H}_n) - \sigma_{\min}(\mathcal{H}_n)]^2, \tag{24}$$

which leads to $\mathcal{F}_Q^{(\rm sep)}(c_1,c_2) = 4(c_1^2+c_2^2)$. Then, the gain can be expressed as

$$g(\varrho) = \max_{c_2} g_{c_1,c_2}(\varrho), \tag{25}$$

where the optimization is only over c_2, and, without the loss of generality, we set $c_1 = 1$. The optimal c_2 can be obtained from an analytical formula [22]. Hence we computed the maximum of the fraction, (5), for local Hamiltonians.

We now stress the following. If we determine the optimal $\mathcal{H}$ for a given M using Observation 2, the eigenvalues of the optimal $\mathcal{H}_n$ satisfying Eq. (20) are $\pm c_n$. Let us assume the contrary. Let us assume that for a state ϱ and for given c_1, c_2 we know the optimal $\mathcal{H}_1$ and $\mathcal{H}_2$, and $\mathcal{H}_n$ fulfill Eq. (20), but not all eigenvalues are $\pm c_n$. We observe that $\langle i[M,\mathcal{H}]\rangle$ is a linear function of the eigenvalues of $\mathcal{H}_n$, thus it takes its maximum at the eigenvalues corresponding to the boundary of the allowed region. Hence, we can always replace the eigenvalues of $\mathcal{H}_n$ by $\pm c_n$ such that $\langle i[M,\mathcal{H}]\rangle$ will not decrease, and $1/(\Delta\theta)^2_M$ will not decrease either.

Using the numerical method above, we obtain a slightly larger value for the noise bounds of metrological usefulness for the state with an ancilla, (12). $g(\varrho^{(\rm anc)}) > 1$ if $p < 0.3941$. The same is true for the case of the two copies of the noisy maximally entangled state, (15). We obtain $g(\varrho^{(\rm tc)}) > 1$ if $p < 0.4170$.

For states with a high symmetry, such as isotropic states [42, 43], and Werner states [44], we obtained the optimal Hamiltonian analytically and determined the subset of these states that are metrologically useful [22]. We also used that to verify our numerical methods.

Activation of a bound entangled state by a separable state.—While bound entangled or non-distillable states [8, 9] are considered weakly entangled, they can share many properties with highly entangled states. For example, there are bound entangled states that can reach the Heisenberg scaling in metrological applications [11]. It has also been shown that bipartite bound entangled states, which have a positive semidefinite partial transposition (PPT), can be useful for metrology [12]. Moreover, bipartite PPT entangled states can even have a high Schmidt-rank [45].

Let us now consider a PPT entangled state $\varrho_{AB}^{(\rm PPT)}$ that is not useful for quantum metrology. Then, we look for a separable state $\varrho^{(\rm sep)}$ such that $\varrho_{AB}^{(\rm PPT)} \otimes \varrho_{A'B'}^{(\rm sep)}$ becomes useful. Hence, in this case we have to optimize not only over $\mathcal{H}$, M, but also over the separable state. Simple convexity arguments show that the maxiumum is taken when we have a pure product state, $\varrho_{A'B'}^{(\rm sep)} = \varrho_{A'}^{(\rm anc)} \otimes \varrho_{B'}^{(\rm anc)}$, which corresponds to two ancillas at the two parties. In fact, even a single ancilla qubit is sufficient for activation.

Activation of a PPT entangled state by an ancilla qubit.—We now consider a PPT entangled state, that is not useful metrologically, and $g(\varrho_{AB}) = 1$. However, with an ancilla it becomes useful, $g(\varrho_{(aA)(B)}) > 1$. We show here examples for $d \times d$ dimensional PPT states found in Ref. [12] for odd dimensions d up to $d \leq 11$. See Table I for the numerical results.

Note that here we fixed $c_i = 1$ for the coefficients of the local Hamiltonians $\mathcal{H}_i$, $i = 1, 2$. However, numerics suggests that optimization over c_i does not help to increase g in the case of two ancillas (last column), due to the permutational symmetry of the states. Optimization over c_i helps only marginally in the case of one ancilla (third column). For instance, in the case of $d = 7$, the g value raises from 1.0096 (corresponding to $c_2 = 1$) to 1.0098 (corresponding to $c_2 \simeq 1.034$) if we optimize over c_2.

d	p^*	Gain with one ancilla	Gain with two ancillas
3	0.0006	1.0007	1.0011
5	0.0960	1.0094	1.0190
7	0.1377	1.0096	1.0195
9	0.1631	1.0090	1.0181
11	0.1807	1.0081	1.0165

TABLE I. Activation of the metrological usefulness found numerically in two-qudit systems. (First column) Local dimension d, where d is odd. For even d up to $d \leq 11$, we did not find activation in the examples of PPT two-qudit states considered. (Second column) White noise fractions of p^* added to the PPT states given by Ref. [12] such that $g_{1,1}(\varrho_{AB}) = 1.0000$, that is, they are not useful metrologically. (Third column) Metrological gain after an ancilla is added to Alice's system, $g_{1,1}(\varrho_{(aA)(B)})$. The states become useful as demonstrated by $g_{1,1}(\varrho_{(aA)(B)}) > 1$. (Fourth column) Metrological gain after a further ancilla is added to Bob's system, $g_{1,1}(\varrho_{(aA)(Bb)})$. The state becomes even more useful metrologically.

Entanglement detection.—Our method can be used for entanglement detection. It identifies the Hamiltonians with which a given quantum state performs better than separable states and hence it is detected as entangled. If we add ancillas or extra copies of the quantum state, the criterion can be even more powerful.

Random states.—We can use our method to determine the distribution of metrological usefulness of random pure or mixed states of a given size. For instance, for 3×3 systems, pure states typically are close to be maximally useful, while this is not the case if we look for the usefulness with respect to a given Hamiltonian. For the numerical result, please see the Supplemental Material [22].

Usefulness of entangled bipartite pure states.—Next we will consider the usefulness of bipartite pure states.

Observation 3.—All entangled bipartite pure states are metrologically useful. (For the two-qubit case, see Ref. [13].)

Proof.—Let us consider a pure state with a Schmidt decomposition

$$|\Psi\rangle = \sum_{k=1}^{s} \sigma_k |k\rangle_a |k\rangle_B, \quad (26)$$

where s is the Schmidt number, and the real positive σ_k Schmidt coefficients are in a descending order. We define

$$\mathcal{H}_a = \sum_{n=1,3,5,\ldots,\tilde{s}-1} |+\rangle\langle+|_{{\rm A},n,n+1} - |-\rangle\langle-|_{{\rm A},n,n+1}, \quad (27)$$

where $\tilde{s}$ is the largest even number for which $\tilde{s} \leq s$, and

$$|\pm\rangle_{\mathrm{A},n,n+1} = (|n\rangle_a \pm |n+1\rangle_a)/\sqrt{2}. \qquad (28)$$

We define $\mathcal{H}_B$ in a similar manner. We also define the collective Hamiltonian

$$\mathcal{H}_{AB} = \mathcal{H}_a \otimes \mathbb{1} + \mathbb{1} \otimes \mathcal{H}_B. \qquad (29)$$

Then, we have $\langle \mathcal{H}_{AB} \rangle_\Psi = 0$. Direct calculation yields

$$\mathcal{F}_Q[|\Psi\rangle, \mathcal{H}_{AB}] = 4(\Delta\mathcal{H}_{AB})^2_\Psi = 8 \sum_{n=1,3,5,...,\tilde{s}-1} (\sigma_n + \sigma_{n+1})^2, \qquad (30)$$

which is larger than the separable bound, $\mathcal{F}_Q^{(\mathrm{sep})} = 8$, whenever the Schmidt rank is larger than 1. For even s, this can be seen noting that

$$\mathcal{F}_Q[|\Psi\rangle, \mathcal{H}_{AB}] > 8 \sum_{n=1}^{s} \sigma_n^2 \qquad (31)$$

holds, where we used Eq. (30) to evaluate the left-hand side of Eq. (31), and we also took into account that $\sigma_n > 0$ for $n = 1, 2, 3, ...,$ and $\sum_{n=1}^{s} \sigma_n^2 = 1$. For odd s, we need that

$$\mathcal{F}_Q[|\Psi\rangle, \mathcal{H}_{AB}] \geq 8 \left(\sum_{n=1}^{s-1} \sigma_n^2 + 2\sigma_1\sigma_2 \right) > 8 \sum_{n=1}^{s} \sigma_n^2 \qquad (32)$$

holds, where we used that $\sigma_1\sigma_2 > \sigma_s^2$. ■

We can even consider several copies of a quantum state. In the Supplemental Material, we prove that for infinite number of copies of entangled pure quantum states the metrological gain is maximal [22].

Conclusions.—We showed that entangled quantum states that cannot outperform separable states in any linear interferometer can still be more useful than separable states, if several copies of them are considered or an ancilla is added to the system. This is a surprising result which shows that the relationship between quantum metrology and the structure of quantum states requires further study. We presented a method to find the Hamiltonian for carrying out metrology in a linear interferometer with a given quantum state that provides the largest gain compared to the precision achievable by separable states. In the Letter we considered bipartite problems, thus it would be important to extend this approach to multipartite systems and examine the scaling of the metrological gain for noisy quantum states. It would be also interesting to look for application in entanglement detection [1], and witnessing the dimension of quantum systems [46–49], where the results of the preliminary analysis seem to be promising. (See the Supplemental Material [22].)

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Supplemental Material for "Activating hidden metrological usefulness"

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The Supplemental Material contains some additional results. We present some properties of the metrological gain. We discuss the relation between the error propagation formula and the quantum Fisher information. We present some details of the optimization over the c_2 parameter of the Hamiltonian. We calculate the optimal Hamiltonian analytically for isotropic states and Werner states. We present concrete calculations for metrology with two-qubit singlets and ancillas. We show how to use our formulas to bound the metrological usefulness by a single operator expectation value. We consider metrology with multi-particle states, if some particles are united into a single party. We consider metrology with an infinite number of copies of arbitrary entangled pure states. We present an alternative optimization over local Hamiltonians. We present numerical results concerning metrology with random pure and mixed states. We determine the maximum achievable precision in a multiparticle system. We define the robustness of metrological usefulness. We show how to witness the dimension of a quantum state based on quantum metrology.

PROPERTIES OF THE METROLOGICAL GAIN IN MULTIPARTITE SYSTEMS

We consider the question, how the metrological gain defined in Eq. (6) behaves if we add an ancilla to the subsystem or provide an additional state, as depicted by Fig. 1. We will now show that it cannot decrease in neither of these cases. We will also show that the metrological gain is convex.

(i) Let us see first adding an ancilla "a" to the system AB. For the gain, we have

$$\begin{aligned} g(\varrho_{AB}) &= g_{\mathcal{H}_{\rm opt}}(\varrho_{AB}) \\ &= g_{\mathcal{H}'_{\rm opt}}(|0\rangle\langle 0|_a \otimes \varrho_{AB}) \le g(|0\rangle\langle 0|_a \otimes \varrho_{AB}), \end{aligned} \quad \text{(S1)}$$

where a Hamiltonian for the aAB system is given as

$$\mathcal{H}'_{\rm opt} = \mathbb{1}_a \otimes (\mathcal{H}_{\rm opt})_{AB}. \quad \text{(S2)}$$

Here, $\mathcal{H}_{\rm opt}$ is the local Hamiltonian acting on AB for which the gain is the largest. The second equality in Eq. (S1) holds, since the quantum Fisher information has the property

$$\mathcal{F}_Q[\varrho_1 \otimes \varrho_2, \mathcal{H}_1 \otimes \mathbb{1} + \mathbb{1} \otimes \mathcal{H}_2] = \mathcal{F}_Q[\varrho_1, \mathcal{H}_1] + \mathcal{F}_Q[\varrho_2, \mathcal{H}_2]. \quad \text{(S3)}$$

For $\mathcal{H}_1 = 0$, we have the special case

$$\mathcal{F}_Q[\varrho_1 \otimes \varrho_2, \mathbb{1} \otimes \mathcal{H}_2] = \mathcal{F}_Q[\varrho_2, \mathcal{H}_2]. \quad \text{(S4)}$$

The inequality in Eq. (S1) holds, since in the extended system there might be a Hamiltonian with a gain larger than that of $\mathcal{H}'_{\rm opt}$. In other words, for any $\mathcal{H}$ and any ϱ, $g_{\mathcal{H}}(\varrho) \le g(\varrho)$ holds.

(ii) For an additional copy of a state, analogously, we have

$$\begin{aligned} g(\varrho_{AB}) &= g_{\mathcal{H}_{\rm opt}}(\varrho_{AB}) \\ &= g_{\mathcal{H}''_{\rm opt}}(\varrho_{AB} \otimes \sigma_{A'B'}) \le g(\varrho_{AB} \otimes \sigma_{A'B'}), \end{aligned} \quad \text{(S5)}$$

where a Hamiltonian for the $ABA'B'$ system is given as

$$\mathcal{H}''_{\rm opt} = (\mathcal{H}_{\rm opt})_{AB} \otimes \mathbb{1}_{A'B'}. \quad \text{(S6)}$$

Here, $\sigma_{A'B'}$ is the additional state provided. In the special case of two copies we have $\sigma = \varrho$. If we replace the role of ϱ_{AB} and $\sigma_{A'B'}$ in Eq. (S5), we arrive at

$$g(\sigma_{AB}) \le g(\sigma_{AB} \otimes \varrho_{A'B'}). \tag{S7}$$

From Eqs. (S5) and (S7), after trivial relabelling of the parties follows

$$g(\varrho_{AB} \otimes \sigma_{A'B'}) \ge \max[g(\varrho_{AB}), g(\sigma_{A'B'})], \tag{S8}$$

where $\max(a, b)$ denotes the maximum of a and b.

(iii) The metrological gain is convex under mixing as can be seen from the series of inequalities

$$\begin{aligned} g(p\varrho + (1-p)\sigma) &= g_{\mathcal{H}_{\rm opt}}(p\varrho + (1-p)\sigma) \\ &\le p g_{\mathcal{H}_{\rm opt}}(\varrho) + (1-p) g_{\mathcal{H}_{\rm opt}}(\sigma) \\ &\le p g(\varrho) + (1-p) g(\sigma), \end{aligned} \tag{S9}$$

where $0 \le p \le 1$. Here, $\mathcal{H}_{\rm opt}$ is the Hamiltonian acting on $p\varrho + (1-p)\sigma$ for which the gain is the largest. The first inequality is due to the convexity of the quantum Fisher information. The second inequality is due to the fact, that in general for any $\mathcal{H}$ and any ϱ, $g_{\mathcal{H}}(\varrho) \le g(\varrho)$ holds.

RELATION BETWEEN THE ERROR-PROPAGATION FORMULA AND THE QUANTUM FISHER INFORMATION

Equation (18) has been described from various point of views in Refs. [36–38]. These ideas have been used in Refs. [2, 23–25]. Related ideas have also been used in Refs. [39, 40] for the optimization of the quantum Fisher information.

For completeness, now we prove Eq. (18) very briefly. Let us consider the uncertainty relation [26, 38]

$$(\Delta A)^2_{\varrho} \mathcal{F}_Q[\varrho, B] \ge \langle i[A, B]\rangle^2_{\varrho}, \tag{S10}$$

where ϱ is a quantum state, and A and B are observables. Ref. [38] stresses the fact that Eq. (S10) is just a strengthening of the Heisenberg uncertainty relation. Then, making the substitutions in Eq. (S10) that $B = \mathcal{H}$, $A = M$, we find that

$$(\Delta\theta)^2_M \equiv \frac{(\Delta M)^2}{\langle i[M, \mathcal{H}]\rangle^2} \ge 1/\mathcal{F}_Q[\varrho, \mathcal{H}] \tag{S11}$$

holds, where the left hand-side is just the error propagation formula. We now show that setting M to the symmetric logarithmic derivative $M_{\rm opt}$ given in Eq. (22) the inequality in Eq. (S11) is saturated. This can be proved using the identities ${\rm Tr}(M_{\rm opt}^2\varrho) = \mathcal{F}_Q[\varrho, \mathcal{H}]$, ${\rm Tr}(M_{\rm opt}\varrho) = 0$, $\langle i[M_{\rm opt}, \mathcal{H}]\rangle = {\rm Tr}(M_{\rm opt}^2\varrho)$.

Note that Eq. (18) is different from the Cramér-Rao bound, (2). The relation between the precision $(\Delta\theta)^2$ for some estimator and the error propagation formula $(\Delta\theta)^2_M$ is not trivial. For any estimator

$$(\Delta\theta)^2 \ge \frac{1}{m}(\Delta\theta)^2_{M=M_{\rm opt}} \tag{S12}$$

holds. In the limit of large number of repetitions m, and if certain further conditions are fulfilled, Eq. (S12) can be saturated by the best estimator. Then, such a $(\Delta\theta)^2$ would also saturate the Cramér-Rao bound, (2) [20].

ANALYSIS OF THE OPTIMIZATION METHOD

The maximization of the error propagation formula can be expressed using a variational formulation as [39]

$$\begin{aligned} &\max_{\mathcal{H}} \max_{M} 1/(\Delta\theta)^2_M \\ &\quad = \max_{\mathcal{H}} \max_{M} \langle i[M, \mathcal{H}]\rangle^2/(\Delta M)^2 \\ &\quad = \max_{\mathcal{H}} \max_{M} \langle i[M, \mathcal{H}]\rangle^2/\langle M^2\rangle \\ &\quad = \max_{\mathcal{H}} \max_{M} \max_{\alpha} \{-\alpha^2\langle M^2\rangle + 2\alpha\langle i[M, \mathcal{H}]\rangle\} \\ &\quad = \max_{\mathcal{H}} \max_{M'} \{-\langle (M')^2\rangle + 2\langle i[M', \mathcal{H}]\rangle\}, \end{aligned} \tag{S13}$$

where M' takes the role of αM. Then, the function is concave in M' and linear in $\mathcal{H}$, and the two-step see-saw algorithm we have described will find better and better Hamiltonians. However, the function in Eq. (S13) is not strictly concave in $(\mathcal{H}, M')$. Hence, our iterative numerical procedure will always lead to Hamiltonians with an increasing quantum Fisher information, however, it is not guaranteed to find a global optimum. Based on extensive numerical experience, for a mixed state in bipartite systems of dimension 3×3 the algorithm converges very fast, and from 10 trials at least 2-3, typically more will lead to the global optimum. The 10 trials of 100 steps can take 5 seconds on a state of the art laptop computer. For larger systems, it is worth to make many trials for few steps, and continue the best one for many steps.

We can understand the expression better as follows. If we subtract a term $4\langle\mathcal{H}^2\rangle$ from the expression appearing on the right-hand side of Eq. (S13), then we will arrive at

$$-\langle ZZ^\dagger\rangle, \tag{S14}$$

where the non-Hermitian matrix is defined as

$$Z = M' + i2\mathcal{H}. \tag{S15}$$

Equation (S14) is clearly concave in $(\mathcal{H}, M')$ but a maximization will converge to $(\mathcal{H}, M') = 0$. The maximization in Eq. (S13) is equivalent to maximizing Eq. (S14) with a quadratic equality constraint $\langle\mathcal{H}^2\rangle = c$, where c is some constant. We can maximize Eq. (S14) for a range of c values, and the largest of these maxima will be the global maximum.

EFFICIENT OPTIMIZATION OVER c_2.

Let us define $\tilde{\mathcal{H}}_k = \mathcal{H}_k/c_k$. Based on Eq. (20),

$$-\mathbb{1} \le \tilde{\mathcal{H}}_k \le \mathbb{1} \tag{S16}$$

hold. Then, the Hamiltonian, (1), becomes

$$\mathcal{H} = c_1\tilde{\mathcal{H}}_1 + c_2\tilde{\mathcal{H}}_2. \tag{S17}$$

In this section, we show how to optimize the metrological performance for Hamiltonians of the form (S17). This will mean an optimization over c_2, while c_1 can be taken to be 1.

For a such $\tilde{\mathcal{H}}_k$ Hamiltonians, the expression in Eq. (19) can be written as

$$\langle i[M,\mathcal{H}]\rangle = c_1\mathrm{Tr}(A_1\tilde{\mathcal{H}}_1) + c_2\mathrm{Tr}(A_2\tilde{\mathcal{H}}_2), \tag{S18}$$

where $A_n = \mathrm{Tr}_{\{1,2\}\setminus n}(i[\varrho, M])$. Then, in order to maximize $\sqrt{(\Delta\theta)^2_M/\mathcal{F}_Q^{(\mathrm{sep})}}$, we need to calculate

$$\max_{c_1,c_2} \frac{c_1\mathrm{Tr}(A_1\tilde{\mathcal{H}}_1) + c_2\mathrm{Tr}(A_2\tilde{\mathcal{H}}_2)}{4\sqrt{c_1^2+c_2^2}}. \tag{S19}$$

The optimal value is at

$$\frac{c_2}{c_1} = \frac{\mathrm{Tr}(A_2\tilde{\mathcal{H}}_2)}{\mathrm{Tr}(A_1\tilde{\mathcal{H}}_1)}. \tag{S20}$$

Without the loss of generality, we set $c_1 = 1$, then c_2 can be obtained from Eq. (S20).

One can add a third step to the two-step procedure of the paper, in which c_2 is updated according to the formula Eq. (S20). For a smoother convergence, one can change c_2 not abruptly, but only by a small value changing it in the direction of the value suggested by Eq. (S20).

METROLOGY WITH ISOTROPIC STATES

We will now consider quantum metrology with isotropic states, which are defined as [42]

$$\varrho_p = pP_d^{(+)} + (1-p)\frac{\mathbb{1}}{d^2}, \tag{S21}$$

where $P_d^{(+)}$ is a projector to the maximally entangled state $|\Psi^{(\mathrm{me})}\rangle$ defined in Eq. (7).

We consider a Hamiltonian of the form

$$\mathcal{H}_{\mathrm{coll}} = \mathcal{H}_1 \otimes \mathbb{1} + \mathbb{1} \otimes \mathcal{H}_2. \tag{S22}$$

The subscript "coll" indicates that the Hamiltonian acts on both subsystems, in contrast to $\mathcal{H}_1$ and $\mathcal{H}_2$ that act only on one of the subsystems. The Hamiltonian is local, since it does not contain interactions terms.

Isotropic states are invariant under transformations of the type

$$U \otimes U^*, \tag{S23}$$

where U is a single-qudit unitary and "*" denotes element-wise conjugation. Hence, isotropic states are invariant under the Hamiltonian

$$\mathcal{H}_{\mathrm{inv}}^{(\mathrm{iso})}(\mathcal{H}) = \mathcal{K} \otimes \mathbb{1} - \mathbb{1} \otimes \mathcal{K}^*, \tag{S24}$$

where $\mathcal{K}$ is a Hermitian operator.

Observation S1.—For short times, the action of the Hamiltonian $\mathcal{H}_{\mathrm{coll}}$ given in Eq. (S22) is the same as the action of

$$\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H}^{(\mathrm{iso})}) = \mathcal{H}^{(\mathrm{iso})} \otimes \mathbb{1} + \mathbb{1} \otimes (\mathcal{H}^{(\mathrm{iso})})^*, \tag{S25}$$

where the single party Hamiltonian is defined as

$$\mathcal{H}^{(\mathrm{iso})} = (\mathcal{H}_1 + \mathcal{H}_2^*)/2. \tag{S26}$$

Proof. Let us define

$$\Delta^{(\mathrm{iso})} = (\mathcal{H}_2^* - \mathcal{H}_1)/2. \tag{S27}$$

In the rest of the section, we omit the superscript "iso" in $\mathcal{H}_{\mathrm{inv}}^{(\mathrm{iso})}, \mathcal{H}^{(\mathrm{iso})}, \Delta^{(\mathrm{iso})}$.

Then, simple algebra shows that

$$\mathcal{H}_{\mathrm{coll}} + \mathcal{H}_{\mathrm{inv}}(\Delta) = \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}. \tag{S28}$$

Hence, for small t

$$e^{-i\mathcal{H}_{\mathrm{coll}}t}e^{-i\mathcal{H}_{\mathrm{inv}}(\Delta)t} \approx e^{-i\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H})t} \tag{S29}$$

holds. The isotropic state is invariant under the action of $\mathcal{H}_{\mathrm{inv}}(\Delta)$, since the corresponding unitary is of the form given in Eq. (S23). Hence, the action of $\mathcal{H}_{\mathrm{coll}}$ is the same as the action of $H_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H})$ for small t. ■.

Note that in the quantum metrology problems we consider we always estimate the parameter t around $t = 0$ assuming that it is small. Hence, the approximate equality in Eq. (S29) is sufficient.

Observation S2.—Replacing the evolution by $\mathcal{H}_{\mathrm{coll}}$ given in Eq. (S22) by the evolution by $\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}$ given in Eq. (S25) does not decrease the metrological gain. Hence, when looking for the Hamiltonian with the largest metrological gain, it is sufficient to look for Hamiltonians of the form (S25).

Proof. When the evolution by $\mathcal{H}_{\mathrm{coll}}$ given in Eq. (S22) is replaced by the evolution by $\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}$ then the quantum Fisher information does not change, while $\mathcal{F}_Q^{(\mathrm{sep})}$ does not increase. The latter can be seen as follows. Let us define

$$f(X) = [\sigma_{\max}(X) - \sigma_{\min}(X)]^2, \tag{S30}$$

where X is some matrix. Then, based on Eq. (24), $\mathcal{F}_Q^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}) = f(\mathcal{H}_1) + f(\mathcal{H}_2)$ holds. On the other hand, we have $\mathcal{F}_Q^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}) = 2f(\mathcal{H})$. Knowing that f is matrix convex, we obtain that

$$\mathcal{F}_Q^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}) \leq \mathcal{F}_Q^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}). \tag{S31}$$

■

We will now use that for a pure state mixed with white noise it is possible to obtain a closed formula for the quantum Fisher information for any operator A as a function of p as [4]

$$\mathcal{F}_Q[\varrho_p, A] = \frac{p^2}{p + 2(1-p)d^{-2}} 4(\Delta A)^2_{\Psi^{(\mathrm{me})}}, \tag{S32}$$

where ϱ_p given in Eq. (S21). Let us simplify Eq. (S32). For the case of $A = \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}$, we can rewrite the variance as

$$(\Delta\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})})^2_{\Psi^{(\mathrm{me})}} = 2\frac{\mathrm{Tr}(\mathcal{H}^2)}{d} + 2\langle\mathcal{H}\otimes\mathcal{H}^*\rangle_{\Psi^{(\mathrm{me})}} - 4\frac{\mathrm{Tr}(\mathcal{H})^2}{d^2}, \tag{S33}$$

where we used that for the reduced state of $|\Psi^{(\mathrm{me})}\rangle$ we have $\rho_{\mathrm{red}1} = \rho_{\mathrm{red}2} = \mathbb{1}/d$. Next, we use the fact that

$$\langle\mathcal{H}\otimes\mathcal{H}^*\rangle_{\Psi^{(\mathrm{me})}} = \frac{1}{d}\mathrm{Tr}(\mathcal{H}^2) \tag{S34}$$

holds. Hence, for the quantum Fisher information we obtain

$$\mathcal{F}_Q[\varrho_p, \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}] = \frac{16p^2}{pd^2 + 2(1-p)}\left[d\mathrm{Tr}(\mathcal{H}^2) - \mathrm{Tr}(\mathcal{H})^2\right]. \tag{S35}$$

Based on Eq. (S35) and on Eq. (24), the metrological gain for a given Hamiltonian $\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}$ is obtained as

$$g(\varrho_p, \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}) = \frac{16p^2}{pd^2 + 2(1-p)} r(\mathcal{H}), \tag{S36}$$

where $r(\mathcal{H})$ is defined as

$$r(\mathcal{H}) = \frac{[d\sum_k h_k^2 - (\sum_k h_k)^2]}{2(h_{\max} - h_{\min})^2}, \tag{S37}$$

and h_k denote the eigenvalues of $\mathcal{H}$.

Let us now consider the metrological gain for the isotropic state for various Hamiltonians.

Observation S3.—Isotropic states have the best metrological performance with respect to separable states with the Hamiltonian given by

$$\mathcal{H}_{\mathrm{best}} = \mathrm{diag}(+1, -1, +1, -1, ...). \tag{S38}$$

Based on Eq. (3), the corresponding metrological performance is described by

$$g(\varrho_p, \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H}_{\mathrm{best}})) = \frac{2p^2[d^2 - \alpha]}{pd^2 + 2(1-p)}, \tag{S39}$$

where α is defined as

$$\alpha = \begin{cases} 0 & \text{for even } d, \\ 1 & \text{for odd } d. \end{cases} \tag{S40}$$

No other Hamiltonian $\mathcal{H}$ corresponds to a better performance.

Equation (S39) is maximal for $p = 1$ and has the value

$$g(\varrho_p, \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H}_{\mathrm{best}})) = 2\frac{d^2 - \alpha}{d^2}, \tag{S41}$$

which is 2 for even d and approaches 2 for large d for odd d.

Proof. Without the loss of generality, let us set $h_{\min} = -1$ and $h_{\max} = +1$. Then, the denominator of Eq. (S37) is 8. Let us consider now the numerator. The maximum of the numerator of Eq. (S37) will be clearly taken by a configuration for which $h_k = \pm 1$. The first term is d^2. Looking at the second term, we see that the numerator is maximized by $\{h_k\}_{k=1}^d = \{+1, -1, +1, -1, ...\}$. We find that the maximum is obtained for the Hamiltonian (S38). ■

Next, we determine which isotropic states are useful metrologically.

Observation S4.—If

$$p > p_{\mathrm{m}} = \frac{d^2 - 2}{4(d^2 - \alpha)} + \sqrt{\frac{(d^2 - 2)^2}{16(d^2 - \alpha)^2} + \frac{1}{d^2 - \alpha}} \tag{S42}$$

holds then the isotropic state ϱ_p is useful for metrology with the Hamiltonian (S38). Otherwise, the isotropic state is not useful with any other Hamiltonian.

Proof. We look for the p for which the righ-hand side of Eq. (S41) is 1. ■

Note that $p_{\mathrm{m}} > 1/2$ for all d while for large d it converges to $1/2$. On the other hand, the isotropic state given in Eq. (S21) is entangled if $p > 1/d$. Hence, for all $d \geq 2$ there are isotropic states there are entangled but not useful for metrology.

Let us now look for the Hamiltonian of the type (S25) with which the isotropic states have the worst metrological performance.

Observation S5.—Isotropic states have the worst metrological performance with respect to separable states with the Hamiltonian given by

$$\mathcal{H}_{\mathrm{worst}} = \mathrm{diag}(1, -1, 0, 0, ..., 0). \tag{S43}$$

The corresponding metrological performance is described by

$$g(\varrho_p, \mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}(\mathcal{H}_{\mathrm{worst}})) = \frac{4p^2 d}{pd^2 + 2(1-p)}. \tag{S44}$$

No other Hamiltonian $\mathcal{H}$ corresponds to a worst performance.

Note that we considered collective Hamiltonians of the type (S25). Other collective Hamiltonians $\mathcal{H}_{\mathrm{coll}}$ can lead to a worse performace and can even have $g(\varrho_p, \mathcal{H}_{\rm coll}) = 0$. In particular, this is the case for Hamiltonians given in Eq. (S24), where $\mathcal{K}$ can be any Hamiltonian.

The metrological gain given in Eq. (S44) is maximal for $p = 1$ and has the value

$$g(\varrho_p, \mathcal{H}_{\rm coll}^{\rm (iso)}(\mathcal{H}_{\rm worst})) = \frac{4}{d}. \tag{S45}$$

If $d \geq 4$, then the right-hand side of Eq. (S45) is not larger than one. Hence, with $\mathcal{H}_{\rm worst}$, no isotropic state can be useful for $d \geq 4$. For $d = 3$, on the other hand the right-hand side of Eq. (S45) is larger than one. Hence, for $d = 3$, the maximally entangled state $|\Psi^{\rm (me)}\rangle$ is useful with the Hamiltonian $\mathcal{H}_{\rm worst}$. We can also see that for $d = 3$ the maximally entangled state $|\Psi^{\rm (me)}\rangle$ is useful with any Hamiltonian $\mathcal{H}_{\rm coll}^{\rm (iso)}$.

In Fig. S1, we plot the results of simple numerics for $d = 3, 4$ and 5. The random mixed states have been generated according to Ref. [28].

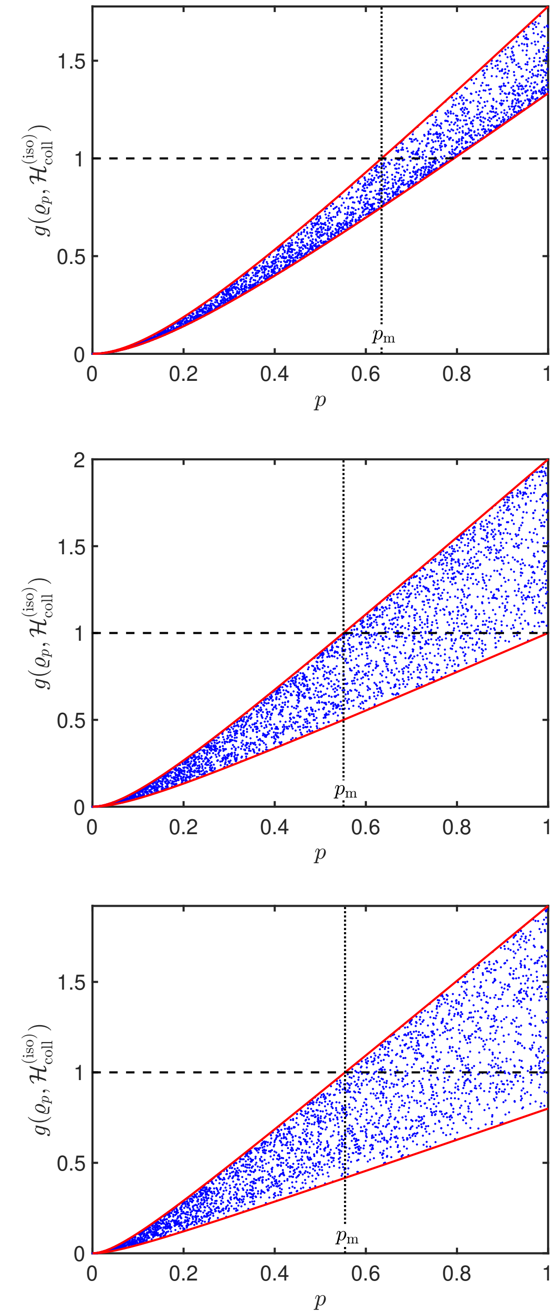

FIG. S1. Metrology with isotropic states given in Eq. (S21) for systems of size (top) 3×3, (middle) 4×4, and (bottom) 5×5. The metrological gain $g(\varrho_p, \mathcal{H}_{\rm coll}^{\rm (iso)})$ is plotted for isotropic states, (S21), of a given p. (dashed) Limit for separable states. (blue dots) Metrological performance of isotropic states for two-body Hamiltonians $\mathcal{H}_{\rm coll}^{\rm (iso)}(\mathcal{H})$ given in Eq. (S25), where $\mathcal{H}$ are chosen randomly. (upper solid red line) Metrology with the best Hamiltonian $\mathcal{H}_{\rm best}$ given in Eq. (S38). (lower solid red line) Metrology with the worst Hamiltonian $\mathcal{H}_{\rm worst}$ given in Eq. (S43). (dotted) Line corresponding the bound $p_{\rm m}$ given in Eq. (S42). Isotropic states with a larger p are useful for metrology.

METROLOGY WITH WERNER STATES

We now examine whether another type of bipartite states with a rotational symmetry, i.e, Werner states defined as [44]

$$\varrho_{\rm W}(\phi) = \frac{\mathbb{1} + \phi V}{d^2 + \phi d}, \tag{S46}$$

outperform separable states in metrology. Here $-1 \leq \phi \leq +1$ and V is the flip operator.

We will consider a general evolution of the type Eq. (S22). Werner states are invariant under transformations of the type

$$U \otimes U, \tag{S47}$$

where U is a single-qudit unitary. Hence, Werner states are invariant under the Hamiltonian

$$\mathcal{H}_{\rm inv}^{\rm (W)}(\mathcal{H}) = \mathcal{J} \otimes \mathbb{1} + \mathbb{1} \otimes \mathcal{J}, \tag{S48}$$

where $\mathcal{J}$ is a Hermitian operator.

Observation S6.—For short times, the action of the Hamiltonian $\mathcal{H}_{\rm coll}$ given in Eq. (S22) is the same as the action of

$$\mathcal{H}_{\rm coll}^{\rm (W)}(\mathcal{H}) = \mathcal{H}^{\rm (W)} \otimes \mathbb{1} - \mathbb{1} \otimes \mathcal{H}^{\rm (W)}, \tag{S49}$$

where the single party Hamiltonian $\mathcal{H}$ is defined as

$$\mathcal{H}^{\rm (W)} = (\mathcal{H}_1 + \mathcal{H}_2)/2. \tag{S50}$$

Proof. Let us define $\Delta^{\rm (W)}$ as

$$\Delta^{\rm (W)} = (\mathcal{H}_2 - \mathcal{H}_1)/2. \tag{S51}$$

In the rest of the section, we omit the superscript "W" in $\mathcal{H}_{\rm inv}^{(\rm W)}, \mathcal{H}^{(\rm W)}, \Delta^{(\rm W)}$. Then, simple algebra shows that

$$\mathcal{H}_{\rm coll} + \mathcal{H}_{\rm inv}^{(\rm W)}\left(\Delta^{(\rm W)}\right) = \mathcal{H}_{\rm coll}^{(\rm W)}. \tag{S52}$$

Hence, for small t

$$e^{-i\mathcal{H}_{\rm coll}t}e^{-i\mathcal{H}_{\rm inv}(\Delta)t} \approx e^{-i\mathcal{H}_{\rm coll}^{(\rm W)}(\mathcal{H})t} \tag{S53}$$

holds. The Werner state is invariant under the action of $\mathcal{H}_{\rm inv}^{(\rm W)}(\Delta)$, since the corresponding unitary is of the form given in Eq. (S47). Hence, the action of $\mathcal{H}_{\rm coll}$ is the same as the action of $H_{\rm coll}^{(\rm W)}(\mathcal{H})$ for small t. ■.

Observation S7.—Replacing the evolution by $\mathcal{H}_{\rm coll}$ given in Eq. (S22) by the evolution by $\mathcal{H}_{\rm coll}^{(\rm W)}$ given in Eq. (S24) does not decrease the metrological gain. Hence, when looking for the Hamiltonian with the largest metrological gain, it is sufficient to look for Hamiltonians of the form (S24).

Proof. The proof is similar to the proof of Observation S2. ■

Werner states, given in Eq. (S46), can also be defined as

$$\varrho_{\rm W}(\phi) = \frac{1+\phi}{d^2+\phi d}P_{\rm s} + \frac{1-\phi}{d^2+\phi d}P_a, \tag{S54}$$

where $P_{\rm s}$ and P_a are the projectors to the symmetric and antisymmetric subspace, respectively. We will be interested in the case $\phi \le 0$. The quantum Fisher information for Werner states for a Hermitian operator A is

$$F_Q[\varrho_{\rm W}, A] = 2\frac{(\lambda_{\rm s}-\lambda_{\rm as})^2}{\lambda_{\rm s}+\lambda_{\rm as}} \times \left(\sum_{k\in\mathcal{S}, l\in\mathcal{A}} |\langle k|A|l\rangle|^2 + \sum_{k\in\mathcal{A}, l\in\mathcal{S}} |\langle k|A|l\rangle|^2\right), \tag{S55}$$

where $k \in \mathcal{S}$ and $l \in \mathcal{A}$ denote the indices of symmetric and antisymmetric eigenstates, respectively. From Eq. (S54), the eigenvalues of the Werner states can be obtained, yielding

$$2\frac{(\lambda_{\rm s}-\lambda_{\rm as})^2}{\lambda_{\rm s}+\lambda_{\rm as}} = \frac{4|\phi|^2}{d^2+\phi d}. \tag{S56}$$

If the operator A is of the form given in Eq. (S24), then for any symmetric states $|\Psi_{\rm s}\rangle$ and antisymmetric states $|\Psi_a\rangle$

$$\langle\Psi_{\rm s}|A|\Psi_{\rm s}\rangle = \langle\Psi_a|A|\Psi_a\rangle = 0 \tag{S57}$$

hold. Hence, we can return to sums over all eigenvectors and write

$$F_Q[\varrho_{\rm W}, \mathcal{H}_{\rm coll}^{(\rm W)}] = \frac{4|\phi|^2}{d^2+\phi d}\sum_{k,l}|\langle k|\mathcal{H}_{\rm coll}^{(\rm W)}|l\rangle|^2 = \frac{8|\phi|^2}{d^2+\phi d}{\rm Tr}((H_{\rm coll}^{(\rm W)})^2). \tag{S58}$$

Then, we need that

$${\rm Tr}((\mathcal{H}_{\rm coll}^{(\rm W)})^2) = 2[d{\rm Tr}(\mathcal{H}^2) - {\rm Tr}(\mathcal{H})^2]. \tag{S59}$$

Hence, we obtain a general formula for the quantum Fisher information for Werner states as

$$F_Q[\varrho_{\rm W}, \mathcal{H}_{\rm coll}^{(\rm W)}(\mathcal{H})] = \frac{8|\phi|^2}{d^2+\phi d}[d{\rm Tr}(\mathcal{H}^2) - {\rm Tr}(\mathcal{H})^2]. \tag{S60}$$

Based on Eq. (S60) and on Eq. (24), the metrological performance is given by

$$g(\varrho_{\rm W}, \mathcal{H}_{\rm coll}^{(\rm W)}(\mathcal{H})) = \frac{8|\phi|^2}{d^2+\phi d}r(\mathcal{H}), \tag{S61}$$

where $r(\mathcal{H})$ is defined in Eq. (S37).

Let us now look for the Hamiltonian that provides the largest metrological gain for Werner states.

Observation S8.—Werner states have the best metrological performance with respect to separable states with the Hamiltonian $\mathcal{H}_{\rm best}$ given in Eq. (S38). The corresponding quantum Fisher information is

$$g(\varrho_{\rm W}, \mathcal{H}_{\rm coll}^{(\rm W)}(\mathcal{H}_{\rm best})) = \frac{|\phi|^2(d^2-\alpha)}{d^2+\phi d}, \tag{S62}$$

where the optimization is carried out over collective Hamiltonians of the form (S24).

No other such collective Hamiltonian corresponds to a better performance. Equation (S62) is maximal for $\phi = -1$ and has the value

$$g(\varrho_{\rm W}, \mathcal{H}_{\rm coll}^{(\rm W)}(\mathcal{H}_{\rm best})) = \frac{d+\alpha}{d+\alpha-1}, \tag{S63}$$

which is close to 1 for large d.

Proof. The best $\mathcal{H}$ operator is the one for which $r(\mathcal{H})$ defined in Eq. (S37) is the largest. In other words, we can look for the $\mathcal{H}$ for a constant $(h_{\max}-h_{\min})^2$ that maximizes $[d{\rm Tr}(\mathcal{H}^2) - {\rm Tr}(\mathcal{H})^2]$. The details of the proof are similar to the proof of Observation S3. ■

Next, we determine which Werner states are useful metrologically.

Observation S9.—If

$$\phi < \phi_{\rm m} := \frac{d}{2(d^2-\alpha)} - \sqrt{\frac{d^2}{4(d^2-\alpha)^2} + \frac{d^2}{d^2-\alpha}} \tag{S64}$$

holds, then the Werner state is useful for metrology with the Hamiltonian (S38). Otherwise, the Werner state is not useful with any other Hamiltonian.

Proof. We look for the ϕ for which the right-hand side of Eq. (S62) is 1. ■

Let us now look for the Hamiltonian of the type (S24) with which the Werner states have the worst metrological performance.

Note that for large d the parameter $\phi_{\rm m}$ converges to 1, while Werner states are entangled if $\phi < -1/d$ [44].

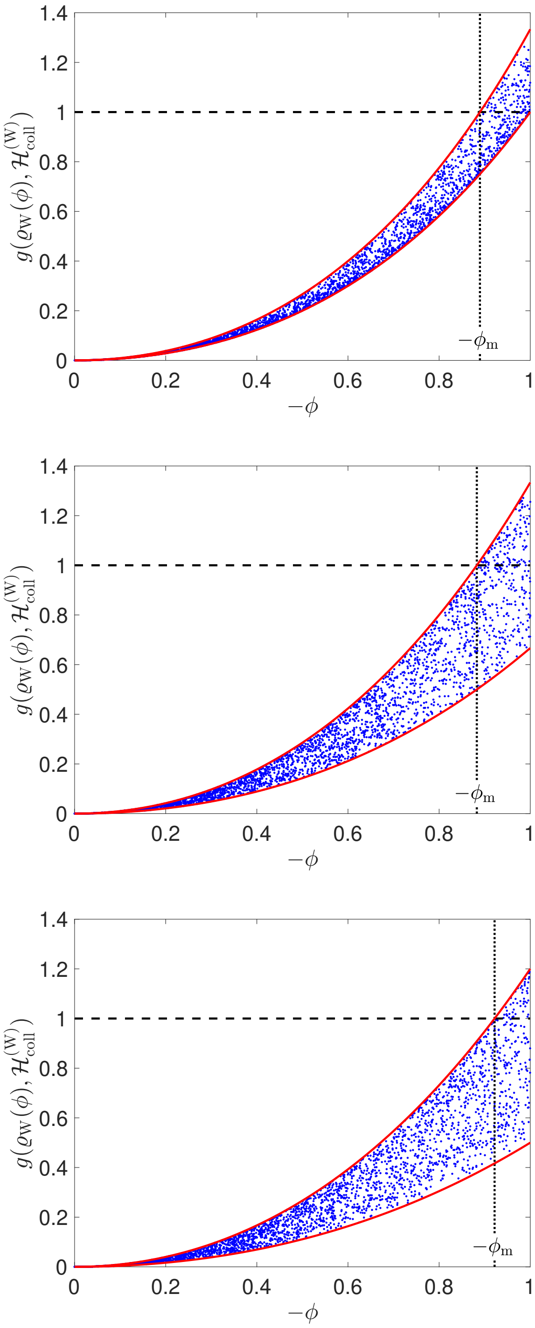

FIG. S2. Metrology with Werner states given in Eq. (S46). (top) 3×3, (middle) 4×4, and (bottom) 5×5 Werner states are considered. The metrological gain $g(\varrho_{\rm W}(\phi),\mathcal{H}_{\rm coll}^{\rm (W)})$ is plotted for Werner states of a given ϕ. (dashed) Limit for separable states. (blue dots) Metrological performance of Werner states for two-body Hamiltonians $\mathcal{H}_{\rm coll}^{\rm (W)}(\mathcal{H})$ given in Eq. (S24), where $\mathcal{H}$ are chosen randomly. (upper solid red line) Metrology with the best Hamiltonian $\mathcal{H}_{\rm best}$ given in Eq. (S38). (lower solid red line) Metrology with the worst Hamiltonian $\mathcal{H}_{\rm worst}$ given in Eq. (S43). (dotted) Line corresponding the bound $\phi_{\rm m}$ given in Eq. (S64). Werner states with $-\phi>-\phi_{\rm m}$ are useful for metrology.

Hence, there are Werner states that are entangled but not useful for metrology.

Observation S10.—Werner states have the worst metrological performance with respect to separable states with the Hamiltonian given in Eq. (S43). The corresponding metrological gain is

$$g(\varrho_{\rm W},\mathcal{H}_{\rm coll}^{\rm (W)}(\mathcal{H}_{\rm worst}))=\frac{2|\phi|^2d}{d^2+\phi d}. \tag{S65}$$

No other Hamiltonian corresponds to a worst performance.

Proof. This can be seen noting that Eq. (S61) is minimized for this case. ∎

Note that we considered Hamiltonians $\mathcal{H}_{\rm coll}^{\rm (W)}(\mathcal{H})$ of the type (S24). Other collective Hamiltonians $\mathcal{H}_{\rm coll}$ can lead to a worse performace and can even reach to $g(\varrho_{\rm W},\mathcal{H}_{\rm coll})=0$. In particular, this is the case for collective Hamiltonian of the form given in Eq. (S24).

Equation (S65) is maximal for $\phi=-1$ and has the value

$$g(\varrho_{\rm W},\mathcal{H}_{\rm coll}^{\rm (W)}(\mathcal{H}_{\rm worst}))=\frac{2}{d-1}. \tag{S66}$$

We can see that for $d\geq3$ the right-hand side of Eq. (S66) is not larger than one, hence the Werner state is not useful with the Hamiltonian $\mathcal{H}_{\rm worst}$. We can also see that the metrological gain, (S66), is close to 0 for large d.

In Fig. S2, we plot the results of simple numerics for $d=3,4$ and 5. The random mixed states have been generated according to Ref. [28].

CONCRETE EXAMPLE WITH TWO-QUBIT SINGLETS

In this Section, we work out in detail the problem of metrology with two-qubit singlets and ancillas. This problem is also interesting, since the Hamiltonians obtained numerically are very simple.

Let us consider the noisy two-qubit singlet

$$\varrho_{AB}^{(p)}=(1-p)|\Psi^-\rangle\langle\Psi^-|+p\mathbb{1}/4, \tag{S67}$$

where

$$|\Psi^-\rangle=\frac{1}{\sqrt{2}}(|01\rangle-|10\rangle). \tag{S68}$$

The state given in Eq. (S67) is a Werner-state given in Eq. (S46) and it is also equivalent to an isotropic state, (S21), under local unitaries. The state is more useful than separable states if the noise is smaller than

$$p_{\rm limit}=\tfrac{1}{8}(7-\sqrt{17})\approx0.3596, \tag{S69}$$

see Eq. (S42) for isotropic states. The optimal local Hamiltonian is

$$\mathcal{H}_{1\ {\rm singlet}}=Z_a-Z_B, \tag{S70}$$

where Z is the Pauli spin matrix diag$(-1,+1)$. Even for a pure singlet, this is the optimal Hamiltonian.

Let us consider two singlets with a bipartition $AA'|BB'$

$$\varrho_{2\text{ singlets}} = \varrho_{AB}^{(p)} \otimes \varrho_{A'B'}^{(p)}. \tag{S71}$$

Then, the optimal Hamiltonian is

$$\mathcal{H}_{2\text{ singlets}} = Z_a Z_{A'} + Z_B Z_{B'}. \tag{S72}$$

Finally, let us consider a singlet in AB and two ancillas in some pure state in $A'B'$

$$\varrho_{AB}^{(p)} \otimes |\Psi_{A'}\rangle\langle\Psi_{A'}| \otimes |\Psi_{B'}\rangle\langle\Psi_{B'}|. \tag{S73}$$

In this case, if $p < p_{\text{limit}}$ then the optimal Hamiltonian is Eq. (S70). That is, the ancillas do not give any advantage, the Hamiltonian does not act on the ancillas. If the singlet is too noisy, that is, $p > p_{\text{limit}}$ then the optimal local Hamiltonian is of the form

$$\mathcal{H}_{A'} + \mathcal{H}_{B'}. \tag{S74}$$

Note that Eq. (S74) acts only on the ancillas.

If we use pure singlets then in all these cases we have $\mathcal{F}_Q = 16$, while the limit for separable states is $\mathcal{F}_Q^{(\text{sep})} = 8$. If we use singlets with p given in Eq. (S69), then

$$\mathcal{F}_Q[\varrho_{2\text{ singlets}}, \mathcal{H}_{2\text{ singlets}}] = 8.1530. \tag{S75}$$

Thus, the state outperfoms separable states. In the case of a single copy, and a single copy with two pure ancillas, $\mathcal{F}_Q = \mathcal{F}_Q^{(\text{sep})} = 8$. On the other hand, the state $\varrho_{2\text{ singlets}}$ remains more useful than separable states if

$$p < 0.3675, \tag{S76}$$

where the limit on the noise fraction has been obtained numerically.

Thus, in the 2×2 case, a singlet mixed with white noise cannot be activated by ancillas. This is also true for isotropic states, since they are locally equivalent to a singlet mixed with white noise.

Finally, we show that if a singlet is mixed with non-white noise, then it can be activated with ancillas. Let us consider the state

$$\frac{1}{2}\left(|\Psi^-\rangle\langle\Psi^-| + |00\rangle\langle 00|\right). \tag{S77}$$

For this state, the optimization over Hamiltonians lead to $\mathcal{F}_Q = 8$, which is also the bound for separable states, i.e., $\mathcal{F}_Q^{(\text{sep})} = 8$. With two ancillas we can reach $\mathcal{F}_Q = 9$. With two copies of the state Eq. (S77), we can reach $\mathcal{F}_Q = 10$. In all these cases, we could use $c_1 = c_2 = 1$ when searching for the optimal Hamiltonian due to the symmetries of the setup. [See Eq. (20) for the definition of c_k.] The state given in Eq. (S77) can be activated even with a single ancilla. By setting $c_1 = c_2 = 1$, we get $\mathcal{F}_Q = 8.4$. On the other hand, the optimal Hamiltonian has $c_1 = 1$ and $c_2 = (1+\sqrt{5})/2 \approx 1.618$ and and the gain reaches $\mathcal{F}_Q/\mathcal{F}_Q^{(\text{sep})} = 3(5+\sqrt{5})/20 \approx 1.0854$.

We considered various multiqubit states in this section. In an application, we have to choose one of them. The basic idea is the following. If the metrological gain of an entangled quantum state is not larger than 1, i.e., $g \leq 1$, then it is better to use product states since they can reach the same precision, but it is easier to create them. Moreover, if we find that an entangled state is more useful than separable states, i.e., $g > 1$, then our algorithm can also tell us the optimal Hamiltonian corresponding to the task where they outperform separable states the most.

ESTIMATION OF THE METROLOGICAL GAIN FOR GENERAL QUANTUM STATES

Recently, there have been several methods presented to find lower bounds on the quantum Fisher information based on few operator expectation values [2, 27]. Our results on isotropic states and Werner states can be used to construct lower bounds for the metrological gain g based on a single operator expectation value.

In order to proceed, we note that any $d \times d$ state can be depolarized into an isotropic state given in Eq. (S21) with the $U \otimes U^*$ twirling operation as

$$\varrho_{\text{iso}}(F) = \int \mathcal{M}(dU)(U \otimes U^*)\varrho(U^\dagger \otimes U^{*\dagger}), \tag{S78}$$

where $\mathcal{M}$ is a unitarily invariant probability measure. The state $\varrho_{\text{iso}}(F)$ is just the isotropic state given in Eq. (S21), defined with a different parametrization as

$$\varrho_{\text{iso}}(F) = F|\Psi^{(\text{me})}\rangle\langle\Psi^{(\text{me})}| + (1-F)\frac{\mathbb{1} - |\Psi^{(\text{me})}\rangle\langle\Psi^{(\text{me})}|}{d^2-1}, \tag{S79}$$

where the maximally entangled state $|\Psi^{(\text{me})}\rangle$ is given in Eq. (7), and

$$F = \text{Tr}(\varrho|\Psi^{(\text{me})}\rangle\langle\Psi^{(\text{me})}|) \tag{S80}$$

is the entanglement fraction of the state ϱ, which is alternatively called the singlet fraction [42, 43]. Based on Eq. (S39), the maximum metrological performance of the isotropic state is given by

$$g(\varrho_{\text{iso}}(F)) = \frac{2(d^2-\alpha)(d^2F-1)^2}{d^2(d^2-1)(1-2F+d^2F)}, \tag{S81}$$

where α is zero for even d, and one otherwise. Here, we remember that the metrological gain is defined in Eq. (6).

Next, we show that $g(\varrho)$ cannot increase under twirling defined in Eq. (S78), i.e.,

$$g(\varrho) \geq g(\varrho_{\text{iso}}(F)). \tag{S82}$$

We use a series of inequalities

$$\begin{aligned}\mathcal{F}_Q[\rho_p,\mathcal{H}] &= \mathcal{F}_Q\left[\int \mathcal{M}(dU)(U\otimes U^*)\varrho(U^\dagger\otimes U^{*\dagger}),\mathcal{H}\right]\\ &\le \int M(dU)\mathcal{F}_Q[(U\otimes U^*)\varrho(U^\dagger\otimes U^{*\dagger}),\mathcal{H}]\\ &\le \mathcal{F}_Q[(U_0\otimes U_0^*)\varrho(U_0^\dagger\otimes U_0^{*\dagger}),\mathcal{H}]\\ &= \mathcal{F}_Q[\varrho,\mathcal{H}'],\end{aligned} \tag{S83}$$

where $\mathcal{H}'=(U_0^\dagger\otimes U_0^{*\dagger})\mathcal{H}(U_0\otimes U_0^*)$ and U_0 is some unitary. To arrive at the second line we used the property of the quantum Fisher information that it is convex in the state, Noting also that the eigenvalues of $\mathcal{H}'$ are the same as that of $\mathcal{H}$, and that $\mathcal{F}_Q^{(\rm sep)}(\mathcal{H})$ in Eq. (24) depends only on the eigenvalues, we arrive at Eq. (S82).

Based on Eq. (S82), the metrological gain of any quantum state can be bounded from below as

$$g(\varrho)\ge g(\varrho_{\rm iso}(F)), \tag{S84}$$

where $g(\varrho_{\rm iso}(F))$ is defined in Eq. (S81) and F is just the entanglement fraction of ϱ. Based on Eq. (S80), F equals the expectation value of the projector to $|\Psi^{(\rm me)}\rangle$. Hence, our lower bound is based on a single operator expectation value.

Similar calculations can be carried out for Werner states, using the fact that any quantum state can be depolarized into a Werner state using the $U\otimes U$ twirling

$$\varrho_{\rm W}(\phi)=\int \mathcal{M}(dU)(U\otimes U)\varrho(U^\dagger\otimes U^\dagger). \tag{S85}$$

Then, we can construct a lower bound

$$g(\varrho)\ge g(\varrho_{\rm W}(\phi)), \tag{S86}$$

where the Eq. (S62) gives the right-hand side of Eq. (S86) as a function of the parameter ϕ. The quantity ϕ is related to the expectation value of the flip operator V as

$$\langle V\rangle=\frac{1+d\phi}{d+\phi}. \tag{S87}$$

UNITING QUDITS

In most of the paper, we considered bipartite examples. In the multipartite case, the usefulness of a quantum state is always relative to the partitioning of the parties. From this point of view, it is worth to look at metrological usefulness of a multipartite state when we put the parties into two groups, and return to the bipartite problem. For instance, the four-qubit ring cluster state is not useful, $\mathcal{F}_Q/\mathcal{F}_Q^{(\rm sep)}=1$ [13]. After uniting two qubits into a ququart it becomes useful, with $\mathcal{F}_Q/\mathcal{F}_Q^{(\rm sep)}=2$. An optimal Hamiltonian with an optimal gain is

$$j_z^{(1)}\otimes j_y^{(2)}+j_y^{(3)}\otimes j_z^{(4)}. \tag{S88}$$

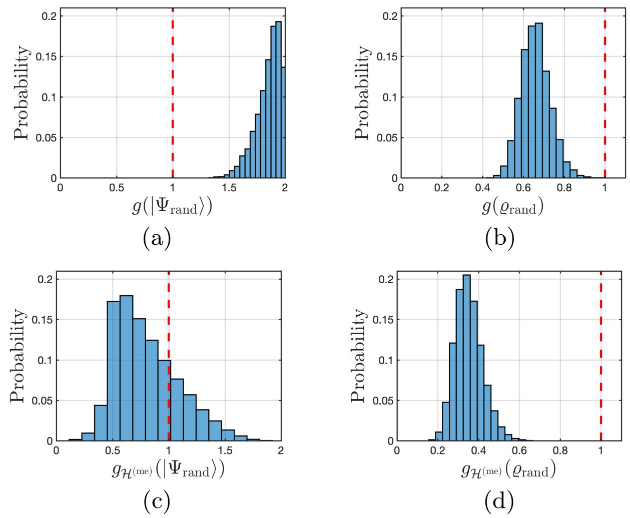

FIG. S3. Distribution of the metrological gain optimized over local Hamiltonians. Results for random states with dimension 3×3 for (a) pure and (b) mixed states. (c) and (d) The same for the Hamiltonian given in Eq. (8). (dashed vertical line) Line corresponding to $g=1$. States are metrologically useful if $g>1$.

We have to measure $M=j_z^{(1)}\otimes j_x^{(2)}\otimes j_x^{(3)}\otimes j_z^{(4)}$ for an optimal estimation precision $(\Delta\theta)^2_M=1/16$. Due to the commutator relations $[j_z^{(n)},M]=[j_z^{(n)},\mathcal{H}]=0$ for $n=1,4$, we can realize the following scheme. We measure j_z on qubits (1) and (4) such that we have a state locally equivalent to a singlet on qubits (2) and (3). Then, we do metrology with qubits (2) and (3). Similar schemes based on preselection have appeared in the theory of entanglement and nonlocality [29, 30].

HOW LARGE PART OF QUANTUM STATES ARE USEFUL

The scaling of the quantum Fisher information with the dimension has been considered for random states and for the best local Hamiltonian in Ref. [31]. We used our optimization algorithm to determine the distribution of the quantum Fisher information and obtain exactly how large part of pure or mixed quantum states are useful. The random pure states and mixed states have been generated according to Ref. [28]. For $d=3$, the results are shown in Fig. S3. It suggests that almost no random mixed states are useful. Pure states are useful almost with a maximal usefulness.

INFINITE NUMBER OF COPIES OF ARBITRARY PURE STATES

It is shown that an infinite number of copies of any entangled pure quantum state of Schmidt rank-s with $s>1$

is maximally useful metrologically. To this end, let us define a pure state in the Schmidt basis with Schmidt rank-s as in Eq. (26). Here, the real non-negative σ_k Schmidt coefficients are in a descending order, and $\sum_{k=1}^{s}\sigma_k^2=1$. In addition, we also assume that $\sigma_1>\sigma_2$.

Then, the n-copy state has the Schmidt coefficients

$$\sigma_{i_1}\sigma_{i_2}\cdots\sigma_{i_n}, \tag{S89}$$

where $i_k\in\{1,2,\ldots,s\}$. The number of equal Schmidt coefficients in the n-copy state follows a multinomial distribution formula. With this and Eq. (30), we obtain the lower bound

$$\mathcal{F}_Q[|\psi\rangle^{\otimes n},\mathcal{H}_{n-\text{copy}}]\geq 8\sum_{k_1+k_2+\ldots+k_s=n}^{n}\left\lfloor\frac{1}{2}\binom{n}{k_1,k_2,\ldots,k_s}\right\rfloor(2\sigma_1^{k_1}\sigma_2^{k_2}\cdots\sigma_s^{k_s})^2, \tag{S90}$$

where

$$\mathcal{H}_{n-\text{copy}}=\left(\bigotimes_{k=1}^{n}\mathcal{H}_{\text{A},k}\right)\otimes\left(\bigotimes_{k=1}^{n}\mathbb{1}_{\text{B},k}\right)+\left(\bigotimes_{k=1}^{n}\mathbb{1}_{\text{A},k}\right)\otimes\left(\bigotimes_{k=1}^{n}\mathcal{H}_{\text{B},k}\right). \tag{S91}$$

Here $\mathcal{H}_{\text{A},k}=\mathcal{H}_{\text{B},k}$ are all equal to the operator given in in Eq. (27). $\mathcal{H}_{\text{A},k}$ and $\mathcal{H}_{\text{B},k}$ act on the kth copy of system, on subsystem A and B, respectively. The meaning of $\mathbb{1}_{\text{A},k}$ and $\mathbb{1}_{\text{B},k}$ is analogous. The expression $\lfloor x\rfloor$ is the floor or integer part of x, and the multinomial coefficients are

$$\binom{n}{k_1,k_2,\ldots,k_s}=\frac{n!}{k_1!k_2!\cdots k_s!}. \tag{S92}$$

Using the multinomial theorem for $\left(\sum_k\sigma_k^2\right)^n=1$ and the relation

$$\left\lfloor\frac{1}{2}\binom{n}{k}\right\rfloor\geq\frac{\binom{n}{k}-1}{2}, \tag{S93}$$

yield a further lower bound

$$\begin{aligned}&\mathcal{F}_Q[|\psi\rangle^{\otimes n},\mathcal{H}_{n-\text{copy}}]\\&\geq 16\sum_{k_1+k_2+\ldots+k_s=n}\left[\binom{n}{k_1,k_2,\ldots,k_s}-1\right]\sigma_1^{2k_1}\sigma_2^{2k_2}\cdots\sigma_s^{2k_s}\\&=16-16\sum_{k_1+k_2+\ldots+k_s=n}\sigma_1^{2k_1}\sigma_2^{2k_2}\cdots\sigma_s^{2k_s}.\end{aligned} \tag{S94}$$

Now we show that for Schmidt rank $s>1$ and in the limit of large n the last sum tends to zero, hence in case of many copies n we get $\mathcal{F}_Q[|\psi\rangle^{\otimes n},\mathcal{H}_{n-\text{copy}}]\to 16$. To this end we set $k_1=n-k$ in the last sum above to get the following series of relations:

$$\begin{aligned}&\sum_{k_1+k_2+\ldots+k_s=n}\sigma_1^{2k_1}\sigma_2^{2k_2}\cdots\sigma_s^{2k_s}\\&=\sum_{k=0}^{n}\left(\sum_{k_2+\ldots+k_s=k}\sigma_1^{2(n-k)}\sigma_2^{2k_2}\cdots\sigma_s^{2k_s}\right)\\&=\sigma_1^{2n}\sum_{k=0}^{n}\left(\sum_{k_2+\ldots+k_s=k}\sigma_1^{-2k}\sigma_2^{2k_2}\cdots\sigma_s^{2k_s}\right)\\&\leq\sigma_1^{2n}\sum_{k=0}^{n}\left(\frac{\sigma_2}{\sigma_1}\right)^{2k}\sum_{k_2+\ldots+k_s=k}1,\end{aligned} \tag{S95}$$

where the inequality above is due to our assumption $\sigma_2\geq\sigma_k$, in the case of $k>2$. Let us now observe that this last upper bound goes to zero in the case of fixed s and n goes to infinity. This comes from the facts that in that case σ_1^{2n} goes to zero, and that $\sum_{k_2+\ldots+k_s=k}1$ is a polynomial function of s, hence owing to the Cauchy ratio test the series

$$\sum_{k=0}^{n}\left(\frac{\sigma_2}{\sigma_1}\right)^{2k}\sum_{k_2+\ldots+k_s=k}1 \tag{S96}$$

converges absolutely. ■

MAXIMAL METROLOGICAL GAIN

In this section, we consider the multiparticle case. For this case, the metrological gain can be define analogously to the bipartite case. We determine the quantum states with a maximum metrological gain.

Let us consider the high-dimensional Greenberger-Horne-Zeilinger (GHZ) state [32, 33]

$$|\text{GHZ}\rangle=\frac{1}{\sqrt{m}}\sum_{n=1}^{m}|n\rangle^{\otimes N}, \tag{S97}$$

where N is the number of particles, d is the dimension of their state space, and $m\leq d$ is the number of the terms in the superposition. We require that m is even. Then, the achievable largest metrological gain

$$g(|\text{GHZ}\rangle)=N \tag{S98}$$

is obtained for the state (S97). Thus, the maximal gain does not increase with the particle dimension d and depends only on the number of particles. In particular, for two particles, the maximal gain is 2.

An optimal Hamiltonian with which the maximal gain can be achieved with the GHZ state given in Eq. (S97) is of the form

$$\mathcal{H}_{\text{opt}}=\sum_{n=1}^{N}\mathbb{1}^{\otimes(n-1)}\otimes D'\otimes\mathbb{1}^{\otimes(N-n-1)}, \tag{S99}$$

where $\mathbb{1}^{\otimes 0}=1$, and the single particle Hamiltonian is defined as

$$D' = \sum_{n=1,3,5,...,m-1} |n\rangle\langle n| - |n+1\rangle\langle n+1|. \quad \text{(S100)}$$

Note that for even d and for $m=d$, the matrix D' equals the matrix D defined in Eq. (9).

In summary, for a given N and d, several of the GHZ states and Hamiltonians $\mathcal{H}_{\rm opt}$ give the maximum metrological gain compared to separable states. Note, however, that this does not mean that $\mathcal{F}_Q[|{\rm GHZ}\rangle, \mathcal{H}_{\rm opt}]$ is maximal in all these cases for a given N and d. It just means that $\mathcal{F}_Q[|{\rm GHZ}\rangle, \mathcal{H}_{\rm opt}]$ is the largest possible compared to what is achievable by separable states with the same Hamiltonian $\mathcal{H}_{\rm opt}$.

ALTERNATIVE OPTIMIZATION METHOD

We present a simple alternative of the two-step iterative optimization method of the paper. We use the following finding proved in the main text. If we determine the optimal $\mathcal{H}$ for a given M using Observation 2, the eigenvalues of the optimal $\mathcal{H}_n$ satisfying Eq. (20) are $\pm c_n$. We assume that $\mathcal{H}_n$ is of the form (21). We set $\tilde{D}_n = c_n {\rm diag}(+1,+1,...,+1,-1,-1,...,-1)$ and then vary U_n in order to get the maximal $\mathcal{F}_Q(\varrho, \mathcal{H}_1\otimes \mathbb{1} + \mathbb{1}\otimes\mathcal{H}_2)$.

ROBUSTNESS OF METROLOGICAL USEFULNESS

We define the quantum metrological robustness, $p_{\rm m}(\varrho, \varrho_{\rm noise})$, where ϱ is some quantum state and $\varrho_{\rm noise}$ a state representing noise. We call $p_{\rm m}$ is the largest noise fraction p for which the noisy state

$$\varrho_p = p\varrho_{\rm noise} + (1-p)\varrho \quad \text{(S101)}$$

have $g(\varrho_p)\ge 1$ [12]. The bound in Eq. (11) for noisy maximally entangled states can be formulated for $d=3$ as

$$p_{\rm m}(\Psi^{(\rm me)}, \mathbb{1}/d^2) = \frac{25-\sqrt{177}}{32} \approx 0.3655. \quad \text{(S102)}$$

In practice, the noise state can be the white noise and $\varrho_{\rm noise} \propto \mathbb{1}$. We can also consider an optimization

$$\min_{\varrho_{\rm noise}\in S_{\rm noise}} p_{\rm m}(\varrho, \varrho_{\rm noise}), \quad \text{(S103)}$$

which gives the noise tolerance against certain types of noise defined by the set $S_{\rm noise}$. For instance, the $S_{\rm noise}$ can contain all states that are metrologically note useful, i.e., for which $g\le 1$.

We can choose another type or parametrization usual in entanglement theory. Given a state ϱ and a metrologically useless state $\varrho_{\rm noise}$, we can call metrological robustness of ϱ relative to $\varrho_{\rm noise}$, the minimal $s\ge 0$ for which

$$R_{\rm m}(\varrho||\varrho_{\rm noise}) = \frac{1}{1+s}(\varrho + s\varrho_{\rm noise}) \quad \text{(S104)}$$

is useless for metrology.

The robustness can be obtained with a numerical search for the noise fraction for which $g=1$. We used a search based on interval halving. That is, we start with an interval given by two noise fractions values $p_{\rm L}$ and $p_{\rm H}$ such that $g(\varrho_{p_{\rm L}}) \le 1 \le g(\varrho_{p_{\rm H}})$. We test the noise fraction corresponding to the center of the interval. Depending on whether for that noise value $g\ge 1$ or $g<1$, we reset the lower or the upper boundary of the interval to the center. We repeat this procedure until the size of the interval is sufficiently small. We used a similar procedure to obtain the noise bounds for states with an extra ancilla and two copies of noise states.

We note that there are general relations between the gain-like and robustness-like quantities, that might be used in our case [22, 34].

WITNESSING DIMENSION

We can use our approach to witness the dimension of the quantum system [46–49], or in general, the type of the interaction that is present. For instance, we can consider the two-qubit singlet state mixed with $p=0.3596$ white noise, see Eq. (S69). Such a state is not more useful than separable states, under any Hamiltonian. Thus,

$$\max_{{\rm local}\mathcal{H}} \mathcal{F}_Q[\varrho,\mathcal{H}] \le \mathcal{F}_Q^{(\rm sep)}. \quad \text{(S105)}$$

If we find that the quantum state is more useful than separable states then it must be connected to an ancilla or a second copy or activated by another quantum state.

Next, we show how to obtain the bound for product states by measurement. We have to create random pure product states ϱ. Then, we can use that [18–21]

$$\mathcal{F}_Q[\varrho,\mathcal{H}] = 1 - F(\varrho,\varrho_t)t^2/2 + O(t^3), \quad \text{(S106)}$$

where $O(t^3)$ respresents terms that are at least third order in t, $F(\varrho,\varrho_t)$ is the fidelity between the initial state ϱ and the evolved state is

$$\varrho_t = e^{-i\mathcal{H}t}\varrho e^{+i\mathcal{H}t}. \quad \text{(S107)}$$

Thus, for a short time evolution, i.e., for small t we have

$$\mathcal{F}_Q[\varrho,\mathcal{H}] \approx 1 - F(\varrho,\varrho_t)t^2/2. \quad \text{(S108)}$$

Since both of these states are pure product states and we know ϱ, we can measure the fidelity, and use it to measure $\mathcal{F}_Q$. We can even look for the product state that maximizes $\mathcal{F}_Q[\varrho,\mathcal{H}]$ by some search algorithm.

We can also test whether the metrological performance is consistent with some particular interaction. We can compute the maximum for Hamiltonians of the form

$$\mathcal{H}_a \mathcal{H}_A + \mathcal{H}_B. \quad \text{(S109)}$$

If the metrological performance is better than this maximum, then the form must be different, i.e., there might be two interaction terms between subsystem A and the ancilla "a".

$$\mathcal{H}_a \mathcal{H}_A + \mathcal{H}'_a \mathcal{H}'_A + \mathcal{H}_B. \quad \text{(S110)}$$

Using ideas similar to the ones in our paper, with our method we can even look for the maximum for such Hamiltonians. If the metrological performance is better than this maximum, the interaction between A and a must contain at least three terms.