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

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

where a Hamiltonian for the aAB system is given as

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

Here, $\mathcal{H}_{\rm opt}$ is the 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

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

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

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

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

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

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

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

Here, $\sigma_{\mathrm{A'B'}}$ 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 $\sigma_{\mathrm{A'B'}}$ in Eq. (S5), we arrive at

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

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

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

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 serious of inequalities

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

where $0 \leq p \leq 1$. Here, $\mathcal{H}_{\mathrm{opt}}$ 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, $g_{\mathcal{H}}(\varrho) \leq g(\varrho)$ 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]

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

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

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

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

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

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

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

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

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),

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

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 $\sqrt{(\Delta\theta)^2_M/\mathcal{F}_{\mathrm{Q}}^{(\mathrm{sep})}}$, 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 $\mathcal{F}_{\mathrm{Q}}^{(\mathrm{sep})}$ 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), $\mathcal{F}_{\mathrm{Q}}^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}) = f(\mathcal{H}_1) + f(\mathcal{H}_2)$ holds. On the other hand, we have $\mathcal{F}_{\mathrm{Q}}^{(\mathrm{sep})}(\mathcal{H}_{\mathrm{coll}}^{(\mathrm{iso})}) = 2f(\mathcal{H})$. Knowing that f is matrix convex, we obtain that

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

■

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]

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

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

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

where we used that for the reduced state of $|\Psi^{(\rm me)}\rangle$ we have $\rho_{\rm red1} = \rho_{\rm red2} = \mathbb{1}/d$. Next, we use the fact that

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

holds. Hence, for the quantum Fisher information we obtain

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

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

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

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

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

Based on Eq. (3), the corresponding quantum Fisher information is

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

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

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

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

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

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 $p_{\rm m} > 1/2$ 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

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

The corresponding quantum Fisher information is

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

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

Note that we considered collective Hamiltonians of the type (S25). Other collective Hamiltonians $\mathcal{H}_{\rm coll}$ 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].

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 $-1\le\phi\le+1$ 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. ■.

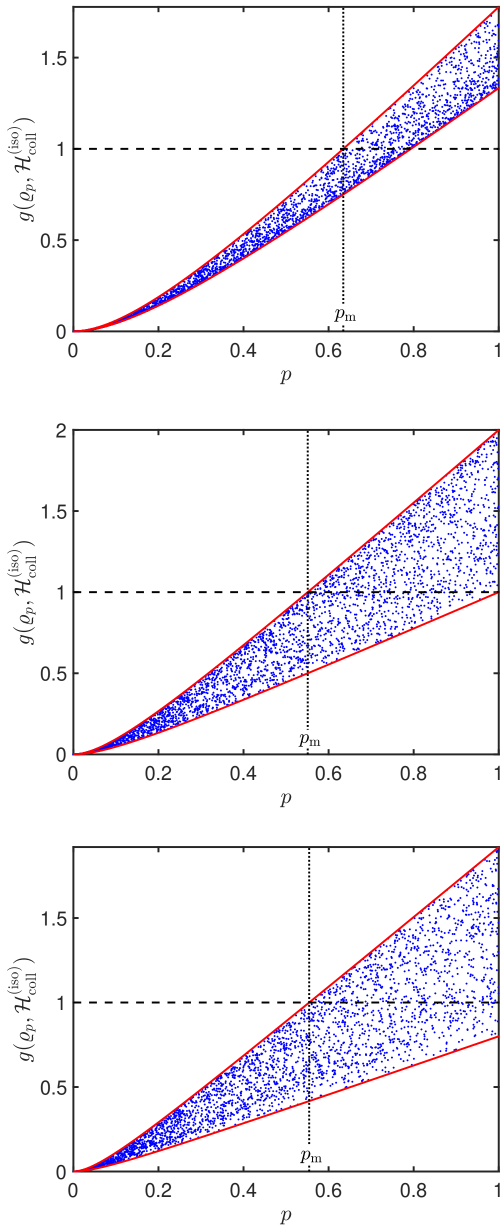

FIG. S1. Metrology with isotropic states given in Eq. (S21) for systems of size (top) 3×3, (middle) 4×4, and (bottom) 5×5. The metrological gain $g(\varrho_p,\mathcal{H}_{\rm coll}^{(\rm iso)})$ is plotted for isotropic states, (S21), of a given p. (dashed) Limit for separable states. (blue dots) Metrological 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.

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

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

where $P_{\rm s}$ and $P_{\rm a}$ 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 $|\Psi_{\rm a}\rangle$

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

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 $(h_{\rm max}-h_{\rm min})^2$ 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]. 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 quantum Fisher information is

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

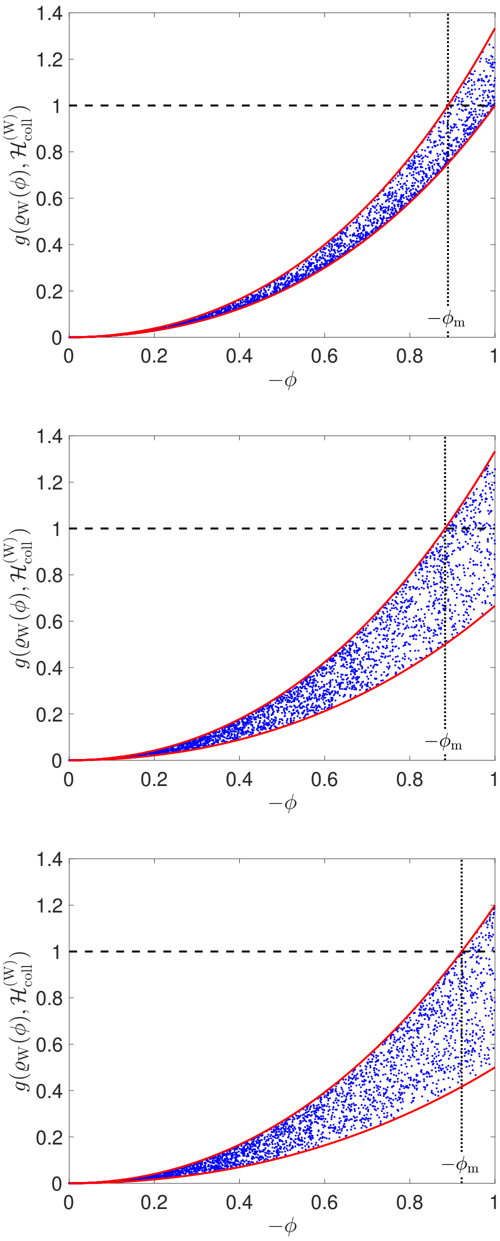

FIG. S2. Metrology with Werner states given in Eq. (S46). (top) 3×3, (middle) 4×4, and (bottom) 5×5 Werner states are considered. The metrological gain $g(\varrho_W(\phi), \mathcal{H}_{coll}^{(W)})$ is plotted for Werner states of a given ϕ. (dashed) Limit for separable states. (blue dots) Metrological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

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 $\mathcal{H}_{coll}^{(W)}(\mathcal{H})$ of the type (S24). Other collective Hamiltonians $\mathcal{H}_{coll}$ can lead to a worse performace and can even reach to $g(\varrho_W, \mathcal{H}_{coll}) = 0$. 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

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

We can see that for $d \geq 3$ the right-hand side of Eq. (S66) is not larger than one, hence the Werner state is not useful with the Hamiltonian $\mathcal{H}_{worst}$. 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

$$p_{limit} = \tfrac{1}{8}(7 - \sqrt{17}) \approx 0.3596, \tag{S69}$$

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

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

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'

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

Then, the optimal Hamiltonian is

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

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

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

In this case, if $p < p_{\rm limit}$ 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, $p > p_{\rm limit}$ then the optimal local Hamiltonian is of the form

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

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

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

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

Thus, the state outperfoms separable states. In the case of a single copy, and a single copy with two pure ancillas, $\mathcal{F}_{\rm Q} = \mathcal{F}_{\rm Q}^{\rm (sep)} = 8$. On the other hand, the state $\varrho_{2\ \rm singlets}$ 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 $\mathcal{F}_{\rm Q} = 8$, which is also the bound for separable states, i.e., $\mathcal{F}_{\rm Q}^{\rm (sep)} = 8$. With two ancillas we can reach $\mathcal{F}_{\rm Q} = 9$. With two singlets, we can reach $\mathcal{F}_{\rm Q} = 10$. 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 $\mathcal{F}_{\rm Q} = 8.4$. 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 $\mathcal{F}_{\rm Q}/\mathcal{F}_{\rm Q}^{\rm (sep)} = 3(5+\sqrt{5})/20 \approx 1.0854$.

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., $g \le 1$, 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

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

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

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

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

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

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

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

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.,

$$g(\varrho) \ge g(\varrho_{\rm iso}(F)). \tag{S82}$$

We use a series of inequalities

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

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 $\mathcal{F}_Q^{(\text{sep})}(\mathcal{H})$ 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

$$g(\varrho) \geq g(\varrho_{\text{iso}}(F)), \tag{S84}$$

where $g(\varrho_{\text{iso}}(F))$ 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 $|\Psi^{(\text{me})}\rangle$. 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

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

Then, we can construct a lower bound

$$g(\varrho) \geq g(\varrho_W(\phi)), \tag{S86}$$

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, $\mathcal{F}_Q/\mathcal{F}_Q^{(\text{sep})} = 1$ [12]. After uniting two qubits into a ququart it becomes useful, with $\mathcal{F}_Q/\mathcal{F}_Q^{(\text{sep})} = 2$. An optimal Hamiltonian with an optimal gain is

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

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 $(\Delta\theta)_M^2 = 1/16$. 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].

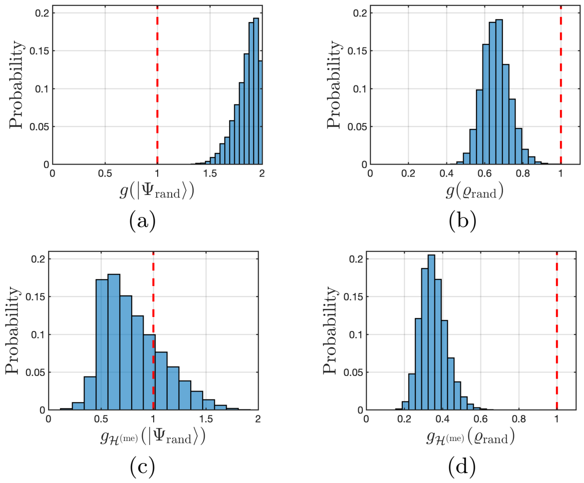

FIG. S3. Distribution of the metrological gain optimized over local Hamiltonians. Results for random states with dimension 3×3 for (a) pure and (b) mixed states. (c) and (d) The same for the Hamiltonian given in Eq. (8). (dashed vertical line) Line corresponding to $g = 1$. States are metrologically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

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

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

where

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

Here $\mathcal{H}_{{\rm A},k}=\mathcal{H}_{{\rm B},k}$ are all equal to the operator given in in Eq. (27). $\mathcal{H}_{{\rm A},k}$ and $\mathcal{H}_{{\rm B},k}$ act on the kth copy of system, on subsystem A and B, respectively. The meaning of $\mathbb{1}_{{\rm A},k}$ and $\mathbb{1}_{{\rm B},k}$ 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

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

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 $\mathcal{F}_{\rm Q}[|\psi\rangle^{\otimes n},\mathcal{H}_{n-{\rm copy}}]\to 16$. To this end we set $k_1=n-k$ in the last sum above to get the following series of relations:

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

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]

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

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

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

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

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

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

$$D'=\sum_{n=1,3,5,\ldots,m-1}|n\rangle\langle n|-|n+1\rangle\langle n+1|. \tag{S100}$$

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 $\mathcal{F}_{\rm Q}[|{\rm GHZ}\rangle,\mathcal{H}_{\rm opt}]$ is maximal in all these cases for a given N and d. It just means that $\mathcal{F}_{\rm Q}[|{\rm GHZ}\rangle,\mathcal{H}_{\rm opt}]$ 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 $\tilde{D}_n = c_n \mathrm{diag}(+1, +1, ..., +1, -1, -1, ..., -1)$ and then vary U_n in order to get the maximal $\mathcal{F}_{\mathrm{Q}}(\varrho, \mathcal{H}_1 \otimes \mathbb{1} + \mathbb{1} \otimes \mathcal{H}_2)$.

ROBUSTNESS OF METROLOGICAL USEFULNESS

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

$$\varrho_p = p\varrho_{\mathrm{noise}} + (1-p)\varrho \tag{S101}$$

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

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

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

$$\min_{\varrho_{\mathrm{noise}} \in S_{\mathrm{noise}}} p_{\mathrm{m}}(\varrho, \varrho_{\mathrm{noise}}), \tag{S103}$$

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

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

$$R_{\mathrm{m}}(\varrho || \varrho_{\mathrm{noise}}) = \frac{1}{1+s}(\varrho + s\varrho_{\mathrm{noise}}) \tag{S104}$$

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_{L} and p_{H} such that $g(\varrho_{p_{\mathrm{L}}}) \leq 1 \leq g(\varrho_{p_{\mathrm{H}}})$. We test the noise fraction corresponding to the center of the interval. Depending on whether for that noise value $g \geq 1$ 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,

$$\max_{\mathrm{local}\,\mathcal{H}} \mathcal{F}_{\mathrm{Q}}[\varrho, \mathcal{H}] \leq \mathcal{F}_{\mathrm{Q}}^{(\mathrm{sep})}. \tag{S105}$$

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]

$$\mathcal{F}_{\mathrm{Q}}[\varrho, \mathcal{H}] = 1 - F(\varrho, \varrho_t)t^2/2 + O(t^3), \tag{S106}$$

where $O(t^3)$ represents 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

$$\varrho_t = e^{-i\mathcal{H}t}\varrho e^{+i\mathcal{H}t}. \tag{S107}$$

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

$$\mathcal{F}_{\mathrm{Q}}[\varrho, \mathcal{H}] \approx 1 - F(\varrho, \varrho_t)t^2/2. \tag{S108}$$

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

$$\mathcal{H}_a \mathcal{H}_A + \mathcal{H}_B. \tag{S109}$$

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".

$$\mathcal{H}_a \mathcal{H}_A + \mathcal{H}_a' \mathcal{H}_A' + \mathcal{H}_B. \tag{S110}$$

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 intaction between A and a must contain at least three terms.