Maximal Quantum Fisher Information for Mixed States

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We study quantum metrology for unitary dynamics. Analytic solutions are given for both the optimal unitary state preparation starting from an arbitrary mixed state and the corresponding optimal measurement precision. This represents a rigorous generalization of known results for optimal initial states and upper bounds on measurement precision which can only be saturated if pure states are available. In particular, we provide a generalization to mixed states of an upper bound on measurement precision for time-dependent Hamiltonians that can be saturated with optimal Hamiltonian control. These results make precise and reveal the full potential of mixed states for quantum metrology.

The standard paradigm of quantum metrology involves the preparation of an initial state, a parameter-dependent dynamics, and a consecutive quantum measurement of the evolved state. From the measurement outcomes the parameter can be estimated [1–3]. Naturally, it is the goal to estimate the parameter as precisely as possible, i.e., to reduce the uncertainty \(\Delta \hat{\alpha} = \text{Var}(\hat{\alpha})^{1/2}\) of the estimator \(\hat{\alpha}\) of the parameter \(\alpha\) that we want to estimate. We consider single parameter estimation in the local regime where one already has a good estimate \(\hat{\alpha}\) at hand (typically from prior measurements) such that this prior knowledge can be used to prepare and control consecutive measurements. Quantum coherence and non-classical correlations in quantum sensors help to reduce the uncertainty \(\Delta \hat{\alpha}\) compared to what is possible with comparable classical resources [4, 5]. The ultimate precision limit for unbiased estimators is given by the quantum Cramér–Rao bound \(\Delta \hat{\alpha} \geq (M I_{\alpha})^{-1/2}\) which depends on the number of measurements \(M\) and the quantum Fisher information (QFI) \(I_{\alpha}\) which is a function of the state [6, 7]. When the number of measurements is fixed, as they correspond to a limited resource, precision is optimal and the QFI is maximal which involves an optimization with respect to the state.

In this Letter, we consider a freely available state \(\rho\), unitary freedom to prepare an initial state from \(\rho\), and unitary parameter-dependent dynamics of the quantum system (see Fig. 1). The parameter-dependent dynamics will be called sensor dynamics in the following in order to distinguish it from the state preparation dynamics. For instance, in a spin system the unitary freedom can be used to squeeze the spin before it is subjected to the sensor dynamics, as it is the case in many quantum-enhanced measurements [8–11]. In the worst case scenario, only the maximally mixed state is available, which does not change under unitary state preparation or unitary sensor dynamics and, thus, no information about the parameter can be gained. In the best-case scenario the available state is pure, when the maximal QFI as well as the optimal state to be prepared are well-known [12, 13].

The appeal and advantage of the theoretical study of unitary sensor dynamics lies in the analytic solutions that can be found that allow fundamental insights in the limits of quantum metrology and the role of resources such as measurement time and system size. The QFI maximized with respect to initial states, also known as channel QFI, can be reached only with pure initial states. If only mixed states are available, as it is usually the case under realistic conditions, this upper bound cannot be saturated and therefore has limited significance. In fact, if pure states are not available, the question for the maximal QFI and optimal state to be prepared is an important open problem [14, 15]. The main result of this Letter, theorem 1 below, is the complete solution of this problem.

The solution is relevant for practically all quantum sensors, as perfect initialization to a pure state can only be achieved to a certain degree that varies with the quantum system and the available technology. For example, nitrogen-vacancy (NV) center arrays [16, 17] or atomic-vapor magnetometers [18, 19] operate with mixed initial states due to imperfect polarization and competing depolarization effects [20, 21]. Particularly relevant is the example of sensors based on nuclear spin ensembles that typically operate with nuclear spins in thermal equilibrium, such that at room temperature the available state is strongly mixed [22]. Hence, the full potential of quantum metrology is exploited only when the mixedness of initial states is taken into account [14, 23–25].

We consider arbitrary, possibly time-dependent Hamiltonians \(H_{\alpha}(t)\) for the sensor dynamics. The corresponding unitary evolution operator is \(U_{\alpha} \equiv e^{-i/H_{\alpha} T} \int_{0}^{T} H_{\alpha}(t) \, dt\), where \(T\) denotes time-ordering, \(T\) is the total time of the sensor dynamics, and...
we set $\hbar = 1$ in the following. In the simplest case, dynamics is generated by a “phase-shift” or “precession” Hamiltonian proportional to the parameter $\alpha$, $H_\alpha = \alpha G$, with some parameter-independent operator $G$. The parameter dependence of the sensor dynamics is characterized by the generator $H_\alpha := iU_\alpha U_\alpha^\dagger \partial \alpha$, which simplifies to $G$ for phase-shift Hamiltonians [12, 26–28].

By introducing the eigenvalue decomposition of the prepared initial state $\rho = \sum_{k=1}^d p_k |\psi_k\rangle \langle \psi_k|$, where $d$ is the dimension of the Hilbert space, the QFI can be expressed as [7], [14]

$$I_\alpha(\rho) := 2 \sum_{k,\ell=1}^d p_k,\ell \langle \psi_k | h_\alpha | \psi_\ell \rangle^2,$$  

(1)

with coefficients

$$p_k,\ell := \begin{cases} 0 & \text{if } p_k = p_\ell = 0, \\ \frac{(p_k-p_\ell)^2}{p_k+ p_\ell} & \text{else}. \end{cases}$$  

(2)

Also, let $U(\alpha)$ denote the set of $d \times d$ unitary matrices.

**Theorem 1.** For any state $\rho$ and any generator $h_\alpha$ with ordered eigenvalues $p_1 \geq \cdots \geq p_d$ and $0 \leq h_1 \cdots \leq h_d$, respectively, the maximal QFI with respect to all unitary state preparations $U \rho U^\dagger$, $U \in U(d)$, is given by

$$I^*_\alpha := \max_U I_\alpha(U \rho U^\dagger) = \frac{1}{2} \sum_{k=1}^d p_{k,d-k+1} |h_k - h_{d-k+1}|^2.$$  

(3)

Let $h_k$ be the eigenvectors of the generator, $h_\alpha |h_k\rangle = h_k |h_k\rangle$. The maximum $I^*_\alpha$ is obtained by preparing the initial state

$$\rho^* = \sum_{k=1}^d p_k |\phi_k\rangle \langle \phi_k|,$$  

(4)

with [29]

$$|\phi_k\rangle = \begin{cases} \frac{|h_k\rangle + |h_{d-k+1}\rangle}{\sqrt{2}} & \text{if } 2k < d + 1, \\ |h_k\rangle & \text{if } 2k = d + 1, \\ \frac{|h_k\rangle - |h_{d-k+1}\rangle}{\sqrt{2}} & \text{if } 2k > d + 1. \end{cases}$$  

(5)

The proof is based on the Bloch–Waston inequality on the Hilbert–Schmidt norm of off-diagonal blocks of a Hermitian matrix [1, 2] and is given in the Supplemental Material [32]. The idea is to construct an upper bound for the QFI in Eq. (3) that exhibits a simpler dependence on the coefficients $p_{k,\ell}$. Then we maximize the upper bound by exploiting the Bloch–Waston inequality. The proof is concluded by showing that at its maximum the upper bound equals the QFI.

It is important to notice that the rank $r$ of the state $\rho$ plays a crucial role both for the maximal QFI and for the optimal state. In order to reach the maximal QFI $I^*_\alpha$, the choice of the $|\phi_k\rangle$ corresponding to vanishing $p_k$, i.e., for $k > r$, is irrelevant. This is best exemplified by considering the well-known case of pure states, characterized by $p_1 = 1$ and $r = 1$ [5, 12, 26, 27, 33]. Then, the maximal QFI in Eq. (3) simply becomes $(h_1 - h_2)^2$ and is obtained by preparing an equal superposition $|\psi_1\rangle + |\psi_2\rangle/\sqrt{2}$ of the eigenvectors corresponding to the minimal and maximal eigenvalues of $h_\alpha$. When the rank is increased but remains less than or equal to $(d+1)/2$, the optimal QFI is equal to $\sum_{i=1}^r p_i (h_i - h_{d-i+1})^2$. This can be seen as a convex sum of pure-state QFIs [35].

The situation changes when the rank is increased even further. For example with $r = 4$ and $d = 5$, the maximal QFI is equal to $p_1(h_1 - h_5)^2 + (2p_4-p_3)^2(h_2 - h_4)^2$. Further, for a Hilbert space of odd dimension, the vector $|\phi_{(d+1)/2}\rangle = |h_{(d+1)/2}\rangle$ is an eigenstate of the generator: it remains invariant under the dynamics and does not contribute to the QFI. For example for both $r = 2$ and $r = 3$ with $d = 5$, the optimal QFI is given by $p_1(h_1 - h_5)^2 + p_2(h_2 - h_4)^2$.

We obtained $I^*_\alpha$ by optimizing with respect to unitary state preparation while keeping the sensor dynamics fixed (see Fig. 1). However, in practice it is often possible not only to manipulate the available state but also the sensor dynamics by adding a parameter-independent control Hamiltonian $H_\alpha(t)$ to the original Hamiltonian $H_\alpha(t)$. While theorem 1 holds for any $H_\alpha(t)$, it is an interesting question to what extent the maximal QFI in Eq. (3) can be increased by adding a time-dependent control Hamiltonian. Again, the answer is only known for pure states [5]. The question, how this generalizes if the available state is mixed, brings us to

**Theorem 2.** For any state $\rho$ with ordered eigenvalues $p_1 \geq \cdots \geq p_d$ and any time-dependent Hamiltonian $H_\alpha(t)$, where $0 \leq \mu_1(t) \cdots \leq \mu_d(t)$ are the ordered eigenvalues of $\partial_\alpha H_\alpha(t) \equiv \partial H_\alpha(t)/\partial \alpha$, an upper bound for the QFI is given by

$$K_\alpha = \frac{1}{2} \sum_{k=1}^d p_{k,d-k+1} \left( \int_0^T [\mu_k(t) - \mu_{d-k+1}(t)] \, dt \right)^2.$$  

(6)

Let $|\mu_k(t)\rangle$ be the time-dependent eigenvectors of $\partial_\alpha H_\alpha(t), \partial_\alpha H_\alpha(t)|\mu_k(t)\rangle = \mu_k(t)|\mu_k(t)\rangle$. The upper bound $K_\alpha$ is reached by preparing the initial state

$$\rho^* = \sum_{k=1}^d p_k |\phi_k\rangle \langle \phi_k|,$$  

(7)

with

$$|\phi_k\rangle = \begin{cases} \frac{|\mu_k(0)| + |\mu_{d-k+1}(0)|}{\sqrt{2}} & \text{if } 2k < d + 1, \\ |\mu_k(0)| & \text{if } 2k = d + 1, \\ \frac{|\mu_k(0)| - |\mu_{d-k+1}(0)|}{\sqrt{2}} & \text{if } 2k > d + 1, \end{cases}$$  

(8)

and choosing the Hamiltonian control $H_\alpha(t)$ such that

$$U_\alpha(t)|\mu_k(0)\rangle = |\mu_k(t)\rangle \quad \forall k = 1, \ldots, d \quad \forall t,$$  

(9)

where

$$U_\alpha(t) = T \left[ \exp \left( -i \int_0^t [H_\alpha(r) + H_\alpha(\tau)] \, d\tau \right) \right].$$  

(10)
The proof (see Supplementary Material [32]) starts by rewriting \( h_α \) as in Ref. [5, Eq. 6] and shows that Eq. (6) is an upper bound for Eq. (3). We use the Schur convexity [4] of Eq. (3) and the inequalities from K. Fan [3, 37] for eigenvalues of the sum of two hermitian matrices.

One of the strengths of the bound \( K_α \) is that it is given by the eigenvalues of \( \partial_α H_α(t) \) and does not depend on the full unitary operator of the sensor dynamics which is hard to calculate for time-dependent Hamiltonians. The optimal initial state with Hamiltonian control in theorem 2 differs from the optimal initial state without Hamiltonian control in theorem 1 by the fact that the eigenvectors of the generator \( h_α \) in Eq. (5) are replaced by those of \( \partial_α H_α(0) \) in Eq. (8). The reason for this is that the optimal initial state of theorem 1 is the most sensitive state with respect to the sensor dynamics \( U_α \). However, if the Hamiltonian is time-dependent, the state which is most sensitive to the sensor dynamics at time \( t \) will also be time-dependent in general. Since the Hamiltonian control is allowed to be time-dependent, we can take this into account and ensure that the optimal initial state evolves such that it is most sensitive to the sensor dynamics for all times \( t \). This corresponds to the condition in Eq. (9). Only in special cases, such as phase-shift Hamiltonians \( H_α = α G \), we have \( h_α = \partial_α H_α \) and, thus, the optimal initial states of theorem 1 and 2 are the same. If they are not the same, a Hamiltonian \( H_α \) can be seen as suboptimal and requires correction by means of the Hamiltonian control in order to reach the upper bound of theorem 2.

Formally, the optimal control Hamiltonian from theorem 2 depends on the (unknown) parameter \( α \). Since we are in the local parameter estimation regime, we have knowledge (from prior measurements) about \( α \) such that \( α \) can be replaced by the estimate \( ˆα \). It was shown that replacing \( α \) by \( ˆα \) in the optimal control Hamiltonian does not ruin the benefits from introducing Hamiltonian control [5], and Hamiltonian control was applied experimentally with great success in Ref. [39]. For a more detailed discussion of control Hamiltonians we refer to the work of Pang et al. [5] [40].

As applications of our theorems we consider two examples: the estimation of a magnetic field amplitude and the estimation of the frequency of an oscillating magnetic field. Both cases can be described with the general Hamiltonian of a system of \( N \) spin-\( j \) particles subjected to a (time-dependent) magnetic field

\[
H(t) = \sum_{k=1}^{N} B f(t) S_z^{(k)} + H_1, \tag{11}
\]

with the magnetic field amplitude \( B \), some time-dependent real-valued modulation function \( f(t) \), and spin operator \( S_z^{(k)} \) in \( z \)-direction of the \( k \)-th spin. We use the standard angular momentum algebra, \( S_z^{(k)} | j, m \rangle = m | j, m \rangle \) with \( m = -j, \ldots, j \). \( H_1 \) is independent of \( B \) and takes into account possible interactions between spins. This rather general Hamiltonian can be seen as an idealization of quantum sensors based on arrays of NV centers [16, 17, 41], nuclear spin ensembles [42], or vapor of alkali atoms [19]. Due to imperfect polarization and competing depolarization effects [20, 21, 43, 44], the available states are mixed.

Here, we consider the available state of each of the \( N \) spins to be described by a spin-temperature distribution (independent of the Hamiltonian in Eq. (11))

\[
\rho_{θh} = \frac{e^{βS_z}}{Z}, \tag{12}
\]

with partition function \( Z = \sum_{m=-j}^{j} e^{βm} \), and inverse (effective) temperature \( β \). Eq. (12) was derived for optically polarized alkali vapors in [20, Eq. 112], and we assume that it is also a good approximation for the other spin-based magnetometers mentioned. \( β \) is related to the degree of polarization \( P \in [0, 1] \) by \( β = \ln \frac{1+P}{1-P} \); \( P = 1 \) corresponds to a perfectly polarized spin in \( z \)-direction, described by a pure state, and \( P = 0 \) corresponds to an unpolarized spin, i.e. a maximally mixed state. The available state of the total system is a tensor product of spin-temperature distributions, \( ρ = ρ_{θh}^N \).

For the estimation of the amplitude \( B \) we assume that the modulation \( f(t) \) is known (the case of unknown \( f(t) \) would correspond to waveform estimation [45, 46]). This is naturally the case for (quasi-)constant magnetic fields, periodic fields of known frequency, or, for example, when the modulation originates from a relative movement of sensor and environment (the source of \( B \)) that is tracked separately with another sensor. The maximal QFI obtained by using control Hamiltonians (cf. theorem 2) for estimating the amplitude \( B \) is found to be

\[
K_B = g^2(T) \sum_{k=-N_j}^{N_j} q(k) \frac{\sin^2(βk)}{Z^N \cosh(βk)} (2k)^2, \tag{13}
\]

where \( q(k) \) takes into account the degeneracy of eigenvalues of \( ρ \) and \( \partial_B H(t) = ≡ \partial H(t)/∂B \). It follows from the definition of the tensor product that the degeneracy of the \( k \)-th eigenvalue of both, \( ρ \) and \( \partial_B H(t) \), where eigenvalues are in weakly decreasing order, equals the number...
of possibilities \( q(k) \) of getting a sum \( k \) when rolling \( N \) fair dice, each having \( 2j+1 \) sides corresponding to values \( \{−j, \ldots, j\} \) (see Supplementary Material [32]) [6, p. 23-24]:

\[
q(k) = \sum_{\ell=0}^{N} (-1)^\ell \binom{N}{\ell} \left( k + N(j + 1) - 1 - \ell(2j + 1) \right) \frac{(N-1)}{2},
\]

where the binomial coefficient \( \binom{n}{k} \) is set to zero if one or both of its coefficients are negative. The dependence on measurement time \( T \) is given by \( q(T) = \int_{0}^{T} |f(t)| dt \).

The QFI in Eq. (13) exhibits a complicated dependence on the number of thermal states \( N \) and their spin size \( j \). However, by deriving a lower bound for Eq. (13), we prove that the QFI scales \( \propto N^2 \) for any \( j \) as well as \( \propto j^2 \) for any \( N \). In particular, we find \( K_B = 4N^2 \langle S_j \rangle^2 + O(N) \) where \( \langle S_j \rangle = \text{tr} [\rho_0 S_j] \), and \( O(N) \) denotes terms of order \( N \) and lower order. In the limit of large temperatures, \( \langle S_j \rangle^2 \) decays as \( \beta^2 \) (see Supplementary Material [32]).

This means that Heisenberg scaling [1, 48, 49], i.e., the quadratic scaling with the system size \( j \) or the number of particles \( N \), is obtained for the optimal unitary state preparation even if only thermal states are available. Note that this also holds in the context of theorem 1 if the generator equals \( S_z \). Importantly, Heisenberg scaling is found for any finite temperature of the thermal state; only in the limit of infinite temperature, the available state is fully mixed and the QFI vanishes.

In order to attain the QFI (13), the conditions (9) must be fulfilled. In particular the Hamiltonian control must cancel interactions between the spins, i.e., \( H_1 \) must be compensated. Also, every time the modulation function \( f(t) \) changes its sign, we must apply a transformation which interchanges the eigenstates corresponding to a (degenerate) eigenvalue \( e^{ik}/N \) of \( \rho \) with the eigenstates corresponding to the (degenerate) eigenvalue \( e^{-ik}/N \) for all \( k = 1, \ldots, Nj \). This is realized, for instance, with a local \( \pi \)-pulse about the \( x \)-axis, which interchanges \( |j, m⟩ \) for every single spin. The \( \pi \)-pulses ensure optimal phase accumulation of the optimal state given by Eq. (7) (cf. Fig. 2).

The degeneracy of eigenvalues of \( \rho \) and \( \partial_B H(t) \) leads to a freedom in preparing the optimal initial state. The special case of qubits, \( j = 1/2 \), constant magnetic field, \( f(t) = 1 \), and no interactions, \( H_1 = 0 \), was studied by Modi et al. [14]. In this case, no Hamiltonian control is required which brings us back to theorem 1. They conjectured that a unitary state preparation consisting of a mixture of GHZ states is optimal in their case and calculated the QFI. Theorem 1 confirms their conjecture.

If, instead of the amplitude, we want to estimate the frequency \( \omega \) of a periodic magnetic field with known amplitude \( B \), \( f(t) = \cos(\omega t) \), the eigenvalues of \( \partial H(t)/\partial \omega \) are modulated not with \( f(t) \) but with \( \partial f(t)/\partial \omega = -t \sin(\omega t) \), see Fig. 2. The maximal QFI \( K_\omega \) equals Eq. (13) with the only difference that \( q(T) \) is replaced by \( g_\omega(T) = \int_{0}^{T} B(t) \sin(\omega t) dt \approx BT^2/\pi \), corresponding to a \( T^2 \)-scaling of QFI, similar to what was reported in Ref. [5]. The optimal control is similar to the estimation of \( B \): interactions must be canceled and local \( \pi \)-pulses about the \( x \)-axis must be applied whenever \( \partial f(t)/\partial \omega \) crosses zero.

Theorem 1 also allows us to study the problem of optimal initial states of given purity \( \gamma = \text{tr} \rho^2 \). Fixing only \( \gamma \) amounts to an additional optimization over the spectrum of the initial state, which we solve numerically. As an example, we consider a two-qubit system with eigenvalues \( p_1 \geq \cdots \geq p_4 \), see Fig. 3. We observe that different levels of degeneracy of the spectrum of the generator results in distinct solutions for the optimal eigenvalues \( p_k \).

In conclusion, theorems 1 and 2 give an answer to the question of optimal unitary state preparation and optimal Hamiltonian control for an available mixed state and given unitary sensor dynamics that encodes the parameter to be measured in the quantum state. In comparison, distilling pure from mixed states at the cost of reducing
the number of available probes would be an alternative. However, probes are typically a valuable resource, that is utilized most efficiently along the lines of theorem 1 and 2. The two theorems allow one to study quantum metrology with mixed states with the same analytical rigor as for pure states, and the well-known results about optimal pure states are recovered as special cases. We find that Heisenberg scaling of the QFI can be reached with thermal states: initial mixedness is not as detrimental as Markovian decoherence during or after the sensor dynamics, which is known to generally destroy the Heisenberg scaling of the QFI [50–52].

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[32] See Supplemental Material at [URL will be inserted by publisher] for proofs of theorem 1 and theorem 2, as well as the proofs of Heisenberg scaling for thermal states.

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SUPPLEMENTAL MATERIAL

Proof of theorem 1

FIG. 4. Schematic sketch of the mechanism used to prove theorem 1 from the Letter. First an upper bound $J_{\alpha}(\rho)$ (red dash-dotted line) for the quantum Fisher information $I_{\alpha}(\rho)$ (black solid line) is constructed. This upper bound is shown to be maximal for $\rho = \rho^*$ (gray dashed line). Then, it is shown that $I_{\alpha}(\rho^*) = J_{\alpha}(\rho^*)$ from which it follows that $I_{\alpha}(\rho^*)$ must be the maximum of $I_{\alpha}(\rho)$.

The idea of the proof of theorem 1 from the Letter is the following, see also Fig. 4: We carefully construct an upper bound $J_{\alpha}(\rho) \geq I_{\alpha}(\rho)$ for the QFI $I_{\alpha}(\rho)$. Then, we show that (i) $J_{\alpha}(\rho)$ is maximized by setting $\rho = \rho^*$ and (ii) $J_{\alpha}(\rho^*) = I_{\alpha}(\rho^*)$. It follows that $I_{\alpha}(\rho^*)$ is the maximum of $I_{\alpha}(\rho)$.

We first give a technical lemma which introduces inequalities for the $p_{i,j}$ coefficients which are defined as

$$p_{i,j} := \begin{cases} 0 & \text{if } p_i = p_j = 0, \\ \frac{(p_i - p_j)^2}{p_i + p_j} & \text{else.} \end{cases} \tag{15}$$

These inequalities will be used to prove proposition 4 about the existence of coefficients $q_{i,j}$ which fulfill specific conditions. Proposition 4 enables us to find the desired upper bound $J_{\alpha}(\rho)$ for the QFI $I_{\alpha}(\rho)$. This is then used in the proof of theorem 5 which corresponds to theorem 1 from the Letter.

To facilitate the understanding of the following lemma and proposition, we introduce a schematic arrangement of a set of coefficients $p_{i,j}$, see Fig. 5. We consider only coefficients with $1 \leq i < j \leq d$ because of the symmetry $p_{i,j} = p_{j,i}$ and because $p_{i,i} = 0$.

FIG. 5. Scheme of $p_{i,j}$ for $1 \leq i < j \leq d = 7$. Coefficients inside the red squared boxes are denoted as central coefficients.
Lemma 3. Let \( p_1 \geq \ldots \geq p_d \geq 0 \). Then, the following inequalities hold:

\[
\begin{align*}
(\text{i}) & \quad p_{i,l} \geq p_{i,j} + p_{j,l} \quad \text{for } 1 \leq i < j < l \leq d, \\
(\text{ii}) & \quad p_{i,l} - p_{i+1,l} \geq p_{i,k} - p_{i+1,k} \quad \text{for } 1 < i + 1 < k < l \leq d, \\
(\text{iii}) & \quad p_{i,l} - p_{i,l-1} \geq p_{j,l} - p_{j,l-1} \quad \text{for } 1 \leq i < j < l - 1 < d.
\end{align*}
\]

Proof. First we prove that

\[
p_{i,l} + p_{j,k} \geq p_{i,k} + p_{j,l} \quad \text{for } 1 \leq i < j < k < l \leq d.
\]

If \( p_i \geq p_j = p_k = p_l = 0 \), inequality (16) holds trivially. Otherwise, we find

\[
p_{i,l} + p_{j,k} - p_{i,k} - p_{j,l} = \frac{4(p_i - p_j)(p_k - p_l)(p_ip_jp_k + p_jp_kp_l + p_i(p_j + p_k)p_l)}{(p_i + p_k)(p_j + p_k)(p_i + p_l)(p_j + p_l)},
\]

which is clearly nonnegative because all factors in the denominator are positive and all factors in the numerator are nonnegative. This proves inequality (16).

Inequalities (i), (ii), and (iii) from the lemma are special cases of inequality (16): If \( p_j = p_k \), inequality (16) holds also for \( j = k \) and it follows inequality (i). Further, from inequality (16) we find \( p_{i,l} - p_{j,l} \geq p_{i,k} - p_{j,k} \) which for \( j = i + 1 \) gives inequality (ii), and we find \( p_{i,l} - p_{i,k} \geq p_{j,l} - p_{j,k} \) which for \( k = l - 1 \) gives inequality (iii). \( \square \)

Proposition 4. For any dimension \( d \geq 2 \) and for any \( p_1 \geq \ldots \geq p_d \geq 0 \), there exist coefficients \( q_{k,k+1} \geq 0 \) with \( 1 \leq k \leq d - 1 \) such that for \( 1 \leq i < j \leq d \):

\[
\begin{align*}
q_{i,j} &= p_{i,j} \quad \text{if } j = d - i + 1 \quad (\text{central coefficients}), \\
q_{i,j} &\geq p_{i,j} \quad \text{else},
\end{align*}
\]

where coefficients \( p_{i,j} \) and \( q_{i,j} \) are defined in Eqs. (15) and (18), respectively.

Proof. The proof works by induction in dimension \( d \), once for even \( d \) and once for odd \( d \).

Even dimension \( d \)

Base case \( d = 2 \): There is only one coefficient \( p_{1,2} \) with \( 1 \leq i < j \leq 2 \), which is \( p_{1,2} \). The proposition for \( d = 2 \) holds because \( q_{1,2} = p_{1,2} \) fulfills conditions (19) and (20) trivially.

Inductive step: Suppose the proposition holds for \( d = n \). We will prove the proposition for \( d = n + 2 \).

First, the induction hypothesis is applied to \( n \) coefficients \( p_{2,2}, \ldots, p_{n+1} \): For any \( p_2 \geq \ldots \geq p_n \geq 0 \), there exist coefficients \( q_{k,k+1} \geq 0 \) for \( 2 \leq k \leq n \) such that for \( 2 \leq i < j \leq n + 1 \):

\[
\begin{align*}
q_{i,j} &= p_{i,j} \quad \text{if } j = n - i + 3 \quad (\text{central coefficients}), \\
q_{i,j} &\geq p_{i,j} \quad \text{else},
\end{align*}
\]

Second, we show that for any \( p_1 \) and \( p_{n+2} \) with \( p_1 \geq p_2 \) and \( p_{n+1} \geq p_{n+2} \geq 0 \) there exist two further coefficients \( q_{1,2} \) and \( q_{n+1,n+2} \) such that

\[
\begin{align*}
q_{1,n+2} &= p_{1,n+2} \quad (\text{central coefficients}), \\
q_{1,j} &\geq p_{1,j} \quad \text{for } j = 2, \ldots, n + 1 \quad (\text{left flank}), \\
q_{1,n+2} &\geq p_{1,n+2} \quad \text{for } i = 2, \ldots, n + 1 \quad (\text{right flank}).
\end{align*}
\]

A graphical visualization of the inductive step is shown in Fig. 6 which explains the terms left flank and right flank used to designate the inequalities above. The existence of \( q_{1,2} \) and \( q_{n+1,n+2} \) such that conditions (23),(24), and (25) hold is shown explicitly by setting

\[
\begin{align*}
q_{1,2} &:= p_{1,n+2} - p_{2,n+2}, \\
q_{n+1,n+2} &:= p_{2,n+2} - p_{2,n+1},
\end{align*}
\]
FIG. 6. Recursion steps from $d = 5$ to $d = 7$ (left) and from $d = 4$ to $d = 6$ (right). In the green squares are the two new elements we need to choose. In the blue (resp. red) rectangles are the new left (resp. right) flanks that need to fulfill conditions (20) ; in the magenta squares are the new central coefficients that need to fulfill condition (19).

and checking conditions (23),(24), and (25): We find

$$q_{1,n+2} = q_{1,2} + q_{2,n+1} + q_{n+1,n+2}$$

[Eq. (18)]

$$= p_{1,n+2} - p_{2,n+2} + p_{2,n+1} + p_{2,n+2} - p_{2,n+1}$$

[Eqs. (26),(27), and Eq. (21) for $i = 2$]

$$= p_{1,n+2}$$

which fulfills the condition for central coefficients [condition (23)]. Further, for $j = 3, \ldots, n+1$:

$$q_{1,j} = q_{1,2} + q_{2,j}$$

[Eq. (18)]

$$\geq p_{1,n+2} - p_{2,n+2} + p_{2,j}$$

[Eq. (26) and inequality (22)]

$$\geq p_{1,j} - p_{2,j} + p_{2,j}$$

[lemma 3 (ii)]

$$= p_{1,j}$$

and

$$q_{1,2} = p_{1,n+2} - p_{2,n+2}$$

[Eq. (26)]

$$\geq p_{1,2} + p_{2,n+2} - p_{2,n+2}$$

[lemma 3 (i)]

which fulfill the conditions for the left flank [condition (24)]. The proof for the right flank is similar: For $i = 2, \ldots, n$:

$$q_{i,n+2} = q_{i,n+1} + q_{n+1,n+2}$$

[Eq. (18)]

$$\geq p_{i,n+1} + p_{2,n+2} - p_{2,n+1}$$

[Eq. (27) and inequality (22)]

$$\geq p_{i,n+1} + p_{i,n+2} - p_{i,n+1}$$

[lemma 3 (iii)]

$$= p_{i,n+2},$$

and

$$q_{n+1,n+2} = p_{2,n+2} - p_{2,n+1}$$

[Eq. (27)]

$$\geq p_{2,n+1} + p_{n+1,n+2} - p_{2,n+1}$$

[lemma 3 (i)]

which fulfill the conditions for the right flank [condition (25)]. This proves the proposition for $d = n + 2$, concluding the proof by induction for even dimensions.

Odd dimension $d$

**Base case $d=3$:** There are only three coefficients $p_{i,j}$ with $1 \leq i < j \leq 3$, which are $p_{1,2}, p_{2,3}$, and $p_{1,3}$. The proposition for $d = 3$ holds because $q_{1,2} = p_{1,3} - p_{2,3}$, $q_{2,3} = p_{2,3}$, and $q_{1,3} = q_{1,2} + q_{2,3}$ fulfill the conditions (19) and (20): $q_{1,2} = p_{1,3} - p_{2,3} \geq p_{1,2} + p_{2,3} - p_{2,3} = p_{1,2}$ where inequality (i) from lemma 3 was used, while the other conditions hold trivially.
**Inductive step:** Analog to the inductive step for even $d$. \hfill $\Box$

Equipped with proposition 4 we can prove theorem 1 from the Letter:

**Theorem 5.** For any state $\rho$ and any generator $h_\alpha$ with ordered eigenvalues $p_1 \geq \cdots \geq p_d$ and $h_1 \geq \cdots \geq h_d$, respectively, the maximal QFI with respect to all unitary state preparations $U \rho U^\dagger$, $U \in U(d)$, is given by

$$I_\alpha^* := \max_U I_\alpha(U \rho U^\dagger) = \frac{1}{2} \sum_{k=1}^d p_k d_{-k+1}^2 (h_k - h_{d-k+1})^2.$$  \hfill (28)

Let $|h_k\rangle$ be the eigenvectors of the generator, $h_\alpha |h_k\rangle = h_k |h_k\rangle$. The maximum $I_\alpha^*$ is obtained by preparing the initial state

$$\rho^* := \sum_{k=1}^d p_k |\phi_k\rangle \langle \phi_k|$$  \hfill (29)

with

$$|\phi_k\rangle := \begin{cases} |h_k\rangle \sqrt{2} & \text{if } 2k < d + 1, \\ |h_k\rangle & \text{if } 2k = d + 1, \\ |h_k\rangle \sqrt{2} & \text{if } 2k > d + 1, \end{cases}$$  \hfill (30)

where $\chi_k$ are arbitrary real phases (the theorem as formulated in the Letter is recovered by setting $\chi_k = 0$).

**Proof.** First we reformulate the optimization problem in a more convenient way:

The unitary state preparation $U \rho U^\dagger$ has invariant eigenvalues for all $U \in U(d)$. However, the unitary freedom $U \in U(d)$ allows one to change the basis from the ordered orthonormal basis of eigenvectors $\{|\psi_i\rangle\}^d_{i=1}$ of $\rho$, where $\rho |\psi_i\rangle = p_i |\psi_i\rangle$, to any other ordered orthonormal basis. Therefore, the optimization problem with respect to unitary transformations $U \in U(d)$ on the state $\rho$ is equivalent to optimizing over ordered bases $B \in S$ where

$$S := \{(|\xi_i\rangle)^d_{i=1} : \langle \xi_i | \xi_j \rangle = \delta_{i,j} \ \forall i, j \in \{1, \ldots, d\}\}.$$  \hfill (31)

Note that the ordering of eigenvectors corresponds to the ordering of eigenvalues $p_i$ which plays a crucial role in the theorem. The basis corresponding to $\rho^*$ is given by $B^* = \{|\phi_i\rangle\}^d_{i=1}$, and the maximization in Eq. (28) is equivalent to

$$I_\alpha^* := \max_{B \in S} I_\alpha(B),$$  \hfill (32)

where the QFI was redefined as a function of $B$:

$$I_\alpha(B) = 2 \sum_{i,j=1}^d p_{i,j} \left| [h_\alpha(B)]_{i,j} \right|^2.$$  \hfill (33)

The coefficients $p_{i,j}$ are defined in Eq. (15) with respect to the eigenvalues $p_i$ and $[h_\alpha(B)]_{i,j} = \langle \xi_i | h_\alpha | \xi_j \rangle$ are the coefficients of $h_\alpha$ with respect to $B = (|\xi_i\rangle)^d_{i=1}$.

In order to prove that the maximum is reached by $B^*$, we introduce an upper bound for the QFI. We start by rewriting the QFI, exploiting the symmetries $p_{i,j} = p_{j,i}$ and $\left| [h_\alpha(B)]_{i,j} \right|^2 = \left| [h_\alpha(B)]_{j,i} \right|^2$:

$$I_\alpha(B) = 2 \sum_{i,j=1}^d p_{i,j} \left| [h_\alpha(B)]_{i,j} \right|^2$$  \hfill (34)

$$= 4 \sum_{i=1}^{d-1} \sum_{j=i+1}^d p_{i,j} \left| [h_\alpha(B)]_{i,j} \right|^2.$$  \hfill (35)

Then, an upper bound for $I_\alpha(B)$ is obtained by replacing coefficients $p_{i,j}$ in Eq. (35) with new coefficients $q_{i,j} \geq p_{i,j}$ for all $1 \leq i < j \leq d$:

$$I_\alpha(B) \leq J_\alpha(B) := 4 \sum_{i=1}^{d-1} \sum_{j=i+1}^d q_{i,j} \left| [h_\alpha(B)]_{i,j} \right|^2,$$  \hfill (36)
where $J_{\alpha}(B)$ denotes the upper bound. We choose coefficients $q_{i,j}$ according to proposition 4, i.e., besides $q_{i,j} \geq p_{i,j}$ they fulfill $p_{i,j} = q_{i,j}$ for $j = d - i + 1$ and $q_{i,j} = \sum_{k=1}^{j-1} q_{k,k+1}$ for all $1 \leq i < j \leq d$. We rewrite the upper bound $J_{\alpha}(B)$:

$$J_{\alpha}(B) = 4 \sum_{i=1}^{d-1} \sum_{j=i+1}^{d} q_{i,j} \left| [h_{\alpha}(B)]_{i,j} \right|^2$$

$$= 4 \sum_{i=1}^{d-1} \sum_{j=i+1}^{d} \sum_{k=1}^{j-1} q_{k,k+1} \left| [h_{\alpha}(B)]_{i,j} \right|^2$$

$$= 4 \sum_{k=1}^{d-1} q_{k,k+1} \sum_{i=1}^{d} \sum_{j=k+1}^{d} \left| [h_{\alpha}(B)]_{i,j} \right|^2$$

$$= 4 \sum_{k=1}^{d-1} q_{k,k+1} \left| [h_{\alpha}(B,k)]_{l} \right|^2,$$

where $h_{\alpha}(B,k)$ denotes the subblock of $h_{\alpha}(B)$ with coefficients from the 1st to the $k$th row and from the $(k+1)$th to the $d$th column, and $\| \cdot \|_2$ denotes the Hilbert–Schmidt norm which is defined for a $m \times n$ matrix $A$ as $\| A \|_2^2 = \text{tr}[A^* A] = \sum_{i,j=1}^{m,n} |A_{i,j}|^2$. Since $h_{\alpha}(B)$ is Hermitian it divides in subblocks as

$$h_{\alpha}(B) = \begin{pmatrix}
\cdot & h_{\alpha}(B,k) \\
h_{\alpha}^*(B,k) & \cdot
\end{pmatrix},$$

where the quadratic subblocks on the diagonal are not further specified.

Next, we maximize the upper bound $J_{\alpha}(B)$ and show that it equals the QFI at its maximum. In order to maximize $J_{\alpha}(B)$, we use the Bloomfield–Watson inequality [1] on the Hilbert–Schmidt norm of off-diagonal blocks such as $h_{\alpha}(B,k)$. We take a convenient formulation of the inequality from Ref.[2, Eqs. (1.14) and (4.3)] and apply it to $h_{\alpha}(B,k)$:

$$\| h_{\alpha}(B,k) \|_2^2 \leq \frac{1}{4} \sum_{i=1}^{m(k)} (h_i - h_{d-i+1})^2,$$

where $m(k) = \min(k,d-k)$. We evaluate the left-hand side of the Bloomfield–Watson inequality (42) for $B = B^*$, where $B^*$ is the eigenbasis of $\hat{\rho}^*$, defined above Eq. (32):

$$\| h_{\alpha}(B^*,k) \|_2^2 = \sum_{i=1}^{k} \sum_{j=k+1}^{d} \left| [h_{\alpha}(B^*)]_{i,j} \right|^2$$

$$= \sum_{i=1}^{k} \sum_{j=k+1}^{d} |\langle \phi_i | h_{\alpha} | \phi_j \rangle|^2$$

$$= \sum_{i=1}^{k} \sum_{j=k+1}^{d} \left( \delta_{i,j} + \delta_{i,j+1} + \delta_{i,j+1} |h_i - h_{d-i+1}|^2 \right)^2$$

$$= \frac{1}{4} \sum_{i=1}^{m(k)} (h_i - h_{d-i+1})^2,$$

where we used the definition of $|\phi_i\rangle$ [Eq. (30)] to get from Eq. (44) to (45). In Eq. (45), the first summand (within the brackets) evaluates always to zero while the second summand is nonzero in $m(k)$ cases as given in Eq. (46). Note, that Eq. (46) equals the right-hand side of inequality (42). Therefore, the Bloomfield–Watson inequality (42) is saturated for $B = B^*$ and, in particular, $\| h_{\alpha}(B,k) \|_2^2 \lesssim \| h_{\alpha}(B^*,k) \|_2^2$ for all $B \in S$.

This implies $J_{\alpha}(B) \lesssim J_{\alpha}(B^*)$ for all $B \in S$ which can be seen from Eq. (40) and by realizing that the coefficients $q_{i,j}$ in $J_{\alpha}(B)$ are nonnegative, which follows from the nonnegativity of $p_{i,j}$. Thus, $J_{\alpha}(B^*)$ is the maximum of $J_{\alpha}(B)$ with respect to $B$. 
Now, we show that $J_\alpha(B^*) = I_\alpha(B^*)$ starting from the definition of $J_\alpha(B)$ in Eq. (36):

\[ J_\alpha(B^*) = 4 \sum_{i=1}^{d-1} \sum_{j=i+1}^{d} q_{i,j} \left( \delta_{i,j} h_{i} + h_{d-i+1} + \delta_{i,d-j+1} \frac{h_{i} - h_{d-i+1}}{2} \right)^2 \]

\[ = \sum_{i=1}^{d-1} \sum_{j=i+1}^{d} q_{i,j} \delta_{i,d-j+1} (h_{i} - h_{d-i+1})^2 \]

\[ = \frac{1}{2} \sum_{i=1}^{d} p_{i,i+d+1} (h_{i} - h_{d-i+1})^2 = I_\alpha(B^*), \]

where we used $q_{i,i-d+1} = p_{i,i-d+1}$ and, to get from Eq. (48) to (49), we first came back to a summation over all $1 \leq i, j \leq d$ before evaluating $\delta_{i,d-j+1}$ which explains the factor $1/2$ in Eq. (49).

It follows from $J_\alpha(B) \succeq I_\alpha(B)$ for all $B \in S$ that $\max_{B \in S} J_\alpha(B) \geq \max_{B \in S} I_\alpha(B)$, and, then, it follows from $\max_{B \in S} J_\alpha(B) = I_\alpha(B^*)$ that $I_\alpha(B^*)$ is the maximum of $I_\alpha(B)$ with respect to $B \in S$.

**Proof of theorem 2**

Let us first introduce some notation. The real, nonnegative coordinate space of $d$ dimensions is denoted by $\mathbb{R}_+^d$. For two vectors $x, y \in \mathbb{R}_+^d$, the element-wise vector ordering $x_i \leq y_i$ for all $i \in \{1, \ldots, d\}$ is denoted as $x \leq y$. For any $x \in \mathbb{R}_+^d$, let $x_{[1]}, \ldots, x_{[d]}$ be the components of $x$ in decreasing order, and let

\[ x_\downarrow := (x_{[1]}, \ldots, x_{[d]}) \]

denote the decreasing rearrangement of $x$. Let

\[ \mathcal{D}_+^d := \{(x_1, \ldots, x_d) : x_1 \geq \cdots \geq x_d \geq 0\} \]

be the set of decreasing rearrangements of elements from $\mathbb{R}_+^d$.

**Definition 6.** For a hermitian matrix $X$ with eigenvalues $x_1 \geq x_2 \geq \cdots \geq x_d$ define

\[ d(X) := (x_1 - x_d, x_2 - x_{d-1}, \ldots, x_{[d/2]} - x_{d-[d/2]+1}) \]

where $[d/2]$ denotes the smallest integer $j$ with $j \geq d/2$.

Note that the entries of $d(X)$ are nonnegative and in decreasing order, i.e., $d(X) \in \mathcal{D}_+^{[d/2]}$.

**Definition 7.** Let $x, y \in \mathbb{R}_+^d$. We say that $x$ is weakly majorized by $y$, denoted by $x \prec_w y$, if

\[ \sum_{i=1}^{k} x_{[i]} \leq \sum_{i=1}^{k} y_{[i]} \quad \forall k = 1, \ldots, d. \]

**Lemma 8.** Let $A$, $B$, and $C = A + B$ be hermitian matrices with eigenvalues $a_1 \geq \cdots \geq a_d$, $b_1 \geq \cdots \geq b_d$, and $c_1 \geq \cdots \geq c_d$, respectively. Then, $d(C) \prec_w d(A) + d(B)$.

**Proof.** The inequalities of K. Fan (see for instance [3, eq.3]) for the eigenvalues of $A$, $B$, and $C = A + B$ are

\[ \sum_{i=1}^{r} c_i \leq \sum_{i=1}^{r} a_i + b_i \quad \forall r = 1, \ldots, d - 1. \]

Subtracting them from the trace condition

\[ \sum_{i=1}^{d} c_i = \sum_{i=1}^{d} a_i + b_i \]

and rearranging the indices gives

\[ \sum_{i=1}^{r} c_{d-i+1} \geq \sum_{i=1}^{r} a_{d-i+1} + b_{d-i+1} \quad \forall r = 1, \ldots, d - 1. \]
Subtracting inequality (56) from inequality (54) gives
\[\sum_{i=1}^{d} c_i - c_{d-i+1} \leq \sum_{i=1}^{d} a_i - a_{d-i+1} + b_i - b_{d-i} \quad \forall r = 1, \ldots, d-1,\] (57)
which are for \(r = 1, \ldots, [d/2]\) the weak majorization conditions for \(d(C) <_w d(A) + d(B)\).

**Definition 9.** For any \(p \in D^d_+\) define
\[\phi_p : \mathbb{R}^d_+ \rightarrow \mathbb{R}, \phi_p(x) := \sum_{i=1}^{d} p_i x_i^2.\] (58)

**Lemma 10.** For any \(p \in D^d_+\), \(\phi_p\) is increasing and Schur convex on \(\mathbb{R}^d_+\), i.e., the following conditions hold [4, part I,ch.3,A.4]:

(i) \(x \leq y \Rightarrow \phi_p(x) \leq \phi_p(y)\) (increasing),

(ii) \(\phi_p(x)\) is invariant under permutation of coefficients of \(x\) for any \(x \in \mathbb{R}^d_+\) (symmetric),

(iii) \((x_i - x_j) \left(\frac{\partial \phi_p(x_i)}{\partial x_i} - \frac{\partial \phi_p(x_j)}{\partial x_j}\right) \geq 0 \forall x \in \mathbb{R}^d_+\) and \(i \neq j\) (Schur’s condition).

**Proof.** From \(x \leq y\) it follows that \(x_i \leq y_i\ \forall i\), which implies \(p_i x_i^2 \leq p_i y_i^2\ \forall i\) for any \(p_i \geq 0\). Finally it follows \(\sum_i p_i x_i^2 \leq \sum_i p_i y_i^2\) which proves condition (i). Condition (ii) follows directly from the definition of \(\phi_p\). Finally, we have
\[(x_i - x_j) \left(\frac{\partial \phi_p(x_i)}{\partial x_i} - \frac{\partial \phi_p(x_j)}{\partial x_j}\right) = (x_i - x_j) 2(q x_i - r x_j),\] (59)
where \(q, r\) are some components of \(p\) with \(q \geq r\) if \(x_i \geq x_j\) and \(q \leq r\) if \(x_i \leq x_j\) due to the definition of \(\phi_p\). It follows condition (iii). \(\square\)

**Lemma 11.** Let \(A, B,\) and \(C = A + B\) be Hermitian matrices. For any \(p \in D^d_+\),
\[d(C) <_w d(A) + d(B) \Rightarrow \phi_p(d(C)) \leq \phi_p(d(A) + d(B)).\] (60)

**Proof.** The proof follows from a theorem given in Ref. [4, part I,ch.3,A.8] about weak majorization and lemma 10. \(\square\)

We are now ready to prove the following inequality:

**Lemma 12.** Let \(p \in D^d_+\), and let \(p_{i,j}\) be defined as in Eq. (15) for the components of \(p\). Let \(A, B,\) and \(C = A + B\) be Hermitian matrices with eigenvalues \(a_1 \geq \cdots \geq a_d, b_1 \geq \cdots \geq b_d,\) and \(c_1 \geq \cdots \geq c_d,\) respectively. Then,
\[\sum_{i=1}^{d} p_{i,d-i+1}(c_i - c_{d-i+1})^2 \leq \sum_{i=1}^{d} p_{i,d-i+1} (a_i - a_{d-i+1} + b_i - b_{d-i})^2.\] (61)

**Proof.** Let us first show that coefficients \(p_{i,d-i+1}\) satisfy
\[(p_{i,d-i+1})_{i=1}^{[d/2]} \in D^d_+ \] (62)
For \(1 \leq i < [d/2]\), where \([d/2]\) denotes the largest integer \(j\) with \(j \leq d/2\), we have
\[p_{i,d-i+1} \geq p_{i,i+1} + p_{i+1,d-i} \geq p_{i,i+1} + p_{i+1,d-i} + p_{d,i,d-i+1},\] (63)
where inequality (i) from lemma 3 was applied twice, and it follows \(p_{i,d-i+1} \geq p_{i+1,d-i}\). For even \(d\) it follows Eq. (62). For odd \(d\), we further have \(p_{d/2},d - [d/2] + 1 \geq p_{d/2},d + [d/2] + 1\) because \(p_{d/2},d - [d/2] + 1 = p_{d/2},d + [d/2] + 1 = 0\) by definition of \(p_{i,j}\). This proves Eq. (62).

Together with lemmata 8 and 11 it follows that
\[\sum_{i=1}^{[d/2]} p_{i,d-i+1}(c_i - c_{d-i+1})^2 \leq \sum_{i=1}^{[d/2]} p_{i,d-i+1} (a_i - a_{d-i+1} + b_i - b_{d-i+1})^2.\] (64)
which, due to the symmetries $p_{i,d-i+1} = p_{d-i+1,i}$ and $(c_i - c_j)^2 = (c_j - c_i)^2$, is equivalent to
\[ \sum_{i=d-[d/2]+1}^{d} p_{i,d-i+1}(c_i - c_{d-i+1})^2 \leq \sum_{i=d-[d/2]+1}^{d} p_{i,d-i+1}(a_i - a_{d-i+1} + b_i - b_{d-i+1})^2. \] (65)

Adding inequalities (64) and (65) proves the lemma since, in case of odd $d$, $p_{[d/2],[d/2]} = 0$. \hfill \square

We are now in the position to prove theorem 2 from the Letter:

**Theorem 13.** For any state $\rho$ with ordered eigenvalues $p_1 \geq \cdots \geq p_d$ and any time-dependent Hamiltonian $H_\alpha(t)$, where $\mu_1(t) \geq \cdots \geq \mu_d(t)$ are the ordered eigenvalues of $\partial_\alpha H_\alpha(t) := \partial H_\alpha(t)/\partial \alpha$, an upper bound for the QFI is given by
\[ K_{\alpha} = \frac{1}{2} \sum_{k=1}^{d} p_{k,d-k+1} \left( \int_{0}^{T} [\mu_k(t) - \mu_d(t)] dt \right)^2. \] (66)

Let $|\mu_k(t)\rangle$ be the time-dependent eigenvectors of $\partial_\alpha H_\alpha(t)$, $\partial_\alpha H_\alpha(t)|\mu_k(t)\rangle = \mu_k(t)|\mu_k(t)\rangle$. The upper bound $K_{\alpha}$ is reached by preparing the initial state
\[ \rho^* = \sum_{k=1}^{d} p_k |\phi_k\rangle \langle \phi_k|, \] (67)
with
\[ |\phi_k\rangle = \begin{cases} |\mu_k(0)\rangle + e^{i\chi_k}|\mu_{d-k+1}(0)\rangle \sqrt{2} & \text{if } 2k < d + 1, \\
|\mu_k(0)\rangle & \text{if } 2k = d + 1, \\
|\mu_k(0)\rangle - e^{i\chi_k}|\mu_{d-k+1}(0)\rangle \sqrt{2} & \text{if } 2k > d + 1, \end{cases} \] (68)
where $\chi_k$ are arbitrary real phases (the theorem as formulated in the Letter is recovered by setting $\chi_k = 0$), and by choosing the Hamiltonian control $H_\epsilon(t)$ such that
\[ U_\alpha(t)|\mu_k(0)\rangle = |\mu_k(t)\rangle \quad \forall k = 1, \ldots, d \quad \forall t, \] (69)
where
\[ U_\alpha(t) = T \left[ \exp \left( -i \int_{0}^{t} [H_\alpha(\tau) + H_\epsilon(\tau)] d\tau \right) \right]. \] (70)

**Proof.** From theorem 5 we have that for any state $\rho$ and any generator $h_\alpha$ with ordered eigenvalues $p_1 \geq \cdots \geq p_d$ and $h_1 \geq \cdots \geq h_d$, respectively, the maximal QFI with respect to all unitary state preparations $U \rho U^\dagger$, $U \in U(d)$, is given by
\[ I_{\alpha}^* := \max_{U} I_{\alpha}(U \rho U^\dagger) = \frac{1}{2} \sum_{j=1}^{d} p_{j,d-j+1}(h_j - h_{d-j+1})^2. \] (71)

Further, the generator can be written as \cite[Eq. 6]{5}
\[ h_\alpha = \int_{0}^{T} U_\alpha(t)^\dagger \partial_\alpha H_\alpha(t) U_\alpha(t) \, dt, \] (72)
Writing the integral as an infinite sum,
\[ h_\alpha = \lim_{n \to \infty} \sum_{\ell=0}^{n} U_\alpha^\dagger(\ell T/n) \partial_\alpha H_\alpha(\ell T/n) U_\alpha(\ell T/n) T/n, \] (73)
repeated application of lemma 12 to bipartitions of the sum yields in the limit of infinite many applications of lemma 12
\[ I_{\alpha}^* = \frac{1}{2} \sum_{j=1}^{d} p_{j,d-j+1}(h_j - h_{d-j+1})^2 \leq \frac{1}{2} \sum_{j=1}^{d} p_{j,d-j+1} \left( \int_{0}^{T} [\mu_j(t) - \mu_{d-j+1}(t)] dt \right)^2 = K_{\alpha}. \] (74)
It remains to show that Eq. (66) can be saturated. In order to show this it suffices to calculate the QFI for the initial state as defined in Eqs. (67) and (68) and a generator as given in Eq. (73) with the unitary transformation fulfilling Eq. (70):

$$I_\alpha(\rho^*) = 2 \sum_{i,j=1}^{d} p_{i,j} |\langle \phi_i | h_\alpha | \phi_j \rangle|^2$$

where $|\phi_j\rangle$ are defined in Eq. (68). More explicitly, in

$$\langle \phi_i | h_\alpha | \phi_j \rangle = \lim_{n \to \infty} \sum_{l=0}^{n} \langle \phi_i | U_\alpha^l(IT/n) \partial_\alpha H_\alpha(IT/n) U_\alpha(IT/n) | \phi_j \rangle \frac{T}{n}$$

we use the definition of $|\phi_j\rangle$ and Eq. (69) which gives, due to

$$\langle \mu_i(IT/n) | \partial_\alpha H_\alpha(IT/n) | \mu_j(IT/n) \rangle = \delta_{i,j} \mu_i(IT/n),$$

the following expression for the matrix coefficients in Eq. (76):

$$\langle \phi_i | U_\alpha^l(IT/n) \partial_\alpha H_\alpha(IT/n) U_\alpha(IT/n) | \phi_j \rangle = \delta_{i,j} \frac{\mu_i(IT/n) + \mu_{d-i+1}(IT/n)}{2} + \delta_{i,d-j+1} \frac{\mu_i(IT/n) - \mu_{d-i+1}(IT/n)}{2}.$$  

Due to $p_{i,i} = 0$ one obtains

$$I_\alpha(\rho^*) = 2 \sum_{j=1}^{d} p_{j,d-j+1} \left| \lim_{n \to \infty} \sum_{l=0}^{n} \frac{\mu_j(IT/n) - \mu_{d-j+1}(IT/n)}{2} \frac{T}{n} \right|^2$$

$$= \frac{1}{2} \sum_{j=1}^{d} p_{j,d-j+1} \left( \int_0^T [\mu_j(t) - \mu_{d-j+1}(t)] dt \right)^2 = K_\alpha.$$  

□

Proof of Heisenberg scaling for thermal states

In this section we will prove that if a product of $N$ thermal spin-$j$ states (at arbitrary finite temperature) is available and sensor dynamics is unitary, one can reach Heisenberg scaling of the QFI $I_\alpha$ for unitary dynamics in $N$ and $j$ by preparing the optimal initial state according theorem 1 in the Letter (or theorem 2, in case of Hamiltonian control). Heisenberg scaling in $N$ and $j$ means $I_\alpha \propto N^2$ for any $j = \frac{1}{2}, 1, \frac{3}{2}, \ldots$ and $I_\alpha \propto j^2$ for any $N = 1, 2, 3, \ldots$.

According to the pinching theorem (also known as squeeze theorem) a function scales with $N^2$ ($j^2$) if there are upper and lower bounds scaling as $N^2$ ($j^2$). Clearly, the QFI of a product of $N$ thermal spin-$j$ states is upper bounded by the pure-state case obtained in the limiting case of zero temperature. For pure states, it is well known that the QFI, optimized over unitary state preparations, scales as $N^2$ ($j^2$). We will find lower bounds for the QFI of a product of $N$ thermal spin-$j$ states that scale as $N^2$ ($j^2$).

Let the QFI be given by (compared to Eq. (14) in the Letter, we set $g(T) = 1$ because we are only interested in the scaling with $N$ and $j$ in the following)

$$K_B = \frac{4}{Z_\beta^N} \sum_{k=N-j}^{N+j} q(k) \frac{\sinh^2(\beta k)}{\cosh(\beta k)} k^2,$$

with $q(k)$ the number of possibilities of getting a sum $k$ when rolling $N$ fair dice, each having $2j+1$ sides corresponding to values $\{-j, \ldots, j\}$, and with $Z_\beta$ the partition function

$$Z_\beta = \sum_{m=-j}^{j} e^{\beta m} = \cosh(\beta j) + \frac{\sinh(\beta j)}{\tanh(\beta/2)},$$
which was rewritten (for $\beta > 0$) making use of the geometric series. First, we find a lower bound $L_B$ for $K_B$:

$$K_B = \frac{4}{Z_\beta^N} \sum_{k=-N_j}^{N_j} q(k) \frac{\sinh^2(\beta k)}{\cosh(\beta k)} k^2 = \frac{4}{Z_\beta^N} \sum_{k=-N_j}^{N_j} q(k) \left( \cosh(\beta k) - \frac{1}{\cosh(\beta k)} \right) k^2$$  \hspace{1cm} (83)

$$\geq \frac{4}{Z_\beta^N} \sum_{k=-N_j}^{N_j} q(k) \left( \cosh(\beta k) - 1 \right) k^2 =: L_B,$$  \hspace{1cm} (84)

where we used that each summand is nonnegative and

$$\frac{\sinh^2(\beta k)}{\cosh(\beta k)} = \frac{\cosh^2(\beta k) - 1}{\cosh(\beta k)} = \cosh(\beta k) - \frac{1}{\cosh(\beta k)} \geq \cosh(\beta k) - 1,$$  \hspace{1cm} (85)

which follows from the trigonometric identity $\sinh^2(x) - \cosh^2(x) = 1$ and $\cosh(x) \geq 1$. Next, we rewrite $L_B$ as

$$L_B = \frac{4}{Z_\beta^N} \sum_{k=-N_j}^{N_j} q(k) \left( e^{\beta k} - 1 \right) k^2,$$  \hspace{1cm} (86)

where we used that $\cosh(x) = (e^x + e^{-x})/2$ and $\sum_{k=-N_j}^{N_j} q(k) e^{\beta k} k^2 = \sum_{k=-N_j}^{N_j} q(k) e^{-\beta k} k^2$ because $q(k)k^2$ is symmetric around $k = 0$.

Then, we make use of the generating function of $q(k)$ [6]:

$$(x^{-j} + x^{-j+1} + \cdots + x^j)^N = \sum_{k=-N_j}^{N_j} q(k)x^k.$$  \hspace{1cm} (87)

By setting $x = e^\beta$, we find $Z_\beta^N = \sum_{k=-N_j}^{N_j} q(k)e^{\beta k}$. Taking the second derivative with respect to $\beta$ yields

$$\frac{\partial Z_\beta^N}{\partial \beta^2} = \sum_{k=-N_j}^{N_j} q(k)e^{\beta k} k^2.$$  \hspace{1cm} (88)

With this, we rewrite $L_B$ as

$$L_B = \left( \frac{\partial^2 Z_\beta^N}{\partial \beta^2} - \frac{\partial^2 Z_\beta^N}{\partial \beta^2} \right)_{\beta=0} \frac{4}{Z_\beta^N},$$  \hspace{1cm} (89)

where the second term is evaluated for $\beta = 0$ and corresponds the negative part of $L_B$ in Eq. (86). Since $Z_\beta \geq Z_{\beta=0}$, we find the lower bound

$$- \frac{\partial^2 Z_\beta^N}{\partial \beta^2} \bigg|_{\beta=0} \frac{4}{Z_\beta^N} \geq - \frac{\partial^2 Z_\beta^N}{\partial \beta^2} \bigg|_{\beta=0} \frac{4}{Z_{\beta=0}^N},$$  \hspace{1cm} (90)

which is readily evaluated:

$$- \frac{\partial^2 Z_\beta^N}{\partial \beta^2} \bigg|_{\beta=0} \frac{4}{Z_{\beta=0}^N} = -4 \left[ N(N - 1) \left( \frac{\partial Z_\beta}{\partial \beta} \bigg|_{\beta=0} \right)^2 + N \frac{\partial^2 Z_\beta}{\partial \beta^2} \bigg|_{\beta=0} \right],$$  \hspace{1cm} (91)

and with $\partial Z_\beta/\partial \beta|_{\beta=0} = \sum_{m=-j}^{j} m = 0$ we find

$$- \frac{\partial^2 Z_\beta^N}{\partial \beta^2} \bigg|_{\beta=0} \frac{4}{Z_{\beta=0}^N} = -4N \sum_{m=-j}^{j} m^2/(2j + 1) = -4N j(j + 1),$$  \hspace{1cm} (92)

again using the geometric series. Together with

$$\frac{\partial^2 Z_\beta^N}{\partial \beta^2} \frac{4}{Z_\beta^N} = 4N(N - 1) \left( \frac{\partial Z_\beta/\partial \beta}{Z_\beta} \right)^2 + 4N \frac{\partial^2 Z_\beta}{\partial \beta^2},$$  \hspace{1cm} (93)
this brings us to another lower bound:

\[ L_B \geq M_B = 4 \left[ N(N - 1) \left( \frac{\partial Z_\beta}{\partial \beta} \right)^2 + N \frac{\partial^2 Z_\beta}{\partial \beta^2} - \frac{1}{3} N j(j + 1) \right] \]  

(94)

Neglecting terms proportional to \( N \) we find after trivial algebraic transformations

\[ M_B \propto 4N^2 \left( \frac{\partial Z_\beta}{\partial \beta} \right)^2 \]

(95)

\[ = N^2 \left\{ (1 + j) \sinh(\beta j) - j \sinh[\beta(1 + j)] \right\}^2 \frac{\sinh^2(\frac{\beta}{2})}{\sinh^2 \left[ \beta \left( j + \frac{1}{2} \right) \right]} , \]

(96)

which is clearly nonnegative for finite temperatures (\( \beta > 0 \)). In order to become zero,

\[ \frac{\sinh[\beta j]}{\beta j} = \frac{\sinh[\beta(j + 1)]}{\beta(j + 1)} \]

(97)

would have to be fulfilled. However, since \( \frac{\sinh(x)}{x} = 1 + \frac{x^2}{3!} + \frac{x^4}{5!} + \ldots \) is an increasing function on \( x \in [0, \infty) \), Eq. (97) leads to a contradiction for \( \beta > 0 \). Therefore, the expression in Eq. (96) is positive for finite temperatures which proves the \( N^2 \) scaling for all \( j = \frac{1}{2}, 1, \frac{3}{2}, \ldots \).

There are two remarks in order:

(i) In summary the lower bound \( M_B \) was obtained from QFI \( K_B \) by finding a lower bound for the negative term of the QFI in Eq. (83),

\[ - \frac{4}{Z_\beta^2} \sum_{k=-N_j}^{N_j} q(k) \frac{k^2}{\cosh(\beta k)} \geq - \frac{4}{3} N j(j + 1) . \]

(98)

Since the right-hand side scales linearly in \( N \), the left-hand side scales at most linearly in \( N \) and, in particular, cannot scale quadratically. Thus, the \( N^2 \)-scaling of the QFI solely comes from the positive term in Eq. (83). Therefore, in leading order of \( N \) we find for the QFI exactly Eq. (95), i.e.,

\[ K_B = 4N^2 \left( \frac{\partial Z_\beta}{\partial \beta} \right)^2 + O(N) = 4N^2 \left( \frac{\partial \ln Z_\beta}{\partial \beta} \right)^2 + O(N) , \]

(99)

where \( O(N) \) denotes terms \( \propto N \) and lower-order terms. With the operator \( S_z \) of a spin \( j \) in \( z \)-direction and corresponding thermal state \( \rho_{th} = e^{\beta S_z}/Z_\beta \), we rewrite \( Z_\beta = \text{tr}[e^{\beta S_z}] \) and

\[ 4N^2 \left( \frac{\partial \ln Z_\beta}{\partial \beta} \right)^2 = 4N^2 \langle S_z \rangle^2 , \]

(100)

where \( \langle S_z \rangle = \text{tr}[\rho_{th} S_z] \).

(ii) Let us identify \( \beta = 1/\chi \) with a temperature \( \chi \). A Taylor expansion of the prefactor \( Q(\beta) = 4 \left( \frac{\partial \ln Z_\beta}{\partial \beta} \right)^2 \) in Eq. (100) around \( \beta = 0 \) yields

\[ Q(\beta) = \frac{4}{9} \left[ j(j + 1) \right]^2 \beta^2 + \mathcal{O}(\beta^4) , \]

(101)

where \( \mathcal{O}(\beta^4) \) denotes terms \( \propto \beta^4 \) and higher-order terms which can be neglected for small \( \beta \). This shows that for small \( \beta \), i.e., for large temperatures \( \chi \), the prefactor decays quadratically, \( \propto \chi^{-2} \). Also, in this regime of high temperatures, the QFI scales with \( j^4 \). However, if the product \( j \beta \) is of order one (or larger), we are no longer in the range of validity of the second-order Taylor expansion in Eq. (101). As we will see in the next section, the QFI scales \( \propto j^2 \) in the limit of large \( j \).
In order to prove $j^2$ scaling, we first consider

$$M_B(N) - NM_B(N = 1) = N(N - 1) \left( \frac{\partial Z_\beta/\partial \beta}{Z_\beta} \right)^2 \geq 0$$

(102)

for any $N \geq 1$. It follows that $M_B(N) \geq NM_B(N = 1)$. We find after some simple algebraic transformations using the partition function as given in Eq. (82),

$$NM_B(1) = 4N \left[ \frac{\partial^2 Z_\beta/\partial \beta^2}{Z_\beta} - \frac{j(j + 1)}{3} \right]$$

(103)

$$= N \left[ \frac{j^2 8}{3} - j \frac{10 \cosh(\beta j) + 2 \cosh[\beta(j + 1)]}{3 \sinh(\beta/2) \sinh[\beta(j + 1/2)]} + \frac{\sinh(\beta) \sinh(\beta j)}{\sinh^3(\beta/2) \sinh[\beta(j + 1/2)]} \right],$$

(104)

which is well defined for finite temperatures, and the third summand as well as the prefactor of $j$ in the second summand clearly converge to a constant in the limit of large $j$. This proves the $j^2$ scaling for finite temperatures for any $N = 1, 2, \ldots$.

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