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.

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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 $\Delta\hat{\alpha} = \mathrm{Var}(\hat{\alpha})^{1/2}$ of the estimator $\hat{\alpha}$ of the parameter α 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 nonclassical 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 $\Delta\hat{\alpha} \geq (MI_\alpha)^{-1/2}$, which depends on the number of measurements M and the quantum Fisher information (QFI) I_α 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 when the QFI is maximal which involves an optimization with respect to the state.

In this Letter, we consider a freely available state ρ, unitary freedom to prepare an initial state from ρ, 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, and 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

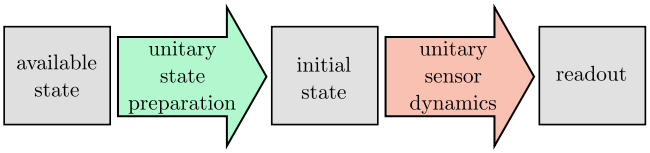

FIG. 1. Schematic representation of the metrology protocol.

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 $U_\alpha := \mathcal{T}(\exp[-i/\hbar \int_0^T H_\alpha(t)dt])$, where $\mathcal{T}$ 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 α, $H_\alpha = \alpha G$, with some parameter-independent operator G. The parameter dependence of the sensor dynamics is characterized by the generator $h_\alpha := iU_\alpha^\dagger(\partial U_\alpha/\partial\alpha)$, which simplifies to G for phase-shift Hamiltonians [12,26–28].

By introducing the eigendecomposition 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]

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

with coefficients

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

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

Theorem 1: For any state ρ and any generator h_α 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 $U\rho U^\dagger$, $U \in \mathcal{U}(d)$, is given by

$$I_\alpha^* := \max_U I_\alpha(U\rho U^\dagger) = \frac{1}{2}\sum_{k=1}^{d} p_{k,d-k+1}(h_k - h_{d-k+1})^2. \tag{3}$$

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

$$\rho^* = \sum_{k=1}^{d} p_k|\phi_k\rangle\langle\phi_k|, \tag{4}$$

with [29]

$$|\phi_k\rangle = \begin{cases} \frac{|h_k\rangle + |h_{d-k+1}\rangle}{\sqrt{2}} & \text{if } 2k < d+1, \\ |h_k\rangle & \text{if } 2k = d+1, \\ \frac{|h_k\rangle - |h_{d-k+1}\rangle}{\sqrt{2}} & \text{if } 2k > d+1. \end{cases} \tag{5}$$

The proof is based on the Bloomfield-Watson inequality on the Hilbert-Schmidt norm of off-diagonal blocks of a Hermitian matrix [30,31] and is given in the Supplemental Material [32]. The idea of the proof 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 Bloomfield-Watson 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 ρ plays a crucial role both for the maximal QFI and for the optimal state: in order to reach the maximal QFI I_α^*, 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 $r = 1$ [12,26,27,33,34]. Then, the maximal QFI in Eq. (3) simply becomes $(h_1 - h_d)^2$ and is obtained by preparing an equal superposition $(|h_1\rangle + |h_d\rangle)/\sqrt{2}$ of the eigenvectors corresponding to the minimal and maximal eigenvalues of h_α. 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 $p_1(h_1 - h_5)^2 + [(p_2 - p_4)^2/(p_2 + p_4)](h_2 - h_4)^2$. 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_α^* 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 $H_c(t)$ 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 [34]. The question, how this generalizes if the available state is mixed, brings us to

Theorem 2: For any state ρ 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

$$K_\alpha = \frac{1}{2}\sum_{k=1}^{d} p_{k,d-k+1}\left(\int_0^T [\mu_k(t) - \mu_{d-k+1}(t)]dt\right)^2. \tag{6}$$

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_α is reached by preparing the initial state

$$\rho^* = \sum_{k=1}^{d} p_k |\phi_k\rangle\langle\phi_k|, \tag{7}$$

with

$$|\phi_k\rangle = \begin{cases} \frac{|\mu_k(0)\rangle + |\mu_{d-k+1}(0)\rangle}{\sqrt{2}} & \text{if } 2k < d+1, \\ |\mu_k(0)\rangle & \text{if } 2k = d+1, \\ \frac{|\mu_k(0)\rangle - |\mu_{d-k+1}(0)\rangle}{\sqrt{2}} & \text{if } 2k > d+1, \end{cases} \tag{8}$$

and choosing the Hamiltonian control $H_c(t)$ such that

$$U_\alpha(t)|\mu_k(0)\rangle = |\mu_k(t)\rangle \quad \forall\ k = 1, \ldots, d \quad \forall\ t, \tag{9}$$

where

$$U_\alpha(t) = \mathcal{T}\left[\exp\left(-i\int_0^t [H_\alpha(\tau) + H_c(\tau)]d\tau\right)\right]. \tag{10}$$

The proof (see the Supplemental Material [32]) starts by rewriting h_α as in Ref. [[34] Eq. (6)] and shows that Eq. (6) is an upper bound for Eq. (3). We use the Schur convexity [38] of Eq. (3) and the inequalities from K. Fan [39,40] 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 $\partial_\alpha H_\alpha(t)$ 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 $\partial_\alpha H_\alpha(0)$ 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 $H_\alpha = \alpha G$, we have $h_\alpha = \partial_\alpha H_\alpha$ and, thus, the optimal initial states of Theorems 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 $\hat{\alpha}$. It was shown that replacing α by $\hat{\alpha}$ in the optimal control Hamiltonian does not ruin the benefits from introducing Hamiltonian control [34], and Hamiltonian control was applied experimentally with great success in Ref. [41]. For a more detailed discussion of control Hamiltonians we refer to the work of Pang *et al.* [34,42].

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

$$H(t) = \sum_{k=1}^{N} B f(t) S_z^{(k)} + H_{\text{int}}, \tag{11}$$

with the magnetic field amplitude B, some time-dependent real-valued modulation function $f(t)$, and spin operator $S_z^{(k)}$ in the z direction of the kth spin. We use the standard angular momentum algebra, $S_z^{(k)}|j,m\rangle = m|j,m\rangle$ with $m = -j, \ldots, j$. H_{int} 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 [16,17,43], nuclear spin ensembles [44], or vapor of alkali atoms [19]. Due to imperfect polarization and competing depolarization effects [20,21,45,46], 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)]

$$\rho_{\text{th}} = \frac{e^{\beta S_z}}{Z}, \tag{12}$$

with partition function $Z = \sum_{m=-j}^{j} e^{\beta m}$, and inverse (effective) temperature β. Equation (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 $\beta = \ln[1 + P/(1-P)]$; $P = 1$ 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, $\rho = \rho_{\text{th}}^{\otimes N}$.

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 [47,48]]. 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

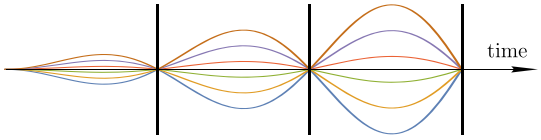

FIG. 2. Exemplary sketch of time-dependent eigenvalues $\mu_1 \geq \cdots \geq \mu_6$ of $\partial H(t)/\partial\omega$ corresponding to $f(t) = \cos(\omega t)$. Vertical black lines indicate the position of single-spin π pulses about the x axis in order to interchange eigenvectors $|j,m\rangle \leftrightarrow |j,-m\rangle$.

$$K_B = g^2(T) \sum_{k=-Nj}^{Nj} q(k) \frac{\sinh^2(\beta k)}{Z^N \cosh(\beta k)} (2k)^2, \qquad (13)$$

where $q(k)$ takes into account the degeneracy of eigenvalues of ρ and $\partial_B H(t) \coloneqq \partial H(t)/\partial B$. It follows from the definition of the tensor product that the degeneracy of the kth 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 the Supplemental Material [32]) [[49] p. 23–24]:

$$q(k) \coloneqq \sum_{\ell=0}^{N} (-1)^\ell \binom{N}{\ell} \binom{k + N(j+1) - 1 - \ell(2j+1)}{N-1}, \qquad (14)$$

where the binomial coefficient $\binom{a}{b}$ is set to zero if one or both of its coefficients are negative. The dependence on measurement time T is given by $g(T) = \int_0^T |f(t)| \mathrm{d}t$.

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 $K_B = 4N^2\langle S_z\rangle^2 + \mathcal{O}(N)$ where $\langle S_z\rangle = \mathrm{tr}[\rho_{\mathrm{th}} S_z]$, and $\mathcal{O}(N)$ denotes terms of order N and lower order. In the limit of large temperatures, $\langle S_z\rangle^2$ decays as β^2 (see the Supplemental Material [32]).

This means that Heisenberg scaling [1,50,51], 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_{int} 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 $e^{\beta k}/Z^N$ of ρ with the eigenstates corresponding to the (degenerate) eigenvalue $e^{-\beta k}/Z^N$ for all $k = 1,\ldots,Nj$. This is realized, for instance, with a local π pulse about the x axis, which interchanges $|j,m\rangle$ and $|j,-m\rangle$ for every single spin. The π pulses ensure optimal phase accumulation of the optimal state given by Eq. (7) (cf. Fig. 2).

The degeneracy of eigenvalues of ρ 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, $H_{\mathrm{int}} = 0$, 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 ω 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_ω equals Eq. (13)

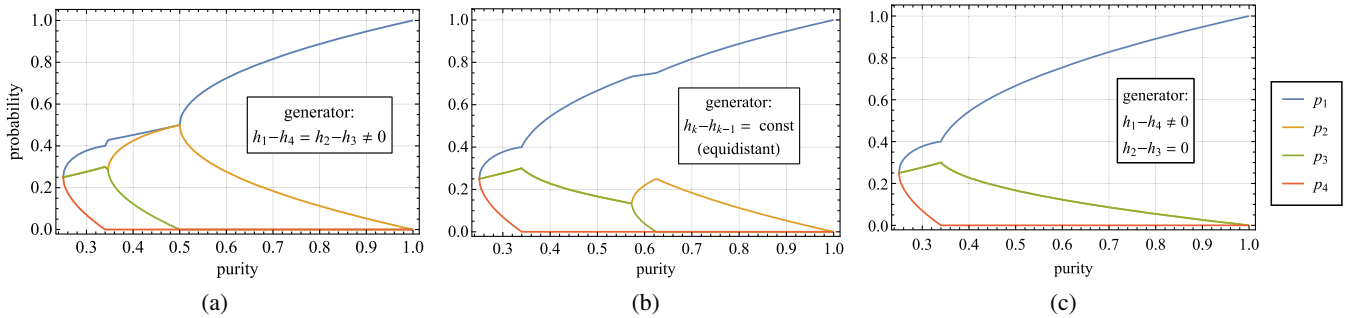

FIG. 3. Eigenvalues $p_1 \geq \cdots \geq p_4$ of initial two-qubit states that maximize the QFI for different values of purity γ. For each value of purity, eigenvalues p_i are found numerically by maximizing the expression for maximal QFI from Theorem 1 in Eq. (3) under the constraints of fixed purity and conservation of probability, $\sum_k p_k = 1$. Different panels correspond to different spectra of the generator with eigenvalues $h_1 \geq \cdots \geq h_4$ as indicated in the insets. The generator used in panel (a) has two degeneracies, the one in panel (b) has an equidistant spectrum, and the one in panel (c) has one degeneracy. In panel (c), the line corresponding to p_3 overlays the line of p_2.

with the only difference that $g(T)$ is replaced by $g_\omega(T) = \int_0^T Bt|\sin(\omega t)|dt \simeq BT^2/\pi$, corresponding to a T^4 scaling of QFI, similar to what was reported in Ref. [34]. The optimal control is similar to the estimation of B: interactions must be canceled and local π 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 $\gamma = \mathrm{tr}\rho^2$. Fixing only γ 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 [52–54].

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[29] More generally, the eigenvectors of ρ^* may be written with a relative phase in the superpositions of $|h_k\rangle$ [$|\mu_k(0)\rangle$ in case of Theorem 2] such that the orthonormality condition remains fulfilled, i.e., $(|h_k\rangle + e^{i\varphi_k}|h_{d-k+1}\rangle/\sqrt{2})$ if $2k < d+1$ and $(|h_k\rangle - e^{i\varphi_k}|h_{d-k+1}\rangle/\sqrt{2})$ if $2k > d+1$, where the same applies for Theorem 2 when replacing the $|h_k\rangle$ by $|\mu_k(0)\rangle$.

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