Supplemental Material for "Efficient adaptive control strategy for multi-parameter quantum metrology in two-dimensional systems"

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This Supplemental Material provides the mathematical derivations and technical details supporting the results in the main text. Specifically, we prove the optimality of the maximally entangled system-extension scheme and derive the conditions for achieving Heisenberg scaling under Hamiltonian control. We also provide analytical derivations of the quantum Fisher information matrix, estimator variances, and the covariance matrix of the adaptive protocol, justify the reparameterization procedure, establish the relation between the adaptive and optimal-control limits, and derive the probability density function of the deviation factor used in the stochastic analysis.

SUPPLEMENTARY NOTE 1. ELIMINATING MEASUREMENT TRADE-OFF VIA SYSTEM EXTENSION

This Supplementary Note proves that the system extension scheme with the initial state being a maximally entangled state eliminates the measurement trade-off.

Consider a d-dimensional system governed by a Hamiltonian $H_{\boldsymbol{\alpha}}$. We introduce an ancillary system with dimension no less than d, whose Hamiltonian is the identity operator I_{A}. The initial state of the joint system is an arbitrary pure state, denoted as $|\psi_0\rangle = \sum_{l=0}^{d-1} \lambda_l |l_{\mathrm{P}}\rangle \otimes |l_{\mathrm{A}}\rangle$, where $\{|l_{\mathrm{P}}\rangle \mid l = 0, 1, \ldots, d-1\}$ forms a complete orthonormal basis of the system, $\{|l_{\mathrm{A}}\rangle \mid l = 0, 1, \ldots, d-1\}$ is a set of mutually orthogonal basis vectors for the ancillary system, and the non-negative coefficients λ_l satisfy $\sum_{l=0}^{d-1} \lambda_l^2 = 1$. The Hamiltonian of the joint system is $H_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}$. From Eq. (8) of the main manuscript, we obtain

$$\langle\psi_0| h_i h_j \otimes I_{\mathrm{A}} |\psi_0\rangle = \langle\psi_0| h_j h_i \otimes I_{\mathrm{A}} |\psi_0\rangle , \tag{S1}$$

where $h_i = -i\left(\partial_{\alpha_i} U_{\boldsymbol{\alpha}}^{\dagger}\right) U_{\boldsymbol{\alpha}}$ and $U_{\boldsymbol{\alpha}} = \exp\left(-iH_{\boldsymbol{\alpha}} t\right)$. Calculating both sides of the equation separately, we obtain

$$\langle\psi_0| h_i h_j \otimes I_{\mathrm{A}} |\psi_0\rangle = \sum_{l=0}^{d-1} \lambda_l^2 \langle l_{\mathrm{P}}| h_i h_j |l_{\mathrm{P}}\rangle , \tag{S2}$$

and

$$\langle\psi_0| h_j h_i \otimes I_{\mathrm{A}} |\psi_0\rangle = \sum_{l=0}^{d-1} \lambda_l^2 \langle l_{\mathrm{P}}| h_j h_i |l_{\mathrm{P}}\rangle . \tag{S3}$$

Since h_i and h_j generally do not commute, Eq. (S1) holds only when $\lambda_l = \frac{1}{\sqrt{d}}$ due to the cyclic property of the trace operator. If the dimension of the ancillary system is equal to that of the system, we conclude that the weak commutativity condition is satisfied when the initial state is the maximally entangled state.

SUPPLEMENTARY NOTE 2. ELIMINATING INITIAL-STATE TRADE-OFF IN TWO-DIMENSIONAL SYSTEMS

This supplementary note proves that, for a two-dimensional system with system extension, choosing the maximally entangled state as the initial state makes the quantum Fisher information matrix optimal.

Suppose the joint Hamiltonian of a two-dimensional system and a two-dimensional ancillary system is $H_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}$ and the initial state is $|\psi_0\rangle = \sqrt{x}\,|0_{\mathrm{P}}0_{\mathrm{A}}\rangle + \sqrt{1-x}\,|1_{\mathrm{P}}1_{\mathrm{A}}\rangle$ $(0 \le x \le 1)$, where $\{|0_{\mathrm{P}}\rangle, |1_{\mathrm{P}}\rangle\}$ and $\{|0_{\mathrm{A}}\rangle, |1_{\mathrm{A}}\rangle\}$ are sets

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of complete orthonormal basis for the system and ancillary system, respectively. According to Eq. (4) in the main manuscript, the entries of quantum Fisher information matrix of the finial state are given by

$$\begin{aligned} F_{ij} = \; & 4\{\mathrm{Re}\left[x\left\langle 0_{\mathrm{P}}\right| h_i h_j \left|0_{\mathrm{P}}\right\rangle + (1-x)\left\langle 1_{\mathrm{P}}\right| h_i h_j \left|1_{\mathrm{P}}\right\rangle\right] \\ & - \left[x\left\langle 0_{\mathrm{P}}\right| h_i \left|0_{\mathrm{P}}\right\rangle + (1-x)\left\langle 1_{\mathrm{P}}\right| h_i \left|1_{\mathrm{P}}\right\rangle\right]\left[x\left\langle 0_{\mathrm{P}}\right| h_j \left|0_{\mathrm{P}}\right\rangle + (1-x)\left\langle 1_{\mathrm{P}}\right| h_j \left|1_{\mathrm{P}}\right\rangle\right]\}. \end{aligned} \tag{S4}$$

Denote the matrix representation of h_i in the basis $\{|0_{\mathrm{P}}\rangle, |1_{\mathrm{P}}\rangle\}$ as

$$h_i^{(\mathrm{M})} = \begin{Bmatrix} h_{i,11}^{(\mathrm{M})} & h_{i,12}^{(\mathrm{M})} \\ h_{i,21}^{(\mathrm{M})} & h_{i,22}^{(\mathrm{M})} \end{Bmatrix}, \tag{S5}$$

we have

$$\begin{aligned} F_{ij} = \; & 4\left\{\mathrm{Re}\left[x\left(h_{i,11}^{(\mathrm{M})}h_{j,11}^{(\mathrm{M})} + h_{i,12}^{(\mathrm{M})}h_{j,21}^{(\mathrm{M})}\right) + (1-x)\left(h_{i,21}^{(\mathrm{M})}h_{j,12}^{(\mathrm{M})} + h_{i,22}^{(\mathrm{M})}h_{j,22}^{(\mathrm{M})}\right)\right]\right. \\ & \left. - \left[xh_{i,11}^{(\mathrm{M})} + (1-x)\,h_{i,22}^{(\mathrm{M})}\right]\left[xh_{j,11}^{(\mathrm{M})} + (1-x)\,h_{j,22}^{(\mathrm{M})}\right]\right\}. \end{aligned} \tag{S6}$$

We define $\delta F = F|_{x=1/2} - F$, the elements of δF are given by

$$\delta F_{ij} = 4\left(h_{i,11}^{(\mathrm{M})} - h_{i,22}^{(\mathrm{M})}\right)\left(h_{j,11}^{(\mathrm{M})} - h_{j,22}^{(\mathrm{M})}\right)\left(x - \frac{1}{2}\right)^2. \tag{S7}$$

For both the two-parameter and three-parameter cases, all principal minors of δF are non-negative, which indicates that δF is positive semi-definite. Therefore, the maximally entangled state is the optimal pure state.

Let $\rho_0 = \sum_i p_i \left|\varphi_i\right\rangle\left\langle\varphi_i\right|$ be an arbitrary mixed initial state. Under the unitary evolution $U_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}$, the final state is $\rho_t = \sum_i p_i \left(U_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}\right)\left|\varphi_i\right\rangle\left\langle\varphi_i\right|\left(U_{\boldsymbol{\alpha}}^{\dagger} \otimes I_{\mathrm{A}}\right)$. According to the convexity of the quantum Fisher information matrix [1], we obtain

$$\begin{aligned} F\left(\rho_t\right) \leq \; & \sum_i p_i F\left[\left(U_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}\right)\left|\varphi_i\right\rangle\left\langle\varphi_i\right|\left(U_{\boldsymbol{\alpha}}^{\dagger} \otimes I_{\mathrm{A}}\right)\right] \\ \leq \; & \sum_i p_i F\left[\left(U_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}\right)\left|\psi_0\right\rangle\left\langle\psi_0\right|\left(U_{\boldsymbol{\alpha}}^{\dagger} \otimes I_{\mathrm{A}}\right)\right]\Big|_{x=\frac{1}{2}} \\ = \; & F|_{x=\frac{1}{2}}, \end{aligned} \tag{S8}$$

demonstrating that the maximally entangled initial state is optimal.

SUPPLEMENTARY NOTE 3. ACHIEVING HEISENBERG SCALING VIA HAMILTONIAN CONTROL

This supplementary note proves that using the counter-Hamiltonian as the control Hamiltonian enables the parameter estimation precision to reach the Heisenberg limit.

Consider a two-dimensional system with Hamiltonian $H_{\boldsymbol{\alpha}}$ and a two-dimensional ancillary system with Hamiltonian I_{A}. The probe and ancilla are initialized in a maximally entangled state and evolve under the joint Hamiltonian $H_{\boldsymbol{\alpha}} \otimes I_{\mathrm{A}}$. The entries of quantum Fisher information matrix for the final evolved state are given by

$$F_{ij} = 2\mathrm{Tr}\left(h_i\left(t\right) h_j\left(t\right)\right) - t^2 \mathrm{Tr}\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)\mathrm{Tr}\left(\partial_{\alpha_j} H_{\boldsymbol{\alpha}}\right), \tag{S9}$$

where $h_i\left(t\right)$ can be expressed as

$$h_i\left(t\right) = \int_0^t \mathrm{e}^{iH_{\boldsymbol{\alpha}}\tau}\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)\mathrm{e}^{-iH_{\boldsymbol{\alpha}}\tau} d\tau \tag{S10}$$

and we have used

$$\mathrm{Tr}\left(h_i\left(t\right)\right) = t\mathrm{Tr}\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right).$$

Suppose the initial Hamiltonian of the system is $H_{\boldsymbol{\alpha}}^{(\mathrm{init})}$, then the control Hamiltonian is $H_{\mathrm{c}} = -H_{\boldsymbol{\alpha}}^{(\mathrm{init})}$. Here, $H_{\boldsymbol{\alpha}}^{(\mathrm{init})}$ is parameter-dependent, while H_{c} is parameter-independent. The total Hamiltonian $H_{\boldsymbol{\alpha}} = H_{\boldsymbol{\alpha}}^{(\mathrm{init})} + H_{\mathrm{c}}$ vanishes, yielding $h_i\left(t\right) = t\partial_{\alpha_i} H_{\boldsymbol{\alpha}}^{(\mathrm{init})}$, and hence

$$F_{ij}^{(\mathrm{c})} = t^2\left\{2\mathrm{Tr}\left[\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}^{(\mathrm{init})}\right)\left(\partial_{\alpha_j} H_{\boldsymbol{\alpha}}^{(\mathrm{init})}\right)\right] - \mathrm{Tr}\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}^{(\mathrm{init})}\right)\mathrm{Tr}\left(\partial_{\alpha_j} H_{\boldsymbol{\alpha}}^{(\mathrm{init})}\right)\right\}. \tag{S11}$$

For sufficiently large n, the estimator variance of the parameter α_i is given by

$$\mathrm{Var}\left(\widehat{\alpha}_i\right) = \left[\left(nF^{(\mathrm{c})}\right)^{-1}\right]_{ii} \propto \frac{1}{nt^2}, \tag{S12}$$

where $\mathrm{Var}\left(\widehat{\alpha}_i\right) = \mathbb{E}\left[\left(\widehat{\alpha}_i - \mathbb{E}\left[\widehat{\alpha}_i\right]\right)^2\right]$.

SUPPLEMENTARY NOTE 4. TIME DEPENDENCE OF QUANTUM FISHER INFORMATION MATRIX

In this Supplementary Note, we derive the explicit time dependence of quantum Fisher information matrix.

To obtain the explicit time dependence of the quantum Fisher information matrix, we first solve for the explicit time dependence of $h_i(t)$ in Eq. (S10). Let $Y(\tau) = \mathrm{e}^{iH_{\boldsymbol{\alpha}}\tau}\left(\partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)\mathrm{e}^{-iH_{\boldsymbol{\alpha}}\tau}$, we obtain

$$\frac{\partial Y(\tau)}{\partial \tau} = i\left[H_{\boldsymbol{\alpha}}, Y\right], \tag{S13}$$

where $Y(0) = \partial_{\alpha_i} H_{\boldsymbol{\alpha}}$. The operator $\mathcal{H}(\cdot) \coloneqq [H_{\boldsymbol{\alpha}}, \cdot]$ is an Hermitian superoperator with four eigenvalues $\Lambda_1, \Lambda_2, \Lambda_3, \Lambda_4$, where $\Lambda_k = 0$ for $k = 1, ..., r$ and $\Lambda_k \neq 0$ for $k = r+1, ..., 4$. The corresponding eigenvectors are $\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_4$, which satisfy $\mathrm{Tr}\left[\Gamma_i^\dagger \Gamma_j\right] = \delta_{ij}$. We can decompose $Y(0)$ based on $\{\Gamma_k\}$ as

$$Y(0) = \sum_{k=1}^{4} c_k \Gamma_k, \tag{S14}$$

where $c_k = \mathrm{Tr}\left(\Gamma_k^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)$. The solution for $Y(\tau)$ is

$$Y(\tau) = \sum_{k=1}^{4} \mathrm{Tr}\left(\Gamma_k^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right) \exp\left(i\Lambda_k \tau\right) \Gamma_k, \tag{S15}$$

leading to

$$h_i(t) = t\sum_{k=1}^{r} \mathrm{Tr}\left(\Gamma_k^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)\Gamma_k - i\sum_{k=r+1}^{4} \frac{\exp\left(i\Lambda_k t\right) - 1}{\Lambda_k} \mathrm{Tr}\left(\Gamma_k^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)\Gamma_k. \tag{S16}$$

Suppose $H_{\boldsymbol{\alpha}}$ has two eigenvalues E_0 and E_1 with $E_0 \neq E_1$, and two corresponding eigenvectors $|E_0\rangle$ and $|E_1\rangle$, the eigenvalues and eigenvectors of $\mathcal{H}(\cdot)$ are

$$\begin{array}{ll} \Lambda_1 = 0 & \Gamma_1 = |E_0\rangle\langle E_0|, \\ \Lambda_2 = 0 & \Gamma_2 = |E_1\rangle\langle E_1|, \\ \Lambda_3 = E_0 - E_1 & \Gamma_3 = |E_0\rangle\langle E_1|, \\ \Lambda_4 = E_1 - E_0 & \Gamma_4 = |E_1\rangle\langle E_0|. \end{array} \tag{S17}$$

The coefficients $\mathrm{Tr}\left(\Gamma_k^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right)$ are

$$\begin{aligned} \mathrm{Tr}\left(\Gamma_1^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right) &= \partial_{\alpha_i} E_0, \\ \mathrm{Tr}\left(\Gamma_2^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right) &= \partial_{\alpha_i} E_1, \\ \mathrm{Tr}\left(\Gamma_3^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right) &= (E_1 - E_0)\langle E_0|\,\partial_{\alpha_i} E_1\rangle, \\ \mathrm{Tr}\left(\Gamma_4^\dagger \partial_{\alpha_i} H_{\boldsymbol{\alpha}}\right) &= (E_0 - E_1)\langle E_1|\,\partial_{\alpha_i} E_0\rangle. \end{aligned} \tag{S18}$$

Substituting Eq. (S17) and Eq. (S18) into Eq. (S16), we obtain

$$h_i(t) = \sum_{l=0}^{1} t\left(\partial_{\alpha_i} E_l\right)|E_l\rangle\langle E_l| + i\left[\exp\left(i\left(E_l - E_{1-l}\right)t\right) - 1\right]\langle E_l|\,\partial_{\alpha_i} E_{1-l}\rangle\,|E_l\rangle\langle E_{1-l}|. \tag{S19}$$

Finally, substituting Eq. (S19) into Eq. (S9) yields

$$F_{ij}(t) = \sum_{l=0}^{1} t^2\left[\left(\partial_{\alpha_i} E_l\right)\left(\partial_{\alpha_j} E_l\right) - \left(\partial_{\alpha_i} E_l\right)\left(\partial_{\alpha_j} E_{1-l}\right)\right] - 8\sin^2\left(\frac{\left(E_l - E_{1-l}\right)t}{2}\right)\langle E_l|\,\partial_{\alpha_i} E_{1-l}\rangle\langle E_{1-l}|\,\partial_{\alpha_j} E_l\rangle. \tag{S20}$$

SUPPLEMENTARY NOTE 5. TIME DEPENDENCE OF ESTIMATOR VARIANCES

This Supplementary Note provides an explicit expression for the estimator variances and analyzes its dependence on time.

The quantum Fisher information matrix can be directly obtained from Eq. (S20). The variance of $\widehat{\alpha}_1$, $\widehat{\alpha}_2$, and $\widehat{\alpha}_3$ are

$$\mathrm{Var}\left(\widehat{\alpha}_1\right)=\frac{t^2\csc^2\left(\frac{1}{2}\delta Et\right)\left[\left(\mu_2\partial_{\alpha_3}\delta E-\mu_3\partial_{\alpha_2}\delta E\right)^2+\left(\nu_2\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_2}\delta E\right)^2\right]+16\left(\mu_3\nu_2-\mu_2\nu_3\right)^2}{16nt^2\left[\mu_1\left(\nu_3\partial_{\alpha_2}\delta E-\nu_2\partial_{\alpha_3}\delta E\right)+\mu_2\left(\nu_1\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_1}\delta E\right)+\mu_3\left(\nu_2\partial_{\alpha_1}\delta E-\nu_1\partial_{\alpha_2}\delta E\right)\right]^2}, \tag{S21}$$

$$\mathrm{Var}\left(\widehat{\alpha}_2\right)=\frac{t^2\csc^2\left(\frac{1}{2}\delta Et\right)\left[\left(\mu_1\partial_{\alpha_3}\delta E-\mu_3\partial_{\alpha_1}\delta E\right)^2+\left(\nu_1\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_1}\delta E\right)^2\right]+16\left(\mu_3\nu_1-\mu_1\nu_3\right)^2}{16nt^2\left[\mu_1\left(\nu_3\partial_{\alpha_2}\delta E-\nu_2\partial_{\alpha_3}\delta E\right)+\mu_2\left(\nu_1\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_1}\delta E\right)+\mu_3\left(\nu_2\partial_{\alpha_1}\delta E-\nu_1\partial_{\alpha_2}\delta E\right)\right]^2}, \tag{S22}$$

and

$$\mathrm{Var}\left(\widehat{\alpha}_3\right)=\frac{t^2\csc^2\left(\frac{1}{2}\delta Et\right)\left[\left(\mu_1\partial_{\alpha_2}\delta E-\mu_2\partial_{\alpha_1}\delta E\right)^2+\left(\nu_1\partial_{\alpha_2}\delta E-\nu_2\partial_{\alpha_1}\delta E\right)^2\right]+16\left(\mu_2\nu_1-\mu_1\nu_2\right)^2}{16nt^2\left[\mu_1\left(\nu_3\partial_{\alpha_2}\delta E-\nu_2\partial_{\alpha_3}\delta E\right)+\mu_2\left(\nu_1\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_1}\delta E\right)+\mu_3\left(\nu_2\partial_{\alpha_1}\delta E-\nu_1\partial_{\alpha_2}\delta E\right)\right]^2}, \tag{S23}$$

respectively, where

$$\begin{aligned}\delta E=&\ E_0-E_1,\\ \mu_i=&\ \mathrm{Re}\left(\langle E_0|\ \partial_{\alpha_i}E_1\rangle\right),\\ \nu_i=&\ \mathrm{Im}\left(\langle E_0|\ \partial_{\alpha_i}E_1\rangle\right).\end{aligned} \tag{S24}$$

Since $\mathrm{Var}\left(\widehat{\alpha}_1\right)$, $\mathrm{Var}\left(\widehat{\alpha}_2\right)$, and $\mathrm{Var}\left(\widehat{\alpha}_3\right)$ are symmetric with respect to each other, we consider only $\mathrm{Var}\left(\widehat{\alpha}_1\right)$ in the following analysis. When $\left(\mu_2\partial_{\alpha_3}\delta E-\mu_3\partial_{\alpha_2}\delta E\right)^2+\left(\nu_2\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_2}\delta E\right)^2=0$, the oscillatory term vanishes. To ensure a nonzero numerator, we have $\partial_{\alpha_2}\delta E=\partial_{\alpha_3}\delta E=0$, leading to

$$\mathrm{Var}\left(\widehat{\alpha}_1\right)=\frac{1}{nt^2\left(\partial_{\alpha_1}\delta E\right)^2}, \tag{S25}$$

where $\partial_{\alpha_1}\delta E\neq 0$. In this case, $\mathrm{Var}\left(\widehat{\alpha}_1\right)$ achieves the Heisenberg limit. For instance, consider a Hamiltonian

$$H_{(B,\theta,\varphi)}=B\left(\cos\left(\theta\right)\cos\left(\varphi\right)\sigma_x+\cos\left(\theta\right)\sin\left(\varphi\right)\sigma_y+\sin\left(\theta\right)\sigma_z\right), \tag{S26}$$

where σ_x, σ_y, and σ_z are Pauli matrices. For estimating B, the fact that the eigenvalues of the Hamiltonian are independent of θ and φ enables the estimation precision to reach the Heisenberg limit.

When $\left(\mu_2\partial_{\alpha_3}\delta E-\mu_3\partial_{\alpha_2}\delta E\right)^2+\left(\nu_2\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_2}\delta E\right)^2\neq 0$, we first consider two extreme cases. When t is sufficiently small such that $t\ll 1/\left|\delta E\right|$, we have $\csc^2\left(\frac{1}{2}\delta Et\right)\approx 4/\left(\delta Et\right)^2$, so

$$\mathrm{Var}\left(\widehat{\alpha}_1\right)\propto\frac{1}{t^2}. \tag{S27}$$

When t is sufficiently large, we have

$$\mathrm{Var}\left(\widehat{\alpha}_1\right)\propto\csc^2\left(\frac{1}{2}\delta Et\right), \tag{S28}$$

which is a periodic function. For general t, its overall trend can be analyzed via periodic sampling. We set the initial sampling time as $t_{\mathrm{s},0}\in\left(0,2\pi/\left|\delta E\right|\right)$, and define $c_0=\csc^2\left(\frac{1}{2}\delta Et_{\mathrm{s},0}\right)$. For $t_{\mathrm{s},k}=t_{\mathrm{s},0}+2k\pi/\left|\delta E\right|$ with $k=1,2,3,\ldots$, we have $\csc^2\left(\frac{1}{2}\delta Et_{\mathrm{s},k}\right)=c_0$. The function

$$\mathrm{Var}\left(\widehat{\alpha}_1\right)_{c_0}=\frac{c_0t^2\xi_1+\xi_2}{nt^2\xi_3}, \tag{S29}$$

passes through all these sampling points, where

$$\begin{aligned}\xi_1=&\ \left(\mu_2\partial_{\alpha_3}\delta E-\mu_3\partial_{\alpha_2}\delta E\right)^2+\left(\nu_2\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_2}\delta E\right)^2,\\ \xi_2=&\ 16\left(\mu_3\nu_2-\mu_2\nu_3\right)^2,\\ \xi_3=&\ 16\left[\mu_1\left(\nu_3\partial_{\alpha_2}\delta E-\nu_2\partial_{\alpha_3}\delta E\right)+\mu_2\left(\nu_1\partial_{\alpha_3}\delta E-\nu_3\partial_{\alpha_1}\delta E\right)+\mu_3\left(\nu_2\partial_{\alpha_1}\delta E-\nu_1\partial_{\alpha_2}\delta E\right)\right]^2.\end{aligned} \tag{S30}$$

Since

$$\partial_t \mathrm{Var}\left(\widehat{\alpha}_1\right)_{c_0} \leq 0, \tag{S31}$$

the variance exhibits an overall decrease. From Eq. (S28), we know that for large t, the minimum variance per period occurs at $t_k = (2k+1)\,\pi/\left|\delta E\right|$, i.e., $c_0 = 1$. Therefore, the infimum of the variance is

$$\inf\ \mathrm{Var}\left(\widehat{\alpha}_1\right) = \frac{\xi_1}{n\xi_3}. \tag{S32}$$

However, since $\left|\partial_t \mathrm{Var}\left(\widehat{\alpha}_1\right)_{c_0}\right|$ decays cubically with time, it is not worthwhile to further increase the evolution time for limited gains in precision.

SUPPLEMENTARY NOTE 6. NECESSITY OF REPARAMETERIZATION

This Supplementary Note shows the necessity for reparameterization. In general, the parameterization of the Hamiltonian for a two-dimensional system can be arbitrary. For example, one can parameterize the Hamiltonian by the coefficients in the Pauli basis or by the strength and angular parameters. For the current problem of reducing δE^2 so as to extend the evolution time for the Heisenberg scaling, it will turn out that the parameterization by the coefficients in the Pauli basis is more convenient, so we will focus on this parameterization.

Usually the unknown parameters can exhibit nontrivial correlations between each other, manifested by the correlation matrix of the estimation. To simplify the estimation task of these parameters, a powerful tool is to transform the unknown parameters to other parameters which have simpler correlations (e.g., a diagonal correlation matrix) [2, 3], sometimes known as reparametrization. If the vector of the parameters $\boldsymbol{\alpha}$ is reparameterized as $\boldsymbol{\beta} = (\beta_1, \beta_2, \ldots, \beta_k)$, the quantum Fisher information matrix of $\boldsymbol{\beta}$ is related to the original quantum Fisher information matrix for $\boldsymbol{\alpha}$ through

$$F_{\boldsymbol{\beta}} = J^{\mathrm{T}} F_{\boldsymbol{\alpha}} J, \tag{S33}$$

where J is the Jacobian matrix defined as $J_{ij} = \partial\alpha_i/\partial\beta_j$.

Suppose the original Hamiltonian of the probe is decomposed in the Pauli basis as

$$H_{\boldsymbol{\alpha}}^{(\mathrm{init})} = \boldsymbol{f}\left(\boldsymbol{\alpha}\right)\cdot\boldsymbol{\sigma}, \tag{S34}$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$, $\boldsymbol{\alpha} = (\alpha_1, \alpha_2, \alpha_3)$, and $\boldsymbol{f}\left(\boldsymbol{\alpha}\right) = \left(f_1\left(\boldsymbol{\alpha}\right), f_2\left(\boldsymbol{\alpha}\right), f_3\left(\boldsymbol{\alpha}\right)\right)$ with $f_1\left(\boldsymbol{\alpha}\right)$, $f_2\left(\boldsymbol{\alpha}\right)$, and $f_3\left(\boldsymbol{\alpha}\right)$ being real-valued functions. The control Hamiltonian at the $(k+1)$-th iteration is denoted by

$$H_{\mathrm{c},k+1} = -\boldsymbol{f}\left(\hat{\boldsymbol{\alpha}}_k\right)\cdot\boldsymbol{\sigma}, \tag{S35}$$

where $\hat{\boldsymbol{\alpha}}_k = (\hat{\alpha}_{k,1}, \hat{\alpha}_{k,2}, \hat{\alpha}_{k,3})$ denotes the control parameters which are essentially the estimates of $\hat{\boldsymbol{\alpha}}$ from the k-th iteration. Suppose $\hat{\boldsymbol{\alpha}}_k$ deviates from the true value of $\boldsymbol{\alpha}$ by $\boldsymbol{\delta\alpha}_k$, i.e.,

$$\hat{\boldsymbol{\alpha}}_k = \boldsymbol{\alpha}_0 + \boldsymbol{\delta\alpha}_k, \tag{S36}$$

with $\boldsymbol{\alpha}_0 = (\alpha_{0,1}, \alpha_{0,2}, \alpha_{0,3})$ being the true value of $\boldsymbol{\alpha}$ and $\boldsymbol{\delta\alpha}_k = (\delta\alpha_{k,1}, \delta\alpha_{k,2}, \delta\alpha_{k,3})$ being the estimation errors from the k-th iteration, we obtain the δE^2 of the $(k+1)$-th iteration as

$$\delta E_{k+1}^2 = 4\left\|\boldsymbol{f}\left(\boldsymbol{\alpha}_0\right) - \boldsymbol{f}\left(\hat{\boldsymbol{\alpha}}_k\right)\right\|^2. \tag{S37}$$

When $\left\|\boldsymbol{\delta\alpha}_k\right\| \ll 1$, Eq. (S37) is simplified to

$$\delta E_{k+1}^2 \approx 4\left\|J\boldsymbol{\delta\alpha}_k\right\|^2, \tag{S38}$$

where J is the Jacobian matrix defined by $J_{ij} = \partial_{\alpha_j} f_i\left(\boldsymbol{\alpha}\right)$ evaluated at $\boldsymbol{\alpha}_0$.

If we reparameterize the Hamiltonian as

$$H_{\boldsymbol{\beta}}^{(\mathrm{init})} = \beta_1\sigma_x + \beta_2\sigma_y + \beta_3\sigma_z, \tag{S39}$$

where the new parameters are defined as $\beta_1 = f_1\left(\boldsymbol{\alpha}\right)$, $\beta_2 = f_2\left(\boldsymbol{\alpha}\right)$, and $\beta_3 = f_3\left(\boldsymbol{\alpha}\right)$. According to Eq. (S37), we obtain

$$\delta E_{k+1}^2 = 4\left\|\boldsymbol{\delta\beta}_k\right\|^2, \tag{S40}$$

which will greatly simplify the subsequent optimization.

SUPPLEMENTARY NOTE 7. DERIVATION OF COVARIANCE MATRIX

In this Supplementary Note, we derive the covariance matrix for the k-th iteration.

Suppose the estimate from the $(k-1)$-th iteration is $\hat{\boldsymbol{\beta}}_{k-1}$. The control Hamiltonian in the k-th iteration is $H_{\mathrm{c},k} = \hat{\boldsymbol{\beta}}_{k-1} \cdot \boldsymbol{\sigma}$ and the total Hamiltonian is $H_{\boldsymbol{\beta},k} = \left(\boldsymbol{\beta} - \hat{\boldsymbol{\beta}}_{k-1}\right) \cdot \boldsymbol{\sigma}$. From Eq. (10) in the main manuscript, the entries of quantum Fisher information matrix are

$$\begin{aligned} F^{(\boldsymbol{\beta})}_{k,ii} &= \frac{4\left(\delta\beta^2_{k-1,i}\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2 t_k^2 + \left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2 - \delta\beta^2_{k-1,i}\right)\sin^2\left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\| t_k\right)\right)}{\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^4}, \\ F^{(\boldsymbol{\beta})}_{k,ij} &= \frac{4\delta\beta_{k-1,i}\delta\beta_{k-1,j}\left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2 t_k^2 - \sin^2\left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\| t_k\right)\right)}{\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^4}, i \neq j. \end{aligned} \tag{S41}$$

The covariance matrix in the k-th iteration is given by

$$C^{(\boldsymbol{\beta})}_k = \left(n_k F^{(\boldsymbol{\beta})}_k\right)^{-1}, \tag{S42}$$

the elements of which turn out to be

$$\begin{aligned} C^{(\boldsymbol{\beta})}_{k,ii} &= \frac{\frac{\delta\beta^2_{k-1,i}}{t_k^2\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2} + \left[\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2 - \delta\beta^2_{k-1,i}\right]\csc^2\left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\| t_k\right)}{4n_k}, \\ C^{(\boldsymbol{\beta})}_{k,ij} &= \frac{\delta\beta_{k-1,i}\delta\beta_{k-1,j}\left(\frac{1}{t_k^2\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2} - \csc^2\left(\left\|\boldsymbol{\delta\beta}_{k-1}\right\| t_k\right)\right)}{4n_k}, i \neq j. \end{aligned} \tag{S43}$$

SUPPLEMENTARY NOTE 8. RELATION BETWEEN ESTIMATION VARIANCES OF ORIGINAL PARAMETERS WITH ADAPTIVE AND OPTIMAL CONTROL

In this Supplementary Note, we derive the relation between estimation variances of the original parameter with adaptive and optimal control strategy.

Suppose the total estimation variance of $\hat{\boldsymbol{\beta}}$ obtained from the adaptive control strategy with m iterations is $V_m = \mathrm{Tr}C^{(\boldsymbol{\beta})}_m$, and that from the optimal control strategy with the true values of the unknown parameters is V_{oc}. If the estimation variance of the original parameter α_i using the adaptive control strategy approaches the variance obtained with the optimal control strategy, analogous to how V_m approaches V_{oc}, it demonstrates the effectiveness of the proposed adaptive control strategy in estimating the original parameters.

For the adaptive control strategy, the covariance matrix after m iterations is $C^{(\boldsymbol{\beta})}_m$, with elements given in Eq. (S43). When the optimal evolution time scheme is applied, the total variance is

$$V_m = \frac{\frac{\left\|\boldsymbol{\delta\beta}_{m-1}\right\|^2}{g_0^2} + 2\left\|\boldsymbol{\delta\beta}_{m-1}\right\|^2 \csc^2\left(g_0\right)}{4n_m}, \tag{S44}$$

where we have used Eq. (25) in the main manuscript. For the optimal control Hamiltonian, V_{oc} is given by Eq. (33) in the main manuscript.

When the Hamiltonian contains three independent unknown parameters, $\boldsymbol{\alpha} = (\alpha_1, \alpha_2, \alpha_3)$, the quantum Fisher information matrix of $\boldsymbol{\alpha}$ is related to the quantum Fisher information matrix of $\boldsymbol{\beta}$ via

$$F_{\boldsymbol{\alpha}} = J^{\mathrm{T}} F_{\boldsymbol{\beta}} J, \tag{S45}$$

where J is given by $J_{ij} = \partial\beta_i/\partial\alpha_j$, with $\beta_i = f_i\left(\boldsymbol{\alpha}\right)$. Therefore, the relation between their covariance matrices is

$$C^{(\boldsymbol{\alpha})} = J^{-1} C^{(\boldsymbol{\beta})} \left(J^{-1}\right)^{\mathrm{T}}. \tag{S46}$$

For the adaptive control strategy, Eq. (S46) yields the variances of the original parameters after m iterations as

$$\begin{aligned} C^{(\boldsymbol{\alpha})}_{m,ii} = &\ \frac{1}{4n_m}\sum_{r,s=1,r\neq s}^{3}\left(J^{-1}\right)_{ir}\left(J^{-1}\right)_{is}\delta\beta_{m-1,r}\delta\beta_{m-1,s}\left(\frac{1}{g_0^2} - \csc^2\left(g_0\right)\right) \\ &+ \frac{1}{4n_m}\sum_{r=1}^{3}\left(J^{-1}\right)^2_{ir}\left[\frac{\delta\beta^2_{m-1,r}}{g_0^2} + \left(\left\|\boldsymbol{\delta\beta}_{m-1}\right\|^2 - \delta\beta^2_{m-1,r}\right)\csc^2\left(g_0\right)\right], \end{aligned} \tag{S47}$$

where we have used the optimal evolution time scheme. Let

$$\begin{aligned}\mu_{\max,i} = & \max\left\{\left|\left(J^{-1}\right)_{ir}\left(J^{-1}\right)_{is}\right| \middle| r,s\in\{1,2,3\}, r\neq s\right\},\\ \nu_{\max,i} = & \max\left\{\left(J^{-1}\right)_{ir}^{2}\middle| r\in\{1,2,3\}\right\}.\end{aligned} \tag{S48}$$

We obtain

$$C_{m,ii}^{(\boldsymbol{\alpha})} \leq \left(2\mu_{\max,i}\frac{g_0^2\csc^2(g_0)-1}{1+2g_0^2\csc^2(g_0)}+\nu_{\max,i}\right)V_m. \tag{S49}$$

For the optimal control strategy, Eq. (S46) yields the variances of the original parameters as

$$\begin{aligned}C_{\mathrm{oc},ii}^{(\boldsymbol{\alpha})} = & \frac{\left(J^{-1}\right)_{i1}^2+\left(J^{-1}\right)_{i2}^2+\left(J^{-1}\right)_{i3}^2}{4n_{\mathrm{oc}}t_{\mathrm{oc}}^2}\\ \geq & \frac{\nu_{\max,i}}{3}V_{\mathrm{oc}}.\end{aligned} \tag{S50}$$

Using

$$V_m = \kappa V_{\mathrm{oc}} \tag{S51}$$

to connect Eq. (S49) and Eq. (S50), we obtain

$$C_{m,ii}^{(\boldsymbol{\alpha})} \leq \frac{12g_0^2\csc^2(g_0)-3}{1+2g_0^2\csc^2(g_0)}\kappa C_{\mathrm{oc},ii}^{(\boldsymbol{\alpha})}, \tag{S52}$$

where $\mu_{\max,i}\leq\nu_{\max,i}$ has been used.

SUPPLEMENTARY NOTE 9. PROBABILITY DENSITY FUNCTION OF DEVIATION FACTOR

In this Supplementary Note, we derive the probability density function of deviation factor D_k for the k-th iteration. Since $\delta E_k^2 = 4\left\|\boldsymbol{\delta\beta}_{k-1}\right\|^2$ and $\boldsymbol{\delta\beta}_{k-1}$ follows a Gaussian distribution $\boldsymbol{\delta\beta}_{k-1}\sim\mathcal{N}\left(\mathbf{0},C_{k-1}^{(\boldsymbol{\beta})}\right)$ in the asymptotic limit of the number of trials, δE_k^2 follows a generalized χ^2 distribution, expressed as a weighted sum of squares of independent standard normal random variables, where the weights are four times the eigenvalues of $C_{k-1}^{(\boldsymbol{\beta})}$. Together with $\left\|\boldsymbol{\delta\beta}_{k-2}\right\|t_{\mathrm{opt},k-1}=g_0$, we obtain

$$\delta E_k^2 = \frac{1}{n_{k-1}t_{\mathrm{opt},k-1}^2}\chi_1^2(1)+\frac{g_0^2\csc^2(g_0)}{n_{k-1}t_{\mathrm{opt},k-1}^2}\chi_2^2(1)+\frac{g_0^2\csc^2(g_0)}{n_{k-1}t_{\mathrm{opt},k-1}^2}\chi_3^2(1). \tag{S53}$$

The mean of δE_k^2 is $\left(2g_0^2\csc^2(g_0)+1\right)/\left(n_{k-1}t_{\mathrm{opt},k-1}^2\right)$. From Eq. (35) of the main manuscript, we have

$$D_k = \frac{\chi_1^2(1)+g_0^2\csc^2(g_0)\left(\chi_2^2(1)+\chi_3^2(1)\right)}{2g_0^2\csc^2(g_0)+1}. \tag{S54}$$

Using the Laplace transform, we obtain the probability density function of D_k as

$$f_{D_k} = \frac{\left(2g_0^2+\sin^2(g_0)\right)\mathrm{e}^{-\frac{D_k\sin^2(g_0)}{2g_0^2}-D_k}\operatorname{erf}\left(\frac{\sqrt{D_k\left(-\frac{\sin^2(g_0)}{g_0^2}+2g_0^2\csc^2(g_0)-1\right)}}{\sqrt{2}}\right)}{2g_0\sqrt{g_0^2-\sin^2(g_0)}}, \tag{S55}$$

where

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}}\int_0^x \mathrm{e}^{-t^2}dt. \tag{S56}$$

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