Supplementary Information for "Conditions for Quantum Violation of Macrorealism in Large-spin Limit"

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A Derivation of the entropic Leggett–Garg inequalities in fig. 2

In the main text, we demonstrate the capacity of entropic Leggett-Garg inequalities (ELGIs) of varying orders to reveal quantum violations, as illustrated in fig. 2. The order of an ELGI corresponds to the number of variables involved in its joint entropy terms; for instance, $H(Q_1, Q_2)$ denotes a second-order entropy, while $H(Q_1, Q_2, Q_3)$ denotes a third-order entropy. In this section, we illustrate the procedure for systematically constructing inequalities of arbitrary order based on the elementary-type ELGIs.

For the case of $n = 3$, elementary-type ELGI is divided into four groups, namely

$$\mathcal{D}_i^{(123)} = H(Q_1, Q_2, Q_3) - H(Q_m, Q_n) \geq 0 \ , \quad (m, n \neq i) \tag{1}$$

$$\mathcal{D}_{ij}^{(123)} = H(Q_i, Q_m) + H(Q_j, Q_m) - H(Q_1, Q_2, Q_3) - H(Q_m) \geq 0 \ , \quad (m \neq i, j) \tag{2}$$

$$\mathcal{D}_k^{(ij)} = H(Q_i, Q_j) - H(Q_m) \geq 0 \ , \quad (m \neq i, j) \tag{3}$$

$$\mathcal{D}_{ij}^{(ij)} = H(Q_i) + H(Q_j) - H(Q_i, Q_j) \geq 0 \ , \tag{4}$$

where $i, j \in \{1, 2, 3\}$ are distinct indices, and the superscripts indicate available time points.

The above elementary-type ELGIs collectively constitute the necessary and sufficient conditions for all third-order ELGIs. It is important to emphasize that restricting the condition to $D_i^{(123)} \geq 0$ and $D_{ij}^{(123)} \geq 0$ alone is insufficient, as distinct third-order ELGIs can be constructed by combining with second-order ELGIs. For example, we obtain

$$\mathcal{D}_k^{(123)} + \mathcal{D}_k^{(ij)} = H(Q_1, Q_2, Q_3) - H(Q_k) \geq 0 \ . \quad (k = i, j) \tag{5}$$

In order to obtain the necessary and sufficient conditions for all second-order ELGIs, we need to remove the third-order term $H(Q_1, Q_2, Q_3)$ from the elementary-type ELGIs. In principle, the elimination of specified variables from a system of linear inequalities can be systematically performed via the Fourier-Motzkin elimination method [S1]. For instance, in the case of $n = 3$, we ultimately obtain all possible combinations of $\mathcal{D}_k^{(123)} + \mathcal{D}_{i,j}^{(123)}$, namely,

$$\mathcal{D}_{i,j|k}^{(123)} := \mathcal{D}_k^{(123)} + \mathcal{D}_{i,j}^{(123)} = H(Q_i, Q_k) + H(Q_j, Q_k) - H(Q_k) - H(Q_{i,j}) \geq 0 \ . \tag{6}$$

For the case $n = 4$, the elementary-type ELGI is divided into six groups, namely

$$\mathcal{D}_i^{(1234)} = H(Q_1, Q_2, Q_3, Q_4) - H(Q_r, Q_s, Q_t) \geq 0 \ , \quad (r, s, t \neq i) \tag{7}$$

$$\mathcal{D}_{ij}^{(1234)} = H(Q_i, Q_r, Q_s) + H(Q_j, Q_r, Q_s) - H(Q_1, Q_2, Q_3, Q_4) - H(Q_r, Q_s) \geq 0 \ , \quad (r, s \neq i, j) \tag{8}$$

$$\mathcal{D}_m^{(ijk)} = H(Q_i, Q_j, Q_k) - H(Q_r, Q_s) \geq 0 \ , \quad (r, s = i, j, k; r, s \neq m) \tag{9}$$

$$\mathcal{D}_{mn}^{(ijk)} = H(Q_m, Q_r) + H(Q_n, Q_r) - H(Q_i, Q_j, Q_k) - H(Q_m, Q_n) \geq 0 \ , \quad (m, n, r = i, j, k; r \neq m, n) \tag{10}$$

$$\mathcal{D}_m^{(ij)} = H(Q_i, Q_j) - H(Q_r) \geq 0 \ , \quad (m, r = i, j; r \neq i, j) \tag{11}$$

$$\mathcal{D}_{ij}^{(ij)} = H(Q_i) + H(Q_j) - H(Q_i, Q_j) \geq 0 \ , \tag{12}$$

where $i, j, k \in \{1, 2, 3, 4\}$ are distinct indices, and the superscripts indicate available time points.

Besides, after Fourier-Motzkin elimination, we obtain

$$\mathcal{D}_{i,j|k}^{(1234)} := \mathcal{D}_k^{(1234)} + \mathcal{D}_{i,j}^{(1234)} = H(Q_i, Q_k, Q_r) + H(Q_j, Q_k, Q_r) - H(Q_k, Q_r) - H(Q_i, Q_j, Q_r) \geq 0 \ , \quad (r \neq i, j, k) \tag{13}$$

$$\mathcal{D}_{m,n|l}^{(ijk)} := \mathcal{D}_l^{(ijk)} + \mathcal{D}_{m,n}^{(ijk)} = H(Q_m, Q_l) + H(Q_n, Q_l) - H(Q_l) - H(Q_{m,n}) \geq 0 \ . \quad (l, m, n \neq i, j, k) \tag{14}$$

Therefore, in fig. 2a, the second-order ELGI contains the inequalities eqs. (3), (4) and (6), and the third-order ELGI contains the inequalities eqs. (1) to (4); in fig. 2b, the second-order ELGI contains the inequalities eqs. (6), (11) and (14), the third-order ELGI contains the inequalities eqs. (9) to (13), and the four-order ELGI contains the inequalities eqs. (7) to (12).

In addition, for the case of $n = 3$, the ELGI formulated by Devi *et al.* is given as

$$H(Q_1, Q_2) + H(Q_2, Q_3) - H(Q_2) - H(Q_1, Q_3) \geq 0 \ ; \tag{15}$$

whereas for $n = 4$, the corresponding ELGIs are

$$H(Q_1, Q_2) + H(Q_2, Q_3) + H(Q_3, Q_4) - H(Q_2) - H(Q_3) - H(Q_1, Q_4) \geq 0 \ , \quad (16)$$
$$H(Q_i, Q_j) + H(Q_j, Q_k) - H(Q_j) - H(Q_i, Q_k) \geq 0 \ , \quad (17)$$

where $i, j, k \in \{1, 2, 3, 4\}$ and $i < j < k$.

B Non-Markovian dynamics in a dissipative qubit system

In this section, we provide the background and model of the trapped dissipative qubit system used to detect non-Markovian effects via ELGIs. The system encodes a qubit in the Zeeman sublevels of the electronic states:

$$\begin{aligned} |0\rangle &= |4^2S_{1/2}, m_J = -1/2\rangle \, , \quad & |1\rangle &= |3^2D_{5/2}, m_J = +1/2\rangle \, , \\ |g\rangle &= |4^2S_{1/2}, m_J = +1/2\rangle \, , \quad & |e\rangle &= |4^2P_{3/2}, m_J = +3/2\rangle \, , \end{aligned} \quad (18)$$

where $|g\rangle$ serves as a tunable sink state. As proposed by Wang *et al.* [S2], the $|0\rangle \leftrightarrow |1\rangle$ transition is coherently driven by a 729-nm laser, while a controllable dissipation channel from $|1\rangle$ to $|g\rangle$ is implemented via a short-lived intermediate state $|e\rangle$ using an 854-nm laser.

The full dynamics of this four-level system is described by the Lindblad master equation

$$\frac{\mathrm{d}\hat{\rho}}{\mathrm{d}t} = -i[\hat{H}, \hat{\rho}] + \hat{L}\hat{\rho}\hat{L}^\dagger - \frac{1}{2}\{\hat{L}^\dagger\hat{L}, \hat{\rho}\}, \quad (19)$$

where the Hamiltonian takes the form

$$\hat{H} = E_0 |0\rangle \langle 0| + E_1 |1\rangle \langle 1| + E_e |e\rangle \langle e| + E_g |g\rangle \langle g| + \omega_0(|0\rangle \langle 1| + |1\rangle \langle 0|) + \omega(|1\rangle \langle e| + |e\rangle \langle 1|), \quad (20)$$

and the dissipative process is captured by the Lindblad jump operator

$$\hat{L} = \sqrt{\gamma} \ |g\rangle \langle e| \, . \quad (21)$$

In the regime where the intermediate state $|e\rangle$ decays much faster than the coherent couplings ($\gamma \gg \omega_0, \omega$), the population of $|e\rangle$ can be adiabatically eliminated, giving rise to an effective three-level model. The Hamiltonian restricted to the qubit subspace is

$$\hat{H}_{\text{eff}} = E_0 |0\rangle \langle 0| + E_1 |1\rangle \langle 1| + \omega_0(|0\rangle \langle 1| + |1\rangle \langle 0|), \quad (22)$$

and the effective decay from $|1\rangle$ to the sink state $|g\rangle$ is described by

$$\hat{L}_{\text{eff}} = \sqrt{\Gamma_{\text{eff}}} \ |g\rangle \langle 1| \, , \quad \Gamma_{\text{eff}} = \frac{|\omega|^2\gamma}{\Delta^2 + (\gamma/2)^2}, \quad (\Delta = E_e - E_1). \quad (23)$$

While the effective model provides an accurate description of the qubit dynamics in the fast-decay (Markovian) limit, deviations arise when the decay of $|e\rangle$ is slow. In such cases, the full four-level dynamics exhibits memory effects that cannot be fully captured by the simple Markovian effective model, leading to observable differences in temporal correlations and positive violations to classical Markovianity conditions.

Our experiment considers an initial state $\hat{\rho} = (|0\rangle \langle 0| + |1\rangle \langle 1|)/2$, which corresponds to the maximally mixed state in the qubit subspace. The first measurement is performed at $\omega t_1 = \pi/4$, which maximizes the expected violation. We adopt $n = 3$ equally spaced measurement times, with $t_{i+1} - t_i = \Delta t$, to evaluate the temporal correlations.

Numerical results indicate that for relatively low decay rates ($\gamma < 0.1\,\omega$), the system exhibits very small quantum (negative) violations of the ELGI under the Markov approximation, $|\mathcal{D}_{1,3}| \sim 10^{-4}$. In contrast, the non-Markovian dynamics of the full four-level system produces a significantly larger violation, $|\mathcal{D}_{1,3}| \sim 10^{-2}$. These results demonstrate that in this regime, where the quantum violation under the Markov approximation is negligible compared to the non-Markovian violation, the protocol can effectively detect and characterize the non-Markovian nature of the system dynamics.

C WKB Approximation Method for Deriving Wigner d-Matrix

In this section, we briefly review the Wenzel–Kramers–Brillouin approximation method [S3, S4, S5] (also known as the Liouville–Green method [S6, S7], especially in mathematical literatures) for approximately deriving the Wigner d-matrix elements. For better handling the behaviour of functions $d^i_{nm}(\beta)$ near the classical-quantum boundary, we are to follow the so-called *uniform* approximation method first established by Cherry [S8] and further discussed by Jeffreys [S9], Erdélyi [S10] and others, with slightly difference from the original WKB method.

C.1 Second-order ordinary differential equation for Wigner d-matrix

The WKB method is generally used to find approximate solutions to second-order ordinary differential equations (in physics, usually the Schrödinger equation). Wigner [S11, Chap. 19] noticed that the functions $D^j_{nm}(\alpha,\beta,\gamma)$ are the eigenfunctions of the symmetric top, and thus satisfy the Schrödinger equation [S12, S13]

$$\left(\frac{\partial^2}{\partial\beta^2}+\frac{\cos\beta}{\sin\beta}\frac{\partial}{\partial\beta}+\big(\frac{I_x}{I_z}+\frac{\cos^2\beta}{\sin^2\beta}\big)\frac{\partial^2}{\partial\gamma^2}+\frac{1}{\sin^2\beta}\frac{\partial^2}{\partial\alpha^2}-\frac{2\cos\theta}{\sin^2\theta}\frac{\partial^2}{\partial\alpha\partial\gamma}+\frac{2I_xE}{\hbar^2}\right)D^j_{nm}(\alpha,\beta,\gamma)=0\ , \tag{24}$$

where I_x and I_z denote for the moments of inertia of the top about the x- and z-axes (the axis of symmetry), respectively. Given that $D^j_{nm}(\alpha,\beta,\gamma)=e^{-im\alpha}d^j_{nm}(\beta)e^{-in\gamma}$, we obtain the differential equation satisfied by the Wigner d-matrix elements,

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}\beta^2}+\frac{\cos\beta}{\sin\beta}\frac{\mathrm{d}}{\mathrm{d}\beta}+j(j+1)-\frac{m^2-2mn\cos\beta+n^2}{\sin^2\beta}\right)d^j_{nm}(\beta)=0\ . \tag{25}$$

where the eigenvalues of energy E obtained in Ref. [S12, S13] have been substituted:

$$E_{j,n}=\frac{\hbar^2}{2I_x}\left(j(j+1)+\big(\frac{I_x}{I_z}-1\big)n^2\right). \tag{26}$$

For simplicity, we denote $J=j+1/2$, $\mu=m/J$, $\nu=n/J$ and assume $0<\beta<\pi$. The other elements of Wigner d-matrix can be derived with the following symmetry:

$$d^j_{m,n}(-\beta)=d^j_{n,m}(\beta)=(-1)^{m-n}d^j_{m,n}(\beta)\ . \tag{27}$$

Thus, the function $w(\beta)=\sqrt{\sin\beta}\cdot d^j_{nm}(\beta)$ obeys the differential equation

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}\beta^2}+\frac{J^2}{\sin^2\beta}\big(\sin^2\beta-\mu^2-\nu^2+2\mu\nu\cos\beta\big)+\frac{1}{4\sin^2\beta}\right)w(\beta)=0\ . \tag{28}$$

C.2 Uniform WKB method

Now we start to deal with eq. (28) following the *uniform* WKB method. The core of this method is to introduce an ancilla parameter ζ, which satisfies equation

$$\zeta\big(\frac{\mathrm{d}\zeta}{\mathrm{d}\beta}\big)^2=-\frac{J^2}{\sin^2\beta}R(\beta)\ ,\qquad \big(R(\beta)=\sin^2\beta-\mu^2-\nu^2+2\mu\nu\cos\beta\big) \tag{29}$$

and the corresponding ancilla function

$$h(\zeta)=\left|\frac{\mathrm{d}\zeta}{\mathrm{d}\beta}\right|^{1/2}\cdot w(\beta)=\sqrt{\frac{J}{\sin\beta}}\Big(\frac{R(\beta)}{-\zeta}\Big)^{1/4}\cdot w(\beta)\ , \tag{30}$$

Here, $R(\beta)$ is the discriminant of eq. (28), with its zeros $\beta_\pm=\arccos\big(\mu\nu\pm\sqrt{(1-\mu^2)(1-\nu^2)}\,\big)$ being the two turning points of the differential equation.

Our intention is to change the variable of eq. (28), thereby obtaining the *standard* form of the second-order differential equation, (cf. [S8, Lem. 1])

$$\frac{\mathrm{d}^2 h}{\mathrm{d}\zeta^2} = \left(\zeta + \left|\frac{\mathrm{d}\beta}{\mathrm{d}\zeta}\right|^{1/2} \frac{\mathrm{d}^2}{\mathrm{d}\zeta^2}\left|\frac{\mathrm{d}\zeta}{\mathrm{d}\beta}\right|^{1/2} + \frac{\zeta}{4J^2 R(\beta)}\right) h(\zeta) = \left(1 + \frac{1}{J^2}\cdot\frac{\delta(\beta)}{4R^3(\beta)}\right)\zeta \cdot h(\zeta) \ , \tag{31}$$

where

$$\delta(\beta) = \frac{5J^2R^3(\beta)}{4\zeta^3(\beta)} + \big(1 - \mu^4 - \nu^4 + 3\mu^2\nu^2 - 6\mu\nu\cos\beta - 2\mu\nu\cos^3\beta - (1 - 4\mu^2 - 4\nu^2 + \mu^2\nu^2)\cos^2\beta\big)\sin^2\beta \ , \tag{32}$$

is bounded and independent of J, as can be seen from eqs. (34) and (35) below. Thus, for J sufficiently large, we can approximately obtain

$$\frac{\mathrm{d}^2 h}{\mathrm{d}\zeta^2} = \zeta \cdot h(\zeta) \quad \rightarrow \quad h(\zeta) = C(\mu,\nu)\cdot \mathrm{Ai}(\zeta) \ , \tag{33}$$

where $\mathrm{Ai}(\zeta)$ represents for the Airy function, the coefficient $C(\mu,\nu)$ is determined through the initial conditions, and we rule out the other solutions with $\mathrm{Bi}(\zeta)$ components, since $h(\zeta)$ should be bounded.

Clearly, ζ is required to have an one-to-one correspondence with β, and $\zeta'(\beta) \neq 0$ for any $\beta \in (0,\pi)$, which is actually impossible as $\zeta(\beta_\pm) = 0$ according to eq. (29). Neverthelss, by allowing $\zeta(\beta)$ to take the value of zero at only one of the two turning points $\beta_\pm$, we obtain a valid solution $\zeta(\beta)$ covering the interval $(0, \beta_+)$ or (β_-, π), respectively.

$$\zeta(\beta) = \begin{cases} -\left(\dfrac{3J}{2}S(\beta)\right)^{2/3} , & R(\beta) > 0 \ ; \\ \left(-\dfrac{3J}{2}S(\beta)\right)^{2/3} , & R(\beta) < 0 \ . \end{cases} \tag{34}$$

Here, we have introduced a function $S(\beta)$ for simplicity, corresponding to the classical action of the trajectory connecting the initial and final state (cf. [S14, Sec. 2]), namely

$$S_\pm(\beta) = \begin{cases} \pm\arctan\dfrac{\cos\beta - \mu\nu}{\sqrt{R(\beta)}} \mp \dfrac{1}{2}(\mu+\nu)\arctan\dfrac{(\cos\beta-\mu\nu)(1+\mu\nu)+(1-\mu^2)(1-\nu^2)}{(\mu+\nu)\sqrt{R(\beta)}} \\ \qquad \pm\dfrac{1}{2}(\mu-\nu)\arctan\dfrac{(\mu\nu-\cos\beta)(1-\mu\nu)+(1-\mu^2)(1-\nu^2)}{(\mu-\nu)\sqrt{R(\beta)}} - \left|\mu+\nu\right|\dfrac{\pi}{4} - \left|\mu-\nu\right|\dfrac{\pi}{4} + \dfrac{\pi}{2} \ , & \big(R(\beta)>0\big) \\ \mp\operatorname{arcoth}\dfrac{\cos\beta-\mu\nu}{\sqrt{-R(\beta)}} \pm \dfrac{1}{2}(\mu+\nu)\operatorname{arcoth}\dfrac{(\cos\beta-\mu\nu)(1+\mu\nu)+(1-\mu^2)(1-\nu^2)}{(\mu+\nu)\sqrt{-R(\beta)}} \\ \qquad \mp\dfrac{1}{2}(\mu-\nu)\operatorname{arcoth}\dfrac{(\mu\nu-\cos\beta)(1-\mu\nu)+(1-\mu^2)(1-\nu^2)}{(\mu-\nu)\sqrt{-R(\beta)}} \ , & \big(R(\beta)<0\big) \end{cases} \tag{35}$$

derived from

$$\frac{\partial S_\pm(\beta)}{\partial\beta} = \mp\frac{\sqrt{|R(\beta)|}}{\sin\beta}, \quad S_\pm(\beta_\pm) = 0 \ , \tag{36}$$

where the subscript denotes which the turning point we have choose.

After substitution, we obtain the approximate expression

$$d^j_{nm}(\beta) \approx \frac{C(\mu,\nu)\cdot\mathrm{Ai}(\zeta)}{\sqrt{J}}\left(\frac{-\zeta}{R(\beta)}\right)^{1/4}. \tag{37}$$

C.3 Boundary conditions of the Wigner *d*-matrix

Now, we are only one step away from a complete approximate expression for $d^j_{nm}(\beta)$, which is to determine the coefficients $C(\mu,\nu)$. Given that for any μ,ν and j, there exists a sufficiently small β to make our approximation valid, we could derive the expression of $C(\mu,\nu)$ by comparing the coefficient of the leading-order term of $d^j_{nm}(\beta)$ when $\beta\to 0$. We start with Wigner's series expansion for $d^j_{nm}(\beta)$ in Ref. [S15, Chap. 15], namely

$$d^j_{nm}(\beta)=\sqrt{(j+m)!(j-m)!(j+n)!(j-n)!}\sum_{s=s_{\min}}^{s_{\max}}\frac{(-1)^{n-m+s}\big(\cos(\beta/2)\big)^{2j+m-n-2s}\big(\sin(\beta/2)\big)^{n-m+2s}}{(j+m-s)!s!(n-m+s)!(j-n-s)!}\ , \tag{38}$$

where $s_{\min}=\max\{0,\,m-n\}$ and $s_{\max}=\min\{j+m,\,j-n\}$, thus we obtain

$$d^j_{nm}(\beta)=\sqrt{\frac{(j+a)!(j-b)!}{(j+b)!(j-a)!}}\frac{(-1)^{a-b}}{(a-b)!}\cdot\big(\frac{\beta}{2}\big)^{a-b}+O(\beta^{a-b+2})\ . \tag{39}$$

where $a=\max(m,n), b=\min(m,n)$.

Given that $S(\beta)\sim O(\ln\beta)$ as $\beta\to 0$ as can be seen from eq. (36), we use the asymptotic formulae for Airy function as the variable tends to be infinity:

$$\mathrm{Ai}(x)=\begin{cases}\dfrac{1}{2\sqrt{\pi}x^{1/4}}\exp\Big(-\dfrac{2}{3}x^{3/2}\Big)\cdot\Big(1-\dfrac{5}{48}\dfrac{1}{x^{3/2}}+O(x^{-3})\Big)\,, & x\to+\infty,\\ \dfrac{1}{\sqrt{\pi}|x|^{1/4}}\Big(\sin\big(\dfrac{2}{3}|x|^{3/2}+\dfrac{\pi}{4}\big)-\cos\big(\dfrac{2}{3}|x|^{3/2}+\dfrac{\pi}{4}\big)\cdot\dfrac{5}{36}\dfrac{1}{x^{3/2}}+O(x^{-3})\Big)\,, & x\to-\infty\ .\end{cases} \tag{40}$$

to obtain (with $R(\beta)<0$ for sufficiently small β)

$$d^j_{nm}(\beta)=\frac{C(\mu,\nu)}{\sqrt{4\pi J}}\Big(\frac{-1}{R(\beta)}\Big)^{1/4}e^{JS(\beta)}\cdot\Big(1+O\big(\frac{1}{\ln\beta}\big)\Big) \tag{41}$$

$$=\frac{C(\mu,\nu)}{\sqrt{4\pi(n-m)}}\cdot e^{JS(\beta)}\cdot\Big(1+O\big(\frac{1}{\ln\beta}\big)\Big)\ . \tag{42}$$

We take the case of where $m<n$ as an example, in which we obtain

$$C(\mu,\nu)=\lim_{\beta\to 0}\sqrt{\frac{(j+n)!(j-m)!}{(j+m)!(j-n)!}}\frac{(-1)^{n-m}}{(n-m)!}\cdot\sqrt{4\pi(n-m)}\cdot\big(\frac{\beta}{2}\big)^{n-m}\cdot\exp\big(-JS(\beta)\big) \tag{43}$$

$$=\sqrt{\frac{(j+n)!(j-m)!}{(j+m)!(j-n)!}}\frac{(-1)^{n-m}}{(n-m)!}\cdot\sqrt{4\pi(n-m)}\cdot(\mu-\nu)^{n-m}\frac{(1+\mu)^{\frac{J+m}{2}}(1-\nu)^{\frac{J-n}{2}}}{(1-\mu)^{\frac{J-m}{2}}(1+\nu)^{\frac{J+n}{2}}}\ , \tag{44}$$

which can be simplified with Stirling's formula to

$$C(\mu,\nu)=(-1)^{n-m}\sqrt{2}\cdot\sqrt{\frac{F(j+n)F(j-m)}{F(j+m)F(j-n)}}\cdot\frac{G(j+n)G(j-m)}{G(j+m)G(j-n)}\ , \tag{45}$$

where

$$F(n)=\frac{n!}{\sqrt{2\pi n}}\Big(\frac{e}{n}\Big)^n=1+\frac{1}{12}n+O(n^{-2}),\quad G(n)=\sqrt{\Big(1+\frac{1}{2n}\Big)^n}=1+\frac{3}{16n}+O(n^{-2})\ . \tag{46}$$

Combined with the symmetry eq. (27) of Wigner d-matrix, for sufficiently large j, we finally obtain

$$d^j_{nm}(\beta)=(-1)^{\max\{0,\,n-m\}}\sqrt{\frac{2}{J}}\cdot\mathrm{Ai}(\zeta)\Big(\frac{-\zeta}{R(\beta)}\Big)^{1/4}. \tag{47}$$

D Asymptotic Analysis on Entropy of Wigner d-matrix

In this section, we turn to calculate the entropy of Wigner d-matrix, which is defined as

$$H_j(\beta):=-\frac{1}{2j+1}\sum_{m,n=-j}^{j}|d^j_{nm}(\beta)|^2\ln|d^j_{nm}(\beta)|^2\ . \tag{48}$$

With the asympotic formulae eq. (40), we have

$$|d^j_{nm}(\beta)| = \begin{cases} \dfrac{2\sin^2(JS+\pi/4)}{\pi J\sqrt{R(\mu,\nu,\beta)}}\Big(1+O(J^{-1})\Big), & R(\beta)>0\ , \\ \dfrac{\exp(2JS)}{2\pi J\sqrt{-R(\mu,\nu,\beta)}}\Big(1+O(J^{-1})\Big), & R(\beta)<0\ . \end{cases} \tag{49}$$

It's worth noting that the boundary $R(\mu,\nu,\beta)=0$ forms an ellipse in μ–ν plane. After affine transformation

$$\begin{cases} x = (\mu+\nu)\sin\dfrac{\beta}{2}\ , \\ y = (\mu-\nu)\cos\dfrac{\beta}{2}\ , \end{cases} \tag{50}$$

and conversion to polar coordinates, we obtain

$$R(r,\beta) = \sin^2\beta - r^2\ . \tag{51}$$

According to eq. (49), we consider dividing the sum eq. (48) into three parts: the classically allowed region (S_{allowed}), the classically forbidden region ($S_{\text{forbidden}}$) and a boundary layer (S_{boundary}) of width ε.

$$H_j(\beta) = S_{\text{allowed}} + S_{\text{boundary}} + S_{\text{forbidden}} \tag{52}$$

$$= -\frac{1}{2J}\sum_{r\le\sin\beta-\varepsilon}|d^j_{nm}|^2\ln|d^j_{nm}|^2 - \frac{1}{2J}\sum_{|r-\sin\beta|<\varepsilon}|d^j_{nm}|^2\ln|d^j_{nm}|^2 - \frac{1}{2J}\sum_{r\ge\sin\beta+\varepsilon}|d^j_{nm}|^2\ln|d^j_{nm}|^2\ . \tag{53}$$

We are to substitute the two asymptotic expressions from into $S_{\text{forbidden}}$ and S_{allowed} respectively, which requires the parameter ε to satisfy $\varepsilon\cdot|\partial\zeta/\partial r|\sim O(1)$. From eq. (35) we obtain ($\varepsilon>0$)

$$S(r,\beta) = -\frac{4\sqrt{2}}{3(\cos\beta-\cos 2\theta)\sqrt{\sin\beta}}\cdot\varepsilon^{3/2} + O(\varepsilon^{5/2})\ , \tag{54}$$

from which $\varepsilon\sim O(J^{-2/3})$. Thus $\varepsilon(J)$ can be set to be an infinitesimal as $J\to+\infty$, which ensures that

$$\lim_{J\to+\infty}\frac{S_{\text{boundary}}}{S_{\text{allowed}}} = 0\ . \tag{55}$$

Here, we implicitly assume that the approximate solution given by eq. (47) remains valid in the boundary region. Although it will be shown subsequently that this assumption does not hold in general, this limitation does not compromise the validity of eq. (55) itself.

Furthermore, given that the Airy function exhibits exponential decay in the classically forbidden region (as demonstrated in eq. (40)), it follows that

$$\lim_{J\to+\infty}\frac{S_{\text{forbidden}}}{S_{\text{allowed}}} = 0\ . \tag{56}$$

Therefore, the summation can be simplified into

$$H_j(\beta) \approx -\frac{1}{2J}\sum_{r<\sin\beta}|d^j_{nm}(\beta)|^2\ln|d^j_{nm}(\beta)|^2 = -\frac{1}{2J}\sum_{r<\sin\beta}\frac{2\sin^2(JS+\pi/4)}{\pi J\sqrt{R(r,\beta)}}\ln\frac{2\sin^2(JS+\pi/4)}{\pi J\sqrt{R(r,\beta)}}\ . \tag{57}$$

D.1 Calculation of Leading and Sub-Leading Order Terms

Considering that the function $\sin^2(\phi)\ln\sin^2(\phi)$ is π-periodic, which can be expressed as the Fourier series expansion

$$\sin^2(\phi)\ln\sin^2(\phi) = \frac{1-2\ln 2}{2} + \big(\ln 2 - \frac{3}{4}\big)\cos 2\phi + \sum_{k=1}^{\infty}\left(\frac{\cos 4k\phi}{8k^3-2k} + \frac{\cos(4k+2)\phi}{8k^3+12k^2+4k}\right) . \tag{58}$$

After substitution, we obtain

$$H_j(\beta) \approx \frac{1}{2J}\sum_{r<\sin\beta}|d^j_{nm}(\beta)|^2\big(\ln(2\pi J)-\frac{3}{2}\big) + \sum_{r<\sin\beta}\frac{1+\ln\sqrt{R(r,\beta)}}{2\pi J^2\sqrt{R(r,\beta)}} + \sum_{r<\sin\beta}\frac{\ln\sqrt{R(r,\beta)}}{2\pi J^2\sqrt{R(r,\beta)}}\cdot\sin(2JS)$$
$$- \sum_{r<\sin\beta}\sum_{k=1}^{\infty}\frac{2\cdot(-1)^k}{2\pi J^2\sqrt{R(r,\beta)}}\left(\frac{\cos(4kJS)}{8k^3-2k} - \frac{\sin\big((4k+2)JS\big)}{8k^3+12k^2+4k}\right) . \tag{59}$$

Now we start to calculate each term in eq. (59), where the first one can be obtained using the normalization condition of the Wigner d-matrix, namely

$$\frac{1}{2J}\sum_{r<\sin\beta}|d^j_{nm}(\beta)|^2\big(\ln(2\pi J)-\frac{3}{2}\big) = \ln(2\pi J) - \frac{3}{2} . \tag{60}$$

And the second summation term can be transformed into an integral over continuous variables, namely

$$\sum_{r<\sin\beta}\frac{1+\ln\sqrt{R(r,\beta)}}{2\pi J^2\sqrt{R(r,\beta)}} = \iint_{r<\sin\beta}\frac{1+\ln\sqrt{R(r,\beta)}}{2\pi J^2\sqrt{R(r,\beta)}}\,\mathrm{d}m\,\mathrm{d}n = \int_0^{\sin\beta}\frac{1+\ln\sqrt{R(r,\beta)}}{2\pi J^2\sqrt{R(r,\beta)}}\cdot\frac{2\pi r\cdot J^2}{\sin\beta}\,\mathrm{d}r = \ln(\sin\beta) - 1 . \tag{61}$$

The most challenging part of this section is managing the summation involving harmonic function factors. Taking the $\sin(2JS)$ term as an example, for the grid point (μ,ν) adjacent to the grid point (μ_0,ν_0), we obtain

$$\sin\big(2JS(\mu,\nu)\big) = \sin\Big(2JS(\mu_0,\nu_0) + \frac{\partial S}{\partial\mu}\Big|_{(\mu_0,\nu_0)}\cdot 2J(\mu-\mu_0) + \frac{\partial S}{\partial\nu}\Big|_{(\mu_0,\nu_0)}\cdot 2J(\nu-\nu_0) + o(1)\Big) , \tag{62}$$

where $2J(\mu-\mu_0), 2J(\nu-\nu_0) = \pm 2$, thus the summation involving harmonic functions cannot be converted into an integral over continous variables as $J\to+\infty$.

Despite lack of means for accurate calculation, we can estimate the magnitude of the harmonic term by considering a coarse-grained grid dividing the $\mu-\nu$ plane into a number of squares, each of which contains $(2J^\delta+1)^2$ grid points, with its center denoted as (μ_0,ν_0). For each square contained in the classical allowed region, we obtain

$$\sum_{r,s=-J^\delta}^{J^\delta}\frac{\ln\sqrt{R(\mu_0+r/J,\nu_0+s/J)}}{2\pi J^2\sqrt{R(\mu_0+r/J,\nu_0+s/J)}}\cdot\sin\big(2JS(\mu_0+r/J,\nu_0+s/J)\big)$$
$$= \frac{\ln\sqrt{R(\mu_0,\nu_0)}}{2\pi J^2\sqrt{R(\mu_0,\nu_0)}}\cdot\sum_{r,s=-J^\delta}^{J^\delta}\left(\sin\Big(2JS(\mu_0,\nu_0) + 2r\cdot\frac{\partial S}{\partial\mu}\Big|_{(\mu_0,\nu_0)} + 2s\cdot\frac{\partial S}{\partial\nu}\Big|_{(\mu_0,\nu_0)}\Big) + O(J^{\delta-1})\right) + O(J^{\delta-2}) \tag{63}$$
$$= \frac{\ln\sqrt{R(\mu_0,\nu_0)}}{2\pi J^2\sqrt{R(\mu_0,\nu_0)}}\cdot\frac{\sin(2JS)\sin\big((2J^\delta+1)(\partial S/\partial\mu)\big)\sin\big((2J^\delta+1)(\partial S/\partial\nu)\big)}{\sin(\partial S/\partial\mu)\sin(\partial S/\partial\nu)}\Big|_{(\mu_0,\nu_0)} + O(J^{3\delta-2}) . \tag{64}$$

Given that $0 \leq \partial S/\partial\mu, \partial S/\partial\nu \leq \pi$, the leading term in the summation over all squares can be bounded as

$$\sum_{\substack{(\mu_0,\nu_0)\\ r<\sin\beta}} \left|\frac{\ln\sqrt{R(\mu_0,\nu_0)}}{2\pi J^2\sqrt{R(\mu_0,\nu_0)}}\right| \cdot \frac{\sin(2JS)\sin\left((2J^\delta+1)(\partial S/\partial\mu)\right)\sin\left((2J^\delta+1)(\partial S/\partial\nu)\right)}{\sin(\partial S/\partial\mu)\sin(\partial S/\partial\nu)}\Big|_{(\mu_0,\nu_0)}$$
$$< \sum_{\substack{(\mu_0,\nu_0)\\ r<\sin\beta}} \left|\frac{\ln\sqrt{R(\mu_0,\nu_0)}}{2\pi J^2\sqrt{R(\mu_0,\nu_0)}}\right| \cdot \frac{1}{\sin(\partial S/\partial\mu)\sin(\partial S/\partial\nu)}\Big|_{(\mu_0,\nu_0)} \xrightarrow{J\to\infty} -\int \frac{\ln\sqrt{R(\mu,\nu)}}{4\pi\sqrt{R(\mu,\nu)}}\frac{J^{1-2\delta}\,\mathrm{d}\mu\,\mathrm{d}\nu}{\sin(\partial S/\partial\mu)\sin(\partial S/\partial\nu)}\,, \tag{65}$$

which is of order $O(J^{1-2\delta})$, while the remainder is $O(J^\delta)$ after summation. Accordingly, setting $\delta = 1/3$ yields an optimal estimate, so that the $\sin(2JS)$ term is bounded by $O(J^{1/3})$. Similar analyses can be applied to other harmonic terms.

D.2 Analysis of Approximate Solutions in Boundary Regions

At the end of this section, we verify the validity of the approximate solution eq. (47) in the boundary layer. In this case $R(\beta) \to 0$, thereby the discarded term in eq. (31) cannot be ignored, as it becomes

$$\frac{1}{J^2}\cdot\frac{\delta(\beta)}{4R^3(\beta)} \approx \frac{1}{J^2x^3}\cdot\frac{\delta(\beta)}{32\sin^3\beta} \to \infty \tag{66}$$

with $x = \sin\beta - r \sim o(J^{-2/3})$. We consider the Taylor series near $\zeta = 0$ and retain the first term, thus obtain

$$\frac{\mathrm{d}^2h}{\mathrm{d}\zeta^2} = \left(1+\frac{1}{J^2}\cdot\frac{\delta(\beta)}{4R^3(\beta)}\right)\zeta\cdot h(\zeta) = \left(1+\frac{3(5+\cos 2\beta_\pm - 6\cos\beta_\pm\cos 2\phi)}{64J^{2/3}\zeta^2\left((\cos\beta_\pm-\cos 2\phi)^2\sin\beta_\pm\right)^{2/3}} + O\big(\frac{1}{J^{4/3}\zeta}\big)\right)\zeta\cdot h(\zeta)\,, \tag{67}$$

where $\beta_\pm$ is the turning point in consideration, and

$$\phi = \arctan\left(\frac{1}{\tan(\beta_\pm/2)}\frac{\mu-\nu}{\mu+\nu}\right)\,. \tag{68}$$

From eq. (67), one could find the uniform approximation method invalidates only if $\zeta \sim O(J^{-1/3})$, or equivalently $\varepsilon \sim O(J^{-1})$. Hence, it is reasonable to take the approximate solution eq. (33) when calculating S_{boundary} in the boundary layer.

Combining the previous analysis, we obtain the asymptotic expression for the entropy of the Wigner d-matrix in the macroscopic limit $j \to \infty$:

$$H_j(\beta) = \ln(2\pi J) + \ln|\sin\beta| - \frac{5}{2} + o(1)\,. \tag{69}$$

E Analysis near the singularity $\beta \to n\pi$ ($n \in \mathbb{Z}$)

According to the previous discussion, the asymptotic expression in eq. (69) diverges as $\beta \to n\pi$ for $n \in \mathbb{Z}$. Such divergence arises because the asymptotic analysis is valid only in the regime where $S_{\text{allowed}} \gg S_{\text{boundary}}$, or equivalently, $|\beta - n\pi| \gg J^{-2/3}$. As β approaches the singularities, the classically allowed region contracts while the boundary region becomes increasingly dominant. Consequently, the WKB approximation and the resulting asymptotic expression eq. (69) breaks down.

Owing to the inherent symmetry of $d^j_{nm}(\beta)$, it is sufficient to consider the limit $\beta \to 0$, without loss of generality. In such regime, an alternative analytical approach is to employ Wigner's series expansion for $d^j_{nm}(\beta)$, as presented in Ref. [S15, Chap. 15], and retaining only the leading-order term, from which we obtain

$$d^j_{nm}(\beta) = \sqrt{\frac{(j+a)!(j-b)!}{(j+b)!(j-a)!}}\frac{(-1)^{a-b}}{(a-b)!}\cdot\big(\frac{\beta}{2}\big)^{a-b} + O(\beta^{a-b+2})\,. \tag{70}$$

where $a = \max(m,n), b = \min(m,n)$.

Consider the summation of $|d^j_{nm}(\beta)|^2 \ln |d^j_{nm}(\beta)|^2$, grouped according to the index difference $k = |m - n|$. For $k = 0$, we obtain

$$S_0 := -\frac{1}{2j+1} \sum_{n=-j}^{j} |d^j_{nn}(\beta)|^2 \ln |d^j_{nn}(\beta)|^2 \tag{71}$$

$$= -\frac{1}{2j+1} \sum_{n=-j}^{j} \Big(1 - \frac{\beta^2}{2}\big(j(j+1) - n^2\big)\Big) \ln \Big(1 - \frac{\beta^2}{2}\big(j(j+1) - n^2\big)\Big) \tag{72}$$

$$= \frac{\beta^2}{2(2j+1)} \sum_{n=-j}^{j} \big(j(j+1) - n^2\big) + O(\beta^2) = \frac{\beta^2}{3}\big(J^2 - \frac{1}{4}\big) + O(\beta^2) \ . \tag{73}$$

For $k \neq 0$, we obtain

$$S_k := -\frac{1}{2j+1} \sum_{n=-j}^{j-k} |d^j_{n,n+k}(\beta)|^2 \ln |d^j_{n,n+k}(\beta)|^2 - \frac{1}{2j+1} \sum_{n=-j+k}^{j} |d^j_{n,n-k}(\beta)|^2 \ln |d^j_{n,n-k}(\beta)|^2$$

$$= -\sum_{n=-j}^{j-k} \frac{2}{2j+1} \Big(\frac{\beta^{2k}}{2^{2k}(k!)^2} \frac{(j+n+k)!}{(j+n)!} \frac{(j-n)!}{(j-n-k)!}\Big) \ln \Big(\frac{\beta^{2k}}{2^{2k}(k!)^2} \frac{(j+n+k)!}{(j+n)!} \frac{(j-n)!}{(j-n-k)!}\Big) + O(\beta^2) \ . \tag{74}$$

Applying the mean value inequality, and defining $\beta = J^{-1-\delta}$ ($\delta > 0$) for simplicity, we obtain

$$\frac{\beta^{2k}}{2^{2k}(k!)^2} \frac{(j+n+k)!}{(j+n)!} \frac{(j-n)!}{(j-n-k)!} \leq \frac{(j+1/2)^{2k}\beta^{2k}}{2^{2k}(k!)^2} = \frac{J^{-2k\delta}}{2^{2k}(k!)^2} \leq \frac{1}{4} \ , \tag{75}$$

from which an upper bound for S_k follows:

$$S_k \leq -\sum_{n=-j}^{j-k} \frac{2}{2j+1} \frac{J^{-2k\delta}}{2^{2k}(k!)^2} \ln \frac{J^{-2k\delta}}{2^{2k}(k!)^2} + O(\beta^2) = -\frac{(2J-k)}{J} \frac{J^{-2k\delta}}{2^{2k}(k!)^2} \ln \frac{J^{-2k\delta}}{2^{2k}(k!)^2} + O(\beta^2) \ . \tag{76}$$

Therefore,

$$H_j(\beta) = \sum_{k=0}^{2j} S_k \leq \frac{\beta^2}{3}\big(J^2 - \frac{1}{4}\big) - \sum_{k=1}^{2J-1} \frac{(2J-k)}{J} \frac{J^{-2k\delta}}{2^{2k}(k!)^2} \ln \frac{J^{-2k\delta}}{2^{2k}(k!)^2} + O(\beta^2) \tag{77}$$

$$\leq \frac{\beta^2}{3}\big(J^2 - \frac{1}{4}\big) - \sum_{k=1}^{\infty} 2 \cdot \frac{J^{-2k\delta}}{2^{2k}(k!)^2} \ln \frac{J^{-2k\delta}}{2^{2k}(k!)^2} + O(\beta^2) \tag{78}$$

$$\leq \frac{\beta^2}{3}\big(J^2 - \frac{1}{4}\big) - \sum_{k=1}^{\infty} 2\Big(\frac{J^{-k\delta}}{2^{k+1}k!} + \frac{J^{-2k\delta}}{2^{2k}(k!)^2}\Big) + O(\beta^2) = \frac{J^{-\delta}}{4} + O(J^{-2\delta}) \ , \tag{79}$$

from which it follows that the leading order of $H_j(\beta)$ does not exceed $O(1)$.

F Ideal negative measurements for higher-order temporal correlations

Ideal negative measurement (INM) protocols discussed in the main text can be naturally extended to measure multi-time joint probabilities. In this section, we focus on the case of three-time joint probabilities for illustration. Each subsection first briefly reviews the implementation for the two-time senario, and then outlines a direct generalization to the three-time scenario. These examples are given for clarity; the same principles apply to correlations of arbitrary order.

F.1 Ancilla-assisted schemes

The invasiveness of quantum measurements arises from the fact that projective measurements inevitably collapse the system state onto an eigenstate associated with the measurement outcome. Thus, unless the density matrix happens to be diagonal in the measurement basis at the time of measurement, the act of measurement will necessarily disturb the system state. A viable strategy to mitigate such disturbance is to avoid projective readout at intermediate times and instead couple the system to an ancilla that becomes entangled with the system in the measurement basis.

Following the approach proposed by Katiyar *et al.* [S16], consider measuring a pure stat

$$|\psi_{\mathrm{S}}\rangle = c_0 |0_{\mathrm{S}}\rangle + c_1 |1_{\mathrm{S}}\rangle + c_2 |2_{\mathrm{S}}\rangle , \tag{80}$$

with respect to the projector $P_0 = |0\rangle\langle 0|$. We introduce an ancillary qubit initially prepared in $|\psi_A\rangle = |0_A\rangle$. At time t_i, we apply to the composite system the controlled gate

$$\mathrm{CG}_0 = |0_{\mathrm{S}}\rangle\langle 0_{\mathrm{S}}| \otimes I_A + |1_{\mathrm{S}}\rangle\langle 1_{\mathrm{S}}| \otimes X_A + |2_{\mathrm{S}}\rangle\langle 2_{\mathrm{S}}| \otimes X_A, \tag{81}$$

which transforms the joint state into

$$|\psi_{SA}\rangle = c_0 |0_{\mathrm{S}}, 0_A\rangle + c_1 |1_{\mathrm{S}}, 1_A\rangle + c_2 |2_{\mathrm{S}}, 1_A\rangle . \tag{82}$$

Hence the ancilla records information about the system's state at time t_i without projecting the system itself, permitting all physical measurements to be deferred to the final time. A joint measurement on the system andancilla then yields the correct joint probabilities.

This construction generalizes straightforwardly to multi-time joint probabilities. For example, to measure the probability $p(q_1 = 0, q_2 = 0, q_3 = 0)$, one applies at times t_1 and t_2 the controlled gates

$$\mathrm{CG}_0^{(1)} = |0_{\mathrm{S}}\rangle\langle 0_{\mathrm{S}}| \otimes I_{\mathrm{A1}} \otimes I_{\mathrm{A2}} + |1_{\mathrm{S}}\rangle\langle 1_{\mathrm{S}}| \otimes X_{\mathrm{A1}} \otimes I_{\mathrm{A2}} + |2_{\mathrm{S}}\rangle\langle 2_{\mathrm{S}}| \otimes X_{\mathrm{A1}} \otimes I_{\mathrm{A2}}, \tag{83}$$

$$\mathrm{CG}_0^{(2)} = |0_{\mathrm{S}}\rangle\langle 0_{\mathrm{S}}| \otimes I_{\mathrm{A1}} \otimes I_{\mathrm{A2}} + |1_{\mathrm{S}}\rangle\langle 1_{\mathrm{S}}| \otimes I_{\mathrm{A1}} \otimes X_{\mathrm{A2}} + |2_{\mathrm{S}}\rangle\langle 2_{\mathrm{S}}| \otimes I_{\mathrm{A1}} \otimes X_{\mathrm{A2}}, \tag{84}$$

and then performs at time t_3 the projective measurement

$$\Pi_{000} = |0_{\mathrm{S}}\rangle\langle 0_{\mathrm{S}}| \otimes |0_{\mathrm{A1}}\rangle\langle 0_{\mathrm{A1}}| \otimes |0_{\mathrm{A2}}\rangle\langle 0_{\mathrm{A2}}| . \tag{85}$$

This procedure yields the correct three-time probability distribution.

The scheme above addresses dichotomic measurements. For multi-outcome sharp measurements one can introduce controlled gates of the form (omitting the identity action on irrelevant subspaces for simplicity)

$$\mathrm{CG}^{(i)} = |0_{\mathrm{S}}\rangle\langle 0_{\mathrm{S}}| \otimes |0_{\mathrm{A}i}\rangle\langle 0_{\mathrm{A}i}| + |1_{\mathrm{S}}\rangle\langle 1_{\mathrm{S}}| \otimes |1_{\mathrm{A}i}\rangle\langle 0_{\mathrm{A}i}| + |2_{\mathrm{S}}\rangle\langle 2_{\mathrm{S}}| \otimes |2_{\mathrm{A}i}\rangle\langle 0_{\mathrm{A}i}| + \cdots . \tag{86}$$

One applies these controlled gates sequentially at the measurement times t_i, and finally at time t_n performs the projective measurement

$$\Pi_{q^{(1)}q^{(2)}\cdots q^{(n)}} = |q_{\mathrm{S}}^{(n)}\rangle\langle q_{\mathrm{S}}^{(n)}| \otimes |q_{\mathrm{A1}}^{(1)}\rangle\langle q_{\mathrm{A1}}^{(1)}| \otimes |q_{\mathrm{A2}}^{(2)}\rangle\langle q_{\mathrm{A2}}^{(2)}| \otimes \cdots . \tag{87}$$

In principle, the observed statistics then directly yield the joint distribution $p(q^{(1)}, q^{(2)}, \ldots, q^{(n)})$.

F.2 Interferometric null-result schemes

A conceptual concern regarding ancilla-assisted schemes is that their claimed non-invasiveness relies on accepting the validity of quantum mechanics, at least insofar as the quantum description of the controlled gates is accurate. Indeed, any experimental protocol that purports to be non-invasive must invoke some underlying physical theory to justify this claim; hence, non-invasiveness can only be assumed based on auxiliary assumptions about the validity of that theory. It is therefore impossible to devise an experimental procedure that is unconditionally non-invasive. Leggett [S17] emphasized this point and further argued that adopting locality as an additional assumption within a classical worldview is a relatively modest and acceptable assumption.

Concretely, in a Mach–Zehnder interferometer, monitoring one arm and observing a non-click event allows the experimenter to infer that the particle traveled through the other arm; under the locality assumption, this inference implies that the system's state remained undisturbed. Within a classical worldview, where measurements are assumed a priori to be local, such a null-result (non-detection) measurement is naturally considered non-invasive.

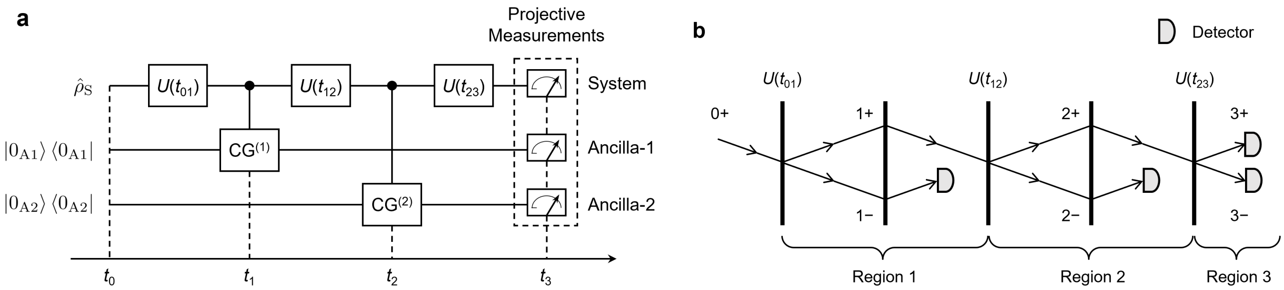

Fig. S1 Schematic representation of the ideal negative measurement protocols. (a) Measurement of three-time joint probabilities via ancilla-assisted schemes. Ancilla-1 and Ancilla-2 are auxiliary systems with the same dimension as the system, both initialized in the state $|0_{Ai}\rangle\langle 0_{Ai}|$. $CG^{(1)}$ and $CG^{(2)}$ denote the controlled quantum gates defined in eq. (86), which implement unitary transformations on the ancilla conditioned on the system state. Projective measurements at intermediate time points are replaced by controlled gates, with a single projective readout performed at the final time t_3. (b) Measurement of three-time joint probabilities via interferometric null-result schemes. The depicted setup directly measures only the parameter $N_{1+2+3\pm}$; the remaining six parameters $N_{1+2-3\pm}$, $N_{1-2+3\pm}$, and $N_{1-2-3\pm}$ are obtained by switching the detector in region 1 and 2.

As a concrete example, we extend the scheme of Kreuzgruber *et al.* [S18] to the case of three measurement times, as illustrated in fig. S1b. We focus on the particle or light intensity detected in Region 3, because any particles detected there must have bypassed the detectors in Region 1 and Region 2. The corresponding paths can therefore be recorded as $(1+, 2+, 3\pm)$, and the counts of the two detectors are denoted as $N_{1+2+3\pm}$. By switching the detector in Region 1 between the 1+ and 1− paths, and the detector in Region 2 between the 2+ and 2− paths, the remaining six measurement outcomes, $N_{1-2+3\pm}, N_{1+2-3\pm}, N_{1-2-3\pm}$, can be obtained. The three-time joint probability distribution is thus given by

$$p(1x, 2y, 3z) = \frac{N_{1x,2y,3z}}{\sum_{i,j,k=\pm} N_{1i,2j,3k}} \qquad (x, y, z = \pm) \tag{88}$$

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