## Work Fluctuations in Slow Processes: Quantum Signatures and Optimal Control

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An important result in classical stochastic thermodynamics is the work fluctuation-dissipation relation (FDR), which states that the dissipated work done along a slow process is proportional to the resulting work fluctuations. We show that slowly driven quantum systems violate this FDR whenever quantum coherence is generated along the protocol, and we derive a quantum generalization of the work FDR. The additional quantum terms in the FDR are found to lead to a non-Gaussian work distribution. Fundamentally, our result shows that quantum fluctuations prohibit finding slow protocols that minimize both dissipation and fluctuations simultaneously, in contrast to classical slow processes. Instead, we develop a quantum geometric framework to find processes with an optimal trade-off between the two quantities.

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Thermodynamics traditionally deals with macroscopic systems at thermal equilibrium, and its laws relate averages of quantities such as work and heat. When bringing the theory to the microscale, fluctuations become significant and can no longer be neglected with respect to average quantities. As a consequence, a stochastic description of thermodynamic processes is needed, which has triggered enormous attention to the understanding of work (and heat) fluctuations [1–4]. In the regime of slow but finite-time classical processes, work fluctuations are governed by a single relation, known as the work fluctuation-dissipation relation (FDR) [5–8]:

$$W_{\rm diss} = \frac{1}{2} \beta \sigma_w^2. \tag{1}$$

Here,  $\sigma_w^2 \equiv \langle w^2 \rangle - \langle w \rangle^2$  is the variance of the work distribution P(w) and  $W_{\text{diss}} \equiv \langle w \rangle - \Delta F \ge 0$  the average dissipated work along the protocol, i.e., the difference between average work done and the change of equilibrium free energy  $\Delta F$ , which is always non-negative due to the second law, and  $\beta = 1/k_B T$ , with *T* being the inverse temperature of the environment. The work FDR (1) is one of the pillars of classical stochastic thermodynamics; it shows that near equilibrium work fluctuations are responsible for dissipation, and conversely that any optimal slow process that minimizes dissipation will subsequently minimize the fluctuations [9,10]. For many slow classical processes the work distribution P(w) is Gaussian [11–15], and if the process also fulfills Jarzynski's equality, then this immediately implies Eq. (1) [8].

For quantum systems, developing a definition of work and understanding how quantum effects influence its statistics has raised much attention recently [16–26].

Previous studies on the nonclassicality of work distributions have considered the emergence of quasiprobabilities due to weak measurement [27–29], contextuality [30], and violations of macrorealism [31,32]. Despite the wealth of research on this topic, a quantum generalization of Eq. (1) has not been addressed.

Based initially on the two-projective-measurement (TPM) distribution P(w) [1,16,17], in this Letter we derive a quantum work FDR and find that it differs from Eq. (1) through an additional contribution arising due to quantum fluctuations generated along the protocol. This extra term is positive definite, implying that slow quantum processes are governed by the *inequality*  $W_{\text{diss}} \leq \beta \sigma_w^2/2$ , with equality obtained when no coherences in energy are created during the dynamics. We further demonstrate that the extra quantum term in the FDR leads to a non-Gaussian P(w), and we show that the same quantum FDR is also valid for work distributions accessed from weak measurements of the system.

While quantum work fluctuations are of fundamental interest, understanding their behavior also provides a method for minimizing them in practical implementations. Indeed, the design of reliable and minimally dissipative thermodynamic engines is of utmost importance in quantum thermodynamics. In the regime of slow processes, the minimization of dissipation can be obtained using techniques from a differential geometry: one can equip the thermodynamic state space with a Riemannian metric [33,34], and optimal protocols can be found by calculating the associated geodesics [9,10,35–40]. Here, we show that the quantum work fluctuations can also be related to a Riemannian metric. However, owing to quantum modifications, this new metric coincides with the metric responsible for minimizing dissipation in the classical

commutative regime only. While this result rules out protocols that simultaneously minimize both  $W_{\text{diss}}$  and  $\sigma_w$  for quantum coherent processes, our framework can be used to find optimal trade-offs between dissipation and fluctuations.

These results are derived under three main assumptions: (i) the coupling between system and bath is weak, (ii) the system reaches thermal equilibrium when interacting with the bath, and (iii) the driving is slow so that we can expand the magnitudes of interest in the driving velocity and keep only the leading terms. Under these assumptions, we now derive a quantum version of the FDR in Eq. (1).

The quantum work FDR.—We study the thermodynamics of an open quantum system S coupled to a thermal bath B with total Hamiltonian  $H_{SB}(t) = H_S(t) + H_B + V_{SB}$ , where  $H_S(t) = H_S(t) \otimes \mathbb{I}_B$  is the driven system Hamiltonian and  $V_{SB}$  a small but finite coupling Hamiltonian. We take a finite-time interval  $t \in [0, \tau]$  and consider processes where the two system Hamiltonian end points are fixed,  $H_{S}(0) = H_{0}$  and  $H_{S}(\tau) = H_{\tau}$ . We assume that the initial density matrix of S and B is a product  $\varrho_{SB}(0) = \pi_S(0) \otimes \pi_B(0), \text{ where } \pi_S(0) = e^{-\beta H_S(0)} / Z_S(0)$ and  $\pi_B = e^{-\beta H_B}/Z_B$  are the respective Gibbs states for the bare system and bath. The compound system evolves as  $\varrho_{SB}(t) = U(t)\varrho_{SB}(0)U^{\dagger}(t)$  with the time-ordered exponential  $U(t) = \overline{T} \exp[-(i/\hbar) \int_0^t dt' H_{SB}(t')]$ . Work is required to perform U(t), and because only the system Hamiltonian changes in time while coupling is weak, this work can be associated with work on the system alone [17]. The work statistics can be defined via the TPM scheme, where a projective energy measurement of the total Hamiltonian is performed at the beginning,  $H_{SB}(0)$ , and the end,  $H_{SB}(\tau)$ , of the process, with the energy differences measured identified as the work values w. From the statistics, the work distribution can then be constructed and becomes  $P(w) = (1/2\pi) \int d\lambda e^{-i\lambda w} G(\lambda)$  with a moment generating function  $G(\lambda) = \operatorname{Tr}_{SB}[U^{\dagger}(\tau)e^{i\lambda H_{SB}(\tau)}U(\tau)e^{-i\lambda H_{SB}(0)}\varrho_{SB}(0)]$ [1,16,17], which directly gives the work moments via  $\langle w^k \rangle = (-i)^k (d^k/d\lambda^k) G(\lambda)|_{\lambda=0}$ . While in the following we will use the TPM work distribution to establish the quantum FDR, we show in Sec. C of the Supplemental Material [41] that our results are also valid for alternative work distributions based on weak measurements [27–29].

From now on we shall use the more compact notation  $X_t \equiv X_S(t)$ , with  $X = \varrho$ , H,  $\pi$ , and denote  $\text{Tr}(\cdot)$  as the trace over the system degrees of freedom. In general, the reduced dynamics of the system can be written as  $\dot{\varrho}_t = -(i/\hbar)\text{Tr}_B\{[H_{SB}(t), \rho_{SB}(t)]\} = \mathcal{L}_t[\varrho_t]$ . Here, we will assume that the system follows an adiabatic Markovian master equation with a unique instantaneous steady state given by the thermal state at each  $t \in [0, \tau]$ :  $\mathcal{L}_t[\pi_t] = 0$ , with  $\pi_t = e^{-\beta H_t}/Z_t$  (a precise form of  $\mathcal{L}_t[\varrho_t]$  is presented in Sec. D of the Supplemental Material [41]). This is well justified whenever the bath dynamics are fast compared to

the driving rate of the system Hamiltonian [42,43], and the coupling between *S* and *B* is weak enough to satisfy the Born-Markov approximation and the rotating wave approximation [44]. Importantly, under these assumptions the TPM statistics can be determined by unravelling the master equation in terms of quantum jump trajectories [45–47]. These trajectories can then be accessed via local measurements of a quantum detector [48], circumventing the need to perform global energy measurements. Under these assumptions, we show in Sec. A of the Supplemental Material [41] that the work fluctuations  $\sigma_w^2 \equiv \langle w^2 \rangle - \langle w \rangle^2$ are given by

$$\sigma_w^2 = 2 \int_0^\tau dt_1 \int_0^{t_1} dt_2 \operatorname{Tr}\{\dot{H}_{t_1} \tilde{P}(t_1, t_2)[\mathbb{S}_{\varrho_{t_2}}(\dot{H}_{t_2})]\}, \qquad (2)$$

where  $\tilde{P}(t_1, t_2) = \tilde{T} \exp\left(\int_{t_2}^{t_1} d\nu \mathcal{L}_{\nu}\right)$  is the propagator for the Lindbladian, and we have introduced the linear mapping

$$\mathbb{S}_{\varrho}(A) \coloneqq \frac{1}{2} \{ \varrho, \Delta_{\varrho} A \}_{+}, \tag{3}$$

with  $\Delta_{\varrho}A = A - \text{Tr}(A\varrho)$  and  $\{\cdot, \cdot\}_+$  denoting the anticommutator. We now assume that the total time  $\tau$  of the process is large with respect to the timescale(s) of thermalization, which are encoded in  $\mathcal{L}_t$ . Since the two end points of the trajectory are fixed at  $H_0$  and  $H_{\tau}$ , one has  $\dot{H}_t \propto \tau^{-1}$ . In this case, we can expand the relevant expressions in terms of  $\tau^{-1}$  and keep the leading orders, which we refer to as the slow driving regime. This assumption allows us to further simplify Eq. (2) in Sec. B of the Supplemental Material [41], using techniques similar to the ones developed in Ref. [49] for classical systems. To first order in  $\tau^{-1}$  the work fluctuations are

$$\sigma_w^2 \simeq -2 \int_0^\tau dt \operatorname{Tr}\{\dot{H}_t \mathcal{L}_t^+[\mathbb{S}_{\pi_t}(\dot{H}_t)]\}.$$
 (4)

Note that the integrand is proportional to  $\tau^{-2}$ , so for the whole integral  $\sigma_w^2 \propto \tau^{-1}$ , as desired. In Eq. (4), we have introduced the so-called Drazin inverse  $\mathcal{L}_t^+$  of the Lindblad operator  $\mathcal{L}_t$  [40,50]. This inverse is defined as

$$\mathcal{L}_{t}^{+}[A] \coloneqq \int_{0}^{\infty} d\nu e^{\nu \mathcal{L}_{t}}[\pi_{t} \operatorname{Tr}(A) - A]$$
(5)

and satisfies three conditions [40]: (i) commutation with the Lindbladian, i.e.,  $\mathcal{L}_t \mathcal{L}_t^+[A] = \mathcal{L}_t^+ \mathcal{L}_t[A] = A - \pi_t \operatorname{Tr}(A)$ , (ii) invariance of the thermal state, i.e.,  $\mathcal{L}_t^+[\pi_t] = 0$ , and (iii) tracelessness, i.e.,  $\operatorname{Tr}(\mathcal{L}_t^+[A]) = 0$ .

An expression similar to Eq. (4) describes the dissipated work,  $W_{\text{diss}}$ , in slow quantum processes [40,51]

$$W_{\text{diss}} = -\beta \int_0^\tau dt \text{Tr}\{\dot{H}_t \mathcal{L}_t^+[\mathbb{J}_{\pi_t}(\dot{H}_t)]\}.$$
 (6)

Note that, in place of  $S_{\pi_t}$  in Eq. (4), the map  $J_{\pi_t}$  appears, with

$$\mathbb{J}_{\varrho}(A) \coloneqq \int_{0}^{1} da \varrho^{a} \Delta_{\varrho} A \varrho^{1-a}.$$
 (7)

In the special case in which *A* commutes with  $\rho$ , the maps  $\mathbb{S}_{\rho}(A)$  and  $\mathbb{J}_{\rho}(A)$  both reduce to  $\mathbb{S}_{\rho}(A) = \rho \Delta_{\rho} A = \mathbb{J}_{\rho}(A)$ .

Taking the expressions for work fluctuations  $\sigma_w^2$  and the dissipated work  $W_{\text{diss}}$  together, we obtain the quantum work FDR:

$$\frac{1}{2}\beta\sigma_w^2 = W_{\rm diss} + Q_w,\tag{8}$$

where  $Q_w = \beta \int_0^{\tau} dt \operatorname{Tr} \{ \dot{H}_t \mathcal{L}_t^+ [(\mathbb{J}_{\pi_t} - \mathbb{S}_{\pi_t})(\dot{H}_t)] \}$  is a quantum correction coming from the difference between the maps  $\mathbb{S}_{\varrho}(A)$  and  $\mathbb{J}_{\varrho}(A)$ .

In Sec. D of the Supplemental Material [41], we show that  $Q_w \ge 0$ , with equality if and only if  $[\dot{H}_t, H_t] = 0$  for  $\beta > 0$  and  $\forall t \in [0, \tau]$ . This implies that, for slow quantum processes with  $[H_t, \dot{H}_t] \ne 0$ , the classical FDR in Eq. (1) breaks down, and the work fluctuations are in fact greater than dissipation. In general, one has an inequality:

$$W_{\rm diss} \le \frac{1}{2} \beta \sigma_w^2. \tag{9}$$

We can then interpret the quantum work FDR (8) as follows. During a slow process where the state remains close to a thermal state  $\pi_t$ , the work fluctuations  $\beta \sigma_w^2/2$  can be divided into two positive contributions: a thermal contribution  $W_{\text{diss}}$ , which arises from the thermal fluctuations in  $\pi_t$ , and a purely quantum contribution  $Q_w$ , which appears whenever quantum fluctuations are created in the dynamics as  $[\pi_t, \dot{H}_t] \neq 0$ .

Let us rewrite  $Q_w = \beta \int_0^x dt \mathcal{I}_t(\pi_t, \dot{H}_t)$ , where we have introduced the *dynamical skew information*  $\mathcal{I}_t(\pi_t, A) :=$  $\operatorname{Tr}\{A\mathcal{L}_t^+[(\mathbb{J}_{\pi_t} - \mathbb{S}_{\pi_t})(A)]\}$  for an arbitrary observable A. To further elaborate on the idea that  $Q_w$  measures the quantum work fluctuations, now suppose that S evolves under a perfectly thermalizing map with a single timescale  $1/\Gamma$ ; i.e., the Lindbladian satisfies

$$\mathcal{L}_t[\varrho_t] = (\pi_t - \varrho_t)\Gamma, \qquad (10)$$

which has the Drazin inverse  $\mathcal{L}_t^+(\cdot) = [\text{Tr}(\cdot)\pi_t - \mathbb{I}(\cdot)]/\Gamma$ . In this case,  $\mathcal{I}_t(\pi_t, A)$  becomes proportional to the average Wigner-Yanase-Dyson skew information [52–54],  $\mathcal{I}_t(\pi_t, A) = -\frac{1}{2\Gamma} \int_0^1 da \text{Tr}([A, \pi^a][A, \pi^{1-a}])$ , which can be understood as a measure of quantum uncertainty in the observable *A* [55]. It is positive and vanishes if and only if  $[A, \pi_t] = 0$ , reduces to the usual variance for pure  $\pi_t = |\psi\rangle\langle\psi|$ , and decreases under classical mixing. For more general Lindbladians,  $\mathcal{I}_t(\pi_t, A)$  also takes into account the presence of different timescales of thermalization through the additional dependence on  $\mathcal{L}_t^+$ . Summarizing, in Eq. (8) we can interpret  $Q_w$  as a measure of the time-integrated quantum fluctuations in the power  $\dot{H}_t$ .

Non-Gaussianity of the work distribution.—Here, we show that these quantum coherences necessarily lead to a non-Gaussian shape of the TPM work distribution P(w). For this P(w) the Jarzynski equality holds [17], which relates the change in equilibrium free energy to the cumulants of work done on the system that are computed from P(w):

$$\Delta F = -\beta^{-1} \ln \langle e^{-\beta_{W}} \rangle = \sum_{k=1}^{\infty} \frac{(-\beta)^{k-1}}{k!} \kappa_{w}^{(k)}.$$
 (11)

Here,  $\kappa_w^{(k)}$  are the cumulants of work, with  $\kappa_w^{(1)} = \langle w \rangle$  and  $\kappa_w^{(2)} = \sigma_w^2$ . After rearranging the terms in Eq. (11) and combining this with the quantum FDR (8), we find that

$$\sum_{k=3}^{\infty} \frac{(-\beta)^{k-1}}{k!} \kappa_w^{(k)} = Q_w \ge 0.$$
 (12)

In fact, as we have seen,  $Q_w$  vanishes if and only if  $[\dot{H}_t, H_t] = 0 \ \forall t \in [0, \tau]$ . Since a Gaussian work distribution has zero cumulants for  $k \ge 3$ , we conclude that P(w) necessarily becomes non-Gaussian whenever the process generates coherences of the power operator with respect to the instantaneous Hamiltonian. This contrasts with the classical expectation that slow processes lead to Gaussian work distributions [7,11]. Equality (12) further demonstrates that measuring the work cumulants can provide a direct witness of quantum fluctuations in power.

*Thermodynamic geometry and optimal paths.*—Now that we have established a relationship between work dissipation and fluctuations, we are in a position to determine optimal protocols. In order to find protocols with minimal fluctuations, one can take a geometric approach similar to Refs. [9,10,40].

We consider a decomposition of the system Hamiltonian of the form  $H_t = X_0 + \vec{\lambda}_t \cdot \vec{X}$ , where  $\vec{\lambda}_t = (\lambda_1(t), \lambda_2(t), ...)$ is the vector of scalar controllable parameters and  $\vec{X} = \partial H_t / \partial \vec{\lambda}_t = (X_1, X_2, ...)$  are the corresponding generalized conjugate forces. Then, Eq. (4) can be recast in the form  $\sigma_w^2 = (2/\beta) \int_0^{\tau} dt [d\vec{\lambda}_t / dt]^T \Lambda(\vec{\lambda}_t) [d\vec{\lambda}_t / dt]$ , where  $\Lambda(\vec{\lambda}_t)$  has the elements

$$\Lambda_{ij}(\vec{\lambda}_t) \coloneqq -\frac{\beta}{2} \operatorname{Tr}\{X_i \mathcal{L}_t^+[\mathbb{S}_{\pi_t}(X_j)] + X_j \mathcal{L}_t^+[\mathbb{S}_{\pi_t}(X_i)]\}.$$
(13)

It follows that since the rate of dissipated work and the dynamical skew information are both positive,  $\Lambda(\vec{\lambda}_t)$  is a positive-definite matrix. Since  $\Lambda(\vec{\lambda}_t)$  is also symmetric and depends smoothly on  $\pi_t$ , it induces a Riemannian metric on the space of quantum thermal states [56]. Differential geometry then provides an efficient and systematic approach to find optimal protocols by solving Euler-Lagrange equations for the functional  $\sigma_w^2$  of the curve  $\vec{\lambda}_t$ . Curves of minimal fluctuations are identified as geodesics of constant velocity.

The work-fluctuation metric  $\Lambda(\vec{\lambda}_t)$  given in Eq. (13) should be compared to the work-dissipation metric  $\boldsymbol{\xi}(\vec{\lambda}_t)$ , for which  $W_{\text{diss}} = \int_0^{\tau} dt [d\vec{\lambda}_t/dt]^T \boldsymbol{\xi}(\vec{\lambda}_t) [d\vec{\lambda}_t/dt]$ , with elements [40]

$$\xi_{ij}(\vec{\lambda}_t) \coloneqq -\frac{\beta}{2} \operatorname{Tr}\{X_i \mathcal{L}_t^+[\mathbb{J}_{\pi_t}(X_j)] + X_j \mathcal{L}_t^+[\mathbb{J}_{\pi_t}(X_i)]\}.$$
(14)

The two metrics  $\Lambda(\vec{\lambda}_t)$  and  $\xi(\vec{\lambda}_t)$  coincide whenever the conjugate forces commute, i.e.,  $[X_i, X_0] = [X_i, X_j] = 0 \forall i, j$ . In this special case both metrics reduce to the classical Fisher-Rao metric over the space of thermal states, multiplied by  $k_B T$  and an integral relaxation time related to the open system dynamics [10].

In general, the fluctuation and dissipation metrics differ, and hence their corresponding geodesics will no longer coincide, in contrast to slow processes in classical thermodynamics. In other words, for quantum processes, any slow protocol  $\vec{\lambda}_t^{\text{opt}}$  that minimizes dissipation will have nonminimal fluctuations, and vice versa. To interpolate between these two extremes, one can resort to minimizing the objective function

$$\mathcal{C}_{\alpha} \coloneqq \alpha \tilde{\sigma}_{w}^{2} + (1 - \alpha) W_{\text{diss}} \quad \text{for } \alpha \in [0, 1], \quad (15)$$

where  $\alpha$  weights the relative importance of the fluctuations versus dissipation and  $\tilde{\sigma}_w^2 = \frac{1}{2}\beta\sigma_w^2$ . The family of metrics minimizing  $C_\alpha$  for weights  $\alpha$  is just the convex sum  $g_\alpha(\vec{\lambda}_t) = \alpha \Lambda(\vec{\lambda}_t) + (1 - \alpha)\xi(\vec{\lambda}_t)$ . In Sec. E of the Supplemental Material [41], we use Euler-Lagrange methods to find the optimal protocol  $\lambda_t^{\text{opt}}(\alpha)$  that minimizes  $C_\alpha$  when  $\lambda_t$  is a one-dimensional control parameter with  $H_t = X_0 + \lambda_t X$ . The optimal velocity takes the form  $\dot{\lambda}_t^{\text{opt}}(\alpha) \propto \sqrt{\xi(\lambda_t) + \alpha\beta \mathcal{I}_t(\pi_t, X)}$ , which clearly depends on  $\alpha$  due to the presence of quantum coherence. This contrasts with the classical case  $[X_0, X] = 0$ , where the optimal protocol can be obtained for any  $\alpha$  by driving the system at a constant dissipation rate [10].

*Example.*—Let us illustrate our results with a slowly driven harmonic oscillator,  $H_t = \hbar \omega_t (a_{\omega_t}^{\dagger} a_{\omega_t} + 1/2)$ , connected to a perfectly thermalizing bath described by the master equation (10). Here,  $\omega_t$  is the time-dependent frequency of the oscillator, and  $a_{\omega_t}$  and  $a_{\omega_t}^{\dagger}$  are the

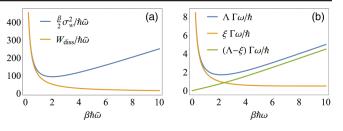


FIG. 1. (a) Dissipated work  $W_{\text{diss}}$  and work fluctuations  $\frac{1}{2}\beta \sigma_w^2$  as a function of initial state inverse temperature  $\beta$  for the harmonic oscillator example. The plots are for a protocol in which the oscillator frequency  $\omega_t$  is increased linearly in time from  $\omega_0 = 0.1\tilde{\omega}$  to  $\omega_1 = 10\tilde{\omega}$  for a fixed reference frequency  $\tilde{\omega}$ . (b) Plot of the metric tensors of fluctuations ( $\Lambda$ ) and dissipation ( $\xi$ ), and of their difference ( $\Lambda - \xi$ ), for the harmonic oscillator example as a function of inverse temperature  $\beta$  at a given energy gap  $\hbar\omega$  (see Sec. F of the Supplemental Material [41]).

frequency-dependent creation and annihilation operators. Taking the time derivative yields the power operator  $\dot{H}_t = \hbar \dot{\omega}_t \{ H_t / \hbar \omega_t + [(a_{\omega_t}^{\dagger})^2 + a_{\omega_t}^2]/2 \},$  which does not commute with the instantaneous Hamiltonian  $H_t$ , i.e.,  $[H_t, H_t] \neq 0$ . In Fig. 1(a), we compare the expressions for  $W_{\rm diss}$  and  $\beta \sigma_w^2/2$  for a slow linear ramp of  $\omega_t$ , and it can be seen that the curves differ substantially at low temperatures (i.e., high  $\beta$ ), where quantum fluctuations become dominant, and become closer for higher temperatures, where thermal fluctuations dominate and classical behavior is recovered. The corresponding metrics  $\Lambda(\omega_t)$  and  $\xi(\omega_t)$ along with their difference,  $\Lambda(\omega_t) - \xi(\omega_t) = \beta \mathcal{I}_t(\pi_t, X)$ , are shown in Fig. 1(b) as a function of inverse temperature. As expected, this difference vanishes in the high temperature limit ( $\beta \rightarrow 0$ ). In the low temperature regime, thermal fluctuations given by the dissipation metric  $\xi(\omega_t)$  decay, while quantum coherences contribute more significantly to the total fluctuations in power that are given by  $\Lambda(\omega_t)$ . The details of all of these calculations are provided in Sec. F of the Supplemental Material [41].

Turning to optimization, we now use the metric  $g_{\alpha}(\omega_t) =$  $\alpha \Lambda(\omega_t) + (1 - \alpha) \xi(\omega_t)$  associated with Eq. (15) to construct geodesics that interpolate between minimally dissipating and minimally fluctuating protocols (see Sec. F of the Supplemental Material [41]). So-called Pareto fronts can be used to bound the region of allowed protocols [57]. This is illustrated in Fig. 2, where Pareto front curves indicate the trade-off between minimal fluctuation  $(\beta \sigma_w^2/2)$ and minimal dissipation ( $W_{diss}$ ) for various values of  $\beta$ . Each curve is obtained by evaluating  $\beta \sigma_w^2/2$  and  $W_{\rm diss}$  for the geodesics minimizing  $C_{\alpha}$  for all values  $\alpha \in [0, 1]$ . If the classical FDR would hold, each curve would collapse into a single point along the diagonal line  $\beta \sigma_w^2/2 = W_{\rm diss}$ . The quantum correction moves each Pareto front above this line and expands it from a single point to a curve, parametrized by  $\alpha$ . As expected, this effect is most significant at low temperatures where quantum fluctuations dominate.

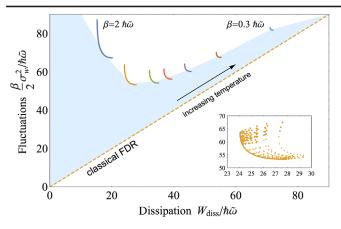


FIG. 2. Pareto fronts limiting the accessible region of fluctuations  $\frac{1}{2}\beta\sigma_w^2$  and dissipation  $W_{\text{diss}}$  for the harmonic oscillator example over all possible protocols  $\{\omega_t\}$  between the end points  $\omega_0 = 0.1\tilde{\omega}$  and  $\omega_1 = 10\tilde{\omega}$  for a fixed reference frequency  $\tilde{\omega}$ . Curves are obtained by varying the weight  $\alpha \in [0, 1]$ , and for each  $\alpha$  by choosing the protocol to follow the geodesic that minimizes  $C_{\alpha}$ . Each curve is for a specific inverse temperature  $\beta = 2\hbar\tilde{\omega}$ (blue),  $\beta = 1\hbar\tilde{\omega}$  (yellow),  $\beta = 0.7\hbar\tilde{\omega}$  (green),  $\beta = 0.6\hbar\tilde{\omega}$  (red),  $\beta = 0.5\hbar\tilde{\omega}$  (purple),  $\beta = 0.4\hbar\tilde{\omega}$  (brown), and  $\beta = 0.3\hbar\tilde{\omega}$  (light blue). The blue shaded region denotes the separation between the quantum optimal protocols (Pareto fronts) and the classical optimal protocols (diagonal) for varying  $\beta$ . (Inset) Magnified Pareto front for  $\beta = \hbar\tilde{\omega}$  and including points for suboptimal protocols, illustrating the accessible part of the fluctuationdissipation plane.

Conclusions .--- In this Letter, we have studied the statistics of work in slowly driven open quantum systems interacting with a thermal environment. We have derived a quantum FDR for work as shown in Eq. (8), which generalizes the well-known classical FDR given by Eq. (1). This result implies that whenever quantum coherence is generated during the dynamics of a slow protocol,  $W_{\rm diss} < \frac{1}{2}\beta\sigma_w^2$ , which is a genuinely quantum effect. Let us briefly comment on the generality of our results. While Eq. (8) has been derived using the TPM approach with thermal initial conditions, we prove in Sec. C of the Supplemental Material [41] that Eq. (8) holds more generally for arbitrary initial states using alternative definitions of work based on weak measurements [22,58–63]. This follows directly because these measurement schemes give rise to the same work average and variance. The validity of the quantum FDR for various work definitions highlights the fact that the quantum effects captured by  $Q_w$  stem from the coherent dynamics of the protocol, rather than arising as the result of a measurement disturbance or a particular choice of work definition (see the discussion in Sec. C of the Supplemental Material [41]).

It is also interesting to discuss how breaking any of the three main assumptions used to derive the quantum FDR namely, (i) slow driving, (ii) thermalization, and (iii) weak coupling—can affect it. Both (i) and (ii) appear to be crucial: in Sec. H of the Supplemental Material [41], we compare  $W_{\text{diss}}$  and  $\sigma_w^2$  for a spin in contact with a bosonic bath and, while we verify the validity of Eq. (8) for sufficiently slow driving, we do find violations of the FDR for faster driving. Regarding assumption (ii), one can demonstrate that the quantum FDR can break down if the system is not close to thermal equilibrium even if the dynamics are slow, as shown in Ref. [64] for closed unitary evolutions. On the other hand, we believe that the quantum FDR can remain valid away from the weak coupling regime [i.e., if (iii) is broken]: a step toward proving this hypothesis is done in Sec. G of the Supplemental Material [41]. By using a discrete model of quasi-isothermal processes [65,66], we derive an analogous quantum FDR for a system strongly coupled to a thermal bath.

The quantum FDR also implies that it is fundamentally impossible to simultaneously minimize dissipation and fluctuations in slow coherent quantum processes. In the second part of the Letter, we have derived a family of metrics whose geodesics interpolate between minimally dissipative and minimally fluctuating thermodynamic protocols, and our results unveil a new geometric structure within quantum thermodynamics. A promising platform to observe these effects experimentally involves quantum dots [67–69] and superconducting qubits [70,71], where slowly driven noncommuting protocols appear to be a realistic possibility [72], and proposals for observing TPM work statistics using a calorimeter have been made [48]. An interesting future direction is to extend the FDR to manybody closed systems [64,73,74], and to investigate how these genuinely quantum effects can modify the thermodynamic uncertainty relations in nonequilibrium steady states [75–77] and FDRs in other contexts such as quantum transport [78].

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- M. Esposito, U. Harbola, and S. Mukamel, Rev. Mod. Phys. 81, 1665 (2009).
- [2] C. Jarzynski, Annu. Rev. Condens. Matter Phys. 2, 329 (2011).
- [3] U. Seifert, Rep. Prog. Phys. 75, 126001 (2012).
- [4] M. Campisi, P. Hänggi, and P. Talkner, Rev. Mod. Phys. 83, 771 (2011).
- [5] J. Hermans, J. Phys. Chem. 95, 9029 (1991).
- [6] R. H. Wood, W. C. F. Muhlbauer, and P. T. Thompson, J. Phys. Chem. 95, 6670 (1991).

- [7] D. A. Hendrix and C. Jarzynski, J. Chem. Phys. 114, 5974 (2001).
- [8] C. Jarzynski, Phys. Rev. Lett. 78, 2690 (1997).
- [9] G. E. Crooks, Phys. Rev. Lett. 99, 100602 (2007).
- [10] D. A. Sivak and G. E. Crooks, Phys. Rev. Lett. 108, 190602 (2012).
- [11] T. Speck and U. Seifert, Phys. Rev. E 70, 066112 (2004).
- [12] P. Chvosta, P. Reineker, and M. Schulz, Phys. Rev. E 75, 041124 (2007).
- [13] E. Šubrt and P. Chvosta, J. Stat. Mech. (2007) P09019.
- [14] T. Speck, J. Phys. A 44, 305001 (2011).
- [15] J. Hoppenau and A. Engel, J. Stat. Mech. (2013) P06004.
- [16] P. Talkner, E. Lutz, and P. Hänggi, Phys. Rev. E 75, 050102
   (R) (2007).
- [17] P. Talkner, M. Campisi, and P. Hänggi, J. Stat. Mech. (2009) P02025.
- [18] C. Jarzynski, H. T. Quan, and S. Rahav, Phys. Rev. X 5, 031038 (2015).
- [19] P. Talkner and P. Hänggi, Phys. Rev. E 93, 022131 (2016).
- [20] H. J. D. Miller and J. Anders, New J. Phys. 19, 062001 (2017).
- [21] M. Perarnau-Llobet, E. Bäumer, K. V. Hovhannisyan, M. Huber, and A. Acin, Phys. Rev. Lett. 118, 070601 (2017).
- [22] P. Solinas, H. J. D. Miller, and J. Anders, Phys. Rev. A 96, 052115 (2017).
- [23] J. Åberg, Phys. Rev. X 8, 011019 (2018).
- [24] E. Bäumer, M. Lostaglio, M. Perarnau-Llobet, and R. Sampaio, in *Thermodynamics in the Quantum Regime: Fundamental Aspects and New Directions*, edited by F. Binder, L. A. Correa, C. Gogolin, J. Anders, and G. Adesso, Fundamental Theories of Physics Vol. 195 (Springer, New York, 2018), pp. 275–300.
- [25] Z. Holmes, S. Weidt, D. Jennings, J. Anders, and F. Mintert, Quantum 3, 124 (2019).
- [26] K.-D. Wu, Y. Yuan, G.-Y. Xiang, C.-F. Li, G.-C. Guo, and M. Perarnau-Llobet, Sci. Adv. 5, eaav4944 (2019).
- [27] A. E. Allahverdyan, Phys. Rev. E 90, 032137 (2014).
- [28] P. Solinas and S. Gasparinetti, Phys. Rev. E 92, 042150 (2015).
- [29] P. P. Potts, Phys. Rev. Lett. 122, 110401 (2019).
- [30] M. Lostaglio, Phys. Rev. Lett. 120, 040602 (2018).
- [31] R. Blattmann and K. Mølmer, Phys. Rev. A 96, 012115 (2017).
- [32] H. J. D. Miller and J. Anders, Entropy 20, 200 (2018).
- [33] F. Weinhold, J. Chem. Phys. 63, 2479 (1975).
- [34] G. Ruppeiner, Phys. Rev. A 20, 1608 (1979).
- [35] J. Nulton, P. Salamon, B. Andresen, and Q. Anmin, J. Chem. Phys. 83, 334 (1985).
- [36] P. Salamon and R. S. Berry, Phys. Rev. Lett. 51, 1127 (1983).
- [37] P. R. Zulkowski, D. A. Sivak, and M. R. DeWeese, PLoS One 8, e82754 (2013).
- [38] P.R. Zulkowski and M.R. DeWeese, Phys. Rev. E 92, 032113 (2015).
- [39] M. V. S. Bonança and S. Deffner, Phys. Rev. E 98, 042103 (2018).
- [40] M. Scandi and M. Perarnau-Llobet, Quantum 3, 197 (2019).
- [41] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.123.230603 for further details on (a) Work moments for open system dynamics, (b) Derivation of Eq. (4), (c) Weak measurements of

fluctuating work, (d) Proof of Eq. (9), (e) Finding geodesics for a single parameter, (f) Thermodynamic metrics for the harmonic oscillator, (g) Work fluctuation-dissipation relation beyond weak-coupling, and (h) Numerical verification of Eq. (8).

- [42] T. Albash, S. Boixo, and D. A. Lidar, New J. Phys. 14, 123016 (2012).
- [43] R. Dann, A. Levy, and R. Kosloff, Phys. Rev. A 98, 052129 (2018).
- [44] H.-P. Breuer, F. Petruccione *et al.*, *The Theory of Open Quantum Systems* (Oxford University Press, New York, 2002).
- [45] M. Silaev, T. T. Heikkilä, and P. Virtanen, Phys. Rev. E 90, 022103 (2014).
- [46] F. Liu, Phys. Rev. E 93, 012127 (2016).
- [47] F. Liu and J. Xi, Phys. Rev. E 94, 062133 (2016).
- [48] J. P. Pekola, P. Solinas, A. Shnirman, and D. V. Averin, New J. Phys. 15, 115006 (2013).
- [49] D. Mandal and C. Jarzynski, J. Stat. Mech. (2016) 063204.
- [50] T. L. Boullion and P. L. Odell, *Generalised Inverse Matrices* (Wiley-InterScience, New York, 1971).
- [51] V. Cavina, A. Mari, and V. Giovannetti, Phys. Rev. Lett. 119, 050601 (2017).
- [52] F. Hansen, Proc. Natl. Acad. Sci. U.S.A. 105, 9909 (2008).
- [53] I. Frérot and T. Roscilde, Phys. Rev. B 94, 075121 (2016).
- [54] H. J. D. Miller and J. Anders, Nat. Commun. 9, 2203 (2018).
- [55] S. Luo, Phys. Rev. A 72, 042110 (2005).
- [56] D. Petz, J. Math. Phys. (N.Y.) 35, 780 (1994).
- [57] A. P. Solon and J. M. Horowitz, Phys. Rev. Lett. 120, 180605 (2018).
- [58] A. A. Clerk, Phys. Rev. A 84, 043824 (2011).
- [59] A. Bednorz, W. Belzig, and A. Nitzan, New J. Phys. 14, 013009 (2012).
- [60] P. P. Hofer and A. A. Clerk, Phys. Rev. Lett. 116, 013603 (2016).
- [61] P. P. Hofer, Quantum 1, 32 (2017).
- [62] A. E. Allahverdyan, Phys. Rev. E 90, 032137 (2014).
- [63] P. Solinas and S. Gasparinetti, Phys. Rev. E 92, 042150 (2015).
- [64] K. Zawadzki, R. M. Serra, and I. D'Amico, arXiv:1908.06488.
- [65] R. Gallego, A. Riera, and J. Eisert, New J. Phys. 16, 125009 (2014).
- [66] M. Perarnau-Llobet, H. Wilming, A. Riera, R. Gallego, and J. Eisert, Phys. Rev. Lett. **120**, 120602 (2018).
- [67] J. P. Pekola, Nat. Phys. 11, 118 (2015).
- [68] M. F. Ludovico, F. Battista, F. von Oppen, and L. Arrachea, Phys. Rev. B 93, 075136 (2016).
- [69] P. T. Alonso, J. Romero, and L. Arrachea, Phys. Rev. B 99, 115424 (2019).
- [70] J. P. Pekola, D. S. Golubev, and D. V. Averin, Phys. Rev. B 93, 024501 (2016).
- [71] N. Cottet and B. Huard, in *Thermodynamics in the Quantum Regime: Fundamental Aspects and New Directions*, edited by F. Binder, L. A. Correa, C. Gogolin, J. Anders, and G. Adesso, Fundamental Theories of Physics Vol. 195 (Springer, New York, 2018), pp. 959–981.

- [72] S. Hacohen-Gourgy, L. S. Martin, E. Flurin, V. V. Ramasesh, K. B. Whaley, and I. Siddiqi, Nature (London) 538, 491 (2016).
- [73] L. Fusco, S. Pigeon, T. J. G. Apollaro, A. Xuereb, L. Mazzola, M. Campisi, A. Ferraro, M. Paternostro, and G. De Chiara, Phys. Rev. X 4, 031029 (2014).
- [74] E. G. Arrais, D. A. Wisniacki, A. J. Roncaglia, and F. Toscano, arXiv:1907.06285 [Phys. Rev. E (to be published)].
- [75] A. C. Barato and U. Seifert, Phys. Rev. Lett. 114, 158101 (2015).
- [76] K. Ptaszyński, Phys. Rev. B 98, 085425 (2018).
- [77] B. K. Agarwalla and D. Segal, Phys. Rev. B 98, 155438 (2018).
- [78] D. V. Averin and J. P. Pekola, Phys. Rev. Lett. 104, 220601 (2010).