Generalized Linear Response Theory for the Full Quantum Work Statistics

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We consider a quantum system driven out of equilibrium via a small Hamiltonian perturbation. Building on the paradigmatic framework of linear response theory (LRT), we derive an expression for the full generating function of the dissipated work. Remarkably, we find that all information about the distribution can be encoded in a single quantity, the standard relaxation function in LRT, thus opening up new ways to use phenomenological models to study nonequilibrium fluctuations in complex quantum systems. Our results establish a number of refined quantum thermodynamic constraints on the work statistics that apply to regimes of perturbative but arbitrarily fast protocols, and do not rely on assumptions such as slow driving or weak coupling. Finally, our approach uncovers a distinctly quantum signature in the work statistics that originates from underlying zero-point energy fluctuations. This causes an increased dispersion of the probability distribution at short driving times, a feature that can be probed in efforts to witness nonclassical effects in quantum thermodynamics.

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At the microscopic level, the traditional laws of thermodynamics fall short at providing an accurate description due to the fact that fluctuations in work, heat, and entropy production play a preponderant role [1–3]. Besides being of fundamental importance, these thermodynamic fluctuations have several direct implications on the performance of small-scale engines and (bio)chemical reactions [4–7]. For these reasons, understanding the statistical aspects of dissipative processes has been one of the overarching themes of the field of *stochastic thermodynamics* [8–10]. At even smaller scales, *quantum mechanics* represents an additional source of fluctuations even in the absence of any thermal agitation. While quantum properties have often been shown to lead to advantages over classical counterparts with regard to expectation values or speedups in total process time [11–13], many open questions still remain surrounding the thermodynamic cost associated to quantum fluctuations (i.e., the process precision) [14–25].

One pressing question is to understand the nature of quantum fluctuations in *finite-time* thermodynamic processes. Recent studies of slowly driven quantum systems have uncovered a wealth of strong results in this direction, ranging from finite-time thermodynamic bounds and trade-offs [25–31], general optimal control strategies [32,33], geometric phase effects [34], and broad identifications of quantum signatures in work statistics [35,36]. However, going beyond slow driving regimes remains a significant challenge, due to the fact that finite-time processes require precise knowledge about the underlying dynamics. This is especially difficult to obtain when a system is driven via time-dependent Hamiltonian driving or in contact with an external environment. One way around this is through *linear response theory* (LRT), which allows one to utilize phenomenological models to make thermodynamic predictions based on the system's response to small perturbations. Since its original formulation by Kubo [37], LRT has remained an indispensable tool for studying systems close to equilibrium [38], with significant applications to quantum transport [39], many-body quantum physics [40], and quantum field theory [41]. In the context of quantum thermodynamics [42], one use of LRT has been the development of general optimization strategies for minimal average work dissipation protocols [43–45].

In this paper, we develop a broader picture by characterizing the full quantum work distribution in LRT. In particular, we derive a universal and model-independent expression for all the statistical cumulants (average, variance, and higher fluctuations) of the dissipated work spent when driving a quantum system out of equilibrium by means of a finite-time, weak perturbation. Crucially, we show that all these higher order fluctuations can be directly obtained through a single well-known quantity; the system's *relaxation function* [37,44]. The latter is one of the central quantities in LRT and often represents the basis for phenomenological thermodynamic descriptions of complex many-body systems. Our main result opens new routes to analyze all the properties of work statistics in complex systems by means of this easily accessible quantity, as we exemplify with examples of systems undergoing overdamped and underdamped Brownian motion.

This result allows us to identify a new set of refined quantum thermodynamic constraints, including a fluctuation theorem and positivity of all work cumulants. These solely rely on minimal assumptions such as unitary dynamics on the full system and small Hamiltonian perturbations. Operationally speaking, our theory moreover predicts the existence of a distinctly quantum effect on the work probability distribution, which results in a significant broadening of the dispersion at low temperatures and prevents saturation of the thermodynamic uncertainty relation. We show that this *nonclassical signature* can be deeply connected to the breakdown of the equipartition theorem in quantum statistical mechanics.

Quantum dissipated work statistics in LRT—We begin by considering a quantum system unitarily driven out of equilibrium by means of a time-dependent Hamiltonian driving, $H_t := H_0 + \lambda_t V$, over a finite interval $t \in [0, \tau]$. Here, $t \mapsto \lambda_t$ is a dimensionless function characterizing a particular driving protocol, and the operator V is treated as a perturbation that is turned on at time $t = 0$ (i.e., $\lambda_0 = 0$). The system is initially prepared in a thermal Gibbs state $\rho_0 = \pi_0$ at inverse temperature $\beta = 1/(k_B T)$, where we denote $\pi_t := e^{-\beta H_t}/\mathcal{Z}_t$. After the driven evolution, the final state is given by $\rho_\tau = U_\tau \pi_0 U_\tau^\dagger$, with $U_\tau = \overleftarrow{\mathcal{T}} \exp(i/\hbar \int_0^\tau dt' H_{t'})$. The main thermodynamic quantity of interest is the *dissipated work* irreversibly spent to drive the system out of equilibrium:

$$W_{\mathrm{diss}} := W - \Delta F, \tag{1}$$

with W denoting the stochastic quantum work defined through a two-time projective energy measurement at the beginning and end of the driving [46,47], and with $\Delta F = -\beta^{-1} \ln(Z_\tau/Z_0)$ being the change in equilibrium free energy. A full stochastic thermodynamic description of the process can be derived from the resulting distribution in dissipated work $P(W_{\mathrm{diss}})$. As shown in Ref. [48], this information is quantified by the *quantum Renyi divergence* between the instantaneous equilibrium state π_τ and the nonequilibrium state ρ_τ, since the *cumulant generating function* (CGF) of the process is found to be

$$K(\eta) := \ln\langle e^{-\eta\beta W_{\mathrm{diss}}}\rangle = (\eta - 1) S_\eta(\pi_\tau || \rho_\tau), \tag{2}$$

where $S_\alpha(\rho_1||\rho_2) := (\alpha - 1)^{-1} \ln \mathrm{Tr}(\rho_1^\alpha \rho_2^{1-\alpha})$ is the Renyi divergence of order $\alpha > 0$, generalizing the quantum relative entropy. From the CGF, we can derive cumulants using the formula $\kappa_W^k := (-k_B T)^k \lim_{\eta\to 0} \partial^k K(\eta)/\partial\eta^k$.

In LRT one assumes a weak perturbation such that $|\lambda_t| \ll 1$ for $\forall\, t \in [0, \tau]$, with normalization $\|V\| = 1$, resulting in a small deviation from the initial equilibrium state at all times given by [49]

$$\rho_t = \pi_0 - \frac{i}{\hbar}\int_0^t dt' \lambda_{t'} [V(t - t'), \pi_0], \tag{3}$$

with notation $A(t) := e^{iH_0 t/\hbar} A e^{-iH_0 t/\hbar}$ indicating the interaction picture.

Under this approximation, it is known that the linear-order correction to the average dissipated work is given in terms of the two-time integral [43,44,50]

$$\beta\langle W_{\mathrm{diss}}\rangle = \frac{1}{2}\int_0^\tau dt \int_0^\tau dt'\, \Psi_0(t - t')\dot{\lambda}_t \dot{\lambda}_{t'}. \tag{4}$$

Here $t \mapsto \Psi_0(t)$ denotes a central object in LRT known as the *relaxation function* [37], which can be expressed using the *Kubo covariance* as follows:

$$\Psi_0(t) := \beta \int_0^\beta ds \langle V(-i\hbar s) V(t)\rangle_0 - \beta^2 \langle V\rangle_0^2, \tag{5}$$

where $\langle . \rangle_0$ denotes the average with respect to the thermal state π_0. Physically, in LRT Ψ_0 allows one to introduce a characteristic timescale $\tau_R := \int_0^\infty dt\, \Psi_0(t)/\Psi_0(0)$ over which two-time correlations in $t \mapsto V(t)$ decay in time. While the calculation of the relaxation function in principle requires knowledge of the exact dynamics of the system, the power of LRT lies in the fact that one may often use phenomenological models of Ψ_0 to investigate the generic behavior of systems where this information may not be available. To do this one must impose certain constraints on any ansatz that would ensure both dynamical and thermodynamic consistency. The two key properties we require are

$$\Psi_0(t) = \Psi_0(-t), \quad \forall\, t \tilde{\Psi}_0(\omega) \geq 0 \quad \forall\, \omega \in \mathbb{R}, \tag{6}$$

where $\tilde{\Psi}_0(\omega) = \mathcal{F}[\Psi_0](\omega)$ denotes the Fourier transform of the relaxation function. The first property reflects *time-reversal symmetry* due to the underlying Hamiltonian dynamics. The second property in (6) expresses the positivity of its Fourier transform, and is equivalent to $\langle W_{\mathrm{diss}}\rangle \geq 0$ in agreement with the second law of thermodynamics [51]. Both conditions (6) follow from a fully Hamiltonian description and thus provide a consistency check for any approximate, phenomenological model of $\Psi_0(t)$.

Going beyond average quantities is however necessary in order to properly characterize the thermodynamics of nanoscale processes, both classical and quantum even within LRT. In what follows we achieve this goal by systematically exploiting the state expansion, Eq. (3), in order to obtain the linear order corrections to the full cumulant generating function of the dissipated work, Eq. (2), and consequently to all its higher statistical cumulants κ_W^k. Expanding the Renyi divergences in Eq. (2) up to second order in the perturbation strength, we arrive at our first main result for the full CGF in LRT,

$$K(\eta) := -\int_0^\tau dt \int_0^t dt' \, \dot{\lambda}_t \dot{\lambda}_{t'} \langle\langle \delta V(t), \delta V(t') \rangle\rangle_0^\eta, \tag{7}$$

with $\delta A := A - \mathrm{Tr}(A\pi_0)$ [52]. The bilinear form in (7) denotes a generalized version of the Kubo correlation function, Eq. (5), defined as

$$\langle\langle A, B \rangle\rangle_0^\eta := \int_0^{\beta\eta} dx \int_{\beta x}^{\beta - \beta x} dy \langle B(-i\hbar y) A \rangle_0. \tag{8}$$

Here the integration over imaginary time relates to the Green-Kubo-Mori-Zwanzig product [55]. This quantity originates from the field of quantum information geometry [56,57] and has recently found applications in stochastic thermodynamics, e.g., in slowly driven processes [35], and in the context of the locality of temperature [58]. It is however worth noting that, at variance with what happens in the slow driving regime where one neglects correlations over long times, the LRT accounts for memory effects due to finite-time driving, as reflected by the double time integral in Eq. (7).

Our second main result establishes a link between Eq. (7) and the Kubo relaxation function $\Psi_0(t)$ in Eq. (5). By using similar arguments of Ref. [59] combined with the time-reversal symmetry $\Psi_0(t) = \Psi_0(-t)$, one can re-express Eq. (7) as

$$K(\eta) = -\int_0^\tau dt \int_0^\tau dt' \, \dot{\lambda}_t \dot{\lambda}_{t'} [g_\eta * \Psi_0](t - t'), \tag{9}$$

where

$$g_\eta(t) := \mathcal{F}^{-1}\left[\frac{\sinh[\beta\hbar\omega(1-\eta)/2]\sinh(\beta\hbar\omega\eta/2)}{\beta\hbar\omega \sinh(\beta\hbar\omega/2)}\right](t) \tag{10}$$

is the inverse Fourier transform of a model- and process-independent function [52]. The key insight of Eq. (9) is that the relaxation function now fully characterizes the stochastic thermodynamics of a process for small perturbations, with g_η acting as a universal generating function for the higher order fluctuations via its convolution with Ψ_0. The benefit of this expression is thus that one can now derive a number of general properties of the dissipated work statistics. Firstly, it is straightforward to see from Eq. (9) that the symmetry $K(\eta) = K(1-\eta)$ holds true for all η. This in turn implies, via an inverse Laplace transform, the validity of the *Evan-Searles fluctuation theorem* [60] for $P(W_{\mathrm{diss}})$:

$$\frac{P(W_{\mathrm{diss}})}{P(-W_{\mathrm{diss}})} = e^{\beta W_{\mathrm{diss}}}. \tag{11}$$

While this has been known to apply to systems driven slowly [35] or via time-symmetric driving protocols [61], here its validity is also demonstrated in LRT.

In contrast to what is typically expected for linear response regimes in classical stochastic thermodynamics [62–64], our result predicts a quantum work distribution that is distinctly *non-Gaussian* at finite temperatures. This can be seen by showing that the cumulants higher than the variance (i.e., skewness, kurtosis, etc.) are nonzero. Taking derivatives of Eq. (9) and using the convolution theorem, we obtain that *all* cumulants in dissipated work are positive and given by

$$\beta^k \kappa_W^k = \int_{\mathbb{R}} \frac{d\omega}{\sqrt{2\pi}} \tilde{\Psi}_0(\omega) \gamma^k(\omega) \left| \int_0^\tau dt \, \dot{\lambda}_t e^{i\omega t} \right|^2 \geq 0, \tag{12}$$

where

$$\gamma^k(\omega) := \begin{cases} \frac{1}{2}(\beta\hbar\omega)^{k-1}\coth(\beta\hbar\omega/2) & \text{if } k \text{ even}, \\ \frac{1}{2}(\beta\hbar\omega)^{k-1} & \text{if } k \text{ odd}. \end{cases} \tag{13}$$

This positivity is a consequence of $\tilde{\Psi}_0(\omega) \geq 0$. These relations provide a refined set of constraints on the shape of $P(W_{\mathrm{diss}})$, demonstrating the presence of non-Gaussian tails for values $W_{\mathrm{diss}} > 0$. These right tails have notable physical implications, since their presence and magnitude quantify an increased likelihood of large dissipation accompanying the realizations of the given driving protocol. An analogous non-Gaussian signature has been shown to occur in slowly driven systems [22,35], and also many-body systems driven in finite time across a phase transition [65]. We emphasize the quantum origin of these tails; classical Gaussian behavior can be recovered by either taking a high temperature limit, or assuming a commuting perturbation $[V, H_0] = 0$ [52].

Statistical interpretation of quantum signatures—As a final result, our linear response approach also highlights a quantum signature in the statistics via a *thermodynamic uncertainty relation* (TUR). It is known that distributions satisfying the fluctuations in relation (11) are constrained by the TUR, which in the LRT regime can be expressed in terms of a lower bound,

$$F_W := \frac{\mathrm{var}(W)}{\langle W_{\mathrm{diss}} \rangle} \geq 2k_B T, \tag{14}$$

on the Fano factor (or relative dispersion) of the work distribution [24]. Classically, for a system that remains close to equilibrium such as in LRT [64,66] or slow driving [63,67] one should expect to saturate the TUR. However, we show here that for finite temperatures the TUR cannot be saturated due to the influence of quantum fluctuations, and we provide a clear-cut statistical interpretation of this effect. To achieve this goal, we use $\tilde{\Psi}_0(\omega)$ to define a normalized probability distribution over the continuum of frequencies $\omega \in [0, \infty)$ associated with the system dynamics, which we refer to as *pseudomodes*:

$$\tilde{P}(\omega) := \frac{\tilde{\Psi}_0(\omega)|\int_0^\tau dt\, \dot{\lambda}_t e^{i\omega t}|^2}{\int_0^\infty d\omega\, \tilde{\Psi}_0(\omega)|\int_0^\tau dt\, \dot{\lambda}_t e^{i\omega t}|^2}, \tag{15}$$

where the positivity is guaranteed by thermodynamic consistency [Eq. (6)]. Crucially, it now becomes apparent that the average energies of these modes determine the dispersion of the work distribution. First one can notice that, since $\Psi_0(t) = \Psi_0(-t)$, then $\tilde{\Psi}_0(\omega)$ is necessarily an even function of ω. It is then straightforward to show from Eq. (12) that the Fano factor, Eq. (14), can be expressed as

$$F_W = \langle \hbar\omega \coth(\beta\hbar\omega/2)\rangle_{\tilde{P}} = 2\langle \mathcal{E}_\omega\rangle_{\tilde{P}}, \tag{16}$$

where $\langle\cdot\rangle_{\tilde{P}}$ denotes an average with respect to the pseudomode distribution $\omega \mapsto \tilde{P}(\omega)$ in Eq. (15). We can now recognize the rhs as twice the average total energy $\mathcal{E}_\omega$ of a *quantum harmonic oscillator* at frequency ω and in thermal equilibrium. Interestingly, this is still true even if the original system at hand is not a harmonic system. This highlights a statistical-mechanical connection between the *physical* dissipated-work fluctuations and the *effective* energy distribution of the pseudomodes associated with the driving protocol. Some key properties of this quantity can then be inferred, such as the following inequality:

$$F_W \geq \hbar\langle\omega\rangle_{\tilde{P}} \coth(\beta\hbar\langle\omega\rangle_{\tilde{P}}/2). \tag{17}$$

This follows from Jensen's inequality for the convex function $x \mapsto x\coth(\beta x/2)$, and is tighter than the TUR (14). The genuine quantum origin of this effect is clear since the term $\hbar\langle\omega\rangle$ represents in fact the average *zero-point energy* of the pseudomodes with respect to $\tilde{P}(\omega)$, and is responsible for preventing any saturation of the TUR. In fact, at low temperatures and short times we may approximate $\hbar\omega\coth(\beta\hbar\omega/2) \simeq \hbar|\omega|$ meaning that $F_W \approx \hbar\langle\omega\rangle$ for $\beta\hbar/\tau_R \gg 1$, and hence the distribution will exhibit nonzero dispersion at absolute zero unlike a classical system. We can relate these nonvanishing fluctuations to the *breakdown of the equipartition* theorem in quantum statistical mechanics [68]. Traditionally, this consideration applies to quantum systems in equilibrium and expresses the fact that energy cannot be equally shared amongst all degrees of freedom due to its discrete nature, implying a frequency dependence on the average energy rather than the classical prediction of $\langle\mathcal{E}_\omega\rangle = kT$ [69]. In the present context, we see that a similar breakdown occurs for nonequilibrium processes in linear response, preventing the saturation of the TUR.

Finally, consistency with classical thermodynamics is ensured in the high-temperature–long-time limit $\beta\hbar/\tau_R \ll 1$, since $x\coth(x) \simeq 1$ for $x \ll 1$, and one recovers $F_W \to 2k_BT$. While a similar-in-spirit quantum signature has been derived for slowly driven *Markovian systems* [33,35], the result obtained here goes significantly beyond that as it applies to arbitrary processes driving a system out of equilibrium in finite-time regimes, provided that the perturbation to the local Hamiltonian remains weak.

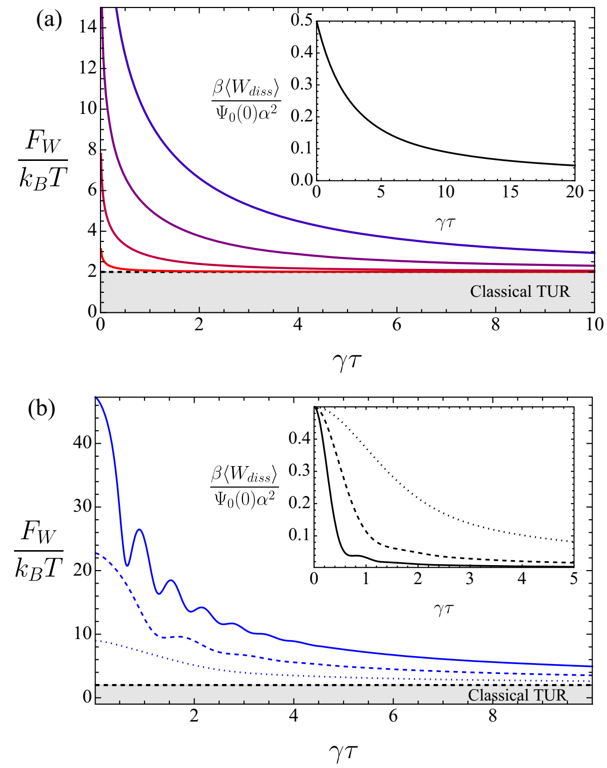

FIG. 1. Fano factor (14) for the work distribution in units of k_BT as a function of the rescaled time $\gamma\tau$. Inset: rescaled dissipated work $\beta\langle W_{\text{diss}}\rangle/[\Psi_0(0)\delta\lambda^2]$ as a function of the rescaled time $\gamma\tau$. Panel (a) depicts the overdamped model $\Psi_0^{(1)}$ in Eq. (18), and the other curves correspond to different values of $\hbar\beta\gamma = 0.5$ (blue), 2 (purple), 5 (magenta), and 10 (red). Panel (b) refers to the underdamped model $\Psi_0^{(2)}$, and the curves correspond to fixed low temperature $\beta = 5$ and different values of the frequency $\nu = 2\gamma$ (blue dotted), 5γ (blue dot-dashed), 10γ (blue dashed), and 15γ (blue solid).

Examples—As mentioned above, one of the most powerful consequences of our main result, Eq. (9), is that it allows one to characterize the full statistics of dissipation in more complex systems, where we may not have solutions to the full Hamiltonian dynamics. This is especially relevant for driven open quantum systems, whose dissipated work's statistics can still be described using our Eq. (9) provided one is simply given a good ansatz or approximation for the relaxation function. Here we benchmark this by considering the following two phenomenological models of Ψ_0, which are thermodynamically consistent according to Eq. (6) [52]:

$$\Psi_0^{(1)}(t) := \Psi_0^{(1)}(0)e^{-\gamma|t|}, \tag{18}$$

$$\Psi_0^{(2)}(t) := \Psi_0^{(2)}(0)e^{-\gamma|t|}\Big(\cos(\nu t) + (\gamma/\nu)\sin(\nu|t|)\Big). \quad (19)$$

In practice, such models can be built from the Kramers-Kronig relations and sum rules by imposing a number of Hamiltonian constraints that adequately characterize the system behavior [70]. In the first instance, $\Psi_0^{(1)}(t)$ provides a reasonable description of an overdamped Brownian dynamics [44], with free parameter $\gamma = 1/\tau_R$ setting the characteristic timescale over which the relaxation function decays. It also can describe many-body quantum systems such as the two-dimensional Ising model in a transverse field [71]. The second model, i.e., $\Psi_0^{(2)}$, includes an oscillatory behavior with an additional degree of freedom ν that quantifies the frequency of an external potential, and should be viewed as a model of underdamped Brownian motion [44]. This model arises, for example, from the quantum dynamics of weakly interacting magnetic systems [72]. A remarkable feature of the linear response regime is that we can predict how the dispersion of the dissipated work distribution changes over time independent of the system-specific features contained in $\Psi_0(0)$. In Fig. 1(a) we plot the dispersion of the overdamped model as a function of time for different temperatures and using a linearly driven protocol. In this case, we see that the correction *monotonically* decays in the long time limit $\gamma\tau \to \infty$, indicating that the quantum fluctuations become less relevant at long times. Conversely, at short times we see a dramatic quantum signature with large dispersion in the work distribution above the classical fluctuation-dissipation relation $F_W = 2k_BT$. While long time decay can be ensured via the damping terms in (18), monotonicity is not necessarily guaranteed. This is clearly seen in the underdamped dynamics in Fig. 1(b), which indeed shows nonmonotonic changes in the dispersion at short times. In the Supplemental Material [52], we also consider a nonexponential model of $\Psi_0(t)$, and use the bound (17) to derive analytic predictions for the Fano factor.

Conclusions—In summary, we have systematically extended a linear response analysis to characterize the full distribution for the quantum dissipated work statistics done along weakly perturbed processes. Our general formula for the CGF, Eq. (9), paves new ways for studying properties of higher order work statistics of complex systems via phenomenological models of the relaxation function. This can be used to explore thermodynamic optimization problems that revolve around stochastic fluctuations such as Pareto optimal work extraction [73] and free energy estimation [74]. The precise connection between the work distribution and relaxation function could also be used to explore the impact of non-Markovianity [75] and phase transitions [76] on work statistics from a general perspective. Our results have also predicted a clear quantum signature (16) that causes an increase in the dispersion of the work distribution at short driving times and finite temperatures, and is universally applicable to composite systems that are driven by local perturbations. Since the Fano factor F_W can be measured experimentally from the work statistics [77], this offers a clear route to detecting truly nonclassical behavior from thermodynamic variables. Finally, it would be interesting to explore generalizations of our formalism to study more general nonequilibrium states [78].

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