## Quantum perspective for the non-equilibrium fluctuation relation and its experimental testing

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Non-equilibrium fluctuation theorems (NFTs) relate work performed on a system as its Hamiltonian varies with time, to equilibrium data of the intial and final states. Testing these remarkable relations is currently restricted to classical systems or to simple quantum systems whose energy state can be directly measured. Here we introduce the concept of a quantum work agent in thermodynamic processes, and suggest that it can allow to probe NFTs in general quantum systems. We elaborate on a simple model in the framework of mesoscopic systems, and discuss future applications.

**Introduction** — Stochastic thermodynamics, as culminated by Jarzynski's equality [1] and the Crooks relation [2], describes non-equilibrium thermodynamics of small systems governed by large fluctuations [3–5]. In classical tests of thermodynamic processes [6, 7], e.g. stretching a single molecule of RNA [8, 9], measurements of the work W yield an intrinsically random result at each realization, but nevertheless allow to verify powerful non-equilibrium fluctuation theorems (NFTs) after gathering sufficient statistics and constructing the work distribution function (WDF) P(W). Quantum extensions of stochastic thermodynamics have been theoretically formulated [4, 10–18], particularly, via the "two-time measurement protocol" [15], which incorporates projective measurements of the energy of the system before and after the non-equilibrium processes. However, measurement of the energy change is not feasible in general systems, such as in many-body systems.

An example of a tunable platform in which one would like to probe NFTs is that of mesoscopic quantum dot systems. Experiments [19–24] in few-electron quantum dot systems have demonstrated NFTs, but their methodology is based on continuous monitoring of the charge state of the QD, and leads to strong backaction, limiting these experiments to the classical regime [25]. Thus, the role of stochastic thermodynamics is basically out of experimental reach except for realizations [26–28] or proposals [29–32] in simple quantum systems.

Furthmore, the "two-time measurement protocol" goes against the ideology of both Themordynamics and Quantum Mechanics. In a thermodynamic formulation, work (W) and heat (Q) are determined via measurements of external bodies. The engineer is probing energy that is transferred to reservoirs  $\mathcal R$  or from work agents  $\mathcal A$ , respectively. The system itself in not measured. Moreover, in Quantum Mechanics, once an initial measurement is done, aka "preparation", the state of the system is no longer that of thermal equilibrium, but a pure state.

**Scope** — In the common formulation of Jarzynski's equality the control parameter X(t), for example the coordinate of a piston, is regarded as a classical coor-

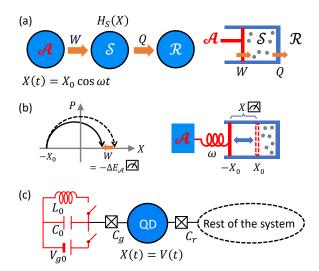


FIG. 1. (a) Thermodynamic processes driven by a work agent  $\mathcal{A}$  performing work on the system  $\mathcal{S}$  via a time dependent parameter X(t) controlling the system's Hamiltonian. Some of it gets dissipated into a reservoir  $\mathcal{R}$ . (b) We generalize the work agent  $\mathcal{A}$  into a dynamical quantum coordinate  $\hat{X}$  which performs work on the system. Its measurement after a half cycle allows to extract work  $W = -\Delta E_{\mathcal{A}}$ . (c) Mesoscopic system:  $\mathcal{S}$  contains a QD. Its energy level  $\epsilon_d(t)$  is driven by an agent  $\mathcal{A}$  realized by an LC circuit such as a microcavity.

dinate, see Fig. 1(a). Here we formulate a quantum demonstration of NFTs which incorporates a dynamical apparatus, referred to as a "quantum work agent" [33]. Of particular interest might be a sweep protocol, say  $X(t) = -X_0 \cos(\omega t)$  for  $t = 0 \to \pi/\omega$ . For the purpose of an experiment we replace X(t) by a quantum dynamical coordinate  $\hat{X}$ , namely, an harmonic oscillator, see Fig. 1(b). The energy of this single degree of freedom can be measured at  $t = \pi/\omega$ , independently of the complexity of the system.

We focus on the feasibility of the quantum work agent concept, considering a generic prototype model. We characterize the ability to extract useful thermodynamic information via Jarzynski's equality, and explore the dependence on generic model parameters. The system is characterized by a typical classical energy scale  $X_0$ , on which we would like to measure work, and a typical quantum energy scale  $\epsilon$  that is related to the internal system dynamics. We distinguish two effects that are ignored within traditional treatment of the NFTs: (a) Back reaction of the system - the dynamics of the system is driven by the work agent coordinate  $\hat{X}(t)$ , but also affects it; (b) Quantum uncertainty of the work agent being a quantum coordinate, the work agent yields an unavoidable uncertainty in the work measurement. In the Born-Oppenheimer limit, where the agent is very heavy, backreaction is minimized. We demonstrate that one can select the parameters of the quantum work agent to minimize also the quantum uncertainty, and then our approach allows to probe the NFTs both in the quasi-static limit and in the nonequilibrium limit.

In the mesoscopic experiments in Ref. [19–21, 23, 24] X(t) is a time dependent voltage applied on a QD, controlling its energy level and driving it across the Fermi level. As a mesoscopic realization of the quantum agent, we propose using an LC circuit, see Fig. 1(c), e.g. a microwave resonator. The measurement is done at the end of the process, only on the external LC circuit (the "agent"), and not on the system. In conclusion we discuss the experimental realization and the novel possibilities that it opens to study stochastic thermodynamics of complex quantum many-body systems.

Two-time protocol for WDF — Consider an external parameter X = X(t), controlling the Hamiltonian  $H_{\mathcal{S}}(X)$  of the quantum system. We assume that the system is initially at thermal equilibrium. One performs projective energy measurements at  $t = t_i$ , and at the end of the process at  $t = t_f$ . Let  $|a\rangle$  and  $|b\rangle$  be eigenstates of  $H_{\mathcal{S}}(X(t_i))$  and  $H_{\mathcal{S}}(X(t_f))$ , with eigenvalue  $E_a^{(i)}$  and  $E_b^{(f)}$ , respectively. According to the two time protocol [15], the WDF is defined as

$$P(W) = \sum_{a,b} p_a |\langle b|U|a\rangle|^2 \delta(W - (E_b^{(f)} - E_a^{(i)})), \quad (1)$$

where  $U = \mathcal{T} \exp \left[ -i \int_{t_i}^{t_f} dt H_{\mathcal{S}}(X(t)) \right]$  is the evolution operator of the closed system, and  $p_a = e^{-E_a^{(i)}/T}/Z_i$ . The initial partition function is  $Z_i = \text{Tr}[e^{-H_{\mathcal{S}}(X(t_i))/T}]$ , and similarly we define the final  $Z_f$ . Jarzynski's equality follows directly from these definition [15, 33], namely,

$$\langle e^{-W/T} \rangle = \int dW P(W) e^{-W/T} = \frac{Z_f}{Z_i} \equiv e^{-\Delta F/T}.$$
 (2)

Remarkably, this identity holds for an arbitrary system and for any non-equilibrium protocol. However, it is unpractical to perform an energy measurement of a many-body quantum system. Below, we determine the work via an energy measurement of an external work agent, which we model as a single degree of freedom oscillator.

**Model system** — We first illustrate the WDF according to the two-time measurement protocol, where X is a classical coordinate. We consider a two level system (TLS) with Hamiltonian

$$H_{\mathcal{S}}(X) = \epsilon \sigma_x + \frac{1}{2}(\sigma_z - 1)X. \tag{3}$$

The instantaneous ground state  $|g\rangle$  and excited state  $|e\rangle$  are schematically shown in Fig. 2(a). The protocol is  $X(t) = -X_0 \cos(\omega t)$  from  $t_i = 0$  to  $t_f = \pi/\omega$ . We start with a thermal state  $\rho_S^{(i)} = (1/Z_i)e^{-H_S(-X_0)/T}$  which can be written as  $p_g|g\rangle\langle g| + p_e|e\rangle\langle e|$ , with probabilities  $p_{g,e} = e^{\pm x}/(e^x + e^{-x})$ , where  $x = \sqrt{\epsilon^2 + (X_0/2)^2}$ . The sweep of X(t) induces a Landau-Zener (LZ) transition, namely,  $|g\rangle \to \sqrt{p_d}|e\rangle + \sqrt{1-p_d}|g\rangle$ , where  $p_d = |\langle e|U|g\rangle|^2$  is the diabatic transition probability. Specifically for  $X_0 \gg \epsilon$ , The well known LZ formula reads  $p_d = e^{-\pi/\alpha}$ , where  $\alpha = \omega X_0/\epsilon^2$ . Accordingly

$$P(W) = \sum_{j=0,1,2} p_j \delta(W + jX_0), \tag{4}$$

which is illustrated in Fig. 2(b). The diabatic peak at W=0 has the weight  $p_0=p_gp_d$ , corresponding to transition form the ground state, as opposed to the thermal peak at  $W=-2X_0$ , that has weight  $p_2=(1-p_g)p_d$ , corresponding to the diabatic transition from the thermal excited state. The adiabtic peak at  $W=-X_0$  is the sum of transitions from both the ground and excited states, and has weight  $p_1=1-p_d$ . In the adiabatic limit  $\alpha\ll 1$  only the adiabatic peak survives, while in the sudden limit  $\alpha\gg 1$  it diminishes. It is easily checked that Eq.(4) satisfies Jarzynski's equality Eq.(2), which is guaranteed in-advance by definition. Note that for large  $X_0$  we get  $Z_f/Z_i\approx e^{X_0/T}$ .

**Quantum work agent** — The variable X in reality is a dynamical coordinate of a work agent. The total Hamiltonian is  $H = H_{\mathcal{S}}(\hat{X}) + H_{\mathcal{A}}$  where

$$H_{\mathcal{A}} = \frac{\omega}{2} \left[ \left( \ell \hat{P} \right)^2 + \left( \frac{\hat{X}}{\ell} \right)^2 \right]. \tag{5}$$

Both  $\hat{X}$  and  $\ell$  have energy units, while  $[\hat{X}, \hat{P}] = 1$ . Our protocol is as follows: (i) We prepare the initial state of the agent in a coherent state at  $X = -X_0$ , decoupled from the system, so that the initial state at  $t_i = 0$  is  $\rho^{(i)} = \rho_{\mathcal{S}}^{(i)} \otimes |-X_0\rangle\langle -X_0|$ , where  $|-X_0\rangle = e^{i\hat{P}X_0}|0\rangle$ . The initial energy of the work agent is  $E_{\mathcal{A}}^{(i)} \approx (1/2)\omega(X_0/\ell)^2$ , where we neglect here and below the numerically negligible zero point energy. Namely, we assume that the oscillator is in a semiclassical state with  $X_0/\ell \gg 1$ . (ii) We let the system and agent evolve according to H till  $t_f = \pi/\omega$ , yielding a final state  $\rho^{(f)} = e^{-iHt_f}\rho^{(i)}e^{iHt_f}$ . After this process, the agent has exchanged energy and got entangled with the system via the thermodynamic process.

(iii) We perform an energy measurement of the agent, rather than that of the system. The energy measurement yields an eigen-energy  $E_{\mathcal{A},n} \approx \frac{\omega}{2}n$  with probability  $p_n^{(f)} = \text{Tr}_{\mathcal{S}}[\langle n|\rho_f|n\rangle]$ , where  $\text{Tr}_{\mathcal{S}}[\cdots]$  is a trace over system degrees of freedom. We define the WDF as

$$P_{\mathcal{A}}(W) = \sum_{n} p_n^{(f)} \delta(W - (E_{\mathcal{A}}^{(i)} - E_{\mathcal{A},n})), \tag{6}$$

We do not perform a projective measurement of the initial energy of the agent because we want it to drive the process as a coherent state. Results are plotted in Fig. 2(c). Although we can identify the diabatic, adiabatic and thermal peaks of P(W), these peaks have been shifted and smeared out in  $P_{\mathcal{A}}(W)$ . Below, we identify the regime within the parameter space  $\{X_0, \ell, \omega, T\}$  where one can accurately use the quantum agent to verify the Jarzynski equality.

Testing the NFT — The above example allows to test the applicability of the NFT within the quantum-work-agent framework. Assume that  $P_{\mathcal{A}}(W)$  of Eq. (6) is experimentally determined, and then used to extract the free energy via  $e^{-\Delta F'/T} = \langle e^{-W/T} \rangle_{\mathcal{A}} = \int dW P_{\mathcal{A}}(W) e^{-W/T}$ . The various distortions of the peaks in Fig. 2(c), result in  $\Delta F' \neq \Delta F$ . To quantify this deviation, in Fig. 3 we plot the quantity  $e^{-(\Delta F' - \Delta F)/T}$  as function of  $\ell$  and T. This quantity tends to unity in the desired validity regime. As we can see that as a function of  $\ell$  there is an intermediate regime of validity of the work agent approach.

The non-monotonic behavior of  $\langle e^{-(W-\Delta F)/T}\rangle_{\mathcal{A}}$  versus  $\ell$  can be understood from the combination of a broadening  $\Delta(\ell)$  and shift  $\delta(\ell)$  of each peak in P(W). Note that the shift is also responsible for the splitting of the adiabatic peak. Consider for the sake of estimate that  $\delta(W+X_0n) \mapsto \frac{1}{\sqrt{\pi}\Delta}e^{-\frac{(W+nX_0-\delta)^2}{\Delta^2}}$ . Then we get  $\left\langle e^{-\frac{W-\Delta F}{T}}\right\rangle = e^{-\frac{\delta(\ell)}{T}}e^{-\frac{\Delta(\ell)^2}{4T^2}}$ . It follows that the validity regime of the NFT is restricted by the condition

$$\{\Delta(\ell), \ \delta(\ell)\} \ll T.$$
 (7)

Next we obtain the following estimates:

$$\Delta(\ell) \approx \frac{1}{2} \omega \frac{X_0}{\ell}, \qquad \delta(\ell) \sim \frac{\ell^2}{\omega}.$$
 (8)

The estimate for  $\Delta(\ell)$  follows from the observation that there is an "error" in W that reflects the quantum uncertainty of X. In order to distinguish the peaks in Fig. 2(c), the width  $\Delta(\ell)$  has to be smaller than T. Irrespective of that, there is a backreaction effect that leads to the shift  $\delta(\ell)$ . Also this shift should be smaller than T. To get the estimate for  $\delta(\ell)$ , consider the adiabatic transition  $|\uparrow\rangle \rightarrow |\downarrow\rangle$ . After the transition the effective Born-Oppenheimer potential in H is shifted, namely it becomes  $V_{\downarrow}(X) = (1/2)\omega(X/\ell)^2 - X$  instead

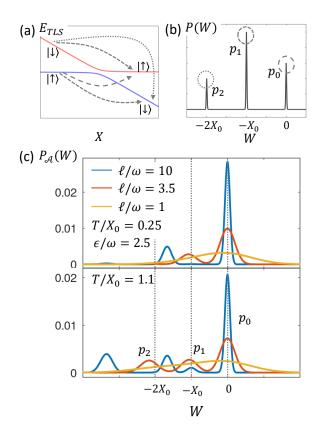


FIG. 2. (a) Energy levels for a TLS  $\mathcal S$  described by Hamiltonian Eq.(3). As a classical work agent  $\mathcal A$  acts on the system by varying X from  $-X_0$  to  $X_0$ , the recorded work gives stochastically one of the values seen in (b), corresponding to a diabatic transition from the thermal state ( $W=-2X_0$ , dotted), adiabatic transitions ( $W=-X_0$ , dashed), and diabatic transition from the ground state (W=0, long dashed). (c) The work agent is now an oscillator with energy quantization  $\omega$  ( $\hbar=1$ ) and coordinate uncertainty  $\ell$ , prepared in a coherent state at position  $-X_0$ . We plot the resulting WDF  $P_{\mathcal A}(W)$  according to Eq.(6) for different  $\ell/\omega$  and  $T/X_0$  denoted with red dots in Fig. 3. We discuss in the main text and in Fig. 3 the regimes in which  $P_{\mathcal A}(W)$  gives a good approximation to P(W) which allows to verify the fluctuation-dissipation theorems.

of  $V_{\uparrow}(X)=(1/2)\omega(X/\ell)^2$ . The positive turning point X' is implied by energy conservation  $V_{\downarrow}(X')=V_{\uparrow}(-X_0)$  and satisfies  $X'>X_0$ . We refer to this deviation as backreaction. Consequently the energy measured by the agent is shifted by  $\delta(\ell)\approx \ell^2/\omega$ . Similarly it can be seen that the thermal peak shifts by  $\delta(\ell)\approx 2\ell^2/\omega$ . In both cases we ignore a negligible change in the turning point time.

The two inequalities of Eq.(7) are plotted by red dashed lines in the regime diagram in Fig. 3 and are highly consistent with the simulations of the protocol. Note that for  $T \sim X_0$  the validity regime is

$$1 \ll \ell/\omega \ll \sqrt{X_0/(2\omega)},\tag{9}$$

which is located between the region washed out by the quantum uncertainty of the energy of A and the region

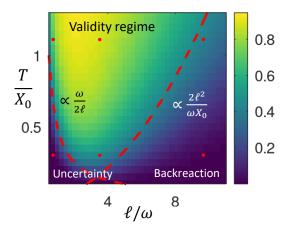


FIG. 3. Regime diagram: we plot  $e^{-(\Delta F - \Delta F')/T}$  for  $X_0/\omega = 150$  and  $\epsilon/\omega = 2.5$ , or  $\alpha = 24$ . The red dashed curves correspond to Eq.(7), which identifies the validity regime of the work agent approach. In Fig. 2(b) we have plotted the WDFs  $P_A(W)$  for the marked red dots. In the "validity regime" both the uncertainty of the agent's coordinate, and the backreaction of the system onto the agent, are small.

with strong backreaction in Fig. 3. The validity regime in Fig. 3 corresponds to the case where  $P_{\mathcal{A}}(W)$  nicely approximates P(W) as seen in the red curves in Fig. 2(b). The WDF in Fig. 2(b) has a dominating diabatic peak at  $W \sim 0$ , which shows that the quantum work agent can operate near the sudden limit where the system is driven strongly out of equilibrium.

Experimental realization — Consider the thermodynamic process of ramping up or down an energy level of a QD coupled to the rest of the system (ROTS), see Fig. 1(c) as in Refs. [19–21, 23, 24], with Hamiltonian  $H_S = X(t)\hat{n}_{\rm QD} + H_t + H_r$ , where  $H_t$  describes tunneling between the QD and the ROTS which has Hamiltonian  $H_r$ . Our TLS example is realized if the ROTS is another QD, where  $\hat{n}_{\rm QD} = c_1^{\dagger}c_1$ , and  $H_t = \epsilon c_1^{\dagger}c_2 + h.c.$ , while  $H_r = vc_2^{\dagger}c_2$  with a single electron residing in the double QD, namely  $\sum_{i=1,2} c_i^{\dagger}c_i = 1$ . We now replace the time dependent gate voltage X(t) by a dynamical variable  $\hat{X} = e\hat{Q}/C_G$ , where  $Q = C_0V$  is the charge of a capacitor of an LC-circuit, and  $C_G = (C_g + C_r)C_0/C_g$ , see Fig. 1(c). The Hamiltonian is

$$H_{\mathcal{A}} = \left[ \frac{1}{2C_0} \hat{Q}^2 + \frac{c^2}{2L_0} \hat{\Phi}^2 \right], \tag{10}$$

where  $[\hat{Q}, \hat{\Phi}] = i$ . Comparing with Eq. (5) we identify  $\omega = 1/\sqrt{L_0C_0}$  and  $\ell^2 = \omega e^2C_0/C_G^2$ . In order to probe the NFT using this LC-circuit we have to satisfy Eq.(9), leading to  $\omega \ll (C_0/C_G)^2[e^2/C_0]$ , and initial voltage  $V_0 \gg e/C_G$ .

The LC circuit could be a microwave resonator as in a recent experiment [34]. One realizes that our protocol involves an experimental challenge. It requires to:

(i) prepare a coherent state in the LC but initially keep it decoupled from the system, which is prepared at a thermal equilibrium with fixed voltage; (ii) start "suddenly" the sweep process by coupling  $\mathcal S$  to  $\mathcal A$ , see switches in Fig. 1(c); and (iii) measure "instantly" the energy of  $\mathcal A$ . In practice the switches have to be fast only compared to the typical scales of the system. For a double quantum dot this would be the maximum of the tunneling rate  $\epsilon$  and dephasing rate which is on the order of GHz [34]. Such time control can be achieved using superconducting qubit technologies.

Exploring Many body physics — The same agent Hamiltonian can be used for probing a general manybody quantum system. Considering Refs. [19–21, 23, 24], the ROTS of Fig. 1(c) is a spinful metallic lead with  $\hat{n}_{\mathrm{QD}} = \sum_{\sigma=\uparrow,\downarrow} d_{\sigma}^{\dagger} d_{\sigma}$ , and  $H_{\mathcal{S}} = \epsilon_{d} \hat{n}_{\mathrm{QD}} + U d_{\uparrow}^{\dagger} d_{\uparrow} d_{\downarrow}^{\dagger} d_{\downarrow} +$  $\sum_{k,\sigma} [\epsilon_k c_{k\sigma}^{\dagger} c_{k\sigma} + t(c_{k\sigma}^{\dagger} d_{\sigma} + h.c.)]$ . The WDF in such manybody systems can be constructed theoretically by applynig various techniques based on the sudden limit [16], linear response [35], and near equilibrium [36], particularly using Green function methods [37]. By coupling this Anderson model to the LC-circuit we can study the WDF in a process connecting two different many-body states, say, an empty state  $n_{QD} = 0$  with a Kondo state at  $n_{QD} = 1$ . As the potential X is swept from  $-X_0$  to  $X_0$ , an electron enters the QD at some X'. The work done is  $W = -X_0 - X'$  with a continuous WDF with typical range of  $\mathcal{O}(2X_0)$ . With a similar reasoning regarding the broadening and shift of the work agent measurement, as long as Eq. (7) is satisfied  $P_{\mathcal{A}}(W)$  gives a good approximation for P(W) even in many-body systems.

Summary — We have introduced a strategy to measure work performed on arbitrary quantum systems, employing a single-coordinate quantum object that plays the role of a "work agent". We illustrated our protocol using an elementary example of a TLS which is trivial in the sense that the two time protocol can be directly realized [27, 28]. Still, this example features the generic aspects of testing NFTs for quantum systems.

There are numerous applications of the possibility to measure work in stochastic processes. For example, by measuring the dissipated work, one can extract relative entropy [38–40], which have been shown to be linked with entanglement in many-body systems [41, 42]. Consequently we have discussed the actual experimental realization of the simple model, as well as the potential applications for exploring many-body physics in QDs realizing the Kondo effect. Even more interestingly, by changing the QD energy one can control an effective magnetic field on the Kondo impurity [43]. These experimentally feasible charge Kondo systems [44] become critical when the ROTS contains multiple leads, yielding highly nontrivial thermodynamics, as seen e.g. by its entropy [45]. The present study could allow to test nontrivial predictions about stochastic thermodynamcis in these systems [46].

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