Exploring quasiprobability approaches to quantum work in the presence of initial coherence: Advantages of the Margenau-Hill distribution

Ji-Hui Pei (裴继辉) [1], Jin-Fu Chen (陈劲夫) [1,*] and H. T. Quan (全海涛) [1,2,3,†]

[1]*School of Physics, Peking University, Beijing 100871, China*
[2]*Collaborative Innovation Center of Quantum Matter, Beijing 100871, China*
[3]*Frontiers Science Center for Nano-optoelectronics, Peking University, Beijing 100871, China*

In quantum thermodynamics, the two-projective-measurement (TPM) scheme provides a successful description of stochastic work only in the absence of initial quantum coherence. Extending the quantum work distribution to quasiprobability is a general way to characterize work fluctuation in the presence of initial coherence. However, among a large number of different definitions, there is no consensus on the most appropriate work quasiprobability. In this article, we list several physically reasonable requirements including the first law of thermodynamics, time-reversal symmetry, positivity of second-order moment, and a support condition for the work distribution. We prove that the only definition that satisfies all these requirements is the Margenau-Hill (MH) quasiprobability of work. In this sense, the MH quasiprobability of work shows its advantages over other definitions. As an illustration, we calculate the MH work distribution of a breathing harmonic oscillator with initial squeezed states and show the convergence to classical work distribution in the classical limit.

I. INTRODUCTION

In stochastic thermodynamics, the studies of thermodynamics were extended from the ensemble average level to the stochastic trajectory level [1,2], where work and other thermodynamic concepts are identified as stochastic quantities [3]. These stochastic quantities were found to satisfy some interesting and strong relations known as fluctuation theorems [4]. On even more microscopic scales, quantum effects play a crucial role in thermodynamic phenomena [5–12]. Due to the absence of the concept of trajectory in quantum mechanics, it is a necessary but challenging task to define the quantum stochastic work [13]. After some early efforts on the operator-of-work scheme [14,15], the stochastic work defined by the two-projective-measurement (TPM) scheme [16,17] is now the standard way provided no quantum coherence in the initial state. Based on this definition, the fluctuation theorems are readily recovered when the quantum system is driven from canonical equilibrium. The TPM scheme is also widely used to define other quantum thermodynamic quantities, including heat and particle number exchange [18]. The consequent quantum thermodynamic theory has flourished in both theoretical and experimental aspects [5–12,19,20].

However, the TPM scheme fails to characterize the quantum stochastic work when the initial state of the system contains quantum coherence, that is, the initial density matrix does not commute with the initial Hamiltonian. The first measurement process becomes invasive and destroys the coherence of the initial state [21,22]. Quantum coherence indeed brings some advantages to quantum thermodynamics. It has been shown that quantum coherence can improve the performance of quantum heat engines [23–30] and can be used to enhance the work extraction [31,32]. To characterize the fluctuation with initial coherence, a generalized definition of stochastic work is needed, which should recover the standard result based on the TPM scheme if there is no initial coherence. Unfortunately, the no-go theorem for quantum work prohibits a genuine probability framework to reconcile the first law and the reproduction of the TPM result [33]. A possible resolution is to adopt the quasiprobability for the work distribution, which allows the distribution to take negative or complex values. It is found that the work quasiprobability is a direct signature of quantum contextuality [34].

Quasiprobability distributions arise naturally in the study of quantum mechanics when treating noncommutative observables [35–38]. Recently, there has been growing interest in studying the quasiprobability of work in quantum thermodynamic phenomena [39,40]. So far, there have been several different proposals on the definitions of quantum work. For example, the Kirkwood-Dirac (KD) quasiprobability of quantum work [41–43] and the Margenau-Hill (MH) quasiprobability of work [44] are based on famous quasiprobability distributions [36,38], which are widely used in quantum optics. The full-counting (FC) quasiprobability of work [45] is another kind of quasiprobability, which is related to full counting statistics [46]. Most recently, a class of quasiprobability distributions that generalizes the MH and FC ones was proposed [47]. A natural question arises: Which definition of quantum work quasiprobability is the most appropriate one?

In this article, we propose several requirements for quantum work and further show that only the MH quasiprobability

*chenjinfu@pku.edu.cn
†htquan@pku.edu.cn

of work meets all these requirements. Hence, it is the most appropriate definition of quantum stochastic work. This paper is organized as follows. In Sec. II, we first briefly review the TPM scheme and the no-go theorem for quantum work. In Sec. III, we list the requirements for an appropriate quantum work and discuss their physical implications. Among these requirements, the support condition is the most conspicuous one. In Sec. IV, we prove the only definition that satisfies all the requirements is the MH quasiprobability of work. In Sec. V, a comparison of some definitions is discussed from the perspective of these requirements. In Sec. VI, we calculate the MH work quasiprobability for a breathing quantum harmonic oscillator starting from a squeezed state and show the quantum-classical correspondence principle for MH work distribution. In Sec. VII, we briefly mention several approaches to measuring the MH quasiprobability of work experimentally. In Sec. VIII, we sketch how the MH quasiprobability can be generalized to open quantum systems.

II. A REVIEW OF NO-GO THEOREM FOR QUANTUM WORK

The isolated driving process is one of the basic processes in the study of nonequilibrium statistical mechanics. Let us consider a nonequilibrium quantum driving process governed by the Schrödinger equation during the time interval $[0, \tau]$. The time-dependent Hamiltonian is $H_S(t), 0 \leqslant t \leqslant \tau$, and the initial density matrix for the system is denoted as ρ. Here we use the subscript S in the Hamiltonian to denote the Schrödinger picture. For simplicity, we set the reduced Planck constant $\hbar = 1$ except in Sec. VI. The time evolution of the system is characterized by the time-evolution operator $U(\tau) = \mathcal{T} \exp(-i \int_0^\tau dt H_S(t))$, where $\mathcal{T}$ is the time-ordering operator. In the Heisenberg picture, the Hamiltonian operator is $H_H(t) = U(t)^{-1} H_S(t) U(t)$ while the quantum state remains invariant. In the later discussion, we will mostly work in the Heisenberg picture and adopt several shorthand notations: $H_0 = H_H(0)$, $H_1 = H_H(\tau)$, $U = U(\tau)$. We diagonalize the initial and final Hamiltonians in the Heisenberg picture as $H_0 = \sum_n \epsilon_n^0 \Pi_n^0$ and $H_1 = \sum_n \epsilon_n^1 \Pi_n^1$, where Π_n^0, Π_n^1 are projective operators corresponding to eigenvalues $\epsilon_n^0, \epsilon_n^1$.

Under the stochastic description, work performed in this process is a random variable, denoted by W. The corresponding probability distribution function is $P(w)$, which is related to the probability by $\Pr(a < W \leqslant b) = \int_a^b P(w) dw$. If the initial state does not contain quantum coherence, that is, $[\rho, H_0] = 0$, then the standard two-projective measurement (TPM) gives the following quantum work probability distribution:

$$P_{\text{TPM}}(w) = \sum_{ij} p_{ji} p_i \delta(w - (\epsilon_j^1 - \epsilon_i^0))$$
$$= \sum_{ij} \text{Tr}[\Pi_j^1 \Pi_i^0 \rho \Pi_i^0 \Pi_j^1] \delta(w - (\epsilon_j^1 - \epsilon_i^0)). \quad (1)$$

The above expression has a clear physical meaning. $p_i = \text{Tr}[\Pi_i^0 \rho \Pi_i^0]$ is the probability of the ith energy level when measuring the initial Hamiltonian. Supposing $\tilde{\rho}_i = \Pi_i^0 \rho \Pi_i^0 / \text{Tr}[\Pi_i^0 \rho \Pi_i^0]$ is the collapsed state after the first measurement, $p_{ji} = \text{Tr}[\Pi_j^1 \tilde{\rho}_i \Pi_j^1]$ is the transition probability from

the ith initial energy level to the jth final energy level while doing the second measurement after the time evolution. Since the system is isolated, the work is identified as the energy difference $\epsilon_j^1 - \epsilon_i^0$ between two measurements with the associated probability $p_{ji} p_i$ of this realization.

For the states without initial coherence, the first measurement does not perturb the overall quantum state. In this case, the TPM work distribution is consistent with the first law of thermodynamics as well as work fluctuation theorems. However, in the presence of initial quantum coherence, the first measurement destroys the initial state, and the off-diagonal terms of the density matrix in the energy basis are eliminated. Due to its invasive nature, the TPM scheme for quantum work fails to describe a process with initial coherence. As a direct consequence, the average work defined by the TPM scheme is in contradiction with the first law of thermodynamics. Despite the success of quantum thermodynamics based on the TPM scheme, its validity is restricted to only a small portion of initial states.

Furthermore, a famous no-go theorem shows that under a genuine probability framework, there is no way to generalize the TPM scheme to incorporate initial coherence [33]. In Ref. [33], a genuine probability distribution implies the following three constraints on $P(w)$. The first two constraints come from the axioms of probability theory and are easy to understand: $P(w) \geqslant 0$, and $\int_{-\infty}^{+\infty} P(w) dw = 1$. The third constraint is a property regarding the initial state known as convexity: For any $0 \leqslant \lambda \leqslant 1$, the distribution satisfies $P(w; \lambda \rho_1 + (1 - \lambda)\rho_2) = \lambda P(w; \rho_1) + (1 - \lambda)P(w; \rho_2)$. An explanation of convexity is given in the next section. The no-go theorem for quantum work [33] states that the following two requirements cannot be simultaneously fulfilled under the above genuine probability framework. One is the first law of thermodynamics for the average work, $\langle W \rangle = \text{Tr}[(H_1 - H_0)\rho]$. The other is the reduction to the standard TPM result for initial states without coherence. These two requirements are both very important in quantum thermodynamics but contradict each other.

III. REQUIREMENTS FOR QUASIPROBABILITY OF QUANTUM WORK

The quasiprobability approach abandons the positivity assumption of the probability distribution function to resolve the contradiction. In the quasiprobability framework, the distribution function $P(w)$ could take negative or even complex values. Other two constraints, namely, the normalization relation and the convexity still hold. We summarize here the modified conditions for a quasiprobability distribution.

(W0) Distribution function is complex-valued (or real-valued) and normalized

$$P(w) \in \mathbb{C},$$
$$\int P(w) dw = 1. \quad (2)$$

(W1) The distribution function exhibits convexity with respect to the initial density operator: For any $0 \leqslant \lambda \leqslant 1$,

$$P(w; \lambda \rho_1 + (1 - \lambda)\rho_2) = \lambda P(w; \rho_1) + (1 - \lambda)P(w; \rho_2). \quad (3)$$

This convexity condition originates from the nature of the classical probability because the mixed density matrix $\lambda\rho_1 + (1-\lambda)\rho_2$ is equivalent to randomly choosing the initial state from ρ_1 and ρ_2 with (classical) probability λ and $1-\lambda$, respectively, and the result for $\lambda\rho_1 + (1-\lambda)\rho_2$ should be a direct combination of two independent parts.

Since the average work in quantum mechanics is well-defined and has been widely used for a long time before the concept of stochastic work, there is no doubt that the first law of thermodynamics at the ensemble average level should be a criterion.

(W2) The first law of thermodynamics for average work:

$$\int wP(w)dw = \mathrm{Tr}[(H_1 - H_0)\rho]. \qquad (4)$$

Physically, the first law requires the average work to be the average of the energy change.

Another condition that appears in the formulation of no-go theorem for quantum work is the reproduction of TPM scheme, which requires the quantum work to match the standard definition by TPM scheme in the case when the initial state contains no quantum coherence. The achievements of quantum thermodynamics such as the quantum fluctuation theorems based on the TPM scheme serve as a justification. Nevertheless, this requirement is not directly used in the later proof but appears as a consequence of other requirements. We regard it as an extra condition that is labeled as requirement (E1) in this paper.

(E1) Reproduction of TPM scheme:

$$P(w) = P_{\mathrm{TPM}}(w), \quad \text{if } [\rho, H_0] = 0. \qquad (5)$$

Before listing other requirements, it is meaningful to introduce the consequences of the above requirements. The convexity shows the distribution exhibits a linear dependence on the initial state, and we assert that $P(w)$ can be expressed as

$$P(w) = \mathrm{Tr}[M(w)\rho], \qquad (6)$$

where $M(w)$ is a bounded operator irrelevant to ρ. This expression is intuitively valid because the right-hand side also keeps the linear property on ρ. Strict proof is given in Appendix A based on functional analysis.

As illustrations, we summarize the expressions of $M(w)$ in some typical definitions for quantum work. From Eq. (1), $M(w)$ for the TPM work distribution is

$$M_{\mathrm{TPM}}(w) = \sum_{ij} \Pi_i^0 \Pi_j^1 \Pi_i^0 \delta\big(w - (\epsilon_j^1 - \epsilon_i^0)\big). \qquad (7)$$

A q class of quasiprobability distributions proposed in Refs. [47,48] generalizes the MH work quasiprobability distribution and FC work quasiprobability distribution. $M(w)$ for q class is

$$M_q(w) = \sum_{ijk} \frac{1}{2}\big[\Pi_k^0 \Pi_j^1 \Pi_i^0 + \Pi_i^0 \Pi_j^1 \Pi_k^0\big]$$

$$\times \delta\big(w - [\epsilon_j^1 - q\epsilon_i^0 - (1-q)\epsilon_k^0]\big), \qquad (8)$$

with a parameter $0 \leqslant q \leqslant 1/2$. The cases with $q = 0$ and $q = 1/2$ reduce to the MH and FC work quasiprobability distributions, respectively.

For initial states without coherence, $[\rho, \Pi_n^0] = 0$, all the above distribution functions coincide with each other. Besides, we can easily verify the first law for the quasiprobability in q class. Thus, in contrast to the no-go theorem, as we abandon the positivity of probability, we arrive at a multiple-choice result: there are many work quasiprobability distributions that satisfy both the first law (W2) and the reproduction of the TPM scheme (E1).

In addition to the above conditions (W0)–(W2) and (E1), from physical consideration, we list several other requirements. They are later used to demonstrate the validity of the MH quasiprobability of work.

The following requirement plays the core role in this paper, which roots in our physical understanding of the concept of work.

(W3) Support condition: The distribution function $P(w)$ is solely determined by H_0, H_1, and ρ. If $P(w) \neq 0$, then there exists i, j such that $w = \epsilon_j^1 - \epsilon_i^0$. And if $k \neq i$, $l \neq j$ for all possible i, j, then $P(w)$ does not depend on ϵ_k^0, Π_k^0 and ϵ_l^1, Π_l^1.

Here is an explanation of this requirement. As is shown in most proposals, the quantum work is viewed as a quantity related to the initial and final Hamiltonians in the Heisenberg picture, H_0 and H_1, with no regard to the evolution details during the process. We adopt this as an assumption for an isolated quantum process; work only depends on H_0, H_1, and the initial state ρ. The distribution function can be explicitly written as $P(w; H_1, H_0, \rho)$. Furthermore, inspired by the physical meaning of the TPM scheme, we assume that the stochastic work characterizes the energy-level transition. Since the energy is distributed only at the eigenvalues, the support (nonzero points) of $P(w)$ is restricted to the differences between the final and initial energy eigenvalues. That is, $P(w) \neq 0$ implies w can be expressed as $\epsilon_j^1 - \epsilon_i^0$. Also, we expect that the transition probability between two energy levels has nothing to do with other energy levels. In consideration of possible degeneracy of the energy difference, let us suppose w belongs to the support of $P(w)$, and (i_1, j_1), (i_2, j_2), ... are all the pairs such that $w = \epsilon_{j_m}^1 - \epsilon_{i_m}^0$. If $k \neq i_m$ for any m, then $P(w)$ has nothing to do with the kth initial energy level, and $P(w)$ does not depend on ϵ_k and Π_k^0. This is also true for the lth final energy level if $l \neq j_m$ for any m. This accounts for the last sentence in (W3).

In this paper, we do not directly require the reproduction of the TPM scheme (E1) but try to extract the physical implication behind the TPM scheme and introduce the support condition (W3). As we will see, the reproduction of the TPM scheme will emerge as a consequence of other requirements.

Furthermore, there should be a linear relation between work and Hamiltonian, which was explicitly proposed in Ref. [48]. Let us consider a global rescaling transform of the time-dependent Hamiltonian, $H_H'(t) = \lambda H_H(t)$. While ρ is fixed, we expect W' in this rescaled protocol to exhibit the same distribution as λW. In another transform, the Hamiltonian is shifted by a c-number (classical number) $H_H'(t) = H_H(t) + e(t)$, which corresponds to different choices of the zero point of the energy. In this case, we naturally expect W' to exhibit the same distribution as $W + e(\tau) - e(0)$. Combining these two kinds of transforms, we have the following requirement.

(W4) A linear rescaling condition on the Hamiltonian: Under the rescaling transform $H_H(t) \rightarrow \lambda H_H(t) + e(t)$ with fixed initial state, the work rescales as $W \rightarrow W' \overset{d}{=} \lambda W + e(t) - e(0)$. Here, $\overset{d}{=}$ means both sides have the same distribution.

The zeroth-order moment and the first-order moment are fixed by the normalization relation and the first law, respectively. Besides, in stochastic thermodynamics, the amplitude of fluctuation can be quantified by the variance, which should be always nonnegative. Equivalently, the second-order moment is also nonnegative. We adopt this as a requirement of work distribution for arbitrary initial states.

(W5) Positivity of the second-order moment:

$$\langle W^2 \rangle = \int w^2 P(w) dw \geqslant 0. \tag{9}$$

The time-reversal symmetry is the last requirement for $P(w)$, which was proposed in Ref. [49]. In the standard formulation, the time-reversed process of an original process is implemented as follows. The antiunitary time-reversal operator Θ is involved, and we will use an overline to denote quantities in the time-reversed process. In the Schrödinger picture, the initial state is set to be the time-reversed final state in the original process $\bar{\rho}(0) = \Theta \rho(\tau) \Theta^{-1}$. The time-reversed Hamiltonian is also equipped with the time-reversal operator, and the time parameter in the Hamiltonian evolves backward: $\bar{H}_S(t) = \Theta H_S(\tau - t) \Theta^{-1}$. In the time-reversed process, thermodynamic quantities including work should exhibit a minus sign compared to the original process according to the time-reversal symmetry.

(E2) Time-reversal symmetry for work: In the time-reversed process, the corresponding work $\bar{W}$ has the same distribution as the minus work $-W$ in the original process:

$$\bar{P}(w) = P(-w). \tag{10}$$

The time-reversal symmetry plays a crucial role in stochastic thermodynamics, especially in the formulation of fluctuation theorems [4]. Requirement (E2) is indispensable for an appropriate work quasiprobability. In the proof in the next section, it will not be directly used but appear as the consequence of other requirements, so we label it as (E2).

IV. VALIDITY OF MARGENAU-HILL QUASIPROBABILITY OF WORK

After the above preparations, we arrive at *our main result*: In an isolated driving process, the MH quasiprobability of work is the only one satisfying all the requirements (W0)–(W5) among all the possible definitions. Meanwhile, the MH quasiprobability of work also satisfies other two requirements (E1)–(E2).

A. Sketch of the proof

We denote the distribution function as $P(w; H_1, H_0, \rho)$. According to the corollary of the convexity condition [shown in Eq. (6)], we have proved that the distribution function can be expressed as $P(w; H_1, H_0, \rho) = \text{Tr}[M(w; H_1, H_0)\rho]$. The proof of Eq. (6) can be found in Appendix A.

Due to the support condition (W3), $P(w)$ only distributes at $\epsilon_j^1 - \epsilon_i^0$, so

$$M(w; H_1, H_0) = \sum_{ij} \delta\big(w - \big(\epsilon_j^1 - \epsilon_i^0\big)\big) N_{ji}. \tag{11}$$

Here the operator N_{ji} is solely dependent on H_1, H_0, which is to be determined in Eq. (12). As operators are fully determined by their eigenvalues and corresponding projectors, we can regard N_{ji} as a function of all the eigenvalues and projectors, namely $\{\epsilon_k^1\}, \{\epsilon_k^0\}, \{\Pi_k^1\}, \{\Pi_k^0\}$. Since the above requirements should be satisfied by all kinds of processes, for simplicity, we assume the energy difference is nondegenerate, that is, $\epsilon_j^1 - \epsilon_i^0$ are different for different pairs of (i, j). Again from (W3), only the relevant energy levels and corresponding $\epsilon_j^1, \epsilon_i^0, \Pi_j^1, \Pi_i^0$ contribute to the work distribution when considering N_{ji}. According to the rescaling requirement (W4), when the eigenvalues $\epsilon_j^1, \epsilon_i^0$ are shifted, the amplitude of the distribution function remains invariant. Therefore, N_{ji} should not depend on eigenvalues, and eventually, $N_{ji}(\Pi_j^1, \Pi_i^0)$ is a function of two projective operators. We assume it to be an analytical function, which can be expanded as the following series with coefficients e_0, a_n, b_n, c_n, d_n:

$$N_{ji}\big(\Pi_j^1, \Pi_i^0\big) = e_0 + \sum_{n=1}^{\infty} a_n \big(\Pi_j^1 \Pi_i^0\big)^n + \sum_{n=1}^{\infty} b_n \big(\Pi_i^0 \Pi_j^1\big)^n$$
$$+ \sum_{n=0}^{\infty} c_n \big(\Pi_i^0 \Pi_j^1\big)^n \Pi_i^0 + \sum_{n=0}^{\infty} d_n \Pi_j^1 \big(\Pi_i^0 \Pi_j^1\big)^n. \tag{12}$$

In each term of the expansion, two different projective operators appear alternatively.

Next, we compare the average of the work to the result from the first law of thermodynamics (W2),

$$\langle W \rangle = \text{Tr}\left[\rho \sum_{ij} \big(\epsilon_j^1 - \epsilon_i^0\big) N_{ji} \right] = \text{Tr}[\rho(H_1 - H_0)]. \tag{13}$$

Since the first law is valid for any state ρ, we get the following relation:

$$\sum_{ij} \big(\epsilon_j^1 - \epsilon_i^0\big) N_{ji} = H_1 - H_0. \tag{14}$$

The next step is to use the above condition to determine the coefficients in the expansion in Eq. (12). This is accomplished by constructing a specific set of projective operators. The detailed treatment is included in the Appendix B. The result shows that the remaining possible choice for N_{ji} is

$$N_{ji} = \frac{1 + \eta}{2} \Pi_j^1 \Pi_i^0 + \frac{1 - \eta}{2} \Pi_i^0 \Pi_j^1. \tag{15}$$

Here η is an arbitrary complex number η. Also, in the Appendix B, we show that the quasiprobability of work defined by the above N_{ji} satisfies the time-reversal symmetry (E2), regardless of the value of the complex number η.

In the last step of the proof, we will show that only when $\eta = 0$, the second-order moment is positive. Direct

TABLE I. Comparison of different definitions of work distributions: ✓ is used only if the requirement is fulfilled under all the possible processes.

	Value (W0)	Linearity on $\rho, H(t)^a$ (W1) (W4)	First law (W2)	Support condition (W3)	Reproduce TPM result (E1)	Time-reversal symmetry (E2)	Positivity of second moment (W5)
Kirkwood-Dirac [41,42]	Complex	✓	✓	✓	✓	✓	×
Margenau-Hill [44]	Real[b]	✓	✓	✓	✓	✓	✓
Full-counting [45]	Real	✓	✓	×	✓	×	✓
q Class ($0 < q < 1/2$) [47]	Real	✓	✓	×	✓	×	✓
Consistent histories [49]	Real	✓	✓	×	×	✓	✓
Operator of work [14,15]	Nonnegative	✓	✓	×	×	✓	✓
TPM [16,17]	Nonnegative	✓	×	✓	✓	×	✓
Hamilton-Jacobi [50][c]	Nonnegative	×	✓	×	×	✓	✓

[a]In this condition, the convexity on initial state (W1) and the linearity with respect to the Hamiltonian (W4) are combined.
[b]"Real" implies possibly negative.
[c]This definition is also known as quantum work in the Bohmian framework.

calculation yields

$$\langle W^2 \rangle = \int w^2 P(w)\, dw$$
$$= \mathrm{Tr}[(H_1 - H_0)^2 \rho] - \eta \mathrm{Tr}[(H_1 H_0 - H_0 H_1)\rho]. \quad (16)$$

If $\eta = 0$, then $\langle W_{\mathrm{MH}}^2 \rangle = \mathrm{Tr}[(H_1 - H_0)^2 \rho]$ is always nonnegative.

For any $\eta \neq 0$, we suppose $H_1 = H_0 + L$, where L is a Hermitian operator characterizing the difference. The expression for $\langle W^2 \rangle$ changes to

$$\langle W^2 \rangle = \mathrm{Tr}(L^2 \rho) - \eta \mathrm{Tr}([L, H_0]\rho). \quad (17)$$

The positivity of the second-order moment should hold for an arbitrary protocol, so we have the freedom to select the value of L, ρ. The first term depends on L quadratically while the second term linearly. Therefore, as long as $\eta \neq 0$, for sufficiently small L, the second term will be dominant, which is not positive definite. As a result, $\langle W^2 \rangle$ cannot always be positive if $\eta \neq 0$.

As an explicit example, $H_0 = \sigma_z$, $L = \nu \sigma_x$, $[L, H_0] = -2\nu i \sigma_y$, where σ_i are Pauli matrices and $0 < \nu \ll \eta$ is a tiny number. By choosing density operators $\rho = \frac{1}{2}(I \pm \sigma_y)$, we can make

$$-\eta \mathrm{Tr}[[L, H_0]\rho] = \pm 2i\eta\nu. \quad (18)$$

Since the term $\mathrm{Tr}[L^2 \rho] = \nu^2$ is negligible, $\langle W^2 \rangle$ can be either complex if $\mathrm{Re}\,\eta \neq 0$ or negative if $\mathrm{Re}\,\eta = 0$.

In conclusion, the following distribution function corresponding to $\eta = 0$ is uniquely determined,

$$P_{\mathrm{MH}}(w) = \frac{1}{2} \sum_{j,i} \mathrm{Tr}\big[\big(\Pi_j^1 \Pi_i^0 + \Pi_i^0 \Pi_j^1\big)\rho\big]\delta\big(w - (\epsilon_j^1 - \epsilon_i^0)\big). \quad (19)$$

This is the familiar Margenau-Hill work quasiprobability [44]. For initial states without coherence, it reduces to the standard TPM scheme. This manifests that the requirements in this paper also justify the TPM scheme in the absence of initial coherence.

V. COMPARISON OF SEVERAL DEFINITIONS

In Table I, we compare different definitions of quantum work and test the requirements for each of them. As is shown in the table, the MH work quasiprobability is the only definition fulfilling all the requirements.

Definitions with positive distribution face severe flaws. In accordance with the no-go theorem, at least one of the following conditions is violated: the first law, the reproduction of TPM, or the convexity. For detailed review of positive-valued work probabilities including Quantum Hamilton-Jacobi [50], Gaussian measurements [13], POVM depending on the initial state [51], please refer to Ref. [52].

In what follows, we will mainly focus on quasiprobability distributions. According to Eq. (8), the quasiprobability distributions in q class ($0 < q \leqslant 1/2$, including FC) demonstrate an asymmetry between the initial and the final Hamiltonians. The initial projector appears twice but the final projector only once. Therefore, the time-reversal symmetry is violated unless $q = 1/2$. Besides, the q class ($0 < q \leqslant 1/2$) does not satisfy the support condition. The work distributes at $\epsilon_j^1 - [q\epsilon_i^0 + (1-q)\epsilon_k^0]$, where the initial energy undergoes a weighted average of two initial eigenvalues.

In contrast, the consistent-history (CH) quasiprobability of work demonstrates time-reversal symmetry [49]. Unlike most definitions, CH work involves the evolution details. In this formulation, a trajectory is built based on the power operator $\partial_t H_H(t)$, and the work is associated with the integrated power during the whole process. The reproduction of the TPM result (E1) and the fluctuation theorems cannot be fulfilled because work is not a quantity related to the initial and final Hamiltonians in this proposal. By the way, we remark that usually, if (E1) is violated, then so is the support condition (W3). This justifies (W3) as a more strict generalization of (E1).

It is noticeable that the Kirkwood-Dirac (KD) quasiprobability of work also satisfies all the requirements, apart from its complex value as well as complex second moment. Its distribution function is given by

$$P_{\mathrm{KD}}(w) = \sum_{ij} \mathrm{Tr}\big[\Pi_j^1 \Pi_i^0 \rho\big]\delta\big(w - (\epsilon_j^1 - \epsilon_i^0)\big). \quad (20)$$

Actually, the MH distribution is just the real part of the KD distribution, and it is wise to first calculate the KD distribution

and take the real part to obtain the MH one in many cases:

$$P_{\mathrm{MH}}(w) = \mathrm{Re}\, P_{\mathrm{KD}}(w). \tag{21}$$

Likewise, if we loosen the positivity of the second moment (W5) to the positivity of the real part of the second moment, then the following KD quasiprobability with a rescaled imaginary part meets all the other requirements:

$$P_{\xi}(w) = \mathrm{Re}\, P_{\mathrm{KD}}(w) + i\xi\, \mathrm{Im}\, P_{\mathrm{KD}}(w), \tag{22}$$

with a real parameter ξ. Since the $\mathrm{Im}\, P_{\mathrm{KD}}(w)$ can be rescaled without violating most requirements, the physical meaning of the imaginary part in the KD distribution is not clear. We suggest that the real part and imaginary part of the KD distribution should be treated separately because of their distinct behaviors. Fortunately, the strict requirement on the positivity of secondary moment (W5) rules out these complex-valued candidates, leaving the MH quasiprobability of quantum work the only appropriate definition.

VI. EXAMPLE: BREATHING HARMONIC OSCILLATOR WITH INITIAL SQUEEZED STATES

We adopt the MH quasiprobability and evaluate the work distribution for the breathing harmonic oscillator. We will further verify the quantum-classical correspondence principle of work distribution for initial states with quantum coherence in this example [53]. The Hamiltonian in the Schrödinger picture is

$$\hat{H}_S(t) = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega(t)^2\hat{x}^2. \tag{23}$$

The frequency $\omega(t)$ is varied in a finite-time process. In this section, we use the symbol "^" to denote operators in quantum mechanics.

Let us adopt a special protocol to vary the frequency [29,54,55],

$$\omega(t) = \frac{\omega_0}{(\omega_0/\omega_\tau - 1)t/\tau + 1}, \tag{24}$$

where $\omega(0) = \omega_0$, and $\omega(\tau) = \omega_\tau$. The final Hamiltonian in the Heisenberg picture can be obtained analytically as [56]

$$\begin{aligned}
\hat{H}_H(\tau) &= \frac{1}{2m}\hat{p}_H(t)^2 + \frac{1}{2}m\omega(t)^2\hat{x}_H(t)^2 \\
&= \frac{\Gamma_{11}}{2}\hat{x}^2 + \frac{\Gamma_{22}}{2}\hat{p}^2 + \frac{\Gamma_{12}}{2}(\hat{x}\hat{p} + \hat{p}\hat{x}). \tag{25}
\end{aligned}$$

The coefficients are

$$\Gamma_{11} = e^r\left[\frac{\cosh(\sqrt{1-g^2}r) - g^2}{1-g^2} + \frac{\sinh(\sqrt{1-g^2}r)}{\sqrt{1-g^2}}\right]m\omega_0^2,$$

$$\Gamma_{22} = e^r\left[\frac{\cosh(\sqrt{1-g^2}r) - g^2}{1-g^2} - \frac{\sinh(\sqrt{1-g^2}r)}{\sqrt{1-g^2}}\right]/m,$$

$$\Gamma_{12} = e^r\frac{g[\cosh(\sqrt{1-g^2}r) - 1]}{1-g^2}\omega_0,$$

where $g = 2\omega_0\omega_\tau\tau/(\omega_\tau - \omega_0)$, and $r = \ln(\omega_\tau/\omega_0)$.

In the following, we consider an initial squeezed state in the form

$$\begin{aligned}
\hat{\rho} &= 2\sinh\left(\tfrac{1}{2}\hbar\sqrt{\gamma_{11}\gamma_{22} - \gamma_{12}^2}\right) \\
&\quad \times \exp\left(-\tfrac{1}{2}[\gamma_{11}\hat{x}^2 + \gamma_{12}(\hat{x}\hat{p} + \hat{p}\hat{x}) + \gamma_{22}\hat{p}^2]\right). \tag{26}
\end{aligned}$$

Here γ_{11}, γ_{22}, and γ_{12} are three parameters characterizing the squeezed state. To evaluate the work distribution, we first calculate the characteristic function of the quantum work under the MH quasiprobability

$$\begin{aligned}
\chi_W(s) &= \int e^{isw} P_{\mathrm{MH}}(w)\, dw \\
&= \mathrm{Tr}\left\{\frac{1}{2}[e^{is\hat{H}_H(\tau)}e^{-is\hat{H}(0)} + e^{-is\hat{H}(0)}e^{is\hat{H}_H(\tau)}]\hat{\rho}\right\}. \tag{27}
\end{aligned}$$

It can be evaluated by the trace formula of a single bosonic mode [57]

$$\mathrm{Tr}\left[\prod_j e^{\frac{1}{2}R^T M_j R}\right] = \left\{-\det\left[\prod_j e^{-\hbar\sigma_y M_j} - I\right]\right\}^{-\frac{1}{2}}, \tag{28}$$

where the position and the momentum are given by $R = (\hat{x} \quad \hat{p})^T$, σ_y is the second Pauli matrix, I is identity 2×2 matrix, and M_j ($j = 1, 2, 3$) are symmetric 2×2 matrices

$$M_1 = is\begin{pmatrix} \Gamma_{11} & \Gamma_{12} \\ \Gamma_{12} & \Gamma_{22} \end{pmatrix}, \tag{29}$$

$$M_2 = -is\begin{pmatrix} m\omega_0^2 & \\ & 1/m \end{pmatrix}, \tag{30}$$

$$M_3 = -\begin{pmatrix} \gamma_{11} & \gamma_{12} \\ \gamma_{12} & \gamma_{22} \end{pmatrix}. \tag{31}$$

The corresponding work distribution $P_{\mathrm{MH}}(w)$ is obtained as the inverse Fourier transform, and it appears to be a discrete distribution.

The classical work distribution can be obtained from the initial distribution in phase space,

$$\rho(x, p) = \frac{\sqrt{\gamma_{11}\gamma_{22} - \gamma_{12}^2}}{2\pi} e^{-\frac{1}{2}(\gamma_{11}x^2 + 2\gamma_{12}xp + \gamma_{22}p^2)}, \tag{32}$$

which represents the classical counterpart of the quantum state given in Eq. (26). The work distribution is obtained as the integral in the phase space

$$P^{\mathrm{cl}}(w) = \iint \rho(x, p)\delta(w - H_H(\tau) + H(0))\, dx\, dp. \tag{33}$$

Here, $H_H(\tau)$ is also given by Eq. (25), but it is a c-number determined by the initial point in phase space. The result of the characteristic function is

$$\chi_W^{\mathrm{cl}}(s) = \iint e^{is(H_H(\tau) - H(0))}\rho(x, p)\, dx\, dp = \sqrt{\frac{\gamma_{11}\gamma_{22} - \gamma_{12}^2}{[\gamma_{11} - is(\Gamma_{11} - m\omega_0^2)][\gamma_{22} - is(\Gamma_{22} - \frac{1}{m})] - (\gamma_{12} - is\Gamma_{12})^2}}.$$
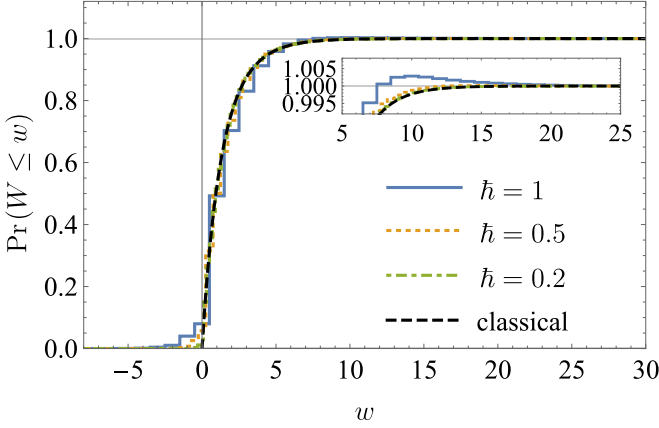
FIG. 1. Accumulated distribution of work $\Pr(W \leqslant w)$. The black dashed curve corresponds to classical work distribution. The discrete distributions with blue solid, orange dotted, and green dot-dashed curves correspond to MH quasiprobability of quantum work with $\hbar = 1, 0.5, 0.2$. The initial squeezed state is prepared with $\gamma_{11} = 1$, $\gamma_{22} = 1.1$, and $\gamma_{12} = 0.9$. The parameters of the harmonic oscillator are chosen as $m = 1$, $\omega_0 = 1$ and $\omega_\tau = 2$, and the operation time $\tau = 1$. In the enlarged subset, the decrease of the accumulated distribution of work with $\hbar = 1$ shows the negative value for roughly $w > 10$. Such negativity is less conspicuous in the case of smaller $\hbar$.

The corresponding work distribution is also obtained by the inverse Fourier transform, but it is a continuous distribution.

In Fig. 1, we show the accumulated distributions of work $\Pr(W \leqslant w) = \int_{-\infty}^{w} P(w')dw'$ with the initial squeezed state in Eq. (26). The parameters are given in the caption of the figure. The discreteness of energy eigenvalues in quantum system leads to discrete quasiprobability distributions (blue solid, orange dotted, and green dot-dashed curves), while the classical work distribution (black dashed curve) is continuous because the energy in classical systems is continuously distributed. With the decrease of $\hbar$, the quasiprobability distributions of quantum work converge to the classical work distribution because the energy levels become denser. This convergence shows the quantum-classical correspondence principle of the MH quasiprobability of quantum work for initial states with quantum coherence [53]. Meanwhile, the figure shows less conspicuous negativity of the distribution with smaller $\hbar$.

We remark that although the Jarzynski equality was originally for initial states in canonical equilibrium, a generalized Jarzynski equality for arbitrary initial states was proposed in classical systems [58]. However, the counterpart in quantum systems is unclear because of the difficulty in defining the stochastic work for systems with initial quantum coherence. By utilizing stochastic work defined by the MH quasiprobability, a reasonable counterpart in quantum regime is obtained [44,53]. The generalized Jarzynski equality for arbitrary initial states with quantum coherence is written as

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F} \operatorname{Re} \operatorname{Tr}[\hat{\rho} \hat{U}^{-1} \hat{\rho}_{\text{eq}}(\tau) U \hat{\rho}_{\text{eq}}(0)^{-1}], \quad (34)$$

where $\hat{\rho}_{\text{eq}}(0) = e^{-\beta \hat{H}_S(0)}/\operatorname{Tr}[e^{-\beta \hat{H}_S(0)}]$ and $\hat{\rho}_{\text{eq}}(\tau) = e^{-\beta \hat{H}_S(\tau)}/\operatorname{Tr}[e^{-\beta \hat{H}_S(\tau)}]$ are equilibrium states with the inverse temperature β at the initial and the final time, and $\hat{U}$ is the evolution

operator. The conventional Jarzynski equality is recovered for an initial equilibrium state $\hat{\rho} = \hat{\rho}_{\text{eq}}(0)$. With the work distribution obtained in Fig. 1, we can verify the generalized Jarzynski equality (34) by evaluating the two sides separately. Through comparing Eq. (26) of Ref. [58] and Eq. (34), one can see the Jarzynski equality for arbitrary initial states with coherence recovers its classical counterpart when $\hbar \to 0$, which shows the quantum-classical correspondence.

VII. EXPERIMENTAL MEASUREMENTS OF MARGENAU-HILL QUASIPROBABILITY

There have been many proposals on experimentally measuring the MH quasiprobability of quantum work, some of which are similar to the techniques of measuring time-ordered correlators. Reference [41] gives a detailed summary of these proposals, including weak two-point-measurement scheme [59], Ramsey interferometric scheme [21,60,61], the cloning approach [62,63], direct reconstruction scheme [64,65], and block-encoding scheme [66]. Particularly, the weak two-point-measurement scheme and the Ramsey interferometric scheme have been realized in experiments to measure the quasiprobability of work in the liquid-state nuclear magnetic resonance platform [21] and in the nitrogen-vacancy center of diamond [67], respectively.

Here, we only give a brief introduction of the weak two-point-measurement scheme following Ref. [41]. It is the simplest scheme, which does not need to couple the system to any ancillas. In this scheme, one carries out three sets of measurements.

The first one is a weak two-point-measurement process, in which time evolution starts from a specific initial state,

$$\rho_i^{\text{NS}} = \Pi_i^0 \rho \Pi_i^0 + (1 - \Pi_i^0)\rho(1 - \Pi_i^0). \quad (35)$$

ρ_i^{NS} can be obtained by a nonselective measurement on ρ with projectors $\{\Pi_i^0, 1 - \Pi_i^0\}$. After the time evolution, the measurement over the final Hamiltonian is performed. In this process, one can obtain the weak two-point-measurement joint probabilities,

$$p_{ji}^{\text{WTPM}} = \operatorname{Tr}[\Pi_j^1 \rho_i^{\text{NS}}]. \quad (36)$$

The second process is the standard two-projective-measurement scheme, and the transition probability is given by

$$p_{ji}^{\text{TPM}} = \operatorname{Tr}[\Pi_j^1 \Pi_i^0 \rho \Pi_i^0]. \quad (37)$$

The third process contains the final measurement only. After evolving from the initial state ρ, one measures the final Hamiltonian and obtains

$$p_j^{\text{END}} = \operatorname{Tr}[\Pi_j^1 \rho]. \quad (38)$$

The MH quasiprobability of work can be constructed by the above results from experiments. We denote the MH joint quasiprobability for energy transitions as

$$q_{ji} = \operatorname{Tr}\left[\tfrac{1}{2}\left(\Pi_j^1 \Pi_i^0 + \Pi_i^0 \Pi_j^1\right)\rho\right], \quad (39)$$

so that the MH quasiprobability distribution of work is $P_{\text{MH}}(w) = \sum_{ji} q_{ji}\delta(w - (\epsilon_j^1 - \epsilon_i^0))$. It is found that q_{ji} can

be expressed as

$$q_{ji} = p_{ji}^{\text{TPM}} + \frac{1}{2}\left(p_j^{\text{END}} - p_{ji}^{\text{WTPM}}\right). \tag{40}$$

The above procedures constitute an experimental scheme of measuring the MH quasiprobability of work.

VIII. MARGENAU-HILL QUASIPROBABILITY IN OPEN QUANTUM SYSTEMS

Although we restrict ourselves to the isolated driving systems up to now, as a unified approach to quantum stochastic thermodynamics, the MH quasiprobability can be readily generalized to open systems. Let us consider a driven quantum system in contact with a heat bath. The total Hamiltonian of the composite system is

$$H^{\text{T}}(t) = H^{\text{S}}(t) + H^{\text{B}} + H^{\text{I}}, \tag{41}$$

where H^{S}, H^{B}, and H^{I} denote the Hamiltonian of the system, the bath, and the interaction. The time-dependent driving only acts on the system. We restrict to the weak coupling regime ($H^{\text{I}} \ll H^{\text{S}}, H^{\text{B}}$). In this case, the total Hamiltonian and system Hamiltonian commute with each other $[H^{\text{T}}(t), H^{\text{S}}(t)] = 0$, and the concepts of work and heat are clear. If there is no initial coherence, then the joint distribution of work performed and heat transferred can be defined via the TPM scheme with the joint measurement over H^{T} and H^{S} [20]. If the initial state contains quantum coherence, then we can replace the TPM scheme with the MH quasiprobability. By doing so, the joint quasiprobability distribution of work and heat is given explicitly by

$$P_{\text{MH}}(w, q) = \text{Re} \sum_{ijkl} \text{Tr}\left[\Pi_l^{1,\text{S}} \Pi_j^{1,\text{T}} \Pi_k^{0,\text{S}} \Pi_i^{0,\text{T}} \rho\right]$$
$$\times \delta\left(w - \epsilon_j^{1,\text{T}} + \epsilon_i^{0,\text{T}}\right) \delta\left(q - \epsilon_l^{1,\text{S}} + \epsilon_k^{0,\text{S}} + \epsilon_j^{1,\text{T}} - \epsilon_i^{0,\text{T}}\right), \tag{42}$$

where Π_i are the projectors of the Hamiltonians in the Heisenberg picture, and ϵ_i are the corresponding eigenvalues. The superscripts 0 or 1 in Π_i and ϵ_i denote the initial or final time, and the superscripts T or S denote the total or system Hamiltonian. It is clear from the expression that the work corresponds to the change of the total Hamiltonian due to the external driving, and the heat corresponds to the subtraction of system energy change and the work, in accordance with the usual stochastic energetics [3].

IX. CONCLUSION AND OUTLOOK

In this paper, we list several requirements for an appropriate definition of work distribution. Building on the physical implication of the TPM scheme, the support condition is introduced, which reveals that the quantum work corresponds to the transition between the initial and final energy levels, and it plays a crucial role in our proof. Other requirements include the convexity with respect to the initial density operator, the first law of thermodynamics, linear relation on the Hamiltonian, the positivity of second-order moment, and the time-reversal symmetry.

We prove that the MH quasiprobability of quantum work is the only definition that satisfies all the requirements. We

also compare some common definitions of quantum work and examine their properties from the perspective of requirements. The FC quasiprobability as well as the q-class quasiprobability fails to fulfill the time-reversal symmetry and the support condition due to the asymmetry between the initial and the final Hamiltonians. The KD quasiprobability is also an acceptable definition, which satisfies most of the requirements. Nevertheless, the physical meaning of its imaginary part is unexplored, and the appearance of the imaginary part violates the positivity requirement of the second-order moment. Under all the requirements in this paper, MH quasiprobability of quantum work shows its advantages over other definitions of quantum work.

We explicitly evaluate the MH quasiprobability of quantum work distribution in the example of a breathing harmonic oscillator. The distribution exhibits slightly negative values at the tail. The Jarzynski equality for initial states with quantum coherence [44,53] is verified in this example.

The MH quasiprobability of quantum work serves as a general way to study work fluctuation, and it also implies useful physical applications. For example, in the study of quantum heat engines, the work distribution provides more detailed information than the average work. The stability of the engine can be characterized by the variance of the work, which is supposed to be small. Moreover, it is worth noticing that negative values of the MH quasiprobability are linked to potential advantages of the work extraction [67,68].

As is briefly shown in Sec. VIII, the MH quasiprobability generally characterizes fluctuations in quantum thermodynamics. In addition to quantum work, the MH quasiprobability can be applied to other thermodynamic quantities, such as heat exchange [69,70], charge transport [71,72], and especially noncommuting charges [73–76]. The requirements in this paper can also help us justify the validity of the MH quasiprobability of these quantities in such cases. In quantum thermodynamics, one often restricts to the process without initial coherence, in contrast to classical stochastic thermodynamics, where no restriction is put on the process. With the help of the MH quasiprobability, it is promising to study thermodynamic processes with initial quantum coherence. We can calculate the full counting statistics and characterize the fluctuation for thermodynamic quantities through a standard way in a generic quantum nonequilibrium process. This will provide a solid foundation for related issues such as quantum heat engines and work extractions. In future studies, we hope that the quasiprobability approach can broaden the scope of quantum stochastic thermodynamics.

APPENDIX A: CONVEXITY ON INITIAL STATE AND ITS COROLLARY

In this Appendix, we are going to prove that the convexity property (W1) of distribution implies the distribution function

$P(w)$ can be expressed as

$$P(w; \rho) = \text{Tr}[M(w)\rho], \qquad (A1)$$

where $M(w)$ is a bounded operator irrelevant to ρ.

Before proceeding, we remark that although in the case with $P \geqslant 0$, this result has been proved [33], there is no strict proof if P is a quasiprobability distribution with real or complex value. Here, we give proof that applies to Hilbert space of any (maybe infinite) dimension.

We now briefly illustrate the outline of the proof. For a specific distribution, we do a linear expansion of $P(w; \rho)$ to $\tilde{P}(w; \tilde{\rho})$, allowing $\tilde{\rho}$ to be any operator that has a definite trace, called trace-class operator. In the trace class, operators are no longer Hermitian or positive. The expanded distribution $\tilde{P}$ becomes a linear function in the trace-class space. Discussions about trace-class space can be found in textbook [77]. The general form of a linear function of the trace-class operator $\tilde{\rho}$ is given by $\text{Tr}[M\tilde{\rho}]$ with M a bounded operator. Therefore, $\tilde{P}$ is represented in this general form, and so is P when restricting to the density operators. In the following, we elaborate on how it works, and the feasibility of the linear expansion will be justified.

We start with a lemma. This lemma is used in the later construction of the linear expansion.

Lemma 1: If a linear combination of several density operators ρ_i with real coefficients $\lambda_i \in \mathbb{R}$ is zero, that is,

$$\lambda_1 \rho_1 + \cdots + \lambda_N \rho_N = 0,$$

then $\lambda_1 P(w; \rho_1) + \cdots + \lambda_N P(w; \rho_N) = 0$.

Proof: ρ_i are all density operators with trace 1, so $0 = \lambda_1 + \cdots + \lambda_N$. The case that all the λ_i equal 0 is trivial, so we suppose at least one element in $\{\lambda_i\}$ is greater than zero. We mark the nonnegative terms as $\lambda_{i_k}, 1 \leqslant k \leqslant M$ and the negative terms as $\lambda_{j_k}, 1 \leqslant k \leqslant N - M$. We move the term with negative λ_{j_k} to the right-hand side and rescaling the equation,

$$\frac{1}{\Lambda}\left(\left|\lambda_{i_1}\right|\rho_{i_1} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}\right)$$
$$= \frac{1}{\Lambda}\left(\left|\lambda_{j_1}\right|\rho_{j_1} + \cdots + \left|\lambda_{j_{N-M}}\right|\rho_{j_{N-M}}\right), \qquad (A2)$$

where $\Lambda = \left|\lambda_{j_1}\right| + \cdots + \left|\lambda_{j_{N-M}}\right| = \left|\lambda_{i_1}\right| + \cdots + \left|\lambda_{i_M}\right|$. The expression in the above equation is a density operator, which we will denote as ρ'. Then we split the first term on the left-hand side from the other terms and write it as

$$\rho' = \frac{\left|\lambda_{i_1}\right|}{\Lambda}\rho_{i_1} + \frac{\Lambda - \left|\lambda_{i_1}\right|}{\Lambda}$$
$$\times \frac{1}{\Lambda - \left|\lambda_{i_1}\right|}\left(\left|\lambda_{i_2}\right|\rho_{i_2} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}\right),$$

where ρ_{i_1} and $\frac{1}{\Lambda - \left|\lambda_{i_1}\right|}\left(\left|\lambda_{i_2}\right|\rho_{i_2} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}\right)$ are both density operators. From the convexity relation, we get

$$P(w; \rho') = \frac{\left|\lambda_{i_1}\right|}{\Lambda}P(w; \rho_{i_1})$$
$$+ \frac{\Lambda - \left|\lambda_{i_1}\right|}{\Lambda}P\left(w; \frac{\left|\lambda_{i_2}\right|\rho_{i_2} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}}{\Lambda - \left|\lambda_{i_1}\right|}\right).$$

Next, for density operator $\frac{\left|\lambda_{i_2}\right|\rho_{i_2} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}}{\Lambda - \left|\lambda_{i_1}\right|}$, we continue to split out the first term. After a similar rescaling and applying the convexity relation, we have

$$P\left(w; \frac{\left|\lambda_{i_2}\right|\rho_{i_2} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}}{\Lambda - \left|\lambda_{i_1}\right|}\right) = \frac{\left|\lambda_{i_2}\right|}{\Lambda - \left|\lambda_{i_1}\right|}P(w; \rho_{i_2})$$
$$+ \frac{\Lambda - \left|\lambda_{i_1}\right| - \left|\lambda_{i_2}\right|}{\Lambda - \left|\lambda_{i_1}\right|}P\left(w; \frac{\left|\lambda_{i_3}\right|\rho_{i_3} + \cdots + \left|\lambda_{i_M}\right|\rho_{i_M}}{\Lambda - \left|\lambda_{i_1}\right| - \left|\lambda_{i_2}\right|}\right).$$

Repeating the steps to the remaining part, we finally split all the terms apart. The final result is simply

$$\Lambda P(w; \rho') = \left|\lambda_{i_1}\right|P(w; \rho_{i_1}) + \cdots + \left|\lambda_{i_M}\right|P(w; \rho_{i_M}).$$

Similar procedure applies to the right-hand side of Eq. (A2),

$$\Lambda P(w; \rho') = \left|\lambda_{j_1}\right|P(w; \rho_{j_1}) + \cdots + \left|\lambda_{j_{N-M}}\right|P(w; \lambda_{j_{N-M}}).$$

Combining the two sides and moving the right-hand side back to the left, we complete the proof of the lemma. ∎

Next, we are going to see that the density operators span the trace class [77], a subspace of the bounded operator space. The trace class is defined by: An operator A belongs to the trace class if and only if $\text{Tr}|A| < \infty$, where $|A| = (A^\dagger A)^{\frac{1}{2}}$. If A belongs to the trace class, then A is called a trace-class operator, which has definite trace $\text{Tr}A$. Obviously, any linear combination of a finite number of density operators is a trace-class operator.

Also, let us assume A is a trace-class operator. Every bounded operator can be decomposed into a Hermitian operator plus an anti-Hermitian operator, so we have

$$A = B + iC,$$

where $B = \frac{1}{2}(A + A^\dagger), C = \frac{1}{2i}(A - A^\dagger)$ are Hermitian operators. Likewise, both B and C are trace-class operators. Furthermore, bounded Hermitian operators can be decomposed into a subtraction of two positive operators [78]. This applies to B and C:

$$B = B_1 - B_2, C = C_1 - C_2.$$

By the way, if $B_1 B_2 = B_2 B_1 = 0$ (or $C_1 C_2 = C_2 C_1 = 0$), then the decomposition is unique. Meanwhile, positive operators B_1, B_2, C_1, C_2 are all trace-class operators with finite trace. After a rescaling, operator A is expressed as a linear combination of four density operators:

$$A = \lambda_1 \rho_1 - \lambda_2 \rho_2 + i\lambda_3 \rho_3 - i\lambda_4 \rho_4, \qquad (A3)$$

where $\rho_1 = B_1/\text{Tr}B_1, \rho_2 = B_2/\text{Tr}B_2, \rho_3 = C_1/\text{Tr}C_1, \rho_4 = C_2/\text{Tr}C_2$, and all the λ_i are nonnegative number. Therefore, any trace-class operator can be decomposed as the linear combination of four density operators (this decomposition is not unique generally), and all the density operators span the trace class.

For a specific distribution function $P(w; \rho)$, we define an expanded distribution $\tilde{P}$ with respect to any trace-class operator A in the following way:

$$\tilde{P}(w; A) = \lambda_1 P(w; \rho_1) - \lambda_2 P(w; \rho_2) + i\lambda_3 P(w; \rho_3)$$
$$- i\lambda_4 P(w; \rho_4), \qquad (A4)$$

provided the decomposition in Eq. (A3). To serve as a good definition, it should be independent of the choice of decomposition. If there is another different decomposition $A = \lambda_1' \rho_1' - \lambda_2' \rho_2' + i\lambda_3' \rho_3' - i\lambda_4' \rho_4'$, then the expanded distribution under this decomposition is

$$
\begin{aligned}
\tilde{P}'(w; A) = & \lambda_1' P(w; \rho_1') - \lambda_2' P(w; \rho_2') + i\lambda_3' P(w; \rho_3') \\
& - i\lambda_4' P(w; \rho_4').
\end{aligned}
\tag{A5}
$$

In the two different decompositions, the Hermitian part and anti-Hermitian part equal each other, respectively, that is, $\lambda_1 \rho_1 - \lambda_2 \rho_2 = \lambda_1' \rho_1' - \lambda_2' \rho_2'$ and $\lambda_3 \rho_3 - \lambda_4 \rho_4 = \lambda_3' \rho_3' - \lambda_4' \rho_4'$. According to lemma 1, $\lambda_1 P(\rho_1) - \lambda_2 P(\rho_2) = \lambda_1' P(\rho_1') - \lambda_2' P(\rho_2')$, $\lambda_3 P(\rho_3) - \lambda_4 P(\rho_4) = \lambda_3' P(\rho_3') - \lambda_4' P(\rho_4')$, so the above P' coincides with P in Eq. (A4). This manifests that the linear expansion is independent of the decomposition. Besides, it is straightforward to verify that the expanded distribution exhibits the standard linear relation that for any complex number μ_1, μ_2 and trace-class operator A_1, A_2,

$$
\tilde{P}(w; \mu_1 A_1 + \mu_2 A_2) = \mu_1 \tilde{P}(w; A_1) + \mu_2 \tilde{P}(w; A_2).
\tag{A6}
$$

Therefore, $\tilde{P}$ is a linear function on the trace class, and we have finished our treatment for the linear expansion.

The dual space (the space of bounded linear function) of the trace class is isomorphic to the bounded operator space [77]. As a linear function of A, the expanded distribution $\tilde{P}$ is represented as

$$
\tilde{P}(w; A) = \text{Tr}[M(w)A].
\tag{A7}
$$

Here $M(w)$ is a bounded operator, which has a one-on-one correspondence with the distribution $\tilde{P}(w)$. If we restrict A to the density matrix ρ, then $\tilde{P}$ reduces to the work distribution P. In this case, we get the final result,

$$
P(w; \rho) = \text{Tr}[M(w)\rho].
\tag{A8}
$$

APPENDIX B: THE FIRST LAW DETERMINES THE EXPANSION COEFFICIENTS

We are going to specify how to determine the expansion coefficients in Eq. (12) under the condition of the first law of thermodynamics so that the form of $N_{ji}(\Pi_j^1, \Pi_i^0)$ is fixed to Eq. (15). In addition, we will verify the time-reversal symmetry for the N_{ji} in Eq. (15).

We have obtained the condition for N_{ij} from the first law in Eq. (14), which is also listed below:

$$
\sum_{ij} (\epsilon_j^1 - \epsilon_i^0) N_{ji} = H_1 - H_0 = \sum_j \epsilon_j^1 \Pi_j^1 - \sum_i \epsilon_i^0 \Pi_i^0. \tag{B1}
$$

Since the first law is valid in arbitrary protocols, we have the liberty to assign the values of $\epsilon_j^1, \Pi_j^1, \epsilon_i^0, \Pi_i^0$. Due to the arbitrariness of $\epsilon_j^1, \epsilon_i^0$, the above equation is equivalent to

$$
\sum_i N_{ji} = \Pi_j^1, \quad \sum_j N_{ji} = \Pi_i^0.
$$

Noticing in the MH work quasiprobability, $N_{ij}^{\text{MH}} = \frac{1}{2}(\Pi_j^1 \Pi_i^0 + \Pi_i^0 \Pi_j^1)$ obviously satisfies the above equation, we adopt a trick here. We consider $\tilde{N}_{ij} = N_{ij} - N_{ij}^{\text{MH}}$ instead, and

a tilde is used for $\tilde{N}_{ij}$ in the same expansion as Eq. (12). That is,

$$
\begin{aligned}
\tilde{N}_{ji}(\Pi_j^1, \Pi_i^0) = & \tilde{e}_0 I + \sum_{n=1}^{\infty} [\tilde{a}_n (\Pi_j^1 \Pi_i^0)^n + \tilde{b}_n (\Pi_i^0 \Pi_j^1)^n] \\
& + \sum_{n=0}^{\infty} [\tilde{c}_n \Pi_i^0 (\Pi_j^1 \Pi_i^0)^n + \tilde{d}_n (\Pi_j^1 \Pi_i^0)^n \Pi_j^1].
\end{aligned}
\tag{B2}
$$

The condition for $\tilde{N}_{ij}$ now changes to

$$
\sum_i \tilde{N}_{ji} = 0, \quad \sum_j \tilde{N}_{ji} = 0.
\tag{B3}
$$

It is enough to find a specific set of Π_j^1, Π_i^0, under which all the $\tilde{a}_n, \tilde{c}_n$ are fixed to be our desired values. We will show how this works in the separable Hilbert space with dimension $2 < d \leqslant \infty$.

For the sake of clarity, in this section, we denote the initial eigenstates as $|\psi_m\rangle$, $1 \leqslant m \leqslant d$, and the final eigenstates as $|\phi_m\rangle$, instead of $|\psi_m^0\rangle, |\psi_m^1\rangle$. The projective operators corresponding to these eigenstates are

$$
\Pi_i^0 = |\psi_i\rangle\langle\psi_i|, \quad \Pi_j^1 = |\phi_j\rangle\langle\phi_j|.
$$

Direct calculation of each term in the expansion yields

$$
\begin{aligned}
(\Pi_j^1 \Pi_i^0)^n &= \langle\phi_j|\psi_i\rangle^n \langle\psi_i|\phi_j\rangle^{n-1} |\phi_j\rangle\langle\psi_i|, \\
(\Pi_i^0 \Pi_j^1)^n &= \langle\phi_j|\psi_i\rangle^{n-1} \langle\psi_i|\phi_j\rangle^n |\psi_i\rangle\langle\phi_j|, \\
\Pi_i^0 (\Pi_j^1 \Pi_i^0)^n &= \langle\phi_j|\psi_i\rangle^n \langle\psi_i|\phi_j\rangle^n |\psi_i\rangle\langle\psi_i|, \\
(\Pi_j^1 \Pi_i^0)^n \Pi_j^1 &= \langle\phi_j|\psi_i\rangle^n \langle\psi_i|\phi_j\rangle^n |\phi_j\rangle\langle\phi_j|.
\end{aligned}
$$

For $2 < d \leqslant \infty$, our specific set of eigenstates are chosen as follows. The initial energy eigenstates are selected to be the standard basis, that is, only the mth component is nonzero,

$$
|\psi_m\rangle = (0, \ldots, 0, \overset{m}{1}, 0, \ldots).
\tag{B4}
$$

As for final energy eigenstates, the first three states are some real vectors:

$$
\begin{aligned}
|\phi_1\rangle &= (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta, 0, \ldots), \\
|\phi_2\rangle &= (\cos\theta\cos\phi, \cos\theta\sin\phi, -\sin\theta, 0, \ldots), \\
|\phi_3\rangle &= (-\sin\phi, \cos\phi, 0, 0, \ldots).
\end{aligned}
\tag{B5}
$$

Other final energy eigenstates are the same as the initial ones,

$$
|\phi_m\rangle = |\psi_m\rangle, \quad \text{for } m > 3.
\tag{B6}
$$

Also, the whole final energy eigenstates form a set of orthogonal basis in the Hilbert space.

Let us consider the following matrix element of the Eq. (B3):

$$
\sum_{j=1}^d \langle\psi_2|\tilde{N}_{j1}|\psi_2\rangle = 0.
\tag{B7}
$$

Substituting Eqs. (B2) and (B5) in the above equation, we find only one kind of term in the expansion survives besides the

identity, and the explicit expression reads

$$
d\tilde{e}_0 + \sum_{n=0}^{\infty} \tilde{d}_n[(\sin\theta\cos\phi)^{2n}(\sin\theta\sin\phi)^2
$$
$$
+ (\cos\theta\cos\phi)^{2n}(\cos\theta\sin\phi)^2 + (-\sin\phi)^{2n}(\cos\phi)^2] = 0. \tag{B8}
$$

To make it clear, we introduce $x = \cos^2\theta$, $1 - x = \sin^2\theta$, $y = \cos^2\phi$, and $1 - y = \sin^2\phi$, and we obtain

$$
d\tilde{e}_0 + \sum_{n=0}^{\infty} \tilde{d}_n F_n(x, y) = 0, \tag{B9}
$$

where $F_n(x, y) = [(1 - x)^{n+1}y^n(1 - y) + x^{n+1}y^n(1 - y) + y(1 - y)^n]$, and the above equation holds for any $0 < x, y < 1$. For a specific value $x = \frac{1}{2}$, we have

$$
F_n\left(\frac{1}{2}, y\right) = \frac{1}{2^n}y^n(1 - y) + y(1 - y)^n. \tag{B10}
$$

For $n > 0$, $F_n(1/2, y)$ is a polynomial of y with degree $n + 1$, while for $n = 0$, $F_0 = 1$. Therefore, F_n with different n are linearly independent. According to Eq. (B9), we find $\tilde{d}_n = 0$ for $n > 0$, and $\tilde{d}_0 = -d\tilde{e}_0$. Here, d is the dimension of the Hilbert space. If considering $\sum_{j=1}^{d}\langle\phi_2|\tilde{N}_{1j}|\phi_2\rangle = 0$, then the similar derivation leads to $\tilde{c}_n = 0$, $n > 0$ and $\tilde{c}_0 = -d\tilde{e}_0$.

Next, we investigate the following matrix element to find the relation of $\tilde{a}_n$,

$$
\sum_{j=1}^{d}\langle\psi_2|\tilde{N}_{j1}|\psi_1\rangle = 0. \tag{B11}
$$

In explicit form, we have

$$
\sum_{n=1}^{\infty} \tilde{a}_n[(\sin\theta\cos\phi)^{2n-1}\sin\theta\sin\phi
$$
$$
+ (\cos\theta\cos\phi)^{2n-1}\cos\theta\sin\phi + (-\sin\phi)^{2n-1}\cos\phi] = 0. \tag{B12}
$$

As long as $0 < \phi < \pi/2$, we can eliminate the common factor $\sin\phi\cos\phi$ in the above equation and obtain

$$
\sum_{n=1}^{\infty} \tilde{a}_n G_n(x, y) = 0, \tag{B13}
$$

where $G_n(x, y) = [x^n y^{n-1} + (1 - x)^n y^{n-1} - (1 - y)^{n-1}]$, and $x = \cos^2\theta$, $y = \cos^2\phi$ again. The above equation is valid for any $0 < x, y < 1$. We find that $G_1 = 0$ for any value of x, y. Assigning a specific value $x = \frac{1}{2}$, G_n reduces to

$$
G_n\left(\frac{1}{2}, y\right) = \frac{1}{2^{n-1}}y^{n-1} - (1 - y)^{n-1}. \tag{B14}
$$

For $n > 1$, $G_n(1/2, y)$ is a polynomial of y with degree $n - 1$. $\{G_n\}$, $(n > 1)$ are linearly independent, so $\tilde{a}_n = 0$ for $n > 1$ according to Eq. (B13). Since $G_1 = 0$, this equation puts no constraint on $\tilde{a}_1$. Likewise, if we consider $\sum_{j=1}^{d}\langle\psi_1|\tilde{N}_{j1}|\psi_2\rangle = 0$ instead, then we can deduce that $\tilde{b}_n = 0$ for $n > 1$ while $\tilde{b}_1$ is free.

The remaining unknown coefficients are $\tilde{a}_1$, $\tilde{b}_1$. Let us consider

$$
\sum_{j=1}^{d}\langle\psi_1|\tilde{N}_{j1}|\psi_1\rangle = 0. \tag{B15}
$$

After some simple calculations, we find the relation between $\tilde{a}_1$, $\tilde{b}_1$:

$$
\tilde{a}_1 + \tilde{b}_1 = d^2\tilde{e}_0. \tag{B16}
$$

Up to now, the expansion for $\tilde{N}_{ji}$ is fixed to

$$
\tilde{N}_{ji}(\Pi_j^1, \Pi_i^0) = \tilde{e}_0 I - d\tilde{e}_0[\Pi_i^0 + \Pi_j^1] + \tilde{a}_1\Pi_j^1\Pi_i^0
$$
$$
+ (-\tilde{a}_1 + d^2\tilde{e}_0)\Pi_i^0\Pi_j^1, \tag{B17}
$$

with free coefficients $\tilde{e}_0$, $\tilde{a}_1$. It can be checked that the above expression satisfies the condition for $\tilde{N}_{ji}$ in Eq. (B3), so it may be regarded as the general form of $\tilde{N}_{ji}$. However, we hope the definition of work is universal and irrespective of the dimension of Hilbert space d. Especially, the above expression should work well in infinite-dimensional space. Therefore, we are forced to choose $\tilde{e}_0 = 0$. By the way, we find that two-dimensional Hilbert space is an exception because we have less freedom to choose the projective operators. Nevertheless, since the definition should be universal irrespective of the dimension, this exception in 2D space does not matter.

In conclusion, under the condition from the first law, the general form for N_{ji} is simply

$$
N_{ji}(\Pi_j^1, \Pi_i^0) = \frac{1 + \eta}{2}\Pi_j^1\Pi_i^0 + \frac{1 - \eta}{2}\Pi_i^0\Pi_j^1, \tag{B18}
$$

where $\eta = 2\tilde{a}_1$ can be an arbitrary complex number. The corresponding distribution function reads

$$
P(w) = \sum_{ji} \delta\left(w - (\epsilon_j^1 - \epsilon_i^0)\right)
$$
$$
\times \text{Tr}\left[\left(\frac{1 + \eta}{2}\Pi_j^1\Pi_i^0 + \frac{1 - \eta}{2}\Pi_i^0\Pi_j^1\right)\rho\right]. \tag{B19}
$$

This form contains the MH quasiprobability for work ($\eta = 0$) and the KD quasiprobability for work ($\eta = 1$).

At the end of this Appendix, we will show that the above distribution satisfies time-reversal symmetry (E2). In the time-reversed process described in requirement (E2), we have the following relations in the Heisenberg picture: $\bar{H}_1 = \Theta U H_0 U^{-1}\Theta^{-1}$, $\bar{H}_0 = \Theta U H_1 U^{-1}\Theta^{-1}$, $\bar{\rho} = \Theta U \rho U^{-1}\Theta^{-1}$. The eigenvalues and corresponding projective operators are: $\bar{\epsilon}_i^0 = \epsilon_i^1$, $\bar{\epsilon}_j^1 = \epsilon_j^0$, $\bar{\Pi}_i^0 = \Theta U \Pi_i^1 U^{-1}\Theta^{-1}$, $\bar{\Pi}_j^1 = \Theta U \Pi_j^0 U^{-1}\Theta^{-1}$. Operator N_{ji} now

changes to

$$N_{ji}(\bar{\Pi}_j^1, \bar{\Pi}_i^0) = \Theta U N_{ji}^*(\Pi_j^0, \Pi_i^1) U^{-1} \Theta^{-1},$$

where superscript $*$ on N_{ji} denotes the complex conjugate over all the coefficients in the expansion of N_{ji}. After some derivation, we get the distribution function in the time-reversed process:

$$\begin{aligned}\bar{P}(w) &= \sum_{ji} \delta(w - (\bar{\epsilon}_j^1 - \bar{\epsilon}_i^0)) \mathrm{Tr}[N_{ji}(\bar{\Pi}_j^1, \bar{\Pi}_i^0)\bar{\rho}] \\ &= \sum_{ji} \delta(w - (\epsilon_j^0 - \epsilon_i^1)) \mathrm{Tr}[N_{ji}^*(\Pi_j^0, \Pi_i^1)\rho]^* \\ &= \sum_{ji} \delta(w + (\epsilon_j^1 - \epsilon_i^0)) \mathrm{Tr}[N_{ij}^{*\dagger}(\Pi_i^0, \Pi_j^1)\rho]. \quad (B20)\end{aligned}$$

In the second equality, please notice that when canceling the antiunitary operator Θ in the trace, there will be an extra complex conjugate. In the third equality, we exchange the summation index i, j. Imposing $\bar{P}(w) = P(-w)$, we find that the time-reversal condition on N_{ji} is

$$N_{ij}^{*\dagger}(\Pi_i^0, \Pi_j^1) = N_{ji}(\Pi_j^1, \Pi_i^0). \quad (B21)$$

In terms of expansion coefficients in Eq. (12), we find that $c_n = d_n$ while there are no constraints on a_n, b_n. N_{ji} in Eq. (B19) satisfies this condition. Particularly, both the MH quasiprobability of work and the KD quasiprobability of work encode the time-reversal symmetry. Thus, the time-reversal symmetry property (E2) can be regarded as a consequence of other requirements.

[1] U. Seifert, Stochastic thermodynamics, fluctuation theorems and molecular machines, Rep. Prog. Phys. **75**, 126001 (2012).

[2] L. Peliti and S. Pigolotti, *Stochastic Thermodynamics: An Introduction* (Princeton University Press, Princeton, NJ, 2021).

[3] K. Sekimoto, *Stochastic Energetics* (Springer, Berlin, 2010), Vol. 799.

[4] C. Jarzynski, Equalities and inequalities: Irreversibility and the second law of thermodynamics at the nanoscale, Annu. Rev. Condens. Matter Phys. **2**, 329 (2011).

[5] S. Vinjanampathy and J. Anders, Quantum thermodynamics, Contemp. Phys. **57**, 545 (2016).

[6] J. P. Pekola, Towards quantum thermodynamics in electronic circuits, Nat. Phys. **11**, 118 (2015).

[7] P. Strasberg, *Quantum Stochastic Thermodynamics: Foundations and Selected Applications* (Oxford University Press, 2022), https://doi.org/10.1093/oso/9780192895585.001.0001.

[8] J. Goold, M. Huber, A. Riera, L. del Rio, and P. Skrzypczyk, The role of quantum information in thermodynamics—A topical review, J. Phys. A: Math. Theor. **49**, 143001 (2016).

[9] J. Åberg, Fully quantum fluctuation theorems, Phys. Rev. X **8**, 011019 (2018).

[10] M. Campisi, J. Pekola, and R. Fazio, Nonequilibrium fluctuations in quantum heat engines: theory, example, and possible solid state experiments, New J. Phys. **17**, 035012 (2015).

[11] J. Millen and A. Xuereb, Perspective on quantum thermodynamics, New J. Phys. **18**, 011002 (2016).

[12] G. Manzano, J. M. Horowitz, and J. M. R. Parrondo, Quantum fluctuation theorems for arbitrary environments: Adiabatic and nonadiabatic entropy production, Phys. Rev. X **8**, 031037 (2018).

[13] P. Talkner and P. Hänggi, Aspects of quantum work, Phys. Rev. E **93**, 022131 (2016).

[14] G. Bochkov and Y. Kuzovlev, General theory of thermal fluctuations in nonlinear systems, Zh. Eksp. Teor. Fiz. **72**, 238 (1977) [Sov. Phys. JETP **45**, 125 (1977)].

[15] A. E. Allahverdyan and T. M. Nieuwenhuizen, Fluctuations of work from quantum subensembles: The case against quantum work-fluctuation theorems, Phys. Rev. E **71**, 066102 (2005).

[16] J. Kurchan, A quantum fluctuation theorem, arXiv:cond-mat/0007360.

[17] H. Tasaki, Jarzynski relations for quantum systems and some applications, arXiv:cond-mat/0009244.

[18] P. Talkner, M. Campisi, and P. Hnggi, Fluctuation theorems in driven open quantum systems, J. Stat. Mech. (2009) P02025.

[19] M. Esposito, U. Harbola, and S. Mukamel, Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems, Rev. Mod. Phys. **81**, 1665 (2009).

[20] M. Campisi, P. Hänggi, and P. Talkner, Colloquium: Quantum fluctuation relations: Foundations and applications, Rev. Mod. Phys. **83**, 771 (2011).

[21] T. B. Batalhão, A. M. Souza, L. Mazzola, R. Auccaise, R. S. Sarthour, I. S. Oliveira, J. Goold, G. De Chiara, M. Paternostro, and R. M. Serra, Experimental reconstruction of work distribution and study of fluctuation relations in a closed quantum system, Phys. Rev. Lett. **113**, 140601 (2014).

[22] S. An, J.-N. Zhang, M. Um, D. Lv, Y. Lu, J. Zhang, Z.-Q. Yin, H. T. Quan, and K. Kim, Experimental test of the quantum Jarzynski equality with a trapped-ion system, Nat. Phys. **11**, 193 (2015).

[23] J. Roßnagel, O. Abah, F. Schmidt-Kaler, K. Singer, and E. Lutz, Nanoscale heat engine beyond the carnot limit, Phys. Rev. Lett. **112**, 030602 (2014).

[24] K. Brandner, M. Bauer, and U. Seifert, Universal coherence-induced power losses of quantum heat engines in linear response, Phys. Rev. Lett. **119**, 170602 (2017).

[25] J. Klatzow, J. N. Becker, P. M. Ledingham, C. Weinzetl, K. T. Kaczmarek, D. J. Saunders, J. Nunn, I. A. Walmsley, R. Uzdin, and E. Poem, Experimental demonstration of quantum effects in the operation of microscopic heat engines, Phys. Rev. Lett. **122**, 110601 (2019).

[26] R. Uzdin, A. Levy, and R. Kosloff, Equivalence of quantum heat machines and quantum-thermodynamic signatures, Phys. Rev. X **5**, 031044 (2015).

[27] G. Watanabe, B. P. Venkatesh, P. Talkner, and A. del Campo, Quantum performance of thermal machines over many cycles, Phys. Rev. Lett. **118**, 050601 (2017).

[28] Y.-H. Ma, C. L. Liu, and C. P. Sun, Works with quantum resource of coherence, arXiv:2110.04550.

[29] J.-F. Chen, C.-P. Sun, and H. Dong, Boosting the performance of quantum Otto heat engines, Phys. Rev. E **100**, 032144 (2019).

[30] J.-F. Chen, C.-P. Sun, and H. Dong, Achieve higher efficiency at maximum power with finite-time quantum Otto cycle, Phys. Rev. E **100**, 062140 (2019).

[31] M. Perarnau-Llobet, K. V. Hovhannisyan, M. Huber, P. Skrzypczyk, N. Brunner, and A. Acín, Extractable work from correlations, Phys. Rev. X **5**, 041011 (2015).

[32] K. Korzekwa, M. Lostaglio, J. Oppenheim, and D. Jennings, The extraction of work from quantum coherence, New J. Phys. **18**, 023045 (2016).

[33] M. Perarnau-Llobet, E. Bäumer, K. V. Hovhannisyan, M. Huber, and A. Acin, No-go theorem for the characterization of work fluctuations in coherent quantum systems, Phys. Rev. Lett. **118**, 070601 (2017).

[34] M. Lostaglio, Quantum fluctuation theorems, contextuality, and work quasiprobabilities, Phys. Rev. Lett. **120**, 040602 (2018).

[35] E. Wigner, On the quantum correction for thermodynamic equilibrium, Phys. Rev. **40**, 749 (1932).

[36] J. G. Kirkwood, Quantum statistics of almost classical assemblies, Phys. Rev. **44**, 31 (1933).

[37] P. A. M. Dirac, On the analogy between classical and quantum mechanics, Rev. Mod. Phys. **17**, 195 (1945).

[38] H. Margenau and R. N. Hill, Correlation between measurements in quantum theory, Prog. Theor. Phys. **26**, 722 (1961).

[39] G. Francica and L. Dell'Anna, Quasiprobability distribution of work in the quantum ising model, Phys. Rev. E **108**, 014106 (2023).

[40] A. Santini, A. Solfanelli, S. Gherardini, and M. Collura, Work statistics, quantum signatures and enhanced work extraction in quadratic fermionic models, Phys. Rev. B **108**, 104308 (2023).

[41] M. Lostaglio, A. Belenchia, A. Levy, S. Hernández-Gómez, N. Fabbri, and S. Gherardini, Kirkwood-Dirac quasiprobability approach to the statistics of incompatible observables, Quantum **7**, 1128 (2023).

[42] A. Ortega, E. McKay, A. M. Alhambra, and E. Martín-Martínez, Work distributions on quantum fields, Phys. Rev. Lett. **122**, 240604 (2019).

[43] A. Teixidó-Bonfill, A. Ortega, and E. Martín-Martínez, First law of quantum field thermodynamics, Phys. Rev. A **102**, 052219 (2020).

[44] A. E. Allahverdyan, Nonequilibrium quantum fluctuations of work, Phys. Rev. E **90**, 032137 (2014).

[45] P. Solinas and S. Gasparinetti, Full distribution of work done on a quantum system for arbitrary initial states, Phys. Rev. E **92**, 042150 (2015).

[46] Y. V. Nazarov and M. Kindermann, Full counting statistics of a general quantum mechanical variable, Eur. Phys. J. B **35**, 413 (2003).

[47] G. Francica, Class of quasiprobability distributions of work with initial quantum coherence, Phys. Rev. E **105**, 014101 (2022).

[48] G. Francica, Most general class of quasiprobability distributions of work, Phys. Rev. E **106**, 054129 (2022).

[49] H. J. D. Miller and J. Anders, Time-reversal symmetric work distributions for closed quantum dynamics in the histories framework, New J. Phys. **19**, 062001 (2017).

[50] R. Sampaio, S. Suomela, T. Ala-Nissila, J. Anders, and T. G. Philbin, Quantum work in the bohmian framework, Phys. Rev. A **97**, 012131 (2018).

[51] T. Sagawa, Second law-like inequalities with quantum relative entropy: An introduction, in *Lectures on Quantum Computing, Thermodynamics and Statistical Physics* (World Scientific, Singapore, 2013), pp. 125–190.

[52] E. Bäumer, M. Lostaglio, M. Perarnau-Llobet, and R. Sampaio, Fluctuating work in coherent quantum systems: Proposals and limitations, 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 (Springer International Publishing, Cham, 2018), pp. 275–300.

[53] R. Pan, Z. Fei, T. Qiu, J.-N. Zhang, and H. T. Quan, Quantum-classical correspondence of work distributions for initial states with quantum coherence, arXiv:1904.05378.

[54] M. Beau, J. Jaramillo, and A. Del Campo, Scaling-up quantum heat engines efficiently via shortcuts to adiabaticity, Entropy **18**, 168 (2016).

[55] S. Lee, M. Ha, and H. Jeong, Quantumness and thermodynamic uncertainty relation of the finite-time Otto cycle, Phys. Rev. E **103**, 022136 (2021).

[56] Z. Fei, J.-F. Chen, and Y.-H. Ma, Efficiency statistics of a quantum Otto cycle, Phys. Rev. A **105**, 022609 (2022).

[57] Z. Fei and H. T. Quan, Group-theoretical approach to the calculation of quantum work distribution, Phys. Rev. Res. **1**, 033175 (2019).

[58] Z. Gong and H. T. Quan, Jarzynski equality, crooks fluctuation theorem, and the fluctuation theorems of heat for arbitrary initial states, Phys. Rev. E **92**, 012131 (2015).

[59] L. M. Johansen, Quantum theory of successive projective measurements, Phys. Rev. A **76**, 012119 (2007).

[60] R. Dorner, S. R. Clark, L. Heaney, R. Fazio, J. Goold, and V. Vedral, Extracting quantum work statistics and fluctuation theorems by single-qubit interferometry, Phys. Rev. Lett. **110**, 230601 (2013).

[61] L. Mazzola, G. De Chiara, and M. Paternostro, Measuring the characteristic function of the work distribution, Phys. Rev. Lett. **110**, 230602 (2013).

[62] H. F. Hofmann, How weak values emerge in joint measurements on cloned quantum systems, Phys. Rev. Lett. **109**, 020408 (2012).

[63] F. Buscemi, M. Dall'Arno, M. Ozawa, and V. Vedral, Universal optimal quantum correlator, Int. J. Quantum Inform. **12**, 1560002 (2014).

[64] J. S. Lundeen, B. Sutherland, A. Patel, C. Stewart, and C. Bamber, Direct measurement of the quantum wave function, Nature (London) **474**, 188 (2011).

[65] J. S. Lundeen and C. Bamber, Procedure for direct measurement of general quantum states using weak measurement, Phys. Rev. Lett. **108**, 070402 (2012).

[66] P. Rall, Quantum algorithms for estimating physical quantities using block encodings, Phys. Rev. A **102**, 022408 (2020).

[67] S. Hernndez-Gmez, S. Gherardini, A. Belenchia, M. Lostaglio, A. Levy, and N. Fabbri, Projective measurements can probe nonclassical work extraction and time-correlations, arXiv:2207.12960.

[68] M. Lostaglio, Certifying quantum signatures in thermodynamics and metrology via contextuality of quantum linear response, Phys. Rev. Lett. **125**, 230603 (2020).

[69] J.-F. Chen, T. Qiu, and H.-T. Quan, Quantum-classical correspondence principle for heat distribution in quantum Brownian motion, Entropy **23**, 1602 (2021).

[70] A. Levy and M. Lostaglio, Quasiprobability distribution for heat fluctuations in the quantum regime, PRX Quantum **1**, 010309 (2020).

[71] H. Kirchberg, M. Thorwart, and A. Nitzan, Charge transfer through redox molecular junctions in nonequilibrated solvents, J. Phys. Chem. Lett. **11**, 1729 (2020).

[72] H. Kirchberg and A. Nitzan, Energy transfer and thermoelectricity in molecular junctions in nonequilibrated solvents, J. Chem. Phys. **156**, 094306 (2022).

[73] G. Manzano, J. M. R. Parrondo, and G. T. Landi, Non-Abelian quantum transport and thermosqueezing effects, PRX Quant. **3**, 010304 (2022).

[74] N. Yunger Halpern, P. Faist, J. Oppenheim, and A. Winter, Microcanonical and resource-theoretic derivations of the thermal state of a quantum system with noncommuting charges, Nat. Commun. **7**, 12051 (2016).

[75] S. Majidy, W. F. Braasch, A. Lasek, T. Upadhyaya, A. Kalev, and N. Y. Halpern, Noncommuting conserved charges in quantum thermodynamics and beyond, Nat. Rev. Phys. **5**, 689 (2023).

[76] T. Upadhyaya, J. William F. Braasch, G. T. Landi, and N. Y. Halpern, What happens to entropy production when conserved quantities fail to commute with each other, arXiv:2305.15480.

[77] J. B. Conway, Normal operators on hilbert space, in *A Course in Functional Analysis* (Springer, New York, NY, 2007), pp. 255–302.

[78] J. B. Conway, Operators on hilbert space, in *A Course in Functional Analysis* (Springer, New York, NY, 2007), pp. 26–62.