Energy exchange statistics and fluctuation theorem for nonthermal asymptotic states

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Energy exchange statistics between two bodies at different thermal equilibria obey the Jarzynski-Wójcik fluctuation theorem. The corresponding energy scale factor is the difference of the inverse temperatures associated to the bodies at equilibrium. In this work, we consider a dissipative quantum dynamics leading the quantum system towards a possibly nonthermal, asymptotic state. To generalize the Jarzynski-Wójcik theorem to nonthermal states, we identify a sufficient condition $\mathcal{I}$ for the existence of an energy scale factor η^* that is unique, finite, and time independent, such that the characteristic function of the energy exchange distribution becomes identically equal to 1 for any time. This η^* plays the role of the difference of inverse temperatures. We discuss the physical interpretation of the condition $\mathcal{I}$, showing that it amounts to an almost complete memory loss of the initial state. The robustness of our results against quantifiable deviations from the validity of $\mathcal{I}$ is evaluated by experimental studies on a single nitrogen-vacancy center subjected to a sequence of laser pulses and dissipation.

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I. INTRODUCTION

The probability distribution of a random variable defined at two times generally depends on the details of the system's dynamical process. A key example is provided by the internal energy variation $\Delta E(t)$. According to the formalism of stochastic thermodynamics [1–6], in a single quantum trajectory the value of $\Delta E(t)$ in the time interval $[t_0, t]$ is defined as the difference of eigenvalues of the quantum system Hamiltonian $\mathcal{H}(t)$ evaluated at the times t_0 and t. The fluctuations can be analyzed by means of the probability distribution $\mathrm{P}(\Delta E(t), t)$, or its characteristic function $\mathcal{G}(u, t)$, with u a complex number, given by the Fourier transform of $\mathrm{P}(\Delta E(t), t)$.

Fluctuation theorems can connect nonequilibrium quantities to equilibrium properties of the dynamical system [7,8]. In the following we are going to address *energy exchange* fluctuation theorems for dissipative quantum systems. In the original formulation of exchange fluctuation theorems [9], two bodies $\mathcal{A}$ and $\mathcal{B}$ with finite dimension are prepared in two equilibrium states at temperatures $T_{\mathcal{A}} \equiv 1/(k_B\beta_{\mathcal{A}})$ and $T_{\mathcal{B}} \equiv 1/(k_B\beta_{\mathcal{B}})$, respectively, and then placed in thermal contact for a given time interval. Fluctuation theorems address the question whether there exists a (single) value of $u = j\eta$ such that the characteristic function (for the separated system) becomes independent of time and equal to 1. Then, such a value acts as an effective "macroscopic" rescaling at all times of the heat exchange fluctuations between the two bodies. Once they are divided again, the question was to understand whether there exists a single "macroscopic" quantity able to rescale at any time the statistics (thus, the fluctuations) of the heat exchange between the two bodies. In Ref. [9], under the assumption of weak interaction between the two bodies, it was shown that for any time t after physical operation of decoupling the systems it is

$$\langle e^{-\Delta\beta\, Q}\rangle_t = 1, \tag{1}$$

where Q is the stochastic value of the heat exchanged by the two bodies, and the average $\langle\cdot\rangle$ is performed over the exchanged heat distribution. In Eq. (1), $\Delta\beta \equiv \beta_{\mathcal{B}} - \beta_{\mathcal{A}}$ denotes the energy scale factor that normalizes the heat fluctuations. The exchanged heat equality discussed above is valid [10] also by taking an initial thermal state and employing the so-called two-point measurement (TPM) scheme [11,12], where quantum measurements are performed at the initial and final times. The TPM scheme is a convenient way to access fluctuations, as experimentally shown in Refs. [13–19]. Recent activity

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focused on how energy exchange statistics could be reconstructed using schemes beyond the TPM scheme [20–27], when quantum coherences in the initial state are taken into account.

Noticeably, Eq. (1) is valid also when a (finite-dimensional) body $\mathcal{A}$, initially in the equilibrium state at temperature $T_{\mathcal{A}}$, is put at a certain time t^* in contact with a *thermal bath* $\mathcal{B}$ at temperature $T_{\mathcal{B}}$ [28]. The detailed balance condition (DBC) is a sufficient condition for the validity of a Crook fluctuation relation [29]; the converse, instead, is not necessarily valid [28]. Independently of the microscopic details of the coupling between $\mathcal{A}$ and $\mathcal{B}$, Eq. (1) is valid at any time later than t^* [28]. This shows that $\Delta\beta$ plays the role of a macroscopic quantity that fully characterizes fluctuations of the energy exchanged at any time, and it is solely determined by the knowledge of the initial and final equilibrium states of the thermodynamic process. In general, this symmetry in the exchanged heat statistics is not present. As an example, in Ref. [30], Eq. (1) is generalized to the case that the initial states of the two bodies $\mathcal{A}$ and $\mathcal{B}$ are nonthermal as an effect of classical correlations. However, the derivation of a fluctuation relation as Eq. (1) in closed form for such a case requires the complete knowledge of both bodies and full access to their whole dynamics. Whether and when one can determine a counterpart of $\Delta\beta$ for nonthermal initial and final states is an open question.

In this paper, we present a fluctuation theorem for the energy exchanged by a quantum system with a dissipative environment, not necessarily at thermal equilibrium. We introduce a sufficient condition for the existence of a unique, finite, and constant (time-independent) energy scale factor η^* such that the characteristic function $\mathcal{G}(u,t) \equiv \int \mathrm{P}(\Delta E, t) e^{ju\Delta E} d\Delta E$ obeys the relation

$$\mathcal{G}(j\eta^*, t) = \langle e^{-\eta^* \Delta E} \rangle_t = 1 \quad \text{for} \quad t \geqslant t^*. \tag{2}$$

In Eq. (2), j denotes the imaginary unit. In general, if the energy scale factor is chosen without a specific criterion, then $\mathcal{G}(j\eta, t)$, with $\eta \in \mathbb{R}$, is not equal to 1 and its value depends on the choice of both the initial state and the whole dynamical process for any time t^*.

The energy exchange fluctuation theorem (2) for nonthermal asymptotic states and the discussion of the sufficient condition making it valid are the main results of the paper. This sufficient condition contains the DBC under two working hypotheses, namely, evaluating fluctuations by means of the TPM scheme, thanks to which a quantum-classical correspondence is possible [31,32], and a dynamical process that asymptotically leads the system into an asymptotic state. A key point of the sufficient condition is related to the memory loss of the exchanged energy from the initial quantum state. For this reason, we motivate the statement of the sufficient condition by making use of Stern-Gerlach protocols [33–35], where the memory loss is induced by quantum measurements. In this way, we can better illustrate the assumptions behind the fluctuation theorem and its physical interpretation.

Equation (2) can find application also to quantum systems under dissipation that do not reach an equilibrium but admit a nonequilibrium steady state. As an example, one may consider the case of time evolutions along isoenergetic trajectories, whereby asymptotically the probabilities to measure the energy values of the system depend neither on time nor on the initial state.

Finally, the robustness of the fluctuation theorem (2) is tested under exemplary experimental conditions where the validity (or not) of the fluctuation theorem's assumption, as well as of the DBC, can be controlled. We also discuss how η^* depends on the values taken by the parameters of the Hamiltonian. Here, the used platform is an autonomous dissipative Maxwell demon formed by a nitrogen-vacancy (NV) center in diamond at room temperature, introduced in Ref. [36].

II. ENERGY SCALE FACTOR IN THE ASYMPTOTIC REGIME

Let us discuss the working hypotheses that will be used in the rest of the paper.

We consider a quantum system with finite-dimensional Hilbert space (having dimension n) subjected to a Hamiltonian with time-independent eigenvalues, and at the same time under the effect of dissipation. We assume that the dynamics is determined by a completely positive trace-preserving (CPTP) map Φ_t that for large times leads the system to an asymptotic, possibly nonequilibrium, state of the form

$$\lim_{t\to\infty} \Phi_t[\rho_0] = \sum_{f=1}^{n} P_f(\infty)\Pi_f(\infty) + \chi[\rho_0, t] \tag{3}$$

that contains fixed values on the diagonal with respect to the energy basis $\{E_f\}$, with $\Pi_f \equiv |E_f\rangle\langle E_f|$, and possibly time-dependent off-diagonal terms χ. The probabilities $P_f(\infty)$, for $f \in \{1, \ldots, n\}$, are independent of both the time and the generic initial state ρ_0, while χ can depend on ρ_0 even in the asymptotic regime.

Exchange energy fluctuations are accessed by employing the TPM scheme. We recall that the TPM scheme provides complete energy exchange statistics in terms of correct marginals and linearity in the state, for any initial state that is diagonal in the system Hamiltonian [37].

In order to demonstrate the existence of a unique, finite, time-independent energy scale factor η^* obeying the fluctuation theorem (2), as a first step we prove the following lemma that is valid in the asymptotic limit:

Lemma 1 (Uniqueness of a nontrivial energy scale factor in the asymptotic limit). For a quantum system with a Hamiltonian with time-independent eigenvalues and subjected to a dissipative map, the equality $\mathcal{G}(j\eta, t) = 1$ is fulfilled asymptotically for $t \to \infty$ only for two distinct values of η at most, namely, $\eta = 0$ (trivial solution) and $\eta = \eta^*$, with $\eta^* \in \mathbb{R}$ a finite real number depending only on the initial and the asymptotic states.

As shown in Appendix A, this result holds as the characteristic function

$$\mathcal{G}(j\eta, t) = \sum_{i,f} P_i P_{f|i}(t) e^{-\eta \Delta E_{i,f}} \tag{4}$$

is analytic and convex in η [38,39]. In Eq. (4), P_i is the probability of measuring E_i at the beginning of the protocol, $P_{f|i}(t)$ is the conditional probability to get E_f at time t after having initially measured E_i, and $\Delta E_{i,f} \equiv E_f - E_i$.

We can now establish the following:

Corollary 1 (Application of Lemma 1 to unital quantum maps). If the initial state is thermal at inverse temperature β and the open quantum dynamics is such that the asymptotic state is the completely mixed state, then the nontrivial solution of $\mathcal{G}(j\eta^*, t) = 1$ at $t \to \infty$ is $\eta^* = \beta$.

The validity of Corollary 1 can be verified by making the substitution $\eta^* = \beta$ and $P_f(\infty) = 1/n$ in $\lim_{t\to\infty} \mathcal{G}(j\eta^*, t)$, where $P_f(\infty)$ is the probability to measure the fth energy value of the system in the correspondence of the asymptotic state.

III. MEMORY LOSS ASSUMPTIONS

In this section we present a discussion of assumptions under which Eq. (2) is valid, not only asymptotically ($t \to \infty$), but for $t \geqslant t^* \geqslant 0$, still using η^* as the energy scale factor. The main motivation in understanding whether and how to rescale energy exchange fluctuations over time with η^*, which we recall depends only on the initial and the asymptotic states, lies within the objective to generalize the Jarzynski-Wójcik fluctuation theorem to a generic dissipative quantum system. In these terms, η^* can be considered as a macroscopic quantity that effectively acts as the proxy of an inverse temperature [40].

One would like to determine necessary and sufficient conditions for having $\mathcal{G}(j\eta^*, t) = \langle e^{-\eta^* \Delta E}\rangle_t = 1$ for $t \geqslant t^*$; see also Eq. (2). Even finding a necessary condition is a very complicated task in the general case, where no specifications on the (thermo)dynamic process under scrutiny are provided. So, we are going to introduce a sufficient condition that guarantees the validity of Eq. (2) depending on properties (specified below) of the energy exchange statistics. In particular, we will focus on dynamics where memory of the initial state is partially lost, which generalizes the case of thermalizing quantum dynamics [28]. In doing this, we are led by the requirement to work within a dynamical regime that (i) can be associated to paradigmatic models with a clear physical meaning and (ii) can be tested in meaningful experimental scenarios.

Regarding point (i), we are going to consider a generalization of Stern-Gerlach protocols where a series of projective measurements is performed sequentially, and among the measurements a dynamical quantum map is included. Moreover, we also give the possibility that each measurement does occur (or fails) with a given probability. For point (ii), concerning an experimental scenario where our findings can be tested, we will consider a dissipative quantum Maxwell demon. Thus, our theory will be experimentally tested on a single NV center in diamond at room temperature that is subjected to a sequence of laser pulses, each of which acts as a combination of a quantum measurement and a dissipative channel. Deviations of the theoretical assumption enabling Eq. (2) from experimental data are analyzed in Sec. V.

A. Generalized Stern-Gerlach protocols

In order to find a sufficient condition for Eq. (2), we start by observing that Eq. (2) is implied by the loss of memory, starting from a time t^*, of the chosen initial state. This is what occurs in the Stern-Gerlach experiment and its variations [41], when a measurement of a certain observable is followed by other measurements of noncommuting observables, so that the later measurements cancel the information about the initial state. Motivated by this observation, here we will consider a Stern-Gerlach protocol in which the first and last measurements are projective measurements and their outcomes are collected, as in the usual Stern-Gerlach protocol. Instead, the second measurement is replaced by a sequence of quantum measurements that all refer to the same observable and take place with a probability $p_m < 1$. We will also study the cases with unitary and dissipative dynamics in between the quantum measurements. We refer to this protocol as a generalized Stern-Gerlach protocol.

We start by considering the simplest Stern-Gerlach protocol for our purposes, namely, the sequential measurement (via projective measurements) of a two-level quantum system (e.g., a spin-$\frac{1}{2}$) with respect to three observables: the first and the third, the Hamiltonian $\mathcal{H}$ of the system (say, the Pauli matrix σ_x), and the intermediate observable an operator that is maximally noncommuting with $\mathcal{H}$ (say, σ_z). Only the outcomes of the first and third measurements are collected, giving origin to the joint probability $P_i P_{f|i}(t)$. For this paradigmatic example, where the probability to measure σ_z (at time t^*) is equal to 1, the conditional probabilities $P_{f|i}(t) = 1/2$ are independent on i, for any f and $t \geqslant t^*$. In fact, whatever is the initial state, consecutively measuring the Pauli matrices σ_x and σ_z leads to a *complete destruction* of the information contained in the initial state [41].

We would be then tempted to set as a sufficient condition for Eq. (2) the following hypothesis:

Hypothesis 1 (*Complete memory loss of the initial energy statistics from a time* t^*). For any final state f, $P_{f|i}(t)$ is independent of all initial states i for $t > t^*$.

However, we will argue that the hypothesis 1 is unnecessarily too restrictive, and possibly unfeasible for quantum systems with dimension larger than 2. Sequential measurements over maximally noncommuting observables lead to complete memory loss, irrespective of the system dimension. In general, determining maximally noncommuting observables that are also experimentally implementable is a nontrivial task. In quantum systems whose dimension is a prime number, or powers of a prime number, two maximally noncommuting observables can be obtained from mutually unbiased bases [42–44]. Whether or not such observables are experimentally implementable depends on the platform. Orthogonal observables, such as the angular momentum operators easily obtained in experimental routines, can provide a readily accessible noncommuting set, albeit no maximal noncommuting. For example, the orthogonal angular momentum operators S_x and S_z are not maximally noncommuting even for a qutrit, a system of relevance to our experiments. Consequently, alternating measurements of S_x and S_z (e.g., measuring S_x, S_z, and S_x) do not completely erase the information about the initial state, and the condition of complete memory loss is no longer valid since $P_{f|i}(t)$ still depends on i. The need of finding maximally noncommuting observables can be overcome by introducing a dynamical evolution given by a map Φ_t interspersed by consecutive measurements, so as to scramble the state of the system and induce memory loss.

Let us further analyze these cases. Consider a protocol in which the first and last measurements are the same as before—

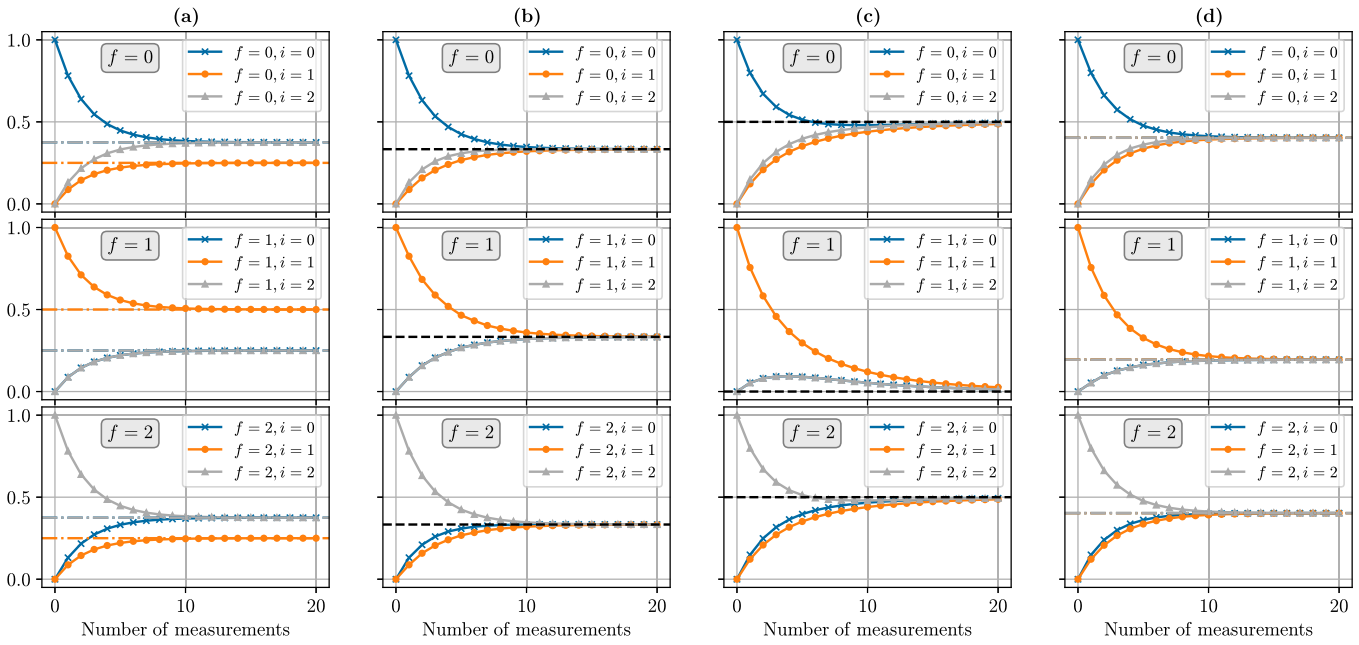

FIG. 1. Energy jump conditional probability $P_{f|i}$ associated with the Stern-Gerlach-like experiment where the Hamiltonian of the three-level system, $\mathcal{H} \propto S_x$, is measured once at the beginning of the protocol and once at the end. In between these measurements, a set of quantum measurements of the observable S_z is considered, and we assume that the system is projectively measured with finite probability ($p = 0.35$). The x axis indicates the number of these intermediate measurements applied to the system. The states 0 and 2 correspond to the eigenstates of S_x with positive and negative eigenvalues, respectively, while the state 1 refers to the eigenstate with associated eigenvalue equal to zero. (a) No dynamics in between measurements. The state of the system in the case the probability of absorption is equal to 1 is indicated by the dot-dashed lines. (b) Unitary evolution in between measurements leading to complete memory loss. The unitary evolution affects the state of the system and, after a series of quantum measurements, it becomes impossible to retrieve information about the initial system's state. The asymptotic state is a completely mixed state, which is denoted by the black dashed lines. [(c) and (d)] Same as (a) and (b), respectively, but with dissipation conditioned on the measurements. The system eventually loses information of the initial state, but the asymptotic state is not completely mixed. In (c), the dissipation pumps the system into the S_z eigenstate with $m_S = 0$ (black dashed lines). In (d), the system is pumped into an out-of-equilibrium steady state indicated by the dot-dashed lines.

projective measurements of the Hamiltonian (e.g., the angular momentum operator S_x)—and we still consider the case where only the outcomes of the first and last measurements are collected. Instead, the second measurement (S_z) is replaced by a sequence of quantum measurements that all refer to the same observable (e.g., S_z) and take place with a probability $p_m < 1$. From an operational point of view, this means that, with periodicity τ, the system is subjected to a projective measurement with probability p_m, thus making the system's wave function collapse (in our example into one of the eigenstates of S_z), while it remains unaltered with probability $1 - p_m$. This specific kind of quantum measurement—occurring after any time interval τ—can be described using the positive operator-valued measure (POVM) formalism. We will focus our attention on the behavior of the energy jump conditional probability $P_{f|i}(t)$ for this Stern-Gerlach-like experiment.

If we consider no evolution between consecutive measurements, i.e., the unitary operator describing the evolution of the system is the identity, then the complete memory loss is not attained. This is shown in Fig. 1(a). In this case, the state of the system asymptotically approaches the state that one would get if we perform a single intermediate projective measurement taking place with probability equal to 1, which corresponds to the Stern-Gerlach experiment for a three-level system with the measurements S_x, S_z, and S_x. The dot-dashed lines in Fig. 1(a) denote the results of this Stern-Gerlach experiment with a three-level system, such that $P_{f|i}(t \to \infty) = \sum_{j=1}^{3} |\langle x_f|z_j\rangle|^2 |\langle z_j|x_i\rangle|^2$, where $|\alpha_k\rangle$ is the kth eigenstate of S_α. As explained before, this case does not represent a complete loss of memory because S_z and S_x are not maximally noncommuting observables.

Complete memory loss is reintroduced if we modify the protocol by including a unitary evolution of the system between the measurements. In such a case, shown in Fig. 1(b), there exists a time t^* (or a finite number of intermediate measurements) after which all the conditional probabilities $P_{f|i}(t)$ tend to $1/n$, with n the dimension of the quantum system ($n = 3$ in this example). The number of different trajectories increases as the power of $n + 1$ with respect to the number of measurements; in our example, four outcomes are possible from each measurement: project into one of the eigenstates of S_z or remain unaltered. With this large number of trajectories affected by the unitary evolution in between measurements, eventually it is impossible to retrieve information about the initial state of the system. This represents a complete loss of memory, thereby fulfilling hypothesis 1, where $P_{f|i}(t)$ is independent of i for any f, for $t \geqslant t^*$. Such a behavior is in agreement with previously known results where dynamics leading to infinite-temperature thermalization (i.e., the state of the system converges towards a completely mixed state)

can be induced by a sequence of projective measurements under specific conditions [45,46]. Notice that an exception occurs when the unitary operator U describing the evolution between measurements commutes with the operators of the intermediate POVMs (in our case when $[U, S_z] = 0$). Such an exceptional case falls back to the case depicted in Fig. 1(a).

We are now in the position to analyze the behavior of the conditional probabilities $P_{f|i}(t)$ in Stern-Gerlach-like experiments that also include *dissipation*. We consider a sequence of intermediate projective measurements of S_z interspersed by unitary operators U not commuting with S_z. As before, the first and the last energy measurements are performed along the basis of S_x, and their outcomes are collected. We still consider that, with periodicity τ, the quantum system is projectively measured with probability p_m, but in this case if the measurement occurs then the state of the system collapses and changes due to a dissipative channel. The system still remains unaffected with probability $1 - p_m$. We will consider a dissipative channel that *pumps* the state of the system into one of the eigenstates of the observable S_z (in this particular example, into the $m_S = 0$ state). This case is particularly interesting since it can be used to interpret the effect of *optical pumping* with short excitation pulses in realistic experimental scenarios, where the probability of light absorption is smaller than 1. An example of this is the implementation of Maxwell's demon on an NV center via a train of short laser pulses [36].

In analogy to the case without dissipation, we will first consider the case where the unitary operator U describing the evolution between measurements is equal to the identity, and then we will consider the case where $[U, S_z] \neq 0$. In the former case, the effect of the dissipation builds up as the number of measurements increases, and eventually the system is pumped into the fixed point of the dissipation, the $m_S = 0$ state. This is shown in Fig. 1(c), where the dashed line represents the projection of the $m_S = 0$ state in the S_x basis. In contrast, when $[U, S_z] \neq 0$, the combination of projective measurements and dissipation, together with unitary evolutions, results in a map with a fixed point that is an out-of-equilibrium steady state. Also in this case the system is pumped into the fixed point, as shown in Fig. 1(d). The fixed point, denoted by the dot-dashed lines in Fig. 1(d), is determined by a nontrivial combination of the probability p_m, the strength of the dissipation, and the strength (Rabi frequency) of the unitary driving U. Further details can be found in Sec. V and in Ref. [36].

In these Stern-Gerlach-like experiments with dissipation, shown in Figs. 1(c) and 1(d), the system eventually loses information of the initial state, but the asymptotic state is not completely mixed. Therefore, these cases correspond to nonunital maps. Moreover, the hypothesis of complete destruction of information is not fulfilled, but a less stringent one holds, which still maintains a symmetry in the indexes of the measurement outcomes. Formally, if the dissipation is present, the following hypothesis, which will be extensively considered in the remainder of the paper, is valid:

Hypothesis 2 (*Almost complete memory loss of the initial energy statistics*). For any f and $t \geqslant t^*$, the conditional probabilities $P_{f|i}(t)$ are independent on all i with $i \neq f$.

Clearly, hypothesis 1 implies hypothesis 2, but not vice versa.

IV. FLUCTUATION THEOREM

We can now provide the statement of the fluctuation theorem formalizing Eq. (2). For this purpose, we express the conditional probabilities $P_{f|i}(t)$ as a function of the probabilities $P_f(\infty)$ in the asymptotic limit (assumed to exist). Specifically, for $i \neq f$, one is always allowed to write the conditional probabilities $P_{f|i}(t)$ as $P_{f|i}(t) = F_{i,f}(t)P_f(\infty)$ with $F_{i,f}(t)$ a generic bounded real function depending on the indices i, f, such that $F_{i,f}(t_0) = 0$ and $\lim_{t\to\infty} F_{i,f}(t) = 1\ \forall\, i, f$. Thus,

Theorem 1 (Fluctuation theorem for dissipative quantum dynamics). Under the validity of both the hypothesis 2 and the detailed balance condition, implying $P_{f|i}(t) = \overline{F}(t)P_f(\infty)\ \ \forall\, i, f, t \geqslant t^*$ with $i \neq f$ and $\overline{F}(t)$ a time-dependent real function, then

$$\mathcal{G}(j\eta^*) = 1 \quad \text{at} \quad t \to \infty \iff \mathcal{G}(j\eta^*, t) = 1 \quad \forall t \geqslant t^*. \tag{5}$$

The proof of Theorem 1 is in Appendix B. It is worth noting that the assumption of Theorem 1 includes the DBC

$$\frac{P_{f|i}(t)}{P_{i|f}(t)} = \frac{P_f(\infty)}{P_i(\infty)} \tag{6}$$

that is obeyed by any reversible Markov process. Moreover, since hypothesis 2 is implied by the complete destruction of information, then also assuming the latter leads to the validity of the theorem's thesis. Theorem 1 provides a sufficient condition such that $\mathcal{G}(j\eta^*, t) = 1$ for $t \geqslant t^*$ using the energy scale factor η^* that only depends on the initial and asymptotic states. Theorem 1 holds independently of the Hamiltonian, as long as its eigenvalues are time independent, and of the dimension of the system. Moreover, Theorem 1 does not require a specific initial state, provided, however, the TPM scheme is applied.

A. Thermal states

Let us discuss a meaningful limit case. When the initial and asymptotic states are thermal with inverse temperatures β and β_∞, respectively, the energy scale factor $\eta^* = \Delta\beta = \beta - \beta_\infty$. Hence, Eq. (2) reduces to the exchange fluctuation relation that is valid for a quantum system under thermalizing dynamics [9,28], or under an effective thermalizing quantum map [15] as it occurs in two-level quantum systems. In the high-temperature limit ($\beta_\infty = 0$), $\eta^* = \beta$. Notice that for two-level quantum systems there always exists an effective temperature such that the elements of any mixed state can be sampled from a Boltzmann distribution. On the contrary, higher-dimensional quantum systems (as in the present paper) provide genuinely quantum states that cannot be mapped into thermal states.

B. Energy extraction power

We conclude this section by showing that the sign of η^* determines whether energy is injected or can be extracted from a dissipative system:

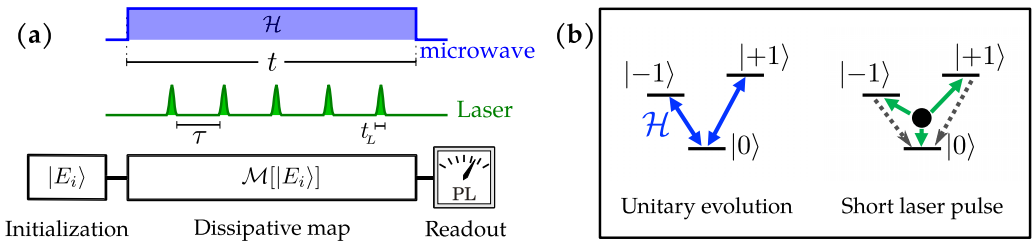

FIG. 2. (a) Sketch of the experimental protocol: The NV spin qutrit ($S_z = [0, \pm 1]$) is initialized in an eigenstate of the Hamiltonian $\mathcal{H}$, and evolves under a map $\mathcal{M}$ given by the combination of $\mathcal{H}$ and short laser pulses [36]. After the evolution for a time t, the spin energy is measured according to the TPM scheme. (b) The map $\mathcal{M}$ operates on the NV through a unitary evolution (generated by $\mathcal{H}$) and a combination of POVMs and dissipation towards the spin state $|0\rangle$ (via laser pulses), sketched in the left and right panels, respectively.

Corollary 2 (η^* as a measure of the energy extraction power). At t large ($t \geqslant t^*$), when the quantum system under dissipation has reached the asymptotic state, the necessary and sufficient condition for energy extraction is $\eta^* < 0$.

Corollary 2 is obtained by applying the Jensen inequality to Eq. (2) that thus implies $\eta^* \langle \Delta E \rangle \geqslant 0$. As a result, energy is extracted from the system when $\eta^* < 0$, and is transferred to the system otherwise ($\eta^* > 0$).

V. EXPERIMENTAL SCENARIO: DISSIPATIVE QUANTUM MAXWELL DEMONS

In this section, we introduce an experimental testbed very relevant for our purposes: the dissipative quantum Maxwell demon, originally introduced in Ref. [36]. The quantum maps realized in this physical platform admit a nonequilibrium steady state that is induced by a sequence of measurements followed by feedback actions. With this setup we test the validity of Theorem 1's assumptions, as well as its thesis.

The experimental platform is based on single NV centers, where the NV electronic spin $S = 1$ is optically initialized and read out, and manipulated with a Hamiltonian $\mathcal{H}$ via a microwave radiation. A detailed discussion of the apparatus, as well as the experimental realization (TPM scheme included) is reported in Ref. [36]. Here we give a summary of the protocol. The NV bare Hamiltonian commutes with the spin operator S_z (with eigenstates $|0\rangle$, $|\pm 1\rangle$). In the presence of a near-resonance microwave field, the system Hamiltonian $\mathcal{H}$ can be written in a rotating frame as a time-independent linear combination of the operators S_x, S_y, and S_z. During the unitary evolution determined by this Hamiltonian, the NV spin state, represented by the density operator ρ, is subjected to a series of short laser pulses, inducing POVMs and dissipation (see Fig. 2). In the terminology used in Sec. III, in the presence of a single laser pulse the NV spin remains unaltered with a probability $1 - p_m$, and it is subjected to a projective measurement (S_z) and dissipation towards $m_S = 0$ with a probability p_m. A formal model of the NV photodynamics involves radiative and nonradiative transitions, including fast phononic relaxation, whose details will not be covered here since they are thoroughly explained in Ref. [36]. The combination of unitary evolution under the Hamiltonian $\mathcal{H}$ and short laser pulses give rise to the CPTP map $\mathcal{M}$. Notably, the quantum dynamical map $\mathcal{M}$, applied to the $\mathcal{H}$ eigenstates, leads the system to a nonequilibrium steady state originated by a nontrivial interplay between quantum measurements and dissipation. The described experiment embodies the right tool to demonstrate the robustness of the theorem presented in this work. In fact, the possibility to design different Hamiltonian operators for the unitary evolution of the spin qutrit allows us to implement various protocols where the DBC may be fulfilled or not.

We would stress that this case study is appropriate for the test of the fluctuation theorem introduced in Sec. IV, since the Hamiltonian of this quantum system can be controlled such that the hypothesis 2 of Theorem 1 as well as the DBC, leading to the assumption $P_{f|i}(t) = \overline{F}(t) P_f(\infty)$ for $t \geqslant t^*$, are valid. Specifically, when $[\mathcal{H}, S_z] = 0$, Theorem 1 is fulfilled. In contrast, when $\mathcal{H} \propto S_x$, hypothesis 2 is only approximately valid, but still $\mathcal{G}(j\eta^*) \simeq 1$ *within experimental precision* for $t \geqslant t^*$. Even in this case where Theorem 1's assumption $P_{f|i}(t) = \overline{F}(t) P_f(\infty)$ holds only approximately or is even not valid, we can experimentally explore the deviations from the predictions of Theorem 1.

A. Robustness analysis

This section is devoted to investigating how much $\mathcal{G}(j\eta^*, t)$ deviates from 1 in case the assumptions of Theorem 1 are not fulfilled. A possible cause for such a condition is that DBC (6) does not hold, which in our case study occurs by setting $\mathcal{H} = \omega S_x$. Here, S_x is the spin operator orthogonal to the natural NV quantization axis z, and ω is the driving Rabi frequency. The energy eigenstates are $|E\rangle_f \in \{|-\omega\rangle, |\emptyset\rangle, |+\omega\rangle\}$, with $|\pm\omega\rangle \equiv (|-1\rangle \pm \sqrt{2}\,|0\rangle + |1\rangle)/2$ and $|\emptyset\rangle \equiv (|-1\rangle - |1\rangle)/\sqrt{2}$. In Fig. 3(a) we report the conditional probabilities $P_{f|+\omega}$ of measuring $|E\rangle_f$ as final states of the TPM scheme (at time t, after N_L laser pulses) once the NV spin is initialized in $|+\omega\rangle$. First, we find a good agreement between the experimental data (colored dots) and theoretical simulations of the evolution under the quantum dissipative model (dashed lines). More interestingly, by imposing

$$P_{f|i}(t) = P_f(\infty)(1 - e^{-t/\tau_D}) + \delta_{i,f}\, e^{-t/\tau_D} \tag{7}$$

with $\delta_{i,f}$ denoting the Kronecker delta between i and f, τ_D a decay time, and $i = +\omega$ in Fig. 3, it is possible to approximate the time evolution of the system to a case where the assumption $P_{f|i}(t) = \overline{F}(t) P_f(\infty)$ holds. Although the validity of this assumption is based on a quite different physical scenario, the agreement between the simulations [solid lines in Fig. 3(a)] and experiments is still valid. Then,

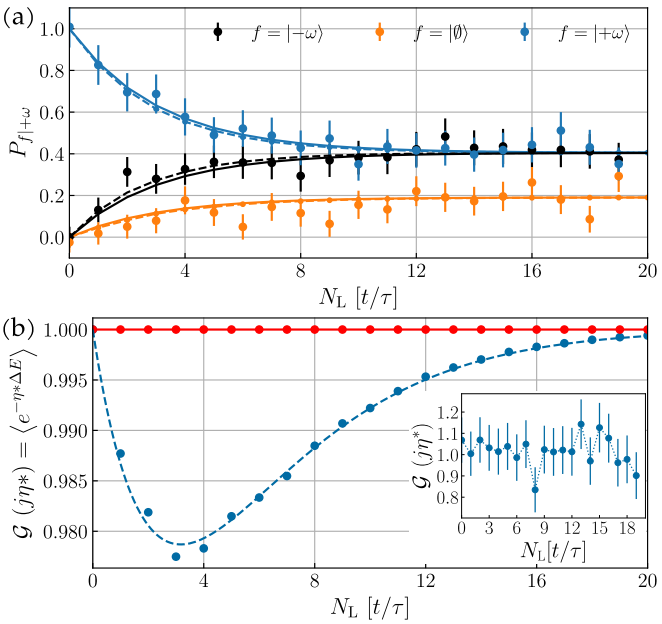

FIG. 3. (a) Conditional probabilities of measuring $|E\rangle_f \in \{|-\omega\rangle, |\emptyset\rangle, |+\omega\rangle\}$ while initializing the qutrit in $|+\omega\rangle$, as a function of the number of laser pulses, $N_L = t/\tau$, with t equal to $\mathcal{M}$ duration and τ the time interval between pulses. Colored scatters and dashed lines correspond to experimental data and numerical simulations, respectively, of the NV spin evolution under the map $\mathcal{M}$, with $\mathcal{H} = \omega S_x$. Solid lines represent the approximated model ensuring the DBC, obtained by fitting the numerical results using the function $P_{f|+\omega}(t) = P_f(\infty)(1 - e^{-t/\tau_D}) + \delta_{+\omega,f}\, e^{-t/\tau_D}$, with $\delta_{+\omega,f}$ the Kronecker delta and $\tau_D = 3.11\tau$. (b) Characteristic function $\mathcal{G}(j\eta^*, t)$ as a function of the number of laser pulses, $N_L = t/\tau$. $\mathcal{G}(j\eta^*, t)$ is evaluated from theoretical simulations (main plot) and experimental data (inset) by considering all the possible combinations $P_{f|t}(t)$, with $i, f \in \{-\omega, 0, +\omega\}$ (see Ref. [36]). The comparison between the approximated model with the DBC (in red) and the original one of Ref. [36] (blue) highlights a discrepancy, as expected from Theorem 1. The characteristic function for the original model can be approximated by $1 + e^{-t/\tau_a} - e^{-t/\tau_b}$, which is given by the blue dashed line. The fitted parameters $\tau_a = 3.07\tau$ and $\tau_b = 3.26\tau$ depend on the decay rate of the conditional probabilities. In the limit the DBC is valid, one obtains $\tau_a = \tau_b$.

by comparing the characteristic functions of the two models for the corresponding η^* value [Fig. 3(b)], we observe that $\mathcal{G}(j\eta^*) = 1$ for any time only for the detailed balanced case, as expected from Theorem 1. On the other hand, their difference is much smaller than our experimental uncertainty [see inset of Fig. 3(b)], proving the robustness of the fluctuation relation (2) against the possible experimental invalidation of the assumptions.

B. Dependence of η^* on the values of the Hamiltonian parameters

We consider now an experimental case where the assumptions used to obtain Theorem 1 hold. This is obtained by turning off the microwave radiation and using the natural NV spin Hamiltonian

$$\mathcal{H} = \Delta S_z^2 + \gamma_e B S_z, \tag{8}$$

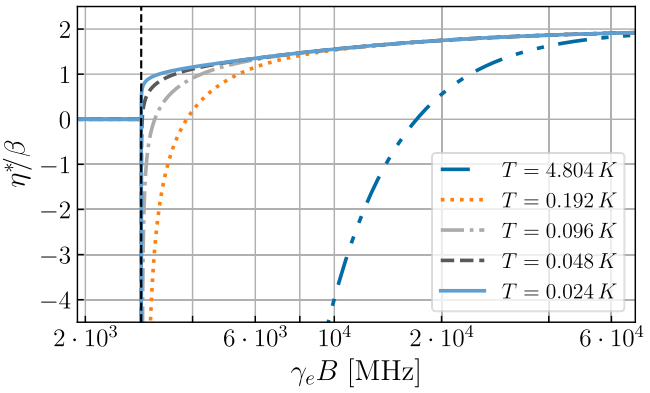

FIG. 4. Ratio between the energy scale factor η^* and the initial temperature T as a function of $\gamma_e B$. Lines are obtained by directly solving the characteristic function $\mathcal{G}$ (as discussed in detail in Appendix C) through a standard optimization routine. At the $|-1\rangle \leftrightarrow |0\rangle$ energy crossing ($\gamma_e B = \Delta$) we find a discontinuity in η^*.

with Δ the zero-field splitting, γ_e the electronic gyromagnetic ratio, and B a bias external magnetic field aligned with the NV quantization axis. Here, the effect of the laser pulse train, always pumping the NV spin in $|0\rangle$, is to make the qutrit collapse in one of the energy eigenstates. This allows us to find a simplified analytical solution to the characteristic function $\mathcal{G}$ with respect to the values of the Hamiltonian parameters (see Appendix C).

We can now distinguish two different regimes: $\gamma_e B < \Delta$ and $\gamma_e B > \Delta$, shown in Fig. 4. Here, for the sake of simplicity, we considered initial thermal states, with $P_i = e^{-\beta E_i}/\sum_k e^{-\beta E_k}$, $k = 1, \ldots, n$. In the first regime, the energy dissipation leads the system in the state corresponding to the energy minimum $E_{|0\rangle} = 0$. This means that the asymptotic energy level is related to a thermal state at zero temperature, $\beta_\infty = \infty$. As a consequence, there is only a trivial solution solving the fluctuation theorem, that is, $\eta^* = 0$.

In the second case ($\gamma_e B > \Delta$), the eigenstate associated to the minimum energy of the system is $|-1\rangle$, and we observe a *nonlinear* relation of η^* as a function of β, meaning that the quantity $\eta^* - \beta$ in general cannot be associated with an inverse temperature. Notably, instead, for a magnetic field $B \to \infty$ the value of η^* becomes constant and is equal to 2β. In this way, by knowing the temperature of the initial thermal state, i.e., the partition function at time $t = 0$, we can evaluate η^* and reconstruct the whole dynamics of the system. This is equivalent to the case with symmetric energy levels, whose analytic solution is studied in Appendix C. Finally, the special condition $\gamma_e B \to \Delta^+$ leads to a degeneracy of $|0\rangle$ and $|-1\rangle$ eigenstates. In this case, η^* shows a discontinuity at $-\infty$ that occurs in correspondence of the energy crossing between $E_{|-1\rangle}$ and $E_{|0\rangle}$ (see Appendix C for more details).

VI. CONCLUSIONS

In this paper, we discussed how to characterize energy exchange statistics in open quantum dynamics (both unital and nonunital) exhibiting dissipation that brings the system towards an asymptotic state not necessarily thermal. In doing this, we derive a quantum fluctuation theorem for a family of

parametrized dynamics that go beyond thermalizing dynamics, hence extending the results by Jarzynski and Wójcik [9]. First of all, we show that in the asymptotic limit ($t \to \infty$) the characteristic function $\mathcal{G}(j\eta)$ of the exchanged energy distribution is independent of the process details and is equal to 1 for a unique, constant (time-independent), nontrivial energy scale factor $\eta = \eta^*$. In addition, we show a sufficient condition under which $\mathcal{G}(j\eta^*, t) = 1$ is valid for any time $t \geqslant t^*$, thus providing us a true fluctuation theorem even for the quantum dissipative case.

The condition that allows for $\mathcal{G}(j\eta^*, t) = 1$ for $t \geqslant t^*$ is $P_{f|i}(t) = \overline{F}(t)P_f(\infty)$, which is implied by the validity of hypothesis 2 and the detailed balance condition (DBC). As shown in Sec. III, hypothesis 2 is related to the almost complete memory loss of the initial energy statistics, and can be obtained (and thus tested) in dissipative instances of Stern-Gerlach protocols. According to hypothesis 2, the conditional probabilities of the exchanged energy distribution have to be uniquely determined by the asymptotic probabilities $P_f(\infty)$ that enter the nonequilibrium state $\lim_{t\to\infty} \Phi_t[\rho_0]$ reached asymptotically by a quantum system under dissipation. Also the DBC is a necessary requirement for $P_{f|i}(t) = \overline{F}(t)P_f(\infty)$. In fact, as shown in Appendix B, given $P_{f|i}(t) = \widehat{F}_f(t)P_f(\infty)$ (implied by hypothesis 2), then the DBC entails $\widehat{F}_f(t) = \overline{F}(t)$.

Experimentally, we took advantage of a platform based on single NV centers to test the robustness of Theorem 1 against the fulfillment of its assumptions. While numerical simulations show a discrepancy between the cases with or without the DBC, the experimental outcomes cannot distinguish them within our statistical uncertainty. This means that, within the error bars of our experimental measurements, the thesis of the fluctuation theorem results are verified approximately, albeit in a strict mathematical sense the theorem may not hold. Moreover, by exploring the behavior of η^* depending on the value of $\gamma_e B$, we observe a nonlinear dependence of η^* from the initial state, and the presence of a discontinuity in correspondence of an energy level crossing.

As an outlook, studies on the energy statistics and tests of the fluctuation theorem at the NV ground state level anticrossing ($\gamma_e B = \Delta$) would highlight the relationship between the thermodynamic properties of the system and its physical counterparts, such as the NV photoluminescence [47]. It is also worth investigating a tighter condition (still sufficient in case) under which the fluctuation theorem [Eq. (2)] is valid, as well as determining a formal relation with the Hatano-Sasa fluctuation theorem [48]. Moreover, one could test the idea that η^* can be seen as an effective inverse temperature by weakly connecting two systems and using one of the two to estimate the value of η^* associated to the other, in analogy with standard thermometry routines of a macroscopic bath. In such a case, the probing system should be assumed to be fully known and maintained at a steady-state solution of its dynamics. Furthermore, we think it could be interesting to explore if and how the quantity $\eta^*\langle\Delta E\rangle$, and its integral along a path on the parameters' space, can be put in relation with the average entropy production that one would determine at the steady state for a reversible thermodynamic transformation.

Our work opens the possibility to evaluate the energetics of stabilizing a qubit by means of a dissipative protocol [49] even with other experimental platforms, or to associate a unique energy scale factor to quantum many-body phases embodied in the local energy statistics of an array of qubits [50,51]. An important application of that would be the dissipative charging of a (many-body) quantum battery [52,53]. Finally, in the case the initial state and the Hamiltonian are not commuting, other protocols than the TPM scheme can be used to characterize energy fluctuations in the quantum regime [20,21,27,37,54–58]. By applying these methodologies, our results might be no longer valid and some "quantum corrections" may be needed as in Ref. [59]. One would thus extend some results in Ref. [60] that concern the interplay of quantum and classical information processes in dissipative dynamics.

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APPENDIX A: PROOF OF LEMMA 1

The characteristic function $\mathcal{G}(j\eta, t)$ of the distribution for the internal energy variation is convex with respect to the scaling factor η [38,39]. Moreover, being given by the finite sum of analytical functions (exponentials), for finite-dimensional systems $\mathcal{G}(j\eta, t)$ is also a twice differentiable real-valued function with positive concavity. As the first step of the proof, our aim is to demonstrate that, at the asymptotic limit $t \to \infty$, the equality $\mathcal{G}(j\eta, \infty) = 1$ holds only for $\eta_0 = 0$ (trivial, constant solution) and for another energy scale factor η^* that is a real number. For the sake of simplicity, from here on in this appendix, we will denote $\mathcal{G}(j\eta, \infty)$ with $\mathcal{G}(j\eta)$. In the second step of the proof, we will demonstrate that η^* depends only on the initial and the asymptotic states.

Let us start by defining $h(\eta, \alpha) \equiv \mathcal{G}(j\eta) - \alpha$, with α being a real and positive number not necessarily equal to 1. We want the roots of $h(\eta, \alpha)$ to be also the solutions of $\mathcal{G}(j\eta) = 1$. This is only achieved for $\alpha = 1$, because $\eta_0 = 0$ represents the trivial solution [see Eq. (2)]. Therefore, we want to find the roots of $g(\eta) \equiv h(\eta, 1) = \mathcal{G}(j\eta) - 1$. Given that $g(\eta)$ is a convex function with positive concavity as well as $\mathcal{G}(j\eta)$, then it can only have a maximum of two roots in the parameter η. But we already know that $\eta_0 = 0$ is a trivial root of $g(\eta)$; hence $g(\eta)$ is only allowed to have at most another root different from zero. This means that there always exist a real number η^* such that $g(\eta^*) = 0$. In general, $\eta^* \neq 0$, except for the specific

case where $\eta_0 = 0$ is the only root of $g(\eta)$. This concludes the first part of the proof, but before moving on to the second part of the proof, let us study the singular case in which $\eta_0 = 0$ is the only root of $g(\eta)$.

The number of roots is determined by the value of the unique minimum of $g(\eta)$. Given that $\eta_0 = 0$ is always a root of $g(\eta)$, then there are only two possible scenarios under which $g(\eta)$ has not a second root: either $\min_\eta g(\eta) = 0$ or $\min_\eta g(\eta) < 0$ but $\eta^* \to \pm\infty$. If we assume that $\min_\eta g(\eta) = 0$, then $\min_\eta g(\eta) = g(\eta_0) = 0$. This happens if and only if the derivative $\partial g(\eta)/\partial\eta = \partial\mathcal{G}(j\eta)/\partial\eta$ evaluated in $\eta_0 = 0$ is equal to zero for any value of the system parameters. Notice also that Eqs. (2) and (4) imply that

$$\left.\frac{\partial g(\eta)}{\partial \eta}\right|_{\eta=0} = -\langle E_\infty\rangle + \langle E_{\rm in}\rangle, \tag{A1}$$

where $\langle E_{\rm in}\rangle \equiv \sum_i P_i E_i$ and $\langle E_\infty\rangle = \lim_{t\to\infty}\sum_f P_f(t)E_f = \sum_f P_f(\infty)E_f$ with $P_f(t) = \sum_i P_{f|i}(t)P_i$. Hence, the case $\min_\eta g(\eta) = 0$ is true if and only if $\langle E_\infty\rangle = \langle E_{\rm in}\rangle$, namely, if the initial and asymptotic values of energy are the same. This can occur, e.g., when the asymptotic state reached by the system is stationary thermal with the same temperature β_∞ as the initial thermal state, i.e., $\beta_\infty = \beta$. On the other hand, the case $\min_\eta g(\eta) < 0$ with $\eta^* \to \pm\infty$ is only valid when $g(\eta)$ is equal to a sum of exponential functions all with the same sign (either negative or positive) in the exponent, as opposed to the usual sum of exponential functions with opposite signs [see Eq. (4)]. This is a quite particular case, but it is not impossible. As an example, we consider the dissipative quantum Maxwell demon addressed in the main text. For the case where $\mathcal{H} \propto S_z$, the characteristic function $\mathcal{G}$ reduces to Eq. (C7). Thus, by initializing the quantum system in a state where either $P_{|+1\rangle}$ or $P_{|-1\rangle}$ is equal to zero, the left-hand side of Eq. (C7) is a simple exponential function. As a consequence, $\eta_0 = 0$ turns out to be the only solution for $\mathcal{G} = 1$.

Let us now focus on the second step of the proof. We want to show that if the probabilities to measure the energies (eigenvalues of the Hamiltonian) of the system do not depend on time, nor on the initial state ρ_0, then the unique finite zero of $g(\eta) = \mathcal{G}(j\eta) - 1$ only depends on the initial and asymptotic states. Such assumption is attained when the state of the quantum system, the solution of the dynamical equation of motion, has constant energy. This is the case, for example, for the asymptotic state considered in the main text [see Eq. (3)]. This condition can be easily translated in a property of the characteristic function $\mathcal{G}(j\eta)$. For this purpose, note that, in correspondence with the asymptotic state, the conditional probabilities $P_{f|i}(\infty)$ are invariant with respect to the index i, resulting in

$$P_{f|1}(\infty) = P_{f|2}(\infty) = \cdots = P_{f|n}(\infty) \equiv P_f(\infty) \tag{A2}$$

for any f, with n denoting the dimension of the system. Thus, by assuming the validity of Eq. (A2), $\mathcal{G}(j\eta)$ admits the decomposition

$$\begin{aligned}\mathcal{G}(j\eta) &= \sum_i P_i e^{\eta E_i}\sum_f P_f(\infty)e^{-\eta E_f}\\ &= {\rm Tr}[e^{\eta\mathcal{H}}\rho_0]{\rm Tr}[e^{-\eta\mathcal{H}}\rho_\infty],\end{aligned} \tag{A3}$$

where ρ_∞ is the asymptotic state of the quantum system. From the decomposition of Eq. (A3), it can be observed that η^* depends only on the set of probabilities $\{P_i\}$ and $\{P_f(\infty)\}$, corresponding respectively to the initial and asymptotic states, and on the initial and final energies of the system.

APPENDIX B: PROOF OF THEOREM 1

Let us set the context for the proof. We consider a dissipative quantum system, whose time evolution is governed by a generic open quantum map Φ_t that admits at least one (nonthermal) map fixed point. Then, the Hamiltonian $\mathcal{H} = \sum_k E_k\Pi_k$ of the system is such that the eigenvalues E_k are time independent. Thus, for $t \to \infty$ the probabilities to measure the energies (eigenvalues of $\mathcal{H}$) of the system do not depend on time (they are indeed constant values) or on the initial state ρ_0. We denote such probabilities as $P_f(\infty)$. Moreover, the fluctuations of energy variations are evaluated by means of the two-point measurement (TPM) scheme. Finally, for our convenience we express the conditional probabilities of the TPM distribution for the internal energy variation as a function of the asymptotic probabilities $P_f(\infty)$. Specifically, for $i \neq f$ (i and f denote, respectively, the indices over the initial and final energies, where the latter are measured at time t where the TPM scheme is stopped), the conditional probabilities $P_{f|i}(t)$ can be written as

$$P_{f|i}(t) = F_{i,f}(t)P_f(\infty), \tag{B1}$$

where $F_{i,f}(t)$ is a generic bounded real function depending on the indices i, f such that $F_{i,f}(t=0) = 0$ and $F_{i,f}(t\to\infty) = 1\ \forall\, i, f$. We note that, since $P_{i|i}(t) = 1 - \sum_{f\neq i}P_{f|i}(t) = 1 - \sum_{f\neq i}F_{i,f}(t)P_f(\infty)$ (valid for any i), we can generally write

$$\begin{aligned}P_{f|i}(t) &= F_{i,f}(t)P_f(\infty)(1-\delta_{i,f})\\ &\quad + \left(1 - \sum_{f\neq i}F_{i,f}(t)P_f(\infty)\right)\delta_{i,f},\end{aligned} \tag{B2}$$

where $\delta_{i,f}$ denotes the Kronecker delta. Equation (B2) holds for any pair (i, f) of indices and time t.

We are now in the position to carry out the proof of Theorem 1. For this purpose, we take the characteristic function of the statistics for the energy variations (obtained from applying the TPM scheme):

$$\mathcal{G}(j\eta, t) = \sum_{i,f} e^{-\eta(E_f - E_i)}P_i\,P_{f|i}(t), \tag{B3}$$

where $P_i \equiv {\rm Tr}[\rho_0\Pi_i]$, $P_{f|i}(t) \equiv {\rm Tr}[\Pi_f\Phi_t[\Pi_i]]$. By using the identity $\sum_f P_{f|i}(t) = 1$ for any t such that $P_{i|i}(t) = 1 - \sum_{f\neq i}P_{f|i}(t)$, we obtain

$$\begin{aligned}\mathcal{G}(j\eta, t) &= \sum_i\left[P_i\left(1 - \sum_{f\neq i}P_{f|i}(t)\right)\right.\\ &\quad\left. + \sum_{f\neq i}P_i\,P_{f|i}(t)\,e^{-\eta(E_f-E_i)}\right]\\ &= 1 + \sum_i\sum_{f\neq i}P_i\,P_{f|i}(t)(e^{-\eta(E_f-E_i)} - 1).\end{aligned} \tag{B4}$$

Hence, by substituting Eq. (B1) in Eq. (B4), we find that

$$\mathcal{G}(j\eta,t) = 1 + \sum_i \sum_{f\neq i} F_{i,f}(t)\, P_i\, P_f(\infty)(e^{-\eta(E_f-E_i)} - 1). \tag{B5}$$

Then, let us apply the assumptions of Theorem 1, namely, the validity of both hypothesis 2 and the detailed balance equation. The first hypothesis means that the functions $F_{i,f}(t)$ do not depend on the index i for $f \neq i$ and $t \geqslant t^*$, i.e.,

$$P_{f|i}(t) = \widehat{F}_f(t) P_f(\infty), \tag{B6}$$

which, together with the detailed balance condition

$$\frac{P_{f|i}(t)}{P_{i|f}(t)} = \frac{P_f(\infty)}{P_i(\infty)}, \tag{B7}$$

implies that

$$P_{f|i}(t) = \overline{F}(t) P_f(\infty) \tag{B8}$$

$\forall\, i, f$ with $f \neq i$ and $t \geqslant t^*$, where $\overline{F}(t)$ is a time-dependent real function. As a result,

$$\mathcal{G}(j\eta,t) = 1 + \overline{F}(t) \sum_i \sum_{f\neq i} P_i\, P_f(\infty)(e^{-\eta(E_f-E_i)} - 1). \tag{B9}$$

Concerning assumption (B8), notice that detailed balance equation (B7) is obeyed by any reversible Markov process, where reversibility here has to be meant with respect to the space of events that define the Markov process of energy outcomes. Instead, hypothesis 2 [i.e., assumption (B6)] stems from the almost complete loss of memory of the initial state, as discussed in the main text in Sec. III.

The last step of the proof is to recall the definition of η^* from Lemma 1's thesis. From Lemma 1, indeed, we already know that η^* is defined as the energy scale factor at $t \to \infty$ such that

$$\lim_{t\to\infty} \mathcal{G}(j\eta^*,t) = 1 \iff$$
$$\sum_i \sum_{f\neq i} P_i\, P_f(\infty)(e^{-\eta^*(E_f-E_i)} - 1) = 0. \tag{B10}$$

As a direct consequence, by substituting Eq. (B10) in Eq. (B9), we determine $\mathcal{G}(j\eta^*,t) = 1$ for $t \geqslant t^*$. This concludes the proof of the thesis of Theorem 1 in the main text.

APPENDIX C: DISSIPATIVE MAXWELL DEMON

Let us consider a generic finite-dimensional quantum system. By diagonalizing the system Hamiltonian at time $t = 0$ as $\mathcal{H} = \sum_k E_k |E_k\rangle\langle E_k|$, the initial state can be written as $\rho_0 = \sum_{k,\ell} \rho_{k\ell} |E_k\rangle\langle E_\ell|$, where $\rho_{k\ell} \equiv \langle E_k|\rho_0|E_\ell\rangle$.

Then, we take again the characteristic function of the TPM probability distribution $\mathrm{P}(\Delta E, t)$, i.e., $\mathcal{G}(j\eta,t) = \langle \exp(-\eta\Delta E)\rangle_t = \sum_{i,f} P_i P_{f|i}(t) e^{-\eta(E_f-E_i)}$, which depends on the energy scale factor η. In the following, we provide the theoretical analysis carried out to test the validity of Theorem 1 in some meaningful cases of the dissipative quantum Maxwell demon shown in the main text. First, we consider a case study with symmetric energy levels of a qutrit, which is applicable to the NV center case for $\mathcal{H} \propto S_z$ and a value of the magnetic field inducing a Zeeman shift $\gamma_e B$ much larger than the so-called ground state level anticrossing (GSLAC). Notably, this can describe also other exemplary cases, e.g., the ground state of a ^{87}Rb atom with $F = 1$. Second, we explicitly study the case of a NV center relaxing the assumption of $\gamma_e B \ll \Delta$, while maintaining $\mathcal{H} \propto S_z$.

We consider that the qutrit has reached the asymptotic regime such that $P_{f|i}(t) = P_f(\infty)$. Hence, the expression of $\mathcal{G}(j\eta^*,t) = 1$ at $t \to \infty$ for a generic qutrit is

$$\begin{aligned} &P_1P_3(\infty)e^{-\eta^*(E_3-E_1)} + P_1P_2(\infty)e^{-\eta^*(E_2-E_1)} \\ &\quad + P_1P_1(\infty) + P_2P_3(\infty)e^{-\eta^*(E_3-E_2)} \\ &\quad + P_2P_2(\infty) + P_2P_1(\infty)e^{-\eta^*(E_1-E_2)} + P_3P_3(\infty) \\ &\quad + P_3P_2(\infty)e^{-\eta^*(E_2-E_3)} + P_3P_1(\infty)e^{-\eta^*(E_1-E_3)} = 1, \end{aligned} \tag{C1}$$

where P_i and $P_f(\infty)$ are the initial and final asymptotic probabilities, respectively. Thus, if we assume that $E_1 = 0$, then

$$\begin{aligned} &P_1P_3(\infty)e^{-\eta^*E_3} + P_1P_2(\infty)e^{-\eta^*E_2} + P_1P_1(\infty) \\ &\quad + P_2P_3(\infty)e^{-\eta^*(E_3-E_2)} + P_2P_2(\infty) + P_2P_1(\infty)e^{\eta^*E_2} \\ &\quad + P_3P_3(\infty) + P_3P_2(\infty)e^{-\eta^*(E_2-E_3)} + P_3P_1(\infty)e^{\eta^*E_3} = 1. \end{aligned} \tag{C2}$$

1. Qutrit with symmetric energies

Assuming that the energy values of the qutrit are symmetric around zero, we define $E_1 = 0$, $E_2 = -\overline{E}$, and $E_3 = \overline{E}$ with $\overline{E} = \hbar\omega/2$. In this way, by means of the substitution

$$x \equiv e^{-\eta^*\overline{E}} \iff \eta^* = -\frac{1}{\overline{E}} \ln x, \tag{C3}$$

the equation $\sum_{i,f} P_i P_f(\infty) e^{-\eta^*(E_f-E_i)} = 1$ can be rewritten as a polynomial equation in x. For a qutrit [see Eq. (C1)] the polynomial equation is

$$\begin{aligned} &(x-1)[P_2P_3(\infty)\,x^3 + (P_1P_3(\infty) + P_2P_1(\infty) + P_2P_3(\infty))x^2 \\ &\quad - (P_1P_2(\infty) + P_3P_1(\infty) + P_3P_2(\infty))x - P_3P_2(\infty)] = 0. \end{aligned}$$

Clearly, the whole equation contains the trivial solution $x = 1$, i.e., $\eta^* = 0$, while solving the third-order algebraic equation

$$\begin{aligned} &P_2P_3(\infty)\,x^3 + (P_1P_3(\infty) + P_2P_1(\infty) + P_2P_3(\infty))x^2 \\ &\quad - (P_1P_2(\infty) + P_3P_1(\infty) + P_3P_2(\infty))x = P_3P_2(\infty) \end{aligned} \tag{C4}$$

provides us the other value of $\eta^* \neq 0$ that obeys the fluctuation relation $\lim_{t\to\infty} \mathcal{G}(j\eta^*,t) = 1$. In this regard, by applying the Routh-Hurwitz criterion to polynomial (C4), we can also prove that only one root of Eq. (C4) has a positive real part different from 1. In fact, according to the Routh-Hurwitz criterion, we recall that each variation (permanence) of the sign of the coefficients of the first column of the Routh table corresponds to a root of the polynomial with a positive (negative) real part. In our case, there are always two sign permanences and only one variation, for any possible value of the probabilities P_i and $P_f(\infty)$. Being $\eta^* \propto -\ln x$, only the unique solution $x \neq 1$ with positive real part is physical, thus returning the unique nontrivial energy scale factor η^* such that the fluctuation relation is valid.

2. NV center with $\mathcal{H} \propto S_z$ subjected to dissipation

As in the main text, for the case $\mathcal{H} \propto S_z$, we consider $\{|0\rangle, |\pm 1\rangle\}$ and $\{E_0, E_{\pm 1}\}$ as the three eigenstates and eigenvalues of the qutrit. Thus, assuming that $P_{\pm 1}(\infty) = 0$ and $P_0(\infty) = 1$, one gets

$$P_0 + P_1 e^{\eta^* E_1} + P_{-1} e^{\eta^* E_{-1}} = 1 \tag{C5}$$

$$\Rightarrow\ 1 - P_1 - P_{-1} + P_1 e^{\eta^* E_1} + P_{-1} e^{\eta^* E_{-1}} = 1 \tag{C6}$$

$$\Rightarrow\ P_1(e^{\eta^* E_1} - 1) + P_{-1}(e^{\eta^* E_{-1}} - 1) = 0. \tag{C7}$$

Low field, $\gamma_e B < \Delta$

$E_1 > 0$ and $E_{-1} > 0$ imply $\eta^* = 0$ as the only possible solution.

As a proof, we can show that, by considering $\eta^* \neq 0$, Eq. (C7) is not satisfied:

$$\text{If } \eta^* < 0 \Rightarrow e^{\eta^* E_1} - 1 < 0 \quad \text{and} \quad e^{\eta^* E_{-1}} - 1 < 0$$
$$\Rightarrow\ P_1(e^{\eta^* E_1} - 1) + P_{-1}(e^{\eta^* E_{-1}} - 1) < 0. \tag{C8}$$

$$\text{If } \eta^* > 0 \Rightarrow e^{\eta^* E_1} - 1 > 0 \quad \text{and} \quad e^{\eta^* E_{-1}} - 1 > 0$$
$$\Rightarrow\ P_1(e^{\eta^* E_1} - 1) + P_{-1}(e^{\eta^* E_{-1}} - 1) > 0. \tag{C9}$$

High field, $\gamma_e B > \Delta$

Recalling that $E_{\pm 1} = \Delta \pm \gamma_e B$, in the limit of $\gamma_e B \gg \Delta$, Eq. (C7) reduces to

$$P_1 e^{\eta^* \gamma_e B}(1 - e^{-\eta^* \gamma_e B}) + P_{-1}(e^{-\eta^* \gamma_e B} - 1) = 0 \tag{C10}$$

$$\Rightarrow\ (P_1 e^{\eta^* \gamma_e B} - P_{-1})(1 - e^{-\eta^* \gamma_e B}) = 0. \tag{C11}$$

The second part of the left-hand side leads to the trivial solution $\eta^* = 0$. Instead, by putting together the conditions $(P_1 e^{\eta^* \gamma_e B} - P_{-1}) = 0$ [from Eq. (C11)] and $P_i = e^{-\beta E_i}/Z$, with the Z partition function valid for any initial thermal state, we obtain

$$\frac{P_{-1}}{P_1} = e^{\eta^* \gamma_e B} \tag{C12}$$

$$\Rightarrow \eta^* = \frac{1}{\gamma_e B} \ln\left[e^{-\beta(E_{-1} - E_{+1})}\right] \tag{C13}$$

$$\Rightarrow \eta^* = \frac{2\gamma_e B \beta}{\gamma_e B} = 2\beta. \tag{C14}$$

This result is confirmed by numerical simulations to evaluate η^*, shown in Fig. 4 of the main text. Simulations are obtained by setting the initial inverse temperature β, and solving the condition $\mathcal{G} = 0$ for the characteristic function in the high-field limit. In addition, for each selected initial temperature, we have iteratively derived η^* while changing B for $\gamma_e B \to \Delta_+$ by using each solution as an initial guess for the subsequent simulation run.

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