

Thermodynamic Framework for Coherently Driven Systems

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The laws of thermodynamics are a cornerstone of physics. At the nanoscale, where fluctuations and quantum effects matter, there is no unique thermodynamic framework because thermodynamic quantities such as heat and work depend on the accessibility of the degrees of freedom. We derive a thermodynamic framework for coherently driven systems, where the output light is assumed to be accessible. The resulting second law of thermodynamics is strictly tighter than the conventional one, and it demands the output light to be more noisy than the input light. We illustrate our framework across several well-established models, and we show how the three-level maser can be understood as an engine that reduces the noise of a coherent drive. Our framework opens a new avenue for investigating the noise properties of driven-dissipative quantum systems.

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Introduction—In the last decades the laws of thermodynamics, traditionally used to describe macroscopic systems, have been extended to small, stochastic systems and even to the quantum regime with great success [1–8]. A major obstacle in this endeavor turned out to be the definition of one of the most central quantities: work [9–12]. At the macroscale, work is carried by the accessible, macroscopic degrees of freedom, while heat is carried by the microscopic, inaccessible degrees of freedom. At the nanoscale, all degrees of freedom are microscopic, resulting in ambiguity when defining work. To remedy this situation, definitions for work that depend on the considered task have been suggested [13–15].

The most prominent definition for work (henceforth called the *conventional* definition) relies on a semiclassical description in terms of a time-dependent Hamiltonian. Work is then described by the energy changes due to its time-dependence, which are induced by controllable degrees of freedom that are described classically [4,7,8]. While often adequate, this approach has conceptual shortcomings for coherently driven systems. To illustrate this, we consider an optical cavity that is driven by coherent light as sketched in Fig. 1. In this scenario, the drive can be described by a time-dependent Hamiltonian, and the conventional definition can be applied. However, all the energy

that leaks out of the cavity is then treated as heat, even if it contains coherence. This may be appropriate if the light leaving the cavity is dissipated, but as pointed out in Ref. [15], this light may be further used. This becomes particularly obvious in the case of an empty cavity, where the light is simply reflected with a shifted phase. The output light leaving the cavity is then just as useful as the input light that induces the time dependence of the Hamiltonian.

In Ref. [15], a resolution of this issue was suggested by considering the coherent part (i.e., the displacement) of the output light as work, not heat. However, it remains unclear if this intuitive definition for work can be embedded in a thermodynamically consistent framework

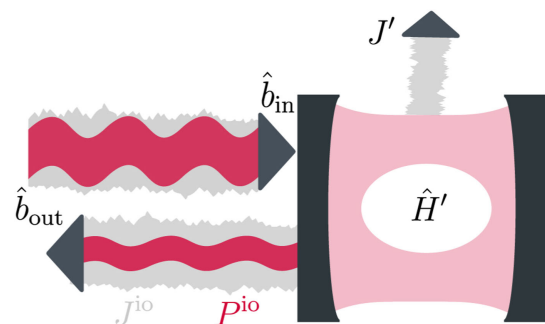


FIG. 1. Setup. A cavity with an embedded quantum system, governed by the Hamiltonian $\hat{H}'$, is driven by the input field, which contains a coherent part, given by the average of the input field $\langle \hat{b}_{\text{in}} \rangle$, as well as thermal fluctuations. Compared to the input, the output field, $\hat{b}_{\text{out}}$, typically has a smaller coherent part and larger fluctuations. Work (P^{io}) and heat (J^{io}) are given by the changes in the coherent part and the fluctuations, respectively. The heat dissipated by the intracavity system is given by J' .

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where a non-negative entropy production is associated to the corresponding definition of heat.

In this Letter, we solve this critical problem by formulating the first and second law of thermodynamics consistent with these new definitions. In this framework, the second law demands that the noise in the output light is larger than the noise in the input light, as long as there is no other source of entropy production. We further show that our new second law is always tighter than the conventional second law. This is consistent with treating the output light as accessible: having more access generally reduces irreversibility. We illustrate our thermodynamic framework with the help of a Kerr oscillator, a two-level system embedded in a cavity, and a three-level maser [13,16]. The last acts as a heat engine, which in our framework corresponds to a machine that can reduce the noise in the light field.

Setup and model—We consider a quantum system placed inside a cavity that is subjected to a coherent drive and couples to a bosonic bath via one channel (see Fig. 1). The Hamiltonian of our setup reads

$$\hat{H}(t) = \Omega \hat{a}^\dagger \hat{a} + \hat{H}' + \hat{H}_d(t), \quad (1)$$

where Ω is the cavity frequency and $\hat{H}'$ describes the system inside the cavity as well as its coupling to the cavity mode. Throughout, primed quantities act on the intracavity system. The drive term is given by

$$\hat{H}_d(t) = i\sqrt{\kappa} \left(\langle \hat{b}_{\text{in}}^\dagger(t) \rangle \hat{a} - \langle \hat{b}_{\text{in}}(t) \rangle \hat{a}^\dagger \right), \quad (2)$$

with κ denoting the linewidth of the cavity. We further introduced the input field, which describes the coherent drive as well as the thermal noise entering the cavity [17],

$$\langle \hat{b}_{\text{in}}(t) \rangle = f(t)e^{-i\omega_d t}, \quad \langle \langle \hat{b}_{\text{in}}^\dagger(t) \hat{b}_{\text{in}}(t') \rangle \rangle = n_c \delta(t - t'), \quad (3)$$

where n_c denotes the thermal occupation and $\langle \langle \hat{A} \hat{B} \rangle \rangle = \langle \hat{A} \hat{B} \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle$. We consider a drive at frequency ω_d with a potential slow modulation, i.e., $|\partial_t f(t)|/|f(t)| \ll \omega_d$. We note that a coherent drive assumes a source with a stable phase. Any phase can then be defined with respect to the source, which provides a phase reference.

We model the dynamics of our setup with the quantum master equation,

$$\partial_t \hat{\rho}(t) = -i[\hat{H}(t), \hat{\rho}(t)] + \mathcal{L}_c \hat{\rho}(t) + \mathcal{L}' \hat{\rho}(t), \quad (4)$$

where $\mathcal{L}'$ is an arbitrary Lindbladian dissipator acting on the intracavity system and the dissipator of the cavity mode reads

$$\mathcal{L}_c \hat{\rho} = \kappa n_c \mathcal{D}[\hat{a}^\dagger] \hat{\rho} + \kappa(n_c + 1) \mathcal{D}[\hat{a}] \hat{\rho}, \quad (5)$$

with $\mathcal{D}[\hat{a}] \hat{\rho} = \hat{a} \hat{\rho} \hat{a}^\dagger - \frac{1}{2} \{ \hat{a}^\dagger \hat{a}, \hat{\rho} \}$. We assume that photons can only be exchanged with the input channel and, thus, demand that $\text{Tr}\{\hat{a}^\dagger \hat{a} \mathcal{L}' \hat{\rho}\} = 0$. Additional dissipation channels for the photons can be included (see Supplemental Material [18]). To describe the light that is reflected by the cavity, we introduce the output operator [17],

$$\hat{b}_{\text{out}} = \hat{b}_{\text{in}} + \sqrt{\kappa} \hat{a}. \quad (6)$$

Thermodynamics—The first law of thermodynamics states that any change in the internal energy U of a system can be divided into heat and work,

$$\partial_t U = P + J, \quad (7)$$

where P denotes power (work per unit time) and J the heat current. The second law constrains the physically allowed processes by demanding a non-negative entropy production defined as

$$\dot{\Sigma} \equiv k_B \partial_t S_{\text{vN}}(\hat{\rho}) - \frac{J_c}{T_c} - \frac{J'}{T'} \geq 0, \quad (8)$$

with the von Neumann entropy $S_{\text{vN}}(\hat{\rho}) = -\text{Tr}\{\hat{\rho} \ln \hat{\rho}\}$. In Eq. (8), the heat current is split into two parts, $J = J_c + J'$, corresponding to the dissipation of the cavity and the intracavity system, respectively. Here we assume one temperature for the environment of the cavity and one for the environment of the intracavity system. Additional temperatures are discussed in Supplemental Material [18].

Before introducing our new thermodynamic framework, we recapitulate how thermodynamic quantities can be consistently defined in the conventional framework, where energy changes due to the time dependence of the Hamiltonian are interpreted as work, i.e., $P = \langle \partial_t \hat{H}(t) \rangle$. Typically, the internal energy is defined as the expectation value of $\hat{H}(t)$, and heat is defined through the first law in Eq. (7) [4,19,20]. This approach may result in violations of the second law for master equations, which do not rely on the secular approximation [21–26], as is the case in our Eq. (4). Here we follow the approach in Ref. [26], which ensures thermodynamic consistency. To this end, we introduce the thermodynamic Hamiltonian,

$$\hat{H}_{\text{TD}} = \omega_d \hat{a}^\dagger \hat{a} + \hat{H}'_{\text{TD}}, \quad (9)$$

which is used to define the internal energy as $U = \langle \hat{H}_{\text{TD}} \rangle$. Note that $\hat{H}_{\text{TD}}$ may differ from the Hamiltonian $\hat{H}(t)$ that governs the dynamics [18]. This choice reflects the fact that the drive populates the cavity with photons at ω_d . Thermodynamic consistency is ensured by choosing $n_c = [e^{\omega_d/(k_B T_c)} - 1]^{-1}$ in Eq. (5). We note that our master equation is only valid when the frequency dependence of the Bose-Einstein distribution within the cavity

linewidth can be neglected. The term $\hat{H}'_{\text{TD}}$ acts on the intracavity system, quantifying its internal energy. A more detailed discussion on the thermodynamic Hamiltonian can be found in Refs. [18,26].

With the thermodynamic Hamiltonian, we find the first law of thermodynamics with

$$P = -i\langle[\hat{H}_{\text{TD}}, \hat{H}(t)]\rangle, \quad J = \text{Tr}\{\hat{H}_{\text{TD}}\mathcal{L}\hat{\rho}(t)\}, \quad (10)$$

where $\mathcal{L} = \mathcal{L}_c + \mathcal{L}'$. We recover the standard form of power when the thermodynamic Hamiltonian obeys $-i[\hat{H}_{\text{TD}}, \hat{H}(t)] \simeq \partial_t \hat{H}(t)$. In our case this holds as long as $-i[\hat{H}_{\text{TD}}, \hat{H}'] = \partial_t \hat{H}'$ [18]. For simplicity, we consider a time-independent $\hat{H}'$ from here on, such that the power is associated only to $\hat{H}_d$.

As above, the heat current may be decomposed in two terms $J = J_c + J'$, with [18]

$$J_c = \text{Tr}\{\omega_d \hat{a}^\dagger \hat{a} \mathcal{L}_c \hat{\rho}(t)\}, \quad J' = \text{Tr}\{\hat{H}'_{\text{TD}} \mathcal{L}' \hat{\rho}(t)\}. \quad (11)$$

For the heat current associated to the cavity and the power, we can evaluate these expressions to find

$$J_c = \omega_d \kappa (n_c - \langle \hat{a}^\dagger \hat{a} \rangle), \quad (12)$$

$$P = -\sqrt{\kappa} \omega_d (\langle \hat{b}_{\text{in}}^\dagger(t) \rangle \langle \hat{a} \rangle + \langle \hat{b}_{\text{in}}(t) \rangle \langle \hat{a}^\dagger \rangle). \quad (13)$$

Typically, the heat characterizes the amount of energy exchanged with the bath, while the power quantifies the energy exchanged with the coherent driving field. This interpretation is challenged in the present scenario, as both the coherent drive and the thermal noise are carried by the same environment. Indeed, one may show that the total energy exchanged with the cavity environment is equal to [18]

$$\omega_d (\langle \hat{b}_{\text{out}}^\dagger(t) \hat{b}_{\text{out}}(t) \rangle - \langle \hat{b}_{\text{in}}^\dagger(t) \hat{b}_{\text{in}}(t) \rangle) = P + J_c. \quad (14)$$

With the definition of heat above, it can be shown by using Spohn's inequality [27,28] that the second law in Eq. (8) is satisfied if $\mathcal{L}' e^{-\hat{H}'_{\text{TD}}/(k_B T')} = 0$ [18].

New thermodynamic framework—We put forward new definitions of heat and work by decomposing the energy flow in Eq. (14) into the coherent part of the fields and their variances,

$$P^{\text{io}} = -\omega_d (|\langle \hat{b}_{\text{out}}(t) \rangle|^2 - |\langle \hat{b}_{\text{in}}(t) \rangle|^2), \quad (15)$$

$$J_c^{\text{io}} = -\omega_d (\langle \hat{b}_{\text{out}}^\dagger(t) \hat{b}_{\text{out}}(t) \rangle - \langle \hat{b}_{\text{in}}^\dagger(t) \hat{b}_{\text{in}}(t) \rangle). \quad (16)$$

In these definitions, the power is determined by the coherent part of the input and output light. Positive power implies that

the input light carries more coherence than the output light. Similarly, a negative heat current implies that the output light is more noisy than the input light. Equation (15) has been suggested previously as a definition for work [15] since the coherent part of the output light can be further used to drive another quantum system. Equations (15) and (16) can, thus, be understood as the relevant definitions of heat and work if the output light is accessible, while Eqs. (12) and (13) are the relevant definitions if all the light leaving the cavity is dissipated. We note that the same definitions have been found in a qubit coupled to a waveguide using bipartite frameworks [29,30]. For an alternative thermodynamic treatment of coherently driven two-level systems, see Ref. [31].

By definition we find $P^{\text{io}} + J_c^{\text{io}} = P + J_c$, guaranteeing that the first law holds for the new definitions. Here we prove the second law and elevate Eqs. (15) and (16) to a consistent thermodynamic framework. To this end, we exploit the fact that the master equation in Eq. (4) can be written in a different form by letting $\mathcal{L}_c \rightarrow \mathcal{L}_s$ and $\hat{H} \rightarrow \hat{H}_s$, with

$$\mathcal{L}_s = \kappa n_c \mathcal{D}[\hat{a}^\dagger - \langle \hat{a}^\dagger \rangle] + \kappa (n_c + 1) \mathcal{D}[\hat{a} - \langle \hat{a} \rangle], \quad (17)$$

$$\hat{H}_s = \hat{H} + \frac{i}{2} \kappa (\langle \hat{a}^\dagger \rangle \hat{a} - \langle \hat{a} \rangle \hat{a}^\dagger). \quad (18)$$

Importantly, this transformation leaves the master equation invariant [32]. Heat and work may then be computed in full analogy to Eq. (10), and we find

$$P^{\text{io}} = -i\langle[\hat{H}_{\text{TD}}, \hat{H}_s(t)]\rangle \quad (19)$$

$$J_c^{\text{io}} = \text{Tr}\{\omega_d \hat{a}^\dagger \hat{a} \mathcal{L}_s \hat{\rho}(t)\} = \omega_d \kappa (n_c - \langle \hat{a}^\dagger \hat{a} \rangle). \quad (20)$$

If $\partial_t \langle \hat{a} \rangle \simeq -i\omega_d \langle \hat{a} \rangle$, we find that the power can again be obtained from the standard expression $P^{\text{io}} = \langle \partial_t \hat{H}_s(t) \rangle$. We note that the rewriting of the master equation only has the effect $P \rightarrow P^{\text{io}}$ and $J_c \rightarrow J_c^{\text{io}}$ and leaves J' invariant.

The second law again follows from Spohn's inequality [18], and we find

$$\dot{\Sigma}^{\text{io}} \equiv k_B \partial_t S_{\text{vN}}(\hat{\rho}) - \frac{J_c^{\text{io}}}{T_c} - \frac{J'}{T'} \geq 0. \quad (21)$$

Furthermore, we find that the second law is strictly tighter than in the conventional framework,

$$\dot{\Sigma} \geq \dot{\Sigma}^{\text{io}} \geq 0. \quad (22)$$

With this we have shown that the new definitions for heat and work are thermodynamically consistent.

We note that the nonunique splitting of master equations into a unitary and a dissipative part has been leveraged before to describe the thermodynamics of systems coupled strongly to their environment [33].

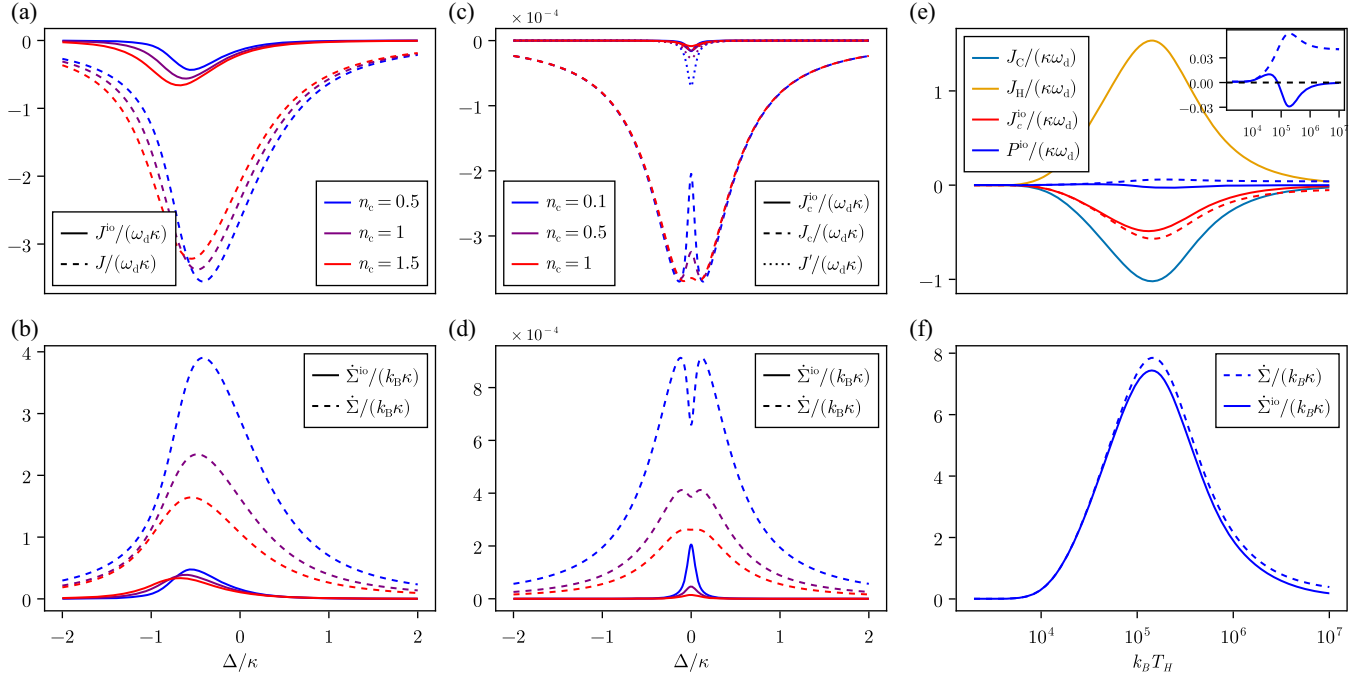


FIG. 2. Conventional and new definitions of heat and work. For (a)–(d) $\Omega/\kappa = 10^4$, $\gamma/\kappa = 0.05$, $g/\kappa = 0.1$, $f/\kappa = 0.01$. (a) Heat current and (b) entropy production for the Kerr oscillator for different occupation numbers n_c as a function of detuning $\Delta = \Omega - \omega_d$. Note that $P^{(io)} = -J^{(io)}$. Parameters: $\Omega/\kappa = 10^4$, $K/\kappa = 0.05$, $f/\kappa = 1$. (c) Heat currents and (d) entropy production for the two-level system for different occupation numbers as a function of detuning. (e), (f) Heat currents and power, and entropy production for the quantum three-level maser; $\Omega/\kappa = 10^4$, $g/\kappa = 0.7$, $f/\kappa = 0.1$, $\omega_2/\kappa = \omega_3/(3\kappa) = \Omega/\kappa$, $\gamma_H/\kappa = 0.5$, $\gamma_C/\kappa = 100$, $n_c = 4.54$, $n_C = 0.007$. (e) We observe that there is a region, $k_B T_H \approx 10^5$, in which the system is a heat engine, $P^{(io)} < 0$, while $P > 0$ (see also inset). The dashed black line in the inset indicates the zero. (f) Entropy production peaks at the heat engine regime.

Empty cavity—As a first example to illustrate our new thermodynamic framework, we consider the case where there is no additional system inside the cavity, i.e., $\hat{H}' = \hat{H}'_{TD} = 0$, and there is no additional loss channel, $\mathcal{L}' = 0$. In our new framework we find that, contrary to the standard definitions, the power and heat current vanish in steady state, $P^{(io)} = J^{(io)} = 0$. This reflects the fact that the input is simply reflected by the cavity and, up to a phase, the output light is identical to it. The output light is, thus, just as valuable as the input light.

Kerr oscillator—As a second, nonlinear example we consider a Kerr oscillator described by $\hat{H}' = K\hat{a}^\dagger\hat{a}^\dagger\hat{a}\hat{a}$, with Kerr parameter K . We do not consider additional losses $\mathcal{L}' = 0$, and we consider the thermodynamic Hamiltonian $\hat{H}_{TD} = \omega_d\hat{a}^\dagger\hat{a}$, which assigns each photon within the cavity the energy ω_d . We note that this requires $K \ll \omega_d$ such that a constant thermal occupation n_c is justified even though the cavity frequency effectively depends on the number of photons within the cavity. We compare the conventional definitions of heat and work with our new definitions (see Fig. 2). As the temperature is increased, the conventional definition predicts a decrease in entropy production because fewer photons can enter the cavity from the drive due to the photon-number dependence of the resonance frequency. In contrast, our new definition is much less sensitive to an increase in n_c since the entropy

production only arises from a reduced coherence due to nonlinear effects. This illustrates how the two definitions provide different insight into irreversibility.

Two-level system—As a next example, we consider a two-level system embedded in the cavity described by

$$\hat{H}' = \frac{\Omega}{2}\hat{\sigma}_z + g(\hat{a}^\dagger\hat{\sigma}_- + \hat{a}\hat{\sigma}_+), \quad (23)$$

$$\hat{H}_{TD} = \omega_d(\hat{a}^\dagger\hat{a} + \hat{\sigma}_+\hat{\sigma}_-), \quad (24)$$

$$\mathcal{L}' = \gamma n_q \mathcal{D}[\hat{\sigma}_+] + \gamma(n_q + 1)\mathcal{D}[\hat{\sigma}_-], \quad (25)$$

where g is the coupling between the cavity and the qubit, γ and $n_q = [e^{\omega_d/(k_B T_q)} - 1]^{-1}$ denote the coupling and the occupation of an additional thermal reservoir coupled to the qubit, and the system operators of the two-level system are given by $\hat{\sigma}_z = |e\rangle\langle e| - |g\rangle\langle g|$, $\hat{\sigma}_- = |g\rangle\langle e| = \hat{\sigma}_+^\dagger$. The thermodynamic Hamiltonian assigns each excitation the same energy ω_d .

We find that the additional heat current for the intracavity system can be expressed as follows:

$$J' = \omega_d \gamma \frac{n_q}{n_F} (n_F - \langle \hat{\sigma}_+\hat{\sigma}_- \rangle), \quad (26)$$

where $n_F = [e^{\omega_d/(k_B T_q)} + 1]^{-1}$ denotes the Fermi-Dirac occupation. Here we focus on a single-temperature environment $n_c = n_q$.

For a weak drive on resonance ($\Delta = 0$), the light emitted by the two-level system destructively interferes with the drive, preventing the cavity from being filled [34]. As a consequence, the conventional entropy production is reduced. In contrast, our new definition shows a peak in the entropy production at $\Delta = 0$ because part of the light is dissipated through the two-level system. Increasing temperature suppresses coherence in the two-level system. As a consequence, the features resulting from destructive interference are suppressed, and we find a finite J^{io} as the coherence of the light is reduced.

Three-level maser—The three-level maser is a canonic example of a quantum heat engine [13,16,20,35–38]. It can be described by

$$H' = \omega_3 |3\rangle\langle 3| + \omega_2 |2\rangle\langle 2| + g(\hat{a}|2\rangle\langle 1| + \hat{a}^\dagger |1\rangle\langle 2|), \quad (27)$$

$$H_{TD} = \omega_d(\hat{a}^\dagger \hat{a} + |2\rangle\langle 2|) + \omega_3 |3\rangle\langle 3|, \quad (28)$$

with $\omega_3 > \omega_2$. The thermodynamic Hamiltonian extends the two-level case by adding the state $|3\rangle$. Dissipation is described by $\mathcal{L}' = \mathcal{L}_H + \mathcal{L}_C$, with

$$\mathcal{L}_H = \gamma_H n_H \mathcal{D}[|3\rangle\langle 1|] + \gamma_H (n_H + 1) \mathcal{D}[|1\rangle\langle 3|], \quad (29)$$

$$\mathcal{L}_C = \gamma_C n_C \mathcal{D}[|3\rangle\langle 2|] + \gamma_C (n_C + 1) \mathcal{D}[|2\rangle\langle 3|], \quad (30)$$

where $n_H = [e^{\omega_3/(k_B T_H)} - 1]^{-1}$, $n_C = [e^{(\omega_3 - \omega_d)/(k_B T_C)} - 1]^{-1}$, and $T_H > T_C$ creates a population inversion between the states $|1\rangle$ and $|2\rangle$. Due to the quantum mechanical treatment of the cavity field, the conventional definitions of heat and work predict $P > 0$ and suggest that the system ceases to operate as a heat engine [see Fig. 2(e)] [13,16]. The reason for this is the energetic cost of maintaining coherence in the cavity. Our new framework resolves this flaw, as there is no energetic cost to maintain coherence when the output light can be further used, identifying regions in which $P_{io} < 0$. In these regions, a heat current between hot and cold baths increases the coherence in the output light.

Entropy production as loss of information—Before concluding, we provide an alternative derivation of our main results, connecting to a broader concept of entropy production that goes beyond Markovian master equations. Following Ref. [39], Eq. (8) may be obtained from

$$\Sigma = k_B S[\hat{\rho}_{\text{tot}} || \hat{\rho} \otimes \hat{\tau}], \quad (31)$$

where $S[\hat{\rho} || \hat{\sigma}] = \text{Tr}\{\hat{\rho} \ln \hat{\rho} - \hat{\rho} \ln \hat{\sigma}\}$, $\hat{\rho}_{\text{tot}}$ describes the density matrix of the system and environment, $\hat{\rho} = \text{Tr}_B\{\hat{\rho}_{\text{tot}}\}$ is obtained by tracing out the environment, and $\hat{\tau}$ denotes the Gibbs state of the environment. In Eq. (31), the entropy can be understood as the information that is lost when

describing the environment using only temperature [8]. For coherently driven systems, the environment also contains the coherent drive, as well as the response of the system. An appropriate state to describe the environment is then a displaced thermal state. We may obtain our new second law through

$$\Sigma^{io} = k_B S[\hat{\rho}_{\text{tot}} || \hat{\rho} \otimes \hat{\tau}(\alpha)], \quad (32)$$

where $\hat{\tau}(\alpha)$ denotes a displaced thermal state, with each bosonic bath mode $\hat{b}_k$ displaced by its average $\alpha_k = \text{Tr}\{\hat{b}_k \hat{\rho}_{\text{tot}}\}$ [18]. Equation (32) may serve as a starting point to extend our framework to strong coupling scenarios, where dissipation is no longer described by Eq. (5) [40,41], or to include further access on the output light, such as its squeezing.

Conclusions and outlook—We have shown how treating the output light of a coherently driven system as accessible leads to a consistent thermodynamic framework. This framework features a second law that is strictly tighter than the conventional approach. Without an additional entropy source, our second law implies that the noise in the output light is always larger than the noise in the input light. Our framework is, thus, particularly promising for investigating the noise properties of cascaded systems and opens up novel ways of investigating the thermodynamics of optical devices such as circulators and amplifiers. Of particular interest is the thermodynamics of nonreciprocal devices [42].

Our formalism only treats the coherent part of the output light as accessible. This requires the presence of a phase reference. Quantifying work in the output light in the absence of a perfect phase reference remains an open problem. Another promising avenue is to investigate fluctuations of heat and work in the new framework, which would allow for investigating fluctuation theorems [1] and thermodynamic uncertainty relations [43].

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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