Quantum Coherence and Ergotropy

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Constraints on work extraction are fundamental to our operational understanding of the thermodynamics of both classical and quantum systems. In the quantum setting, finite-time control operations typically generate coherence in the instantaneous energy eigenbasis of the dynamical system. Thermodynamic cycles can, in principle, be designed to extract work from this nonequilibrium resource. Here, we isolate and study the quantum coherent component to the work yield in such protocols. Specifically, we identify a coherent contribution to the ergotropy (the maximum amount of unitarily extractable work via cyclical variation of Hamiltonian parameters). We show this by dividing the optimal transformation into an incoherent operation and a coherence extraction cycle. We obtain bounds for both the coherent and incoherent parts of the extractable work and discuss their saturation in specific settings. Our results are illustrated with several examples, including finite-dimensional systems and bosonic Gaussian states that describe recent experiments on quantum heat engines with a quantized load.

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Introduction.—The Thomson [1] formulation of the second law is a constraint on the ability of an external agent to extract work from a system. More precisely, it states that no work can be extracted from a closed equilibrium system during a cyclic variation of a parameter by an external source [2,3]. This formulation was influential in mathematical physics, leading to a definition of the condition of thermal equilibrium for quantum states through the notion of passivity and complete passivity [4,5]. A state $\hat{\rho}$ is said to be passive with respect to a Hamiltonian when no work can be extracted from it by means of a cyclical variation of a Hamiltonian parameter, while it can be shown that a Gibbs state is the unique completely passive state such that $\hat{\rho}^{\otimes N}$ remains passive for all N. In other words, passivity allows us to derive Thomson's formulation of the second law as a constraint on unitary work extraction from quantum systems [6]. If a state is nonpassive with respect to a Hamiltonian, work can be extracted and, upon maximization over the space of cyclical unitaries, the optimal yield is known as the *ergotropy* [7,8]. The ergotropy has been established as an important quantity in the emerging field of quantum thermodynamics [10–14] and has recently been measured in two experiments which explore work deposition to external loads coupled to microscopic engines [15,16]. In the limit of many copies, the ergotropy converges to the conventional non-equilibrium part of the free energy [17] and it has also been incorporated into an open system thermodynamic description of finite quantum systems, recovering first and second laws [18].

A central theme in the field of quantum thermodynamics over the last decade has been the identification of uniquely quantum signatures in thermodynamic settings. This includes the identification of quantum effects in thermal machines [19–34], in work extraction protocols [17,35–48], in fluctuations of work [49–55], and in work deposition processes [56–62], to name but a few examples. Arguably the most fundamental of all nonclassical features is quantum coherence, yet precise mathematical techniques for its quantification have only recently been formulated in quantum information theory [63,64]. From the perspective of quantum thermodynamics, many studies have aimed at highlighting the nontrivial role that coherence may play [14,65–74]. Coherence is a basis-dependent quantity that can be expressed in terms of the relative entropy between the state of the system at hand and its dephased counterpart in the relevant basis [63]. This provides a connection to the finite-time thermodynamics of quantum systems, where the relative entropy is ubiquitous in the assessment of irreversible entropy production of closed [75–77] and open systems [78–85]. This connection was recently exploited in order to isolate a coherent contribution to the entropy production in quantum dynamics [86–89]. Here, the relevant coherence is defined relative to the energy eigenbasis, which plays a distinguished role in thermodynamics.

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In this work, we focus on the role of such coherence in ergotropic work extraction. We believe the simplicity of our approach, together with its operational significance will be of particular interest to those interested in isolating nonclassical signatures in quantum thermodynamics.

Preliminaries.—Given a quantum system in an initial state $\hat{\rho}$, and a Hamiltonian $\hat{H} = \sum_k \varepsilon_k |\varepsilon_k\rangle\langle\varepsilon_k|$, we are interested in the amount of coherence in the energy eigenbasis. In what follows, we will quantify the coherence with the relative entropy of coherence $C(\hat{\rho})$ [63,64]. This is motivated from the description of coherence as a quantum resource theory [64,90].

A quantum resource arises when there is a naturally restricted set of operations $\mathcal{O}$ which are significantly easier to implement than operations outside $\mathcal{O}$—e.g., local operations and classical communication (LOCC) in entanglement theory [91]. If these free operations $\mathcal{O}$ only allow some free states $\mathcal{F}$ to be created "for free," all other states become a resource whose creation requires the (costly) implementation of operations outside $\mathcal{O}$. We may quantify the resourcefulness of a nonfree state by means of a function μ that maps states to non-negative reals. We call μ a resource monotone if (i) its value cannot increase under application of any free operation $\Omega \in \mathcal{O}$ to any state $\hat{\rho}$: $\mu(\hat{\rho}) \geq \mu[\Omega(\hat{\rho})]$; and if (ii) $\mu(\hat{\varphi}) = 0$ for all $\hat{\varphi} \in \mathcal{F}$. One way of constructing a monotone μ is to minimize a (contractive) distance function d on the space of quantum states with respect to $\mathcal{F}$: $\mu_d(\hat{\rho}) \coloneqq \min_{\hat{\varphi}\in\mathcal{F}} d(\hat{\rho}, \hat{\varphi})$. The usefulness of such a distance-based μ_d then depends not least on its ease of computation—i.e., if it can be expressed as a closed-form function.

Returning to coherence, various viable classes of free operations have been considered for which the free states $\mathcal{F}$ are the set of incoherent states I_H, i.e., density matrices $\hat{\delta}$ that are diagonal in the energy eigenbasis [64]. For all of these classes, valid coherence monontones may be obtained based on suitable distance measures such as Tsallis relative α-entropies D_α for which succinct expressions have been found [92]: $C_\alpha \coloneqq \min_{\hat{\delta}\in I_H} D_\alpha(\hat{\rho}\|\hat{\delta})$, where the normalized state $\hat{\delta}_{\rho,\alpha} \propto \sum \langle\varepsilon_j|\hat{\rho}^\alpha|\varepsilon_j\rangle^{1/\alpha}|\varepsilon_j\rangle\langle\varepsilon_j|$ obtains the minimum. We here focus on the limit $\alpha \to 1$ as, in this case, the minimal state $\hat{\delta}_\rho \equiv \hat{\delta}_{\rho,\alpha} = \Delta(\hat{\rho})$ is directly connected to the original state $\hat{\rho}$ by a physical operation—dephasing with Δ. In this limit, D_α becomes the standard quantum relative entropy $D(\hat{\rho}\|\hat{\delta}) = \mathrm{Tr}\{\hat{\rho}(\log\hat{\rho} - \log\hat{\delta})\}$ and C_α becomes the entropy of coherence $C(\hat{\rho}) = S(\hat{\delta}_\rho) - S(\hat{\rho})$, with the Von Neumann entropy $S(\hat{\sigma}) = -\mathrm{Tr}\{\hat{\sigma}\log\hat{\sigma}\}$ [63].

Following the seminal paper [7], we are now interested in extracting work from the quantum system at hand by using a cyclic unitary transformation $\hat{U} \in \mathcal{U}_c$, where $\mathcal{U}_c$ denotes the set of unitary transformations generated in a given interval $(0, \tau)$ by a time-dependent Hamiltonian $\hat{H}(t)$ such that $\hat{H}(0) = \hat{H}(\tau) = \hat{H}$. In this context, one typically assumes complete control over the system [70]: that is, the possibility of generating any unitary evolution through suitable control fields applied to the system, which are switched off at the end of the transformation. Under the action of the unitary $\hat{U}$, the state transforms as $\hat{\rho} \to \hat{U}\,\hat{\rho}\,\hat{U}^\dagger$, and the average work extracted from the system is $W(\hat{\rho}, \hat{U}) = \mathrm{Tr}\{\hat{H}(\hat{\rho} - \hat{U}\,\hat{\rho}\,\hat{U}^\dagger)\}$. The maximum of W over the set $\mathcal{U}_c$ is called ergotropy, $\mathcal{E}$. After ordering the labels of eigenstates of $\hat{H}$ and of $\hat{\rho}$ such that $\hat{H} = \sum_{k=1}^d \varepsilon_k|\varepsilon_k\rangle\langle\varepsilon_k|$, with $\varepsilon_k < \varepsilon_{k+1}$, and $\hat{\rho} = \sum_{k=1}^d r_k|r_k\rangle\langle r_k|$, with $r_k \geq r_{k+1}$, we define the optimal *ergotropic* transformation $\hat{E}_\rho$ as the one that maps $\hat{\rho}$ into the passive state $\hat{P}_\rho = \hat{E}_\rho\hat{\rho}\hat{E}_\rho^\dagger = \sum_k r_k|\varepsilon_k\rangle\langle\varepsilon_k|$. We notice that the optimal unitary $\hat{E}$ depends on the state $\hat{\rho}$, and that the ergotropy is then given by

$$\mathcal{E}(\hat{\rho}) = \max_{\hat{U}\in\mathcal{U}_c} W(\hat{\rho}, \hat{U}) \equiv W(\hat{\rho}, \hat{E}_\rho) = \mathrm{Tr}\{\hat{H}(\hat{\rho} - \hat{P}_\rho)\} \equiv \sum_k \varepsilon_k(\rho_{kk} - r_k), \tag{1}$$

where ρ_{kk} (the population of $\hat{\rho}$ in the kth energy eigenstate) can be expressed as $\rho_{kk} = \sum_{k'} r_{k'}|\langle r_{k'}|\varepsilon_k\rangle|^2$. Our main aim is to demonstrate a precise connection between $\mathcal{E}$ and the amount of coherence in the initial state $C(\hat{\rho})$ [93]. In the following section, we show how to split the ergotropy into two contributions, one of which directly connected to the presence of energetic coherence in the state $\hat{\rho}$.

Coherent and incoherent contributions to ergotropy.—We start by introducing the incoherent part of the ergotropy $\mathcal{E}_i$, which can be defined in two equivalent ways. One can think of $\mathcal{E}_i$ as the maximum work extractable from $\hat{\rho}$ without altering its coherence. To formalize this idea, we can imagine breaking the transformation $\hat{E}_\rho$ into an incoherent operation followed by a second, coherence-consuming, cyclic unitary. To this end, we define the subset $\mathcal{U}_c^{(i)} \subset \mathcal{U}_c$ of incoherent, cyclic, unitary transformations, such that any $\hat{V} \in \mathcal{U}_c^{(i)}$ is coherence preserving: $C(\hat{\rho}) = C(\hat{V}\,\hat{\rho}\,\hat{V}^\dagger)$. Such $\hat{V}$ is in fact a member of the class of strictly incoherent operations (SIOs), which admit a very operational structure [64,94,95]; $\hat{V}$ amounts to a reshuffling of the energy basis, up to irrelevant phase factors, of the form $\hat{V} = \sum_k e^{-i\varphi_k}|\varepsilon_k\rangle\langle\varepsilon_{\pi_k}| \equiv \hat{V}_\pi$, where π_k is the kth element in the result of the permutation π of the indices [96]. The incoherent contribution to ergotropy is then defined as

$$\mathcal{E}_i = \max_{\hat{V}\in\mathcal{U}_c^{(i)}} W(\hat{\rho}, \hat{V}) \equiv \max_\pi W(\hat{\rho}, \hat{V}_\pi). \tag{2}$$

The optimal permutation $\tilde{\pi}$, realizing the maximum in the equation above, is the one that rearranges the populations

$\{\rho_{kk}\}_{k=1,\ldots d}$ in descending order: $\rho_{\tilde{\pi}_j\tilde{\pi}_j} \geq \rho_{\tilde{\pi}_{j+1}\tilde{\pi}_{j+1}}$, $\forall$ j. Letting $\hat{\sigma}_\rho = \hat{V}_{\tilde{\pi}}\hat{\rho}\hat{V}^\dagger_{\tilde{\pi}} = \sum_k \sum_{k'} \rho_{\tilde{\pi}_k,\tilde{\pi}_{k'}} |\varepsilon_k\rangle\langle\varepsilon_{k'}|$, the incoherent contribution to ergotropy is

$$\mathcal{E}_i(\hat{\rho}) = \mathrm{Tr}\{\hat{H}(\hat{\rho} - \hat{\sigma}_\rho)\} = \sum_k \varepsilon_k(\rho_{kk} - \rho_{\tilde{\pi}_k\tilde{\pi}_k}). \quad (3)$$

The state $\hat{\sigma}_\rho$ possesses the same coherence as $\hat{\rho}$, but less average energy. Therefore, $\mathcal{E}_i$ is the maximum amount of work that can be extracted from $\hat{\rho}$ without changing its coherence, and, among the states having the same amount of coherence as $\hat{\rho}$, $\hat{\sigma}_\rho$ is singled out as the one that possesses the least possible average energy [97]. In particular, we notice that, when trying to extract work from the state $\hat{\sigma}_\rho$ through the optimal cyclic unitary $\hat{E}_\sigma$, one arrives at the very same passive state that is obtained from $\hat{\rho}$. In our notation, $\hat{P}_\sigma = \hat{P}_\rho$. This is because $\hat{\sigma}_\rho$ has the same eigenvalues as $\hat{\rho}$.

An alternative (but equivalent) route to the identification of the incoherent contribution to ergotropy is provided by defining $\mathcal{E}_i$ as the maximum amount of work extractable from $\hat{\rho}$ after having erased all of its coherences via the dephasing map Δ. This amounts to defining $\mathcal{E}_i$ as the full ergotropy of the dephased state, $\mathcal{E}_i = \mathcal{E}(\hat{\delta}_\rho)$, where $\hat{\delta}_\rho = \Delta[\hat{\rho}]$ has the same energy populations as $\hat{\rho}$ (and, thus, the same average energy) but zero coherence. The ergotropy of $\hat{\delta}_\rho$ can be written by first defining the passive state $\hat{P}_\delta$ obtained from $\hat{\delta}_\rho$ after rearranging the populations in decreasing order, and then letting

$$\mathcal{E}_i(\hat{\rho}) \equiv \mathcal{E}(\hat{\delta}_\rho) = \mathrm{Tr}\{\hat{H}(\hat{\delta}_\rho - \hat{P}_\delta)\}. \quad (4)$$

This definition is fully equivalent to the one given in Eq. (3) [98]. Indeed, $\hat{\delta}_\rho$ has the same populations as $\hat{\rho}$ in the energy basis; consequently, the optimal reshuffling unitary that maps $\hat{\delta}_\rho$ into $\hat{P}_\delta$ is given by the very same $\hat{V}_{\tilde{\pi}}$ introduced above. This implies that $\hat{P}_\delta$ has the same populations as $\hat{\sigma}_\rho$ (in the same order!), but no coherence. As a result of these considerations, one immediately realizes that $\hat{P}_\delta$ can be obtained by applying the dephasing map to $\hat{\sigma}_\rho$, and that the two states share the same average energy:

$$\hat{P}_\delta \equiv \Delta[\hat{\sigma}_\rho] \Rightarrow \mathrm{Tr}\{\hat{H}\hat{\sigma}_\rho\} \equiv \mathrm{Tr}\{\hat{H}\hat{P}_\delta\}.$$

Having defined the incoherent part of $\mathcal{E}(\hat{\rho})$, the coherent contribution to ergotropy is simply given by the difference

$$\mathcal{E}_c = \mathcal{E} - \mathcal{E}_i = \mathrm{Tr}\{\hat{H}(\hat{\sigma}_\rho - \hat{P}_\rho)\} \equiv \sum_k \varepsilon_k(\rho_{\tilde{\pi}_k\tilde{\pi}_k} - r_k). \quad (5)$$

This is a non-negative quantity as, in general, $\hat{\sigma}_\rho$ is an active state. Notice further that it coincides with the full ergotropy of $\hat{\sigma}_\rho$.

The coherent ergotropy $\mathcal{E}_c$ can be understood as that part of extractable work which cannot be obtained by means of incoherent operations applied to state $\hat{\rho}$, and it is due to the presence of coherence in the initial state. Despite this, $\mathcal{E}_c$ is not a coherence monotone, as the inequality $\mathcal{E}_c(\hat{V}\,\hat{\rho}\,\hat{V}^\dagger) \leq \mathcal{E}_c(\hat{\rho})$ is not satisfied for every incoherent operation $\hat{V}$ (see Supplemental Material [101] for an illustrative example). Nevertheless, both the state $\hat{\sigma}_\rho$ and the coherent part of the ergotropy, $\mathcal{E}_c$, are uniquely defined once the state $\hat{\rho}$ and the Hamiltonian $\hat{H}$ are given, and they result entirely from the initial coherence, implying that $\hat{\sigma}_\rho$ is not passive.

Figure 1 summarizes these considerations and relationships. It shows the various states and operations defined up to now in the coherence-versus-average-energy plane.

Bounds for coherent ergotropy.—Given the form of the coherent ergotropy, we can provide upper and lower bounds to its value and show their tightness. Indeed, by introducing the Gibbs state $\hat{\rho}_\beta = e^{-\beta\hat{H}}/Z$ with inverse temperature β, we can exploit the identity $D(\hat{\sigma}\|\hat{\rho}_\beta) = \beta\mathrm{Tr}\{\hat{H}(\hat{\sigma} - \hat{\rho}_\beta)\} - S(\hat{\sigma}) + S(\hat{\rho}_\beta)$, valid for any state $\hat{\sigma}$, in order to obtain the following chain of relations

$$\begin{aligned}\beta\mathcal{E}_c &= \beta(\mathcal{E} - \mathcal{E}_i) = \beta\mathrm{Tr}\{\hat{H}(\hat{P}_\delta - \hat{P}_\rho)\} \\ &= \beta\mathrm{Tr}\{\hat{H}(\hat{P}_\delta - \hat{\rho}_\beta)\} - \beta\mathrm{Tr}\{\hat{H}(\hat{P}_\rho - \hat{\rho}_\beta)\} \\ &= [D(\hat{P}_\delta\|\hat{\rho}_\beta) + S(\hat{P}_\delta) - S(\hat{\rho}_\beta)] \\ &\quad - [D(\hat{P}_\rho\|\hat{\rho}_\beta) + S(\hat{P}_\rho) - S(\hat{\rho}_\beta)].\end{aligned}$$

After taking into account that $S(\hat{P}_\rho) = S(\hat{\rho})$, and that $S(\hat{P}_\delta) = S(\hat{\delta}_\rho)$ (due to the fact that they are connected by unitary transformations), and, finally, using $C(\hat{\rho}) = S(\hat{\delta}_\rho) - S(\hat{\rho})$, we obtain

$$\beta\mathcal{E}_c = C(\hat{\rho}) + D(\hat{P}_\delta\|\hat{\rho}_\beta) - D(\hat{P}_\rho\|\hat{\rho}_\beta), \quad (6)$$

which is valid for every finite β.

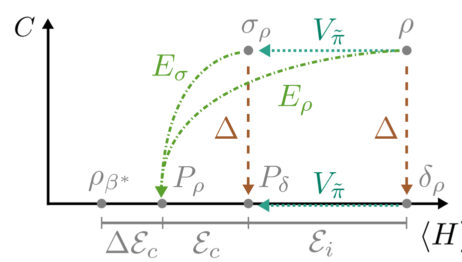

FIG. 1. Position of the various states (see main text) in a coherence-versus-average-energy diagram. Gray dots represent quantum states—e.g., arising from the initial state $\hat{\rho}$ after the transformations $\hat{E}_\rho$, Δ, $\hat{V}_{\tilde{\pi}}$ are performed. The arrows representing transformations are intended merely to point from the initial to the final state, without implying a precise path in the plane. For example, the transformation $\hat{V}_{\tilde{\pi}}$ is represented by a horizontal line because it connects states with the same amount of coherence; however, coherence may change *during* the transformation. The horizontal distance $\Delta\mathcal{E}_c$ between the thermal state $\hat{\rho}_{\beta^*}$ and $\hat{P}_\rho$ is the bound ergotropy (see Sec. IV). It may be zero, depending on the system at hand (i.e., iff the eigenvalues of ρ and ρ_{β^*} are related by a permutation).

From this relation, using the fact the $D \geq 0$, one easily obtains bounds for $\mathcal{E}_c(\hat{\rho})$:

$$C(\hat{\rho}) - D(\hat{P}_\rho \| \hat{\rho}_\beta) \leq \beta \mathcal{E}_c(\hat{\rho}) \leq C(\hat{\rho}) + D(\hat{P}_\delta \| \hat{\rho}_\beta). \quad (7)$$

One can saturate the upper bound if $\hat{P}_\rho = \hat{\rho}_\beta$. This requires that the ergotropic transformation $\hat{E}_\rho$ takes $\hat{\rho}$ to the thermal state $\hat{\rho}_\beta$. Because of unitarity of this transformation, a necessary condition on β is that $S(\hat{\rho}) = S(\hat{\rho}_{\beta^*})$. We label the specific value of β for which this entropic equality holds β^*, and note that it exists for any $\hat{\rho}$. Moreover, for a single qubit, as well as for the important class of bosonic or fermionic states of Gaussian form, the condition $\beta = \beta^*$ is not just necessary, but also sufficient for the saturation of the upper bound in Eq. (7) (see examples in Sec. V).

More generally, however, the choice $\beta = \beta^*$ does not imply saturation of the bound. That is, the difference

$$\begin{aligned} \Delta\mathcal{E}_c &:= \frac{1}{\beta^*}[C(\hat{\rho}) + D(\hat{P}_\delta \| \hat{\rho}_{\beta^*})] - \mathcal{E}_c(\hat{\rho}) \\ &= \frac{1}{\beta^*} D(\hat{P}_\rho \| \hat{\rho}_{\beta^*}) \geq 0 \end{aligned} \quad (8)$$

does not generally vanish. In fact, by expressing it as $\Delta\mathcal{E}_c = \mathrm{Tr}\{\hat{H}(\hat{P}_\rho - \hat{\rho}_{\beta^*})\}$ we note that it equates to what is called the bound ergotropy $\mathcal{E}_b$ [17]—, i.e., the amount of additional ergotropy that a global unitary transformation could retrieve from $\hat{\rho}^{\otimes n}$, per system, in the limit $n \to \infty$ (in addition to the single-system ergotropy $\mathcal{E}$).

The saturation of the upper bound of Eq. (7) is, furthermore, equivalent to the results of Ref. [86] where the irreversible work W_{irr} performed on a quantum system was analyzed for a unitary transformation taking an initial thermal state $\hat{\rho}_{\beta^*}$ to a final state $\hat{\rho} = \hat{U}\hat{\rho}_{\beta^*}\hat{U}^\dagger$. It was shown that $\beta^* W_{\mathrm{irr}} = C(\hat{\rho}) + D(\hat{\delta}_\rho \| \hat{\rho}_{\beta^*})$. For a cyclic transformation, W_{irr} coincides with the average work performed on the system, whose absolute value, in turn, coincides with the work extracted from it by the cyclic unitary $\hat{U}^\dagger$, when it is prepared in the state $\hat{\rho}$. If we take $\hat{U}^\dagger = \hat{E}_\rho$, then the result of Ref. [86] is translated into our notation as

$$\beta^* \mathcal{E}(\hat{\rho}) = C(\hat{\rho}) + D(\hat{\delta}_\rho \| \hat{\rho}_{\beta^*}), \quad \text{if } \hat{E}_\rho \hat{\rho} \hat{E}_\rho^\dagger = \hat{\rho}_{\beta^*}. \quad (9)$$

But, with the same argument as given above, the incoherent ergotropy, Eq. (4), can be rewritten (for any β) as

$$\beta \mathcal{E}_i(\hat{\rho}) = D(\hat{\delta}_\rho \| \hat{\rho}_\beta) - D(\hat{P}_\delta \| \hat{\rho}_\beta). \quad (10)$$

Taking $\beta = \beta^*$, and subtracting this relation from Eq. (9), we obtain the saturation of the upper bound of Eq. (7):

$$\beta^* \mathcal{E}_c(\hat{\rho}) = C(\hat{\rho}) + D(\hat{P}_\delta \| \hat{\rho}_{\beta^*}), \quad \text{if } \hat{E}_\rho \hat{\rho} \hat{E}_\rho^\dagger = \hat{\rho}_{\beta^*}. \quad (11)$$

The lower bound in Eq. (7), on the other hand, is saturated iff $\hat{P}_\delta = \hat{\rho}_\beta$ for some inverse temperature β. For $\mathcal{E}_c > 0$, this requires that the populations of $\hat{\rho}$ in the energy basis (coinciding with those of $\hat{\delta}_\rho$) are indeed thermal, but in the wrong order, and that the state $\hat{\rho}$ contains some coherence in the energy basis. An example is provided by the following qutrit density matrix, written in the energy basis:

$$\hat{\rho} = \begin{pmatrix} g_1 & c & 0 \\ c^* & g_3 & 0 \\ 0 & 0 & g_2 \end{pmatrix}, \qquad g_i = \frac{e^{-\beta\varepsilon_i}}{\sum_j e^{-\beta\varepsilon_j}}, \qquad |c| \leq \sqrt{g_1 g_3}.$$

For such a state, the three populations r_i are obtained by decreasingly ordering the set of numbers $\{(g_1 + g_3/2) + \sqrt{[(g_1 - g_3)^2/4] + |c|^2}; g_2; [(g_1 + g_3)/2] - \sqrt{[(g_1 - g_3)^2/4] + |c|^2}\}$, and the passive state $\hat{P}_\rho$ is obtained by taking the ordered set as energy level populations. On the other hand, $\hat{P}_\delta \equiv \hat{\rho}_\beta = \mathrm{diag}\{g_1, g_2, g_3\}$; but this thermal state does not have the same entropy as $\hat{\rho}$ (and β has nothing to do with β^*). Using the definitions above, we obtain $\mathcal{E} = \varepsilon_1(g_1 - r_1) + \varepsilon_2(g_3 - r_2) + \varepsilon_3(g_2 - r_3)$, while $\mathcal{E}_i = (\varepsilon_3 - \varepsilon_2)(g_2 - g_3)$. The difference between these two quantities gives $\mathcal{E}_c$, which saturates the lower bound in Eq. (7) [i.e., for these states, $D(\hat{P}_\delta \| \hat{\rho}_\beta) = 0$].

Lastly, we can exploit Eq. (6) to investigate the convertibility of the states $\hat{P}_\delta$ and $\hat{P}_\rho$ under thermal operations, and endow this problem with an operational interpretation thanks to the definition of ergotropy. Since both these states commute with the Hamiltonian and are passive, their convertibility may be addressed within the resource theory of athermality [102–104]. In particular, if a thermal operation [103,105] exists that takes $\hat{P}_\delta$ to $\hat{P}_\rho$ ($\hat{P}_\rho$ to $\hat{P}_\delta$), it follows that $D(\hat{P}_\delta \| \hat{\rho}_\beta) - D(\hat{P}_\rho \| \hat{\rho}_\beta) \equiv \beta\mathcal{E}_c - C(\hat{\rho}) \geq 0$ (≤ 0, respectively).

Examples.—In order to illustrate our results, we consider first the simple case of a qubit, having energy eigenvalues $\varepsilon_1 = 0$ and ε_2. In this case, any initial state $\hat{\rho}$ is transformed by the ergotropic transformation $\hat{E}$ into a passive state with a thermal structure $\hat{P}_\rho \equiv \hat{\rho}_{\beta^*}$, for a suitably chosen inverse temperature β^*. Then, $\Delta\mathcal{E}_c$ vanishes and the upper bound in Eq. (7) is saturated. Moreover, in this case, the coherent part of ergotropy can be directly expressed in terms of the purity of the state, $p(\hat{\rho}) = \mathrm{Tr}\{\hat{\rho}^2\}$ and of another coherence quantifier, the l_1 norm of coherence [64], defined as $C_{l_1}(\hat{\rho}) = 2|\langle\varepsilon_1|\hat{\rho}|\varepsilon_2\rangle|$. Indeed, some simple manipulations lead to

$$\mathcal{E}_c(\hat{\rho}) = \frac{\varepsilon_2}{2}\left[\sqrt{2p(\hat{\rho}) - 1} - \sqrt{2p(\hat{\rho}) - 1 - C_{l_1}^2(\hat{\rho})}\right]. \quad (12)$$

This is proved by noticing that $\mathcal{E}_c(\hat{\rho}) = \varepsilon_2(\rho_{22} - r_2)$, where the smallest eigenvalue of $\hat{\rho}$ is $r_2 = [1 - \sqrt{2p(\hat{\rho}) - 1}]/2$, and where the smallest population of $\hat{\rho}$ is $\rho_{22} = [1 - \sqrt{2p(\hat{\rho}) - 1 - C_{l_1}^2}]/2$.

If follows from Eq. (12) that the ergotropy increases for any operation Ω with $p[\Omega(\hat{\rho})] < \frac{1}{2} + \frac{1}{2}\{[\mathcal{E}_c(\hat{\rho})/(\varepsilon_2)] + \frac{1}{4}C_{l_1}^2[\Omega(\hat{\rho})][(\varepsilon_2)/\mathcal{E}_c(\hat{\rho})]\}^2$. In the Supplemental Material [101] we provide an example of an incoherent such operation—generalized amplitude damping—to prove that E_c is not a coherence monotone.

For a given value of the purity p, the coherence takes its maximum value for mixed states $\hat{\rho}$ with equal populations, $\rho_{11} = \rho_{22} = 1/2$, for which $p = (1 + C_{l_1}^2)/2$ and $\mathcal{E}_c = C_{l_1}/2$. It follows that $\mathcal{E}_c(\rho)$ is maximized if the initial state is a maximally coherent pure state with $C_{l_1} = 1$ and $p = 1$.

This latter observation is, in fact, more general: for a d-level system, we get the maximum value of $\mathcal{E}_c(\hat{\rho})$ (with, correspondingly, a null incoherent contribution $\mathcal{E}_i$) when $\hat{\rho}$ is a maximally coherent pure state, $\hat{\rho} = |\psi\rangle\langle\psi|$, with $|\psi\rangle = \sum_i |\varepsilon_i\rangle/\sqrt{d}$. In such a case, indeed, any incoherent unitary $\hat{V}_\pi$ preserves the average energy.

To discuss a less trivial case, where the upper bound in Eq. (7) is not always saturated, we now consider the behavior of the coherent part of ergotropy for a three-level system with energy eigenvalues $\varepsilon_1 = 0$, and $\varepsilon_2 = R\varepsilon_3$ [with $R \in (0, 1)$]. In particular, we ask under what conditions the bound is saturated (i.e., $\Delta\mathcal{E}_c = 0$). Selecting $\beta = \beta^*$ as required for saturation, Eq. (11) implies that once the energy values are fixed, what really matters are just the first two eigenvalues of the density matrix, r_1, r_2 (which fix the third one as $r_3 = 1 - r_1 - r_2$). For our three-level system, the bound ergotropy can be written as $\Delta\mathcal{E}_c = \varepsilon_3[r_2(R-1) + 1 - r_1 - Z^{-1}(Re^{-\beta^* R\varepsilon_3} + e^{-\beta^*\varepsilon_3})]$, where $Z = 1 + e^{-\beta^* R\varepsilon_3} + e^{-\beta^*\varepsilon_3}$. Looking for the values of r_1 and r_2 that give rise to a vanishing $\Delta\mathcal{E}_c$, we obtain the numerical results reported in Fig. 2, where we can appreciate that only under very stringent conditions on the eigenvalues of $\hat{\rho}$ one obtains a saturation of the inequality. For fixed R, all suitable eigenvalue pairs are confined to a single curve within the total (r_1, r_2) plane.

Beyond finite-dimensional systems, our results can also be directly applied to bosonic Gaussian states. By

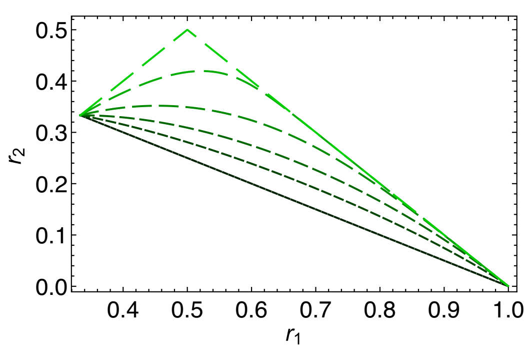

FIG. 2. In a three-level system, we identify the class of states that allow us to saturate the right inequality in Eq. (7) by looking at a pair of eigenvalues, r_1 and r_2, for which $\Delta\mathcal{E}_c = 0$. The various lines refer to the cases in which the ratio of the second and third energy eigenvalues is given by $R = 0$, 0.1, 0.3, 0.5, 0.7, 1 (lighter dashed to darker solid lines).

definition, these are related to a thermal state by a unitary transformation. As a consequence, they saturate the upper bound in Eq. (7). See the Supplemental Material [101] for a detailed example.

Summary and conclusions.—In summary, in this Letter we have highlighted the role of quantum coherence in work extraction processes, by identifying a contribution to the ergotropy that precisely corresponds to initial coherence in the energy basis. This is obtained by breaking the optimal, ergotropic, unitary cycle into an initial incoherent unitary operation, followed by a second unitary cycle through which one extracts work by exhausting the coherence. We have analyzed this coherent ergotropic contribution by exploring its range of possible values, which we have identified in terms of two bounds that can be saturated in specific cases. In particular, we discovered that the tightness of the upper bound is intimately related to the concept of bound ergotropy—a form of work potential that becomes available only when processing multiple identical copies of the system together. Finally, we have illustrated our results with the simplest nontrivial examples of a qubit and a qutrit, as well as a single-mode bosonic Gaussian state. The latter opens the possibility for future analysis of work extraction in continuous variable systems beyond unconstrained unitaries on single modes, considering, for instance, Gaussian operations, multiple modes, or both [106–110].

As quantum coherence is arguably the most primordial nonclassical effect in nature, we expect the framework described here to prove useful for the experimental characterization of work production in quantum heat engines [15,16], and, more generally, to help reveal and quantify the delicate fingerprints of genuinely quantum effects in nonequilibrium thermodynamic processes.

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[97] We note that, when the state $\hat{\rho}$ does not commute with $\hat{H}$ it evolves in the time. Thus, the application of the ergotropic unitary cyle $\hat{E}$ requires a very precise timing. On the other hand, this is not the case for the incoherent operation $\hat{V}_\pi$.

[98] This equivalence is a direct consequence of SIOs being a subset of dephasing-covariant incoherent operations (DIOs), which are those quantum channels that commute with full dephasing [99,100]. Here, this implies that $\hat{V}_{\tilde{\pi}}$ followed by Δ and Δ followed by $\hat{V}_{\tilde{\pi}}$ both result in the same state $\hat{P}_\rho$, and hence the same definition of $\mathcal{E}_i$.

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