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Experimental Investigation of Coherent Ergotropy in a Single Spin System

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Ergotropy is defined as the maximum amount of work that can be extracted through a unitary cyclic evolution. It plays a crucial role in assessing the work capacity of a quantum system. Recently, the significance of quantum coherence in work extraction has been theoretically identified, revealing that quantum states with more coherence possess more ergotropy compared to their dephased counterparts. However, an experimental study of the coherent ergotropy remains absent. Here, we report an experimental investigation of the coherent ergotropy in a single spin system. Based on the method of measuring ergotropy with an ancilla qubit, both the coherent and incoherent components of the ergotropy for the nonequilibrium state were successfully extracted. The increase in ergotropy induced by the increase in the coherence of the system was observed by varying the coherence of the state. Our work reveals the interplay between quantum thermodynamics and quantum information theory, future investigations could further explore the role other quantum attributes play in thermodynamic protocols.

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Quantum thermodynamics bridges two pillars of physics: quantum mechanics and thermodynamics [1–4]. One of its central topics is the work extraction from an out-of-equilibrium system. As a fundamental process in the thermodynamics of quantum systems, the work extraction has been extensively studied [5–12]. The concept of ergotropy, defined as the maximal amount of work that is extractable via cyclic unitary evolution, was brought up to describe the work capacity of a quantum state [13]. Beyond the mean energy of a quantum state, ergotropy reflects how much usable energy a quantum system can deliver to external systems. It has been measured recently in several experiments to showcase the performance of their thermodynamic devices [14–16]. The connection between the ergotropy of a quantum state and its quantum signatures has been identified theoretically [17–27]. One of the most fundamental nonclassical features of a quantum system is the coherence, its contribution to the ergotropy is isolated by dividing the optimal working-extracting operation into a coherence-preserving cycle and a coherence-consuming one [17,18,28]. Despite the theoretical advancements, an experimental investigation to demonstrate how coherence yields larger ergotropy remains absent. It is essential to experimentally study the relationship between coherence and ergotropy, which provides insights to both theoretical investigation and potential applications in thermodynamic protocols.

Here, we report an experimental investigation of the coherent ergotropy in a single spin system. We developed a method for ergotropy measurement with an ancilla qubit, which avoids the usage of complicated quantum state tomography. An isotopically purified diamond ($[^{13}\mathrm{C}] = 0.001\%$) was synthesized so that the electron spin in the nitrogen-vacancy (NV) center [29] with sufficiently long coherence time can be used to demonstrate the relationship between ergotropy and coherence (see Supplemental Material [30] for detailed information on the impact of decoherence on experimental results). In our experiment, the coherent and incoherent components of the ergotropy were extracted separately by dividing the optimal operation that extracts the ergotropy of the state into a coherence-preserving operation and a coherence-consuming one. Moreover, by adding coherence into a totally dephased state in energy basis, we observed a corresponding increase of the coherent ergotropy. Thus, the contribution of the coherence to the ergotropy was systematically revealed.

We study the work extraction process by considering a quantum system in state ρ subjected to Hamiltonian $H_S = \sum_n \epsilon_n |\epsilon_n\rangle\langle\epsilon_n|$, where ϵ_n ($|\epsilon_n\rangle$) is the energy

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eigenvalue (eigenstate). The mean energy can be evaluated simply as $\langle H_S\rangle_\rho = \mathrm{Tr}[\rho H_S]$. However, this is not the quantity of work that can be extracted to external systems. Thus, the ergotropy of a quantum state, which is defined as the maximal amount of extractable work under cyclic unitary evolution, is introduced to quantify the usable work of a quantum state. We consider a cyclic evolution in which the total Hamiltonian of the system can be written as $H_{\mathrm{total}} = H_S + V_{\mathrm{ext}}(t)$ $(0 \le t \le T)$, with $V_{\mathrm{ext}}(t)$ being the external driving or coupling to other systems, it satisfies $V_{\mathrm{ext}}(0) = V_{\mathrm{ext}}(T) = 0$. The work extracted is $W = \mathrm{Tr}[\rho H_S] - \mathrm{Tr}[U\rho U^\dagger H_S]$, where U is the evolution operator corresponding to the work extraction protocol. Because any unitary evolution can be generated through a suitable choice of $V_{\mathrm{ext}}(t)$ [35], the ergotropy can be found by taking the maximum of the extracted work over all possible evolution operators:

$$\mathcal{E}(\rho) = \max_U(\mathrm{Tr}[\rho H_S] - \mathrm{Tr}[U\rho U^\dagger H_S]). \tag{1}$$

The optimal operation is denoted as E_ρ and the corresponding final state is $P_\rho = E_\rho \rho E_\rho^\dagger$. P_ρ is called the passive state of ρ, indicating that no additional work can be extracted via any unitary operations [36].

To establish the connection between the coherence and the work extraction, the coherence of state ρ is quantified by the quantum relative entropy of ρ and its totally dephased state in energy basis [37,38]: $C(\rho) = D(\rho||\delta_\rho) = \mathrm{Tr}[\rho(\log\rho - \log\delta_\rho)]$, where $\delta_\rho = \sum_n |\epsilon_n\rangle\langle\epsilon_n|\langle\epsilon_n|\rho|\epsilon_n\rangle$. Subsequently, the incoherent component of the ergotropy is defined as the maximum work extractable from ρ without altering its coherence. Specifically, the incoherent ergotropy is given by $\mathcal{E}_i(\rho) = \max_V(\mathrm{Tr}[\rho H_S] - \mathrm{Tr}[V\rho V^\dagger H_S])$, where V is a unitary transformation that satisfies $C(\rho) = C(V\rho V^\dagger)$ [38–40]. The optimal coherence-preserving operation that extracts maximum work is denoted as V_π, where π signifies the permutation of energy basis up to irrelevant phase factors. If the eigenvalues of Hamiltonian H_S are in a descending order, V_π rearranges the population of ρ in an ascending order. Therefore, state ρ and its totally dephased counterpart δ_ρ have the same amount of incoherent ergotropy, as they share the same population distribution in energy basis [41]. The resulting state after the optimal incoherent extraction is denoted as $\sigma_\rho = V_\pi \rho V_\pi^\dagger$. State σ_ρ maintains the amount of coherence of ρ but stores less energy. To extract the rest of its remaining extractable energy, the operations that alter its coherence are introduced. The coherent ergotropy of ρ is thus isolated as the maximum extractable work of σ_ρ: $\mathcal{E}_c(\rho) = \mathcal{E}(\sigma_\rho)$. The process of extracting the ergotropy of ρ is equivalent to a sequential extraction of its incoherent and coherent ergotropy, formalized as

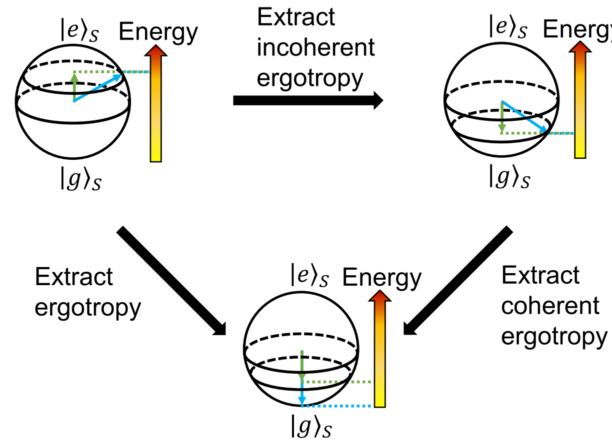

FIG. 1. Schematic diagram of the optimal extractions in a Bloch sphere in energy basis. $|e\rangle_S$ corresponds to the high-energy level. The energy of initial state with coherence (blue) and its totally dephased counterpart (green) can be extracted by first extracting the incoherent ergotropy and then the coherent part or directly extracting the ergotropy. The totally dephased state yields no contribution in the coherent extraction process.

$$\mathcal{E}_c(\rho) = \mathcal{E}(\rho) - \mathcal{E}_i(\rho). \tag{2}$$

The relation of the coherence and the coherent ergotropy can be quantitively expressed by introducing an inverse temperature parameter β and a Gibbs state ρ_β: $\beta\mathcal{E}_c(\rho) = C(\rho) + D(P_{\delta_\rho}||\rho_\beta) - D(P_\rho||\rho_\beta)$ [17], where P_{δ_ρ} is the passive state of δ_ρ, this relation is valid for every finite β.

We study the coherent ergotropy in energy basis such that the system Hamiltonian is diagonal. Without loss of generality, we choose our model Hamiltonian as $H_S = \left(\begin{smallmatrix}\epsilon & 0\\ 0 & 0\end{smallmatrix}\right)$, where $\epsilon = 1.05$ MHz. The initial system state is chosen as $\rho_S = |\psi_0\rangle\langle\psi_0|$, $|\psi_0\rangle = (\sqrt{2}|e\rangle_S + |g\rangle_S)/\sqrt{3}$, and its totally dephased state is $\delta_{\rho_S} = (2|e\rangle_{SS}\langle e| + |g\rangle_{SS}\langle g|)/3$. The states and the ergotropic extractions are depicted on Bloch spheres in Fig. 1. The blue (green) arrows represent ρ_S (δ_{ρ_S}) and the states after it is manipulated. Two approaches to the passive state are shown in Fig. 1. The first one is directly applying the optimal operation that transforms ρ_S (δ_{ρ_S}) into its passive state P_{ρ_S} ($P_{\delta_{\rho_S}}$). The total ergotropy of the initial system state is extracted, and the coherence of ρ_S is consumed completely at the same time. The total ergotropy of the two states, $\mathcal{E}(\rho_S)$ and $\mathcal{E}(\delta_{\rho_S})$, are obtained. The second approach to the passive states consists of two steps. The optimal incoherent operation $V_\pi = |e\rangle_{SS}\langle g| - |g\rangle_{SS}\langle e|$ is first applied to swap the order of population in energy basis. The resulting state after the incoherent extraction of ρ_S still stores the coherence and the coherent ergotropy of ρ_S. Meanwhile, the totally dephased state possesses zero extractable work after the incoherent extraction. The second step is to apply a coherence-consuming operation that transforms σ_{ρ_S}

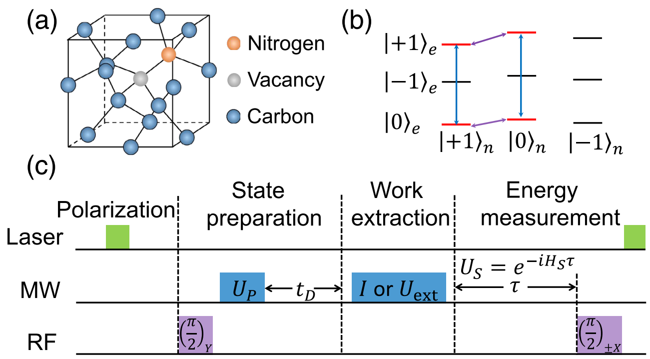

FIG. 2. The NV center system and the experimental pulse sequence. (a) Schematic atomic structure of the NV center. (b) Ground state energy levels of the NV center. The red lines denote the energy levels utilized in this experiment. The transitions between different electron (nuclear) spin states can be steered by microwave (radio frequency) pulses represented by blue (purple) arrows. (c) Experimental pulse sequence of ergotropy measurement, which includes polarization, state preparation, work extraction, and energy measurement. MW pulses are applied to prepare the electron state and extract work from it. rf pulses transform the nuclear spin state to assist the ergotropy measurement.

into P_{ρ_S} to extract the coherent ergotropy of ρ_S. Meanwhile, the dephased state remains unchanged and no work is extracted from it.

An NV center in diamond was utilized to investigate the coherent ergotropy of a quantum state. The NV center consists of a substitutional nitrogen atom adjacent to a vacancy shown in Fig. 2(a). When an external magnetic field is applied along the symmetric axis of the NV center, the Hamiltonian of the NV center can be written as

$$H_{\rm NV} = 2\pi(DS_z^2 + \omega_e S_z + QI_z^2 + \omega_n I_z + AS_z I_z), \quad (3)$$

where S_z (I_z) is the spin-1 operator of the electron (nuclear) spin, $D = 2.87$ GHz is the electronic zero-field splitting, $Q = -4.95$ MHz is the nuclear quadrupolar interaction constant, and $A = -2.16$ MHz is the hyperfine coupling constant. ω_e (ω_n) corresponds to the Zeeman frequency of the electron (nuclear) spin. The energy levels utilized in this experiment are represented by red bars in Fig. 2(b) with $|\ldots\rangle_e$ ($|\ldots\rangle_n$) encoding the electron (nuclear) spin state. In this work, the electron spin was considered as the system and the nuclear spin served as an ancilla qubit to readout the ergotropy of the system state. In the construction of the model Hamiltonian H_S, $|0\rangle_e$ was mapped to the high-energy level $|e\rangle_S$ of H_S. The magnetic field was set to 500 G and the NV center was polarized into state $|0\rangle_e|1\rangle_n$ via a green laser pulse [42] with the electron spin polarization being 0.97(1). The population of the nuclear spin state $|1\rangle_n$ is measured to be 0.99(1), which is near unity in our experiment after the polarization (see details in Supplemental Material [30]). As shown in Fig. 2(b), microwave (MW) pulses represented by blue arrows and radio-frequency (rf) pulses represented by purple arrows were applied to manipulate the quantum states of the electron spin and nuclear spin, respectively.

The pulse sequence for the measurement of the ergotropy is shown in Fig. 2(c). Practically, the ergotropy of a state is obtained by calculating the difference of the results of two energy measurements: the mean energy of the initial state and the mean energy of the state after an extraction operation is applied. Therefore, experimentally measuring the ergotropy requires implementing the pulse sequence twice. The whole sequence consists of four parts: polarization, state preparation, work extraction, and energy measurement. The NV center was polarized into state $|0\rangle_e|1\rangle_n$ by a green laser pulse. The nuclear spin was prepared in an equal superposition state $(|0\rangle_n + |1\rangle_n)/\sqrt{2}$ to facilitate the measurement of the mean energy [32] (see details in Supplemental Material [30]). The preparation of the initial state and the following manipulation of the system state were realized by applying microwave pulses [33,34]. The electron spin was first prepared in state ρ_S by U_P. The fidelity between the experimental initial state and the theoretical one is 1.00(1) (see details in Supplemental Material [30]). An extra free evolution time $t_D = 3T_2^*$ was required to dephase the coherence when preparing the dephased state δ_{ρ_S} with $T_2^* = 56(3)$ μs being the dephasing time of the electron spin. The following pulses differ according to the target of the experiment trial: (i) to measure the mean energy of ρ_S or δ_{ρ_S}, the work extraction stage was only an identity evolution; (ii) to measure the mean energy of the states after work extraction, a proper operation $U_{\rm ext}$ was applied in the work extraction stage depending on which component of the ergotropy was to be measured (detailed pulses are in Supplemental Material [30]). In our experiment, the operators are mainly affected by the spin-lattice relaxation, the interaction between the electron spin and the spin bath, and the fluctuation in the amplitude of the controlling field. We estimated the fidelities of the operations in the experiment taking these effects into account. The fidelities between the experimental operations and the ideal operations are higher than 99%, which shows that the operations in our experiment are very close to unitary (see detailed discussion in Supplemental Material [30]). Then a conditional unitary transformation $U_C = |1\rangle_{nn}\langle 1| \otimes I + |0\rangle_{nn}\langle 0| \otimes U_S$ was applied, where $U_S = e^{-iH_S\tau}$ with the evolution time $\tau = 500$ ns. After the evolution time, the nuclear spin state is $\rho_n = (|0\rangle_{nn}\langle 0| + |1\rangle_{nn}\langle 1| + \mathrm{Tr}[\rho_f U_S^\dagger]|0\rangle_{nn}\langle 1| + \mathrm{Tr}[\rho_f U_S]|1\rangle_{nn}\langle 0|)/2$, where ρ_f is the state of the electron spin before energy measurement. The information of the mean energy is encoded in the off-diagonal element of the nuclear spin state. Specifically, its imaginary part of the off-diagonal element is $\mathrm{Im}\mathrm{Tr}[\rho_f U_S^\dagger] \approx \mathrm{Tr}[\rho_f H_S]\tau = \langle H_S\rangle_{\rho_f}\tau$ with small evolution time approximation. This method to extract the mean energy can be applied to quantum systems

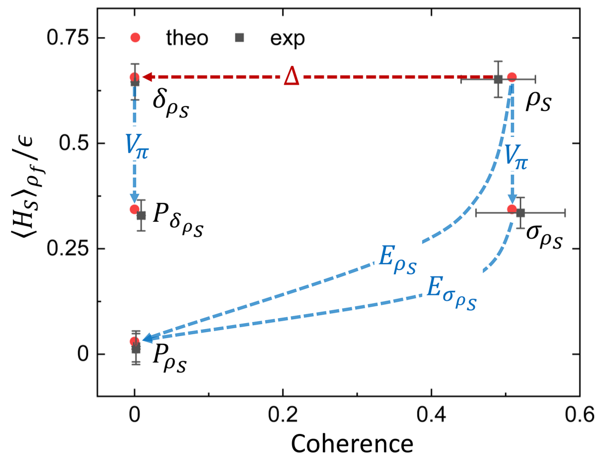

FIG. 3. Normalized mean energy and coherence of the system state before and after work extraction. Red dots (black squares) are the theoretical prediction (experimental result) of the mean energy and coherence. The influence of the imperfect polarization of the system spin is considered in the theoretical prediction. The green lines represent the unitary work-extracting operations, the red line represent the dephasing process. Two experimental data points correspond to one theoretical point of P_{ρ_S} because two trajectories lead to this point.

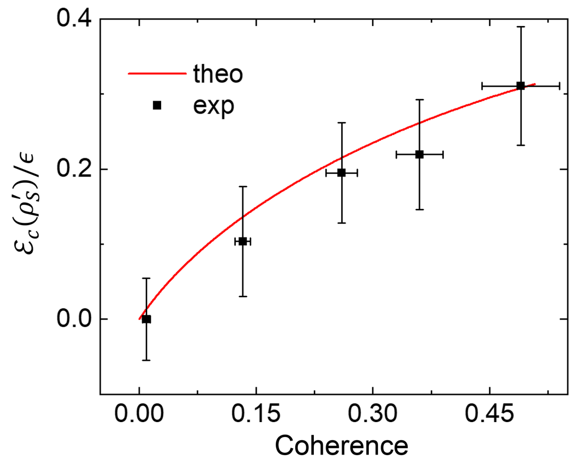

FIG. 4. The dependence of the normalized coherent ergotropy of the system state on the coherence. The black points are the experimental date and the red solid line represents the theoretical prediction of the coherent ergotropy and coherence. Error bars show one standard deviation.

of arbitrary dimension. Finally, another rf pulse was applied to measure the mean energy from the nuclear spin combined with the last laser pulse.

Figure 3 displays the energy and coherence change with the initial state being ρ_S or its totally dephased counterpart δ_{ρ_S}. The influence of the imperfect electron spin polarization was considered in the theoretical prediction. The coherence was evaluated via state tomography. The ergotropy components can be calculated by subtracting the exhibited data points. The normalized incoherent ergotropy of ρ_S is $\mathcal{E}_i(\rho_S) = \langle H_S \rangle_{\rho_S} - \langle H_S \rangle_{\sigma_{\rho_S}} = 0.32(6)$, while the coherence remains unchanged in accordance with the definition of incoherent extraction. The totally dephased counterpart δ_{ρ_S} was transformed into its passive state by the same extraction operation V_π. The resulting incoherent optimal yield is $\mathcal{E}(\delta_{\rho_S}) = \langle H_S \rangle_{\delta_{\rho_S}} - \langle H_S \rangle_{P_{\delta_S}} = 0.32(6)$, which matches the value of the incoherent ergotropy of ρ_S. After the incoherent extraction of ρ_S, the resulting state σ_{ρ_S} experienced one more extraction $E_{\sigma_{\rho_S}}$ to deliver the coherent ergotropy: $\mathcal{E}_c(\rho_S) = \langle H_S \rangle_{\sigma_{\rho_S}} - \langle H_S \rangle_{P_{\rho_S}} = 0.32(5)$. Finally, the work was directly optimally extracted from ρ_S to give the total ergotropy: $\mathcal{E}(\rho_S) = \langle H_S \rangle_{\rho_S} - \langle H_S \rangle_{P_{\rho_S}} = 0.63(6)$. An alternative assessment that should give the same total ergotropy is summing the incoherent and coherent components of the ergotropy, which is $\mathcal{E}(\rho_S) = \mathcal{E}_i(\rho_S) + \mathcal{E}_c(\rho_S) = 0.63(8)$. Our experimental results agree well with Eq. (2).

To further investigate the dependence of the coherent ergotropy on the coherence, we varied the coherence of the system state (see details in Supplemental Material [30]) and measured its coherent ergotropy. We prepared state ρ_S', which has an identical population distribution as δ_{ρ_S}, but with different off-diagonal elements in energy basis. The increase of coherent ergotropy is given by $\mathcal{E}_c(\rho_S') = [C(\rho_S') - D(P_{\rho_S'}||\rho_\beta)]/\beta$. The coherence and coherent ergotropy of different ρ_S' were experimentally obtained. The theoretical prediction and experiment result are displayed in Fig. 4. For each state ρ_S', the optimal extraction operation was applied to evaluate the total ergotropy $\mathcal{E}(\rho_S')$. The coherent ergotropy was obtained by taking the difference $\mathcal{E}_c(\rho_S') = \mathcal{E}(\rho_S') - \mathcal{E}_i(\rho_S')$. The equality $\mathcal{E}_i(\rho_S') = \mathcal{E}_i(\rho_S)$ was utilized, since the incoherent ergotropy solely depends on the population distribution in energy basis. The experiment result aligns well with the theoretical prediction within one standard deviation. The positive contribution of the coherence to the ergotropy of a state is clearly confirmed.

In summary, we demonstrated the role of quantum coherence in the ergotropic work extraction in a single spin system. Both the incoherent and coherent ergotropy were experimentally measured, and the positive dependence of the coherent ergotropy on the coherence of the state is observed. The interplay between quantum information theory and quantum thermodynamics is revealed by studying the work extraction process. Future studies could further investigate the relationship between the ergotropy of a quantum state and other distinctive quantum properties, such as entanglement [24,25] and quantum discord [27]. Additionally, the concept of the local and global ergotropy can also be studied by extending the system size [43]. The ergotropy has also been employed to study the thermodynamics of quantum systems subjected to environment, the work storing and extracting process can be studied by considering the effects of the surrounding environment [19,44]. Our work can potentially guide the enhancement of the capacity and efficiency of quantum devices operationally.

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