Experimental verification of a Jarzynski-related information-theoretic equality using a single trapped ion

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Most non-equilibrium processes in thermodynamics are quantified only by inequalities, however the Jarzynski relation presents a remarkably simple and general equality relating non-equilibrium quantities with the equilibrium free energy, and this equality holds in both classical and quantum regimes. We report a single-spin test and confirmation of the Jarzynski relation in quantum regime using a single ultracold \(^{40}\text{Ca}^+\) ion trapped in a harmonic potential, based on a general information-theoretic equality for a temporal evolution of the system sandwiched between two projective measurements. By considering both initially pure and mixed states, respectively, we verify, in an exact and fundamental fashion, the non-equilibrium quantum thermodynamics relevant to the mutual information and Jarzynski equality.

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Since the original proposals of the celebrated ideas of Maxwell’s demon \([1]\) and Szilárd’s engine \([2]\), much effort has been devoted to incorporating information into thermodynamics by reconsidering the meaning of thermodynamical entropic and energetic costs. So far, the field of information thermodynamics has reformulated the restrictions of the original thermodynamics, e.g., the second law of thermodynamics, in the light of the interplay between the amount of information and its thermodynamic utility. Reconsideration of the second law based on the notion of information and further clarifications of the physical nature of information are typically expected to reconcile any apparent contradictions we might have regarding our understanding of the laws of thermodynamics \([3]\).

In addition to this, there has been a parallel line of development because the conventional equilibrium thermodynamics cannot reasonably treat most natural or engineered processes that occur far from equilibrium. Namely, the non-equilibrium processes in thermodynamics are usually described by inequalities (or equalities that only hold in the linear regime, which means not far from equilibrium). In contrast, the Jarzynski relation presents a simple and general equality to calculate the free-energy difference between two states from Boltzmann-weighted statistics of the irreversible work done along the trajectories arbitrarily out of equilibrium \([4]\). As the only equality in non-equilibrium thermodynamics, the Jarzynski relation can also be understood from the fluctuation theorem \([5]\) under the assumption of microscopically reversible and thermostated dynamics. The ensuing investigations \([4, 5]\) have further confirmed that the Jarzynski equality promises to correctly predict any behavior, adiabatic or arbitrarily fast, in the presence of the Boltzmann statistics. A comprehensive review of thermodynamic experiments regarding the fluctuation theorem can be found in \([6]\).

Understanding thermodynamical process at the quantum level is currently a topic attracting much attention \([7, 8]\). Several attempts have been paid to extend the Jarzynski relation to quantum regime \([9, 10]\). From the quantum perspective, the origin of fluctuations is no longer just thermal but also quantum, and most thermodynamical quantities should be retracted. For example, the amount of work itself is not an observable in quantum thermodynamics, and its quantification therefore needs to be reconsidered \([11, 12, 13, 14]\). Besides this, the quantum entropy is actually an indication of the entanglement between the system and its environment \([15]\), and is no longer simply the thermodynamical arrow of time.

Here we show a single-spin verification of an information-theoretic equality relevant to Jarzynski re-
We first review briefly the main points in \cite{28}. The scheme gets started from a quantum state $\rho$, followed by a measurement on the basis $\{P\}$. Then the ensuing evolution is governed by the most general completely positive trace preserving (CPTP) map, $\sum_i \Lambda_i(\cdot) \Lambda_i^\dagger$, followed by another measurement on the basis $\{Q\}$. Such a process, under the Born rule, can be described by the joint probability

$$p_{nm} = \text{tr}\{Q_m \sum_i \Lambda_i(P_n \rho P_n) \Lambda_i^\dagger Q_m\} = p_m|n_p_n, \quad (1)$$

where $p_n = \text{tr}\{P_n \rho\}$ is the probability regarding the measurement $\{P\}$, and $p_m|n = \text{tr}\{Q_m \sum_i (\Lambda_i P_n \Lambda_i^\dagger)\}$ is the conditional probability implying the result of the second measurement dependent on the first measurement outcome. These quantities are associated with the mutual information

$$I_{nm} = -\ln q_m + \ln p_m|n, \quad (2)$$

which witnesses the difference between the entropy of the $m$th outcome without the knowledge of $n$ (given by $-\ln q_m$ with $q_m = \text{tr}\{Q_m \sum_i (\Lambda_i P_n \Lambda_i^\dagger)\}$) and the $m$th outcome when $n$ is known (given by $-\ln p_m|n$). Based on the mutual information $I_{nm}$, an information-theoretic equality is proposed, which satisfies the equality below,

$$\langle e^{-I_{nm}} \rangle := \sum_{nm} p_{nm} e^{-I_{nm}} = 1. \quad (3)$$

The equation not only gives a simple expression of the probability conservation, but also represents a relation to the Jarzynski equality \cite{4}, if the system is initially prepared as a Gibbs state. The relation is stated as

$$I_{nm} = -\beta (W - \Delta F), \quad (4)$$

where $W$ represents the work the system performs between the initial and final states with the free energy difference $\Delta F$. The free energy is defined as $F = -\ln Z/\beta$ with the partition function $Z = \sum_n e^{-\beta E_n}$, where $\beta = 1/k_BT$ is the temperature parameter with the Boltzmann constant $k_B$ and the temperature $T$, and $E_n$ is the eigenenergy under the measurement.

Before presenting our experimental observations, we introduce briefly our system involving a single $^{40}\text{Ca}^+$ ion confined stably in a linear Paul trap \cite{30}, whose axial and radial frequencies are $\omega_z/2\pi = 1.01$ MHz and $\omega_r/2\pi = 1.2$ MHz, respectively. Under the magnetic field of 6 Gauss, we encode the qubit in $|4S_1/2, m_J = +1/2\rangle$ as $|\downarrow\rangle$ and in $|3S_5/2, m_J = +3/2\rangle$ as $|\uparrow\rangle$, where $m_J$ is the magnetic quantum number. Although our investigation below only focuses on this qubit, cooling the ion to be ultracold is still necessary because thermal phonons yield offsets of Rabi oscillation. As such, the Doppler cooling and the resolved sideband cooling are executed in order, which leads to the $z$-axis motional mode to be cooled down to the vibrational ground state with the final average phonon number $\bar{n}_z < 0.1$.
The numbers in parentheses represent the standard errors of the mean, i.e., the RMS error.

\[ \sum_{n,m} p_{nm} I_{nm} \] is to check whether the summation of all the possible mutual information is positive, and \( e^{-\tau_{nm}} \) should be close to unit. The numbers in parentheses represent the standard errors of the mean, i.e., the RMS error.

\[
\begin{array}{cccccc}
\alpha & t = \pi/5\Omega & t = 2\pi/5\Omega & t = 3\pi/5\Omega & t = 4\pi/5\Omega \\
1 & 0.001(21) & 0.002(6) & 0.001(18) & 0.978(25) & 0.978(8) & 0.979(11) & 0.973(20) \\
\sqrt{2/3} & 0.937(54) & 0.560(23) & 0.508(19) & 0.509(46) & 0.985(39) & 0.985(61) & 1.015(63) & 0.974(29) \\
\sqrt{1/3} & 0.520(36) & 0.540(24) & 0.553(25) & 0.930(51) & 0.983(59) & 1.021(78) & 1.023(55) & 1.009(29) \\
\end{array}
\]

The qubit is initialized to \( |\downarrow\rangle \) with a probability of 99.3(2)\%.

With the 729-nm laser pulses, we realize the carrier-transition Hamiltonian \( H_c = \Omega \left( \sigma_z e^{i\phi} + \sigma_x e^{-i\phi} \right)/2 \) and the system evolves under the government of the carrier-transition operator

\[
U_C(\theta, \phi) = \cos(\theta/2)I - i\sin(\theta/2)\left( \sigma_x \cos \phi - \sigma_y \sin \phi \right),
\]

where \( \theta = \Omega t \) is determined by the evolution time with the laser-ion coupling strength \( \Omega/2\pi = 47.0(5) \) kHz, and \( \phi \) represents the laser phase. Each experimental cycle is synchronized with the 50-Hz AC power line and repeated 40,000 times. The 729-nm laser beam is controlled by a double pass acousto-optic modulator. The frequency sources for the acousto-optic modulator are based on a direct digital synthesizer controlled by a field programmable gate array. Employment of the direct digital synthesizer helps the phase- and frequency-control of the 729-nm laser during each experimental operation.

In the first part of our scheme, we focus on pure states to verify Eq. (5) and the second part is to test the Jarzynski equality related to Eq. (4) by exemplifying the thermal states as the Gibbs states. Our operations in each part consist of four steps [24]. For example, for the pure state case, the steps include: From \( |\downarrow\rangle \) to \( |\xi\rangle \) - state preparation; From \( |\xi\rangle \) to \( |\zeta\rangle \) - CPTP map; From \( |\zeta\rangle \) to \( |\z\rangle \) - state measurement; Finally a projection measurement on \( |\uparrow\rangle \). The first three steps are achieved, respectively, by \( U_c(\theta_0, \phi_0) \), \( U_c(\theta_1, \phi_1) \) and \( U_c(\theta_2, \phi_2) \), based on Eq. (5).

The projectors, in the Bloch representation, are generally described as \( P_\pm = (I \pm \vec{\sigma} \cdot \vec{\sigma})/2 \) and \( Q_\pm = (I \pm \vec{q} \cdot \vec{\sigma})/2 \) with \( \vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z) \).

For the case of pure states, we choose \( \vec{p} = (0, 0, 1) \) and \( \vec{q} = (1, 0, 0) \). We first produce a pure state \( \rho \) by \( U_c(\theta_0, \phi_0) \), followed by a measurement \( P_\pm = (I \pm \sigma_z)/2 \), an ensuing evolution under \( U_c(\theta_1, \phi_1) \) and another measurement \( Q_\pm = (I \pm \sigma_y)/2 \). Since our measurements are performed by detecting the population in the state \( |\downarrow\rangle \langle\uparrow| \), execution of \( P_\pm \) or \( Q_\pm \) is accomplished by a measurement pulse under the unitary operator \( U_c(\theta_2, \phi_2) \) in addition to a projective measurement. For example, the measurement pulse for \( P_- \) is performed by \( U_c^\dagger(\theta_2, \phi_2) |\uparrow\rangle \langle\downarrow| U_c(\theta_2, \phi_2) \) with \( \theta_2 = \pi, \phi_2 = 0 \), as specified in Table I. To verify Eq. (3), we need three measurement results \( p_n, p_m \) and \( p_{mn} \), which are obtained, respectively, by the three steps as shown in Fig. 1.

With the pure state \( \rho = |\psi\rangle\langle\psi| \) with \( |\psi\rangle = \alpha |\downarrow\rangle - i \beta |\uparrow\rangle \) and \( \alpha^2 + \beta^2 = 1 \), we have accomplished experimental measurements \( p_n, p_m \) and \( p_{mn} \) by choosing three diff-
different pure states with $\alpha = 1$, $\sqrt{2/3}$ and $1/\sqrt{3}$. Fig. 2 demonstrates the results for $\alpha = \sqrt{2/3}$. In our case, since the first measurement is made on the eigenstates of $\sigma_z$, the results strongly depend on the initial state of the system and remain unchanged with time. But the second measurement is different due to outcomes from the eigenstates of $\sigma_y$. As such, the results of both $Q_\pm$ and the conditional probability $p_{m|n}$ are time dependent. Based on the measurement results as listed in Table II, we confirm Eq. (3) under root-mean-square (RMS) error $\leq 0.078$, in which the error is induced dominantly by quantum projection noise, relevant to vacuum fluctuation, rather than the thermal noise in conventional thermodynamics. This evidently indicates that Eq. (3) is robust against vacuum fluctuation in quantum thermodynamical process. Besides, in terms of quantum information theory, the total mutual information should be never negative. But subject to quantum projection noise, individual observations of $I_{nm}$ in our experiment are sometimes negative. Nevertheless, our observation of the total mutual information $\sum_{nm} p_{nm} I_{nm}$, as listed in Table II, is always positive, in agreement with the results from the fluctuation theorem based on the probability distributions.

Considering a more general situation with the mixed states, we initially prepare a thermal state in the system, followed by a temporal evolution sandwiched by two projective measurements. In this way, we confirm a Jarzynski equality [28, 31] relevant to the mutual information $I_{nm}$ as tested above. To this end, we may start from a thermal state $\rho_i = \exp(-\beta H_i)/Z_i$ with the partition function $Z_i = tr\{\exp(-\beta H_i)\}$, where $H_i$ is the Hamiltonian of the system after the projective measurement on $\{P\}$. So we assume $H_i = \sum_{\pm} E_{\pm} P_{\pm}$ with $|E_{\pm}| = E_i$. For another projective measurement on $\{Q\}$, we have the Hamiltonian $H_f = \sum_{\pm} E_{\pm} Q_{\pm}$ with $|E_{\pm}| = E_f$. In our experiment, due to only two levels involved, we simply have $E' = E = E_f$. So the work is defined as $W = E_i - E_f$ where $E_i$ and $E_f$ are the corresponding eigenvalues regarding the measurements $\{P\}$ and $\{Q\}$. The free energy difference is $\Delta F = F_i - F_f$, where $F_k = - ln Z_k/\beta$ with $Z_k = tr\{\exp(-\beta H_k)\}$. Thus we have $p_{nm} = tr\{Q_{m} U_{C} P_{n} \rho_{i} P_{n} U_{C}^{\dagger} Q_{m}\} = tr\{Q_{m} P_{n} \rho_{i} P_{n}\} = tr\{Q_{m} P_{n} \rho_{i} P_{n}\} e^{-\beta E_{i}}/Z_i$, where we have used the fact that $Q_{m}$ commutes with $U_{C}$ and $P_{n}$ commutes with $P_{n}$. Based on above processes, Eq. (3) works and Eq. (6) can be rewritten as [28, 31]

$$\langle e^{\beta W - \Delta F} \rangle = 1,$$

which is termed the Jarzynski equality to be verified below.

In our operations below, we choose $\vec{p} = (0, 0, 1)$, implying $H_i = \sigma_x$, and we consider three different forms of $H_f$ with $\vec{q} = (1, 0, 0)$, $(0, 1, 0)$ and $(1/2, \sqrt{3}/2, 0)$, respectively, corresponding to $H_f = \sigma_x, \sigma_y$ and $\sigma_z/\sqrt{2}$. Then we obtain a two-level Gibbs state $\rho_i = \exp(-\beta E_{i})/Z_i = [e^{\beta E_i} |\downarrow\rangle\langle\downarrow| + e^{-\beta E_i} |\uparrow\rangle\langle\uparrow|]/Z_i$ with $Z_i = e^{-\beta E_i} + e^{\beta E_i}$. In this case, we find that $Z_i = Z_f$ implying $\Delta F = 0$.

By means of the qubit dephasing, we experimentally prepare the Gibbs state, and then carry out operations following similar steps to the pure state case. Accomplishment of the measurements regarding $P_\pm$ and $Q_\pm$ also depends on the measurement operator $U_{C}^{\dagger}(\theta_2, \phi_2) |\uparrow\rangle\langle\uparrow| U_{C}(\theta_2, \phi_2)$, where the values of $\theta_2$ and $\phi_2$ are listed in Table III. By considering the initial states regarding $\beta E = 0.2, 0.5$ and $1.0$, respectively, we have carried out the above steps and confirmed Eq. (6) with high precision, see Table. IV where the RMS errors are smaller than $0.03$. Different from the case of pure states, both thermal noise and quantum projection noise exist in this case, where the latter is dominant as analyzed in [29]. We have a smaller RMS error here than the pure state case just because the measurements made here are simpler [29]. The observation values in Table. IV indicate that the Jarzynski equality holds under the influence of vacuum fluctuation and on the other hand, our operations are precise enough to witness a single-spin thermodynamic process governed by the Jarzynski equality.

The experimentally determined errors are partly from imperfection of initial state preparation ($0.7(2)\%$) and final state detection ($0.22(8)\%$). Decoherence effects are negligible due to our short-time implementation: $50 \mu$s operation time for pure states and $3$ ms operation time for mixed states. The dominant errors, as mentioned above, due to quantum projection noise are inevitable in any quantum mechanical measurement, but can be reduced by more measurements. As such, we have tried to repeat our measurements by $40,000$ times, suppressing the relevant errors for individual point to be below $2\%$.

In summary, our experiment has provided the first single-spin evidence confirming a simple and general

| $\beta E$ | $\sum_{nm} p_{nm} (\Delta F - W)$ | $\langle e^{\beta W - \Delta F} \rangle$ |
|---------|---------------------------------|----------------------------------|
| 0.2     | 0.046(3) 0.044(4) 0.048(3)      | 0.987(14) 0.998(17) 0.999(14)    |
| 0.5     | 0.234(8) 0.231(12) 0.240(8)     | 0.990(17) 1.002(20) 1.002(17)    |
| 1       | 0.766(13) 0.761(25) 0.779(15)   | 0.963(23) 0.977(26) 0.976(24)    |
equality involving the expectation value of the exponential of mutual information. Since the equality relies on the properties of classical probabilities (that arise from the projective quantum measurements) and is concomitant to the quantum Jarzynski equality, our experimental implementation at this fundamental level of a single spin will be helpful for further understanding thermodynamic processes in quantum regime, particularly when quantum information and more degrees of freedom are involved \[5\] \[51\].

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