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Batalhão, T. B., Souza, A. M., Mazzola, L., Auccaise, R., Sarthour, R. S., Oliveira, I. S., Goold, J., De Chiara, G., Paternostro, M., & Serra, R. M. (2014). Experimental Reconstruction of Work Distribution and Study of Fluctuation Relations in a Closed Quantum System. *Physical Review Letters*, 113(14), Article 140601. <https://doi.org/10.1103/PhysRevLett.113.140601>

Published in:
Physical Review Letters

Document Version:
Publisher's PDF, also known as Version of record

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Experimental Reconstruction of Work Distribution and Study of Fluctuation Relations in a Closed Quantum System

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(Received 1 March 2014; revised manuscript received 15 August 2014; published 3 October 2014)

We report the experimental reconstruction of the nonequilibrium work probability distribution in a closed quantum system, and the study of the corresponding quantum fluctuation relations. The experiment uses a liquid-state nuclear magnetic resonance platform that offers full control on the preparation and dynamics of the system. Our endeavors enable the characterization of the out-of-equilibrium dynamics of a quantum spin from a finite-time thermodynamics viewpoint.

DOI: 10.1103/PhysRevLett.113.140601

PACS numbers: 05.70.Ln, 05.30.-d, 05.40.-a, 64.60.Ht

Research on the out-of-equilibrium dynamics of quantum systems has so far produced important statements on the thermodynamics of small systems undergoing quantum mechanical evolutions [1,2]. Key examples are provided by the Crooks and Jarzynski relations [3,4]: taking into account fluctuations in nonequilibrium dynamics, such relations connect equilibrium properties of thermodynamical relevance with explicit nonequilibrium features. Although the experimental study of such fundamental relations in the classical domain has encountered considerable success [5–9], their quantum mechanical versions [10] require the assessment of the statistics of work performed by or onto an evolving quantum system, a step that has so far shown hurdles due to the practical difficulty to perform reliable projective measurements of instantaneous energy states [2,11], which embodies a key experimental challenge.

Albeit a few interesting proposals to overcome such bottlenecks have been made [11,12], including an ingenious calorimetric one [13], the experimental reconstruction of the work statistics for a quantum protocol has so far remained elusive. Recently, an alternative approach to this problem has been devised, based on well-known interferometric schemes of the estimation of phases in quantum systems, which bypasses the necessity of direct projective measurements on the instantaneous state for the system [14,15] (see Ref. [16] for an interesting development of the original proposal).

In this Letter we exploit such a scheme to study the out-of-equilibrium thermodynamics of a spin-1/2 system undergoing a closed quantum nonadiabatic evolution, realized in a liquid-state nuclear magnetic resonance

(NMR) setup [17–22], and thus achieve sufficient information to assess both the Tasaki-Crooks and Jarzynski identities. To the best of our knowledge, our Letter reports the first experimental assessment of fluctuation relations for quantum dynamics.

Work statistics in the quantum domain.—When addressing quantum dynamics, the concept of work done by or on a system needs to be reformulated [23] so as to include *ab initio* both the inherent nondeterministic nature of quantum evolution and the effects of quantum fluctuations. In this sense, work acquires a meaning only as a statistical variable W . In order to introduce the associated probability distribution, let us consider a quantum system undergoing a transformation that changes its Hamiltonian as $\hat{\mathcal{H}}(0) \rightarrow \hat{\mathcal{H}}(\tau)$ in a time τ . We refer to this as the *forward* protocol, with corresponding distribution $P^F(W) = \sum_{n,m} P_n^0 P_{m|n}^\tau \delta[W - (\bar{\epsilon}_m - \epsilon_n)]$ [23]. We have introduced the probability p_n^0 to find the system in the n th eigenstate of $\hat{\mathcal{H}}(0)$ (with energy ϵ_n) at the start of the protocol, and the transition probability $p_{m|n}^\tau$ to find it in the m th eigenstate of $\hat{\mathcal{H}}(\tau)$ (with energy $\bar{\epsilon}_m$) at time τ if it were in the n th state at initial time. One can then define a *backward* protocol that implements the transformation $\hat{\mathcal{H}}(\tau) \rightarrow \hat{\mathcal{H}}(0)$ with an inverted control sequence. It is worth mentioning that recent years have seen the proposal of other formulations of quantum work that explicitly bypass the two-time energy measurements illustrated above [24].

While the initial state of the system can be arbitrary, in this Letter we will be concerned with initial thermal-equilibrium states at a given temperature. Moreover, it is often convenient to use the Fourier transform of the work distribution, or

work characteristic function. For the forward protocol, this is defined as $\chi_F(u) = \int P_F(W) e^{iuW} dW$ and takes the form

$$\begin{aligned} \chi_F(u) &= \sum_{m,n} p_n^0 p_m^\tau e^{iu(\bar{\epsilon}_m - \epsilon_n)} \\ &= \text{Tr}[(\hat{U} e^{-iu\hat{\mathcal{H}}(0)}) \rho_0 (e^{-iu\hat{\mathcal{H}}(\tau)} \hat{U})^\dagger], \end{aligned} \quad (1)$$

with ρ_0 being the initial equilibrium state of the system, \hat{U} the time propagator generated by the forward protocol, and u the conjugate variable to W . The characteristic function of the backward protocol is defined analogously. The work probability distributions introduced above (or equivalently their characteristic functions) allow for the formulation of quantum versions of the aforementioned fundamental fluctuation theorems [1,2]. As we discuss in what follows, the inference of the statistics of work and the study of such theorems is the focus of our experimental efforts.

Experimental setup and reconstruction of the work statistics.—Our experiment was carried out using liquid-state NMR spectroscopy of the ^1H and ^{13}C nuclear spins of a chloroform-molecule sample. This system can be regarded as a collection of identically prepared, noninteracting spin-1/2 pairs [17,18]. As discussed in Ref. [25], which also addresses the main sources of imperfections of the setup, this allows us to describe the state of the system with a single-spin density matrix. The rest of the molecule can be disregarded, providing mild environmental effects that, within the time span of our experiments, are inessential to our results. The ^{13}C nuclear spin is the driven system, while the ^1H one embodies an ancilla that will be instrumental to the reconstruction of $\chi_{F,B}(u)$. The protocol implemented in our experiment consists of a rapid change in a time-modulated radio frequency (rf) field resonant with the ^{13}C nuclear spin. Formally, this can be described by the time-dependent Hamiltonian (in the rotating frame at the frequency of the rf pulse [17] and for the forward protocol only) $\hat{\mathcal{H}}^F(t) = 2\pi\hbar\nu(t)(\hat{\sigma}_x^C \sin(\pi t/2\tau) + \hat{\sigma}_y^C \cos(\pi t/2\tau))$, where $\hat{\sigma}_{x,y,z}^C$ are the Pauli operators for the ^{13}C spin and $\nu(t) = \nu_1(1 - t/\tau) + \nu_2 t/\tau$ is a linear ramp (taking an overall time $\tau = 0.1$ ms) of the rf field frequency, from $\nu_1 = 2.5$ kHz to $\nu_2 = 1.0$ kHz, $t \in [0, \tau]$. The chosen value of τ is in the nonadiabatic regime. The reverse quench (realizing the B protocol) is described by $\hat{\mathcal{H}}^B(t) = -\hat{\mathcal{H}}^F(\tau - t)$.

In order to reconstruct the work distribution of both the forward and the backward protocols, we make use of the proposals put forward in Refs. [14,15], which rely on the Ramsey-like interferometric scheme illustrated in Fig. 1. Through a series of one- and two-body operations, which are presented in Ref. [25], this protocol maps the characteristic function of the work distribution for a system S (the ^{13}C nuclear spin, in our case) prepared in a pseudoequilibrium state ρ_S and undergoing the protocol $\hat{\mathcal{H}}^\alpha(0) \rightarrow \hat{\mathcal{H}}^\alpha(\tau)$ onto the transverse magnetization of an ancillary system A (the ^1H nuclear spin), initialized in $|0\rangle_A$.

In the first step of our experiment, we used spatial averaging methods to prepare the ^1H - ^{13}C nuclear-spin pair in the joint (factorized) state $\rho_{HC}^0 = |0\rangle\langle 0|_H \otimes \rho_C^0$, with ρ_C^0 being a diagonal state of the ^{13}C nuclear spin that can be interpreted as the equivalent equilibrium state $\rho_C^0 = e^{-\beta\hat{\mathcal{H}}^C(0)}/Z_0$ at the spin (pseudo)temperature T , which can be controlled and varied by suitably initializing the state of the system. We have introduced the logical states of the ^1H nuclear spin $\{|0\rangle, |1\rangle\}_H$, the inverse pseudotemperature $\beta = (k_B T)^{-1}$ (k_B is the Boltzmann constant), and the partition function $Z_0 = \text{Tr}[e^{-\beta\hat{\mathcal{H}}^C(0)}]$.

The structure of Eq. (1) suggests that one can reconstruct the characteristic function using simple single-spin operations and only two joint gates, each controlled by the ancilla state, and reading

$$\begin{aligned} \hat{G}_1 &\equiv |0\rangle\langle 0|_H \otimes e^{-iu\hat{\mathcal{H}}^\alpha(0)} + |1\rangle\langle 1|_H \otimes \hat{1}^C, \\ \hat{G}_2 &\equiv |0\rangle\langle 0|_H \otimes \hat{1}^C + |1\rangle\langle 1|_H \otimes e^{-iu\hat{\mathcal{H}}^\alpha(\tau)}. \end{aligned} \quad (2)$$

The full sequence of operations needed to reconstruct $\chi_F(u)$ is illustrated in Fig. 1, and the implementation of the operations based on our NMR device is discussed in detail, for both the forward and the backward protocols, in Ref. [25] (cf. Fig. S3). The completion of the protocol, which requires the exploitation of the natural coupling $\hat{\mathcal{H}}_J = 2\pi J \hat{\sigma}_z^H \hat{\sigma}_z^C$ (with J being the coupling rate) between the ^1H and ^{13}C nuclear spins (cf. Fig. 1) [17,18], encodes the characteristic function in Eq. (1) in the coherences of the final ^1H state as $\text{Re}[\chi(u)] = 2\langle \hat{\sigma}_x^H \rangle$ and $\text{Im}[\chi(u)] = 2\langle \hat{\sigma}_y^H \rangle$ [25]. This shows that the full form of $\chi(u)$ can be obtained from the x and y components of the ^1H transverse magnetization, a quantity that is straightforwardly accessed in our NMR setup.

The experiments were performed for states with different initial pseudotemperatures, sampling the characteristic

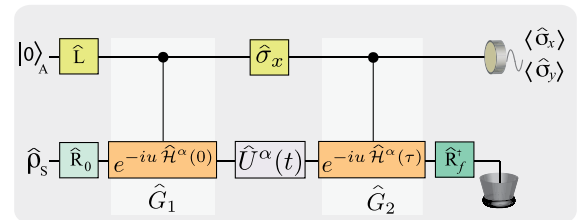


FIG. 1 (color online). Scheme for the interferometric reconstruction of $\chi_\alpha(u)$ ($\alpha = F, B$) [14,15]. We show the conditional joint gates given in Eq. (2) and the single-spin operations needed to complete the protocol. Here, ρ_S is a generic initial state of a driven system (the ^{13}C nuclear spin in our experiment), while $|0\rangle_A$ is an initial preparation for the ancilla (the ^1H nuclear spin). We have $\hat{R}_0 = \hat{K}$ ($\hat{R}_f = \hat{L}$) and $\hat{R}_f = \hat{L}$ ($\hat{R}_f = \hat{K}$) for the forward and backward protocols, respectively, with $\hat{K} = (\hat{\sigma}_y + \hat{\sigma}_z)/\sqrt{2}$ and $\hat{L} = (\hat{\sigma}_x + \hat{\sigma}_z)/\sqrt{2}$. Finally, $\hat{U}^\alpha(t)$ is the time evolution operator determined by the time-dependent Hamiltonian $\hat{\mathcal{H}}^\alpha(t)$ of the $\alpha = F, B$ protocol.

function at the 17.9 kHz rate. The interaction time s in Figs. 2(a) and 2(b) was varied through 360 equally spaced values. Each realization corresponds to an independent experiment with an average over our sample. The measured transverse magnetization (characteristic function), for the F protocol and two different values of T , is shown in Fig. 2. The amplitude of the oscillations of $\text{Re}[\chi_{F,B}(u)]$ (proportional to the x component of the magnetization) is approximately the same for all values of the pseudotemperature, while a clear decrease can be seen in the imaginary part (the y component of the magnetization) as the spin pseudotemperature increases. Similar considerations hold for the B protocol, whose experimental data are presented in [25].

The sample is processed in an environment at room temperature. However, the experimental data acquisition time (for each initial thermal state), which varies from 0.1 to 327 ms, is much shorter than the thermal relaxation time, which in NMR is associated with the spin-lattice relaxation occurring in a characteristic time \mathcal{T}_1^H . We have measured $(\mathcal{T}_1^H, \mathcal{T}_1^C) \approx (7.36, 10.55)$ s. Transverse relaxation at the characteristic times $(\mathcal{T}_2^H, \mathcal{T}_2^C) \approx (4.76, 0.33)$ s affects, in principle, the coherences of both the system and the ancilla state. Nevertheless, the characteristic dephasing time on ^1H is longer than the acquisition time. The situation for the ^{13}C spin is somewhat more complicated due to the shorter value of \mathcal{T}_2^C . However, the diagonal nature of the initial

equilibrium state of ^{13}C and the fact that the system-ancilla coupling commutes with the map responsible for the dephasing of its nuclear spin state reveal that, as long as we perform measurements only on the ancilla, the data acquired are unaffected by the system's transverse relaxation. As time increases, we observe an exponential decay of the magnetization, which is mainly due to transverse relaxation [cf. Figs. 2(a) and 2(b)]. This decay only limits how long we can track the characteristic function oscillations, which will ultimately bound the precision in the Fourier spectrum of the characteristic function.

The work distribution of the experimental protocol is obtained from the inverse Fourier transform of $\chi_{F,B}(u)$. For each value of T we observe well-defined peaks in the corresponding $P^F(W)$ [cf. Figs. 2(c) and 2(d)]. For the F protocol, the amplitudes of the two peaks in the $W/h < 0$ ($W/h > 0$) semiaxis in Figs. 2(c) and 2(d) are proportional to the excited-state (ground-state) population in the initial thermal state, which increases (decreases) with the pseudotemperature. For the backward protocol, this analysis still holds, but for a different location of the peaks. The experimental data are well fitted by a sum of four Lorentzian peaks centered at $(\pm 1.5 \pm 0.1)$ and $(\pm 3.5 \pm 0.1)$ kHz (solid lines), in agreement with the theoretical expectation [for both $\hat{\mathcal{H}}^{F(B)}(0)$ and $\hat{\mathcal{H}}^{F(B)}(\tau)$] that predict the peaks' location to be at $\pm(\nu_1 + \nu_2)$ and $\pm(\nu_1 - \nu_2)$. The peaks' amplitudes for the forward protocol are proportional to the probabilities $p_{1|0}^0 p_{0|1}^\tau$, $p_{1|1}^0 p_{1|1}^\tau$, $p_{0|0}^0 p_{0|0}^\tau$, $p_{0|1}^0 p_{1|0}^\tau$, respectively. The values of $p_{m|n}^\tau$ are set by the quench and do not depend on the value of the pseudotemperature. We have estimated $p_{1|1}^\tau \approx 0.71 \pm 0.01$, $p_{0|0}^\tau \approx 0.69 \pm 0.01$, and $p_{0|1}^\tau \approx p_{1|0}^\tau \approx 0.31 \pm 0.01$ for both protocols (cf. Ref. [25] for details on the error analysis). The relevance of this estimate is twofold. On one hand, these probabilities are key for the inference of the statistics of work and thus the study of out-of-equilibrium thermodynamics, as commented above. On the other hand, the set $\{p_{n|0}^0 p_{m|n}^\tau\}$ that we have experimentally gathered provides full information on the dynamics of our system, which is subjected to a fast quench generated by a time-dependent Hamiltonian. The corresponding Schrödinger equation does not admit an analytical solution, in general, a problem that is here bypassed experimentally through our technique. Finally, as illustrated in Ref. [25], we have checked that $p_{m|n}^{\tau,F} = p_{n|m}^{\tau,B}$, which is strong evidence of the validity of the microreversibility hypothesis [2].

Study of the fluctuation theorems.—The reconstructed work distributions for both protocols can now be used to study the fluctuation relations for the system at hand [2]. The protocols that we have implemented are genuinely quantum mechanical, being embodied by Hamiltonians consisting of noncommuting terms. As such, our experiment represents an important step towards the assessment of out-of-equilibrium dynamics in quantum systems subjected to a time-dependent protocol. We start by computing

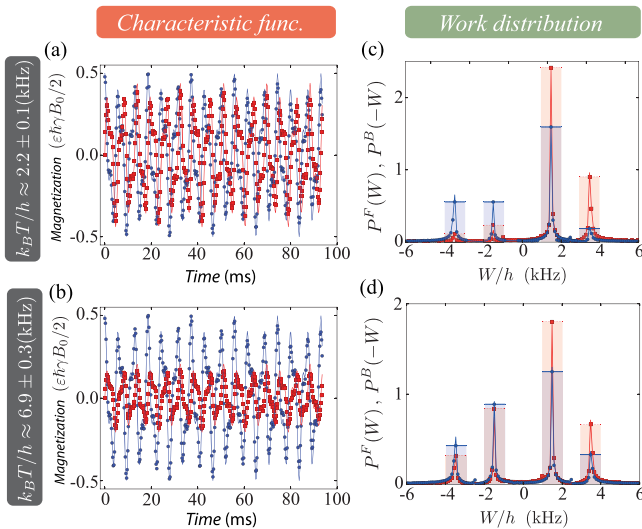


FIG. 2 (color online). (a) and (b): Experimental data for the x and y components of the ^1H transverse magnetization (blue circles and red squares, respectively) at two different values of the spin pseudotemperature, plotted against the time length $s = \mu\pi\nu_2/J$ of the controlled operations \hat{G}_1 and \hat{G}_2 . The solid lines show Fourier fittings, which are in agreement with the theoretical simulation of the protocol. The error bars are smaller than the size of the symbols and are not shown (cf. Ref. [25] for the definition of ϵ, γ and B_0). (c) and (d): The experimental points for the distribution corresponding to the forward (backward) protocol are shown as red squares (blue circles).

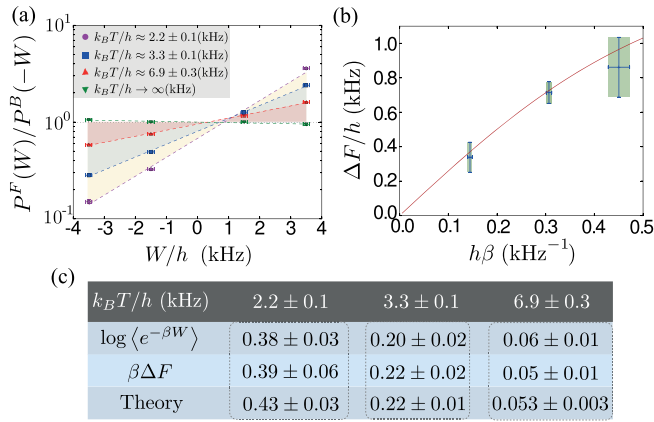


FIG. 3 (color online). (a): The ratio $P^F(W)/P^B(-W)$ is plotted in logarithmic scale for four values of the spin pseudotemperature. The data are determined using the values of $P^F(W)$ and $P^B(-W)$ at the peaks shown in Figs. 2(c) and 2(d) and in Ref [25]. (b): Mean values and uncertainties for ΔF and β obtained using a linear fit of the data corresponding to $T > 0$ in panel (a). The full red line represents the theoretical expectation, $\Delta F = (1/\beta) \ln(\cosh(\beta\nu_1)/\cosh(\beta\nu_2))$. (c): We report the experimental values of the left- and right-hand sides of the Jarzynski identity, measured for three choices of pseudotemperature, together with the respective uncertainties. The experimental results are compared to the theoretical predictions for $\ln(Z_\tau/Z_0)$.

the ratio $\eta(W) = P^F(W)/P^B(-W)$ and use it to verify the Tasaki-Crooks relation $\ln \eta(W) = \beta(W - \Delta F)$, where $\Delta F = -(1/\beta) \ln(Z_\tau/Z_0)$ [26,27]. We plot the left-hand side of this relation in Fig. 3(a), for four values of T . The trend followed by the data associated with each pseudotemperature is in very good agreement with the expected linear relation, thus confirming the predictions of the Crooks theorem. The point at which $\eta(W) = 1$ can be used to determine the value of ΔF experimentally. In Fig. 3(b), we show β and ΔF , obtained from a linear fitting according to the aforementioned strategy.

We can now investigate the Jarzynski identity at the quantum regime. We use the formulation of the equality $\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}$ [3], where the average is taken over $P^F(W)$ and is determined through the relation $\langle e^{-\beta W} \rangle = \chi(u = i\beta)$, obtained by analytical continuation of the characteristic function, based on the experimental data on the transverse magnetization of the ^1H nuclear spin. Second, we use the linear fit of the Tasaki-Crooks relation, thus combining forward and backward protocols. Finally, we have calculated the theoretical expectation value of the ratio Z_τ/Z_0 and have used the relation $Z_\tau/Z_0 = e^{-\beta \Delta F}$ [26,27] to provide a theoretical benchmark for the results obtained as described above. Figure 3(c) shows the mutual agreement among these approaches, which provide consistent results within the respective associated uncertainties.

Conclusions.—We have explored experimentally the statistics of work following a quasiunitary quantum

protocol in a spin-1/2 system using an ancilla-based interferometric approach adapted to NMR technology. Our experimental methodology has allowed us to address fluctuation relations at the quantum level and to go significantly beyond the current experimental state of the art, which was previously constrained to the classical domain. Despite addressing the finite-time thermodynamics of a simple single particle, our results might inspire further experimental endeavors towards the study of the thermodynamics of out-of-equilibrium quantum systems. In this respect, it will be particularly relevant to investigate possible interesting extensions to the quantum many-body context [27–29].

We thank M. Campisi, L. Céleri, J. Eisert, S. F. Huelga, K. Modi, G. M. Palma, M. B. Plenio, F. L. Semião, D. O. Soares-Pinto, and V. Vedral for the valuable discussions. We acknowledge financial support from CNPq, CAPES, FAPERJ, and FAPESP. This Letter was performed as part of the Brazilian National Institute of Science and Technology for Quantum Information (INCT-IQ). L. M. is supported by the EU through the Marie Curie Action. M. P. thanks the CNPq Ciência sem Fronteiras program for a grant under the Pesquisador Visitante Especial initiative (Grant No. 401265/2012-9), and the Alexander von Humboldt Stiftung. G. D. C. and M. P. acknowledge the United Kingdom EPSRC for a First Grant award, a Career Acceleration Fellowship, and grants awarded under the New Directions for Research Leaders initiative (Grants No. EP/L005026/1 and No. EP/G004579/1), the John Templeton Foundation (Grant No. 43467), and the EU Collaborative Project TherMiQ (Grant No. 618074). L. M. and M. P. thank the Universidade Federal do ABC and the CBPF labs for the kind hospitality during the early stages of this Letter.

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