

## Experimental Validation of Fully Quantum Fluctuation Theorems Using Dynamic Bayesian Networks

Kaonan Micadei,<sup>1</sup> John P. S. Peterson,<sup>2,3</sup> Alexandre M. Souza<sup>2</sup>,<sup>3</sup> Roberto S. Sarthour,<sup>2</sup> Ivan S. Oliveira,<sup>2</sup> Gabriel T. Landi,<sup>4</sup> Roberto M. Serra<sup>5,6</sup> and Eric Lutz<sup>1</sup>

<sup>1</sup>*Institute for Theoretical Physics I, University of Stuttgart, D-70550 Stuttgart, Germany*

<sup>2</sup>*Centro Brasileiro de Pesquisas Físicas, Rua Dr. Xavier Sigaud 150, 22290-180 Rio de Janeiro, Rio de Janeiro, Brazil*

<sup>3</sup>*Institute for Quantum Computing and Department of Physics and Astronomy, University of Waterloo, Waterloo N2L 3G1, Ontario, Canada*

<sup>4</sup>*Instituto de Física, Universidade de São Paulo, C.P. 66318, 05315-970 São Paulo, São Paulo, Brazil*

<sup>5</sup>*Centro de Ciências Naturais e Humanas, Universidade Federal do ABC, Avenida dos Estados 5001, 09210-580 Santo André, São Paulo, Brazil*

<sup>6</sup>*Department of Physics, University of York, York YO10 5DD, United Kingdom*



(Received 3 February 2021; revised 10 March 2021; accepted 5 October 2021; published 29 October 2021)

Fluctuation theorems are fundamental extensions of the second law of thermodynamics for small systems. Their general validity arbitrarily far from equilibrium makes them invaluable in nonequilibrium physics. So far, experimental studies of quantum fluctuation relations do not account for quantum correlations and quantum coherence, two essential quantum properties. We here apply a novel dynamic Bayesian network approach to experimentally test detailed and integral fully quantum fluctuation theorems for heat exchange between two quantum-correlated thermal spins-1/2 in a nuclear magnetic resonance setup. We concretely verify individual integral fluctuation relations for quantum correlations and quantum coherence, as well as for the sum of all quantum contributions. We further investigate the thermodynamic cost of creating correlations and coherence.

DOI: [10.1103/PhysRevLett.127.180603](https://doi.org/10.1103/PhysRevLett.127.180603)

A defining property of out-of-equilibrium systems is that they dissipate energy, leading to an irreversible increase of their entropy. The irreversible entropy production is thus a central quantity of nonequilibrium thermodynamics in the same way that entropy is a central quantity of equilibrium physics [1]. In small systems dominated by thermal or quantum fluctuations, the entropy production  $\Sigma$  is a stochastic variable [2,3]. Detailed fluctuation relations quantify the occurrence of negative entropy production events via the general equality  $P(\Sigma)/P(-\Sigma) = \exp(\Sigma)$  for the distribution  $P(\Sigma)$  [4–6]. Integral fluctuation theorems of the form  $\langle \exp(-\Sigma) \rangle = 1$  are obtained after integration over  $\Sigma$ . Both relations imply the second law of thermodynamics,  $\langle \Sigma \rangle \geq 0$ , and are therefore regarded as its far-from-equilibrium generalization. They are among only a few exact equalities known to be valid beyond the linear-response regime [4–6].

A standard procedure to investigate quantum fluctuation theorems, both theoretically and experimentally, is the two-projective-measurement approach [7,8]. In this framework, the energy change of a quantum system, and accordingly its stochastic entropy production, are determined by projectively measuring the energy at the beginning and at the end of a nonequilibrium process [9]. Equivalent schemes based on generalized measurements [10,11] and Ramsey-like interferometry [12,13] have additionally been developed.

These methods have been successfully implemented to test quantum fluctuation relations for mechanically driven [14–18] and thermally driven [19,20] systems, using a variety of experimental platforms, such as nuclear magnetic resonance, trapped ions, cold atoms, nitrogen-vacancy centers, and superconducting qubits [14–20]. The two-projective-measurement approach successfully captures the discrete quantum energy spectrum of the system, as well as its nonequilibrium quantum coherent dynamics between the two measurements [21]. However, due to its inherent projective nature, it fails to quantify the thermodynamic effects of quantum correlations and quantum coherence that may be present in initial and final states. Since these are two central quantum features [22,23] that affect the expression of the second law, such fluctuation theorems may be viewed as not fully quantum [24–31].

In this Letter, we report the first experimental study of fully quantum fluctuation relations for heat exchange between two initially quantum-correlated qubits prepared in local thermal states at different temperatures using nuclear magnetic resonance techniques [32,33]. To that end, we apply a general dynamic Bayesian network approach that fully accounts for quantum correlations and quantum coherence at all times [34]. After initiating thermal coupling between the two qubits, we analyze the statistics of the exchanged heat by tracking the evolution of

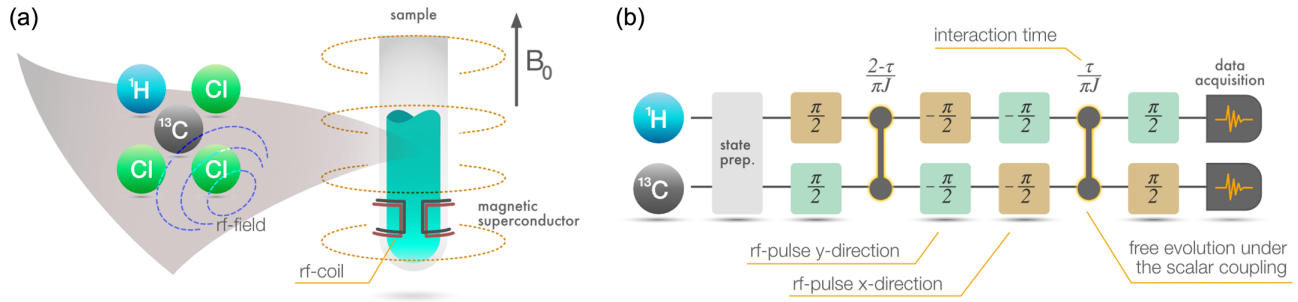


FIG. 1. Schematic representation of the experimental system. (a) Two qubits made of the nuclear spins-1/2 of  $^1\text{H}$  and  $^{13}\text{C}$  of  $^{13}\text{C}$ -labeled chloroform diluted in Acetone- $d_6$  are placed in a NMR magnetometer that produces a high intensity magnetic field ( $B_0$ ) in the longitudinal direction using a superconducting magnet. The liquid sample inside a 5 mm glass tube and maintained at room temperature (about 298 K) is placed at the center of the magnet within the radio frequency coil of the probe head. The magnet is composed of a superconducting coil (that produces a magnetic field strength of 11.75 T) placed in a thermally shielded vessel and immersed in liquid He (consequently, the temperature along the coil is lower than 10 K). Vacuum chambers and a liquid N chamber are part of the thermal isolation, and are used to maintain the temperature of the superconducting coil stable. (b) Experimental pulse sequence used to implement thermal interaction between the two qubits. The brown (green) square represents  $x$  ( $y$ ) rotations by the indicated angle. The black vertical lines indicate free evolution under the scalar coupling,  $\mathcal{H}_J^{\text{HC}} = (\pi\hbar/2)J\sigma_z^{\text{H}}\sigma_z^{\text{C}}$ , between the  $^1\text{H}$  and  $^{13}\text{C}$  nuclear spins with durations  $2\tau/\pi J$  and  $\tau/\pi J$ . We perform a total of 22 samplings of the interaction time  $\tau$  in the interval 0 to 2.32 ms.

the global two-qubit state with the help of quantum state tomography [32]. We reconstruct the heat distribution, at any time, during a forward nonequilibrium heat exchange process, as well as during its (time) reverse, with the help of the Bayesian network scheme. We use these distributions to examine the effect of correlations throughout the dynamics, and verify detailed fluctuation relations for heat, both with [34] and without [35] initial correlations between the qubits. We further demonstrate the validity of independent integral fluctuation relations for classical correlations (in the form of a stochastic classical mutual information [36]), quantum correlations (in the form of a stochastic quantum mutual information [36]) and quantum coherence (by means of a stochastic relative entropy of coherence [37]), as well as for the sum of all the quantum contributions. Finally, we employ these refined formulations of the second law to study the energetic cost of generating quantum correlations and quantum coherence [38–40].

*Experimental setup.*—We consider two qubits consisting of the nuclear spins-1/2 of  $^1\text{H}$  (qubit A) and  $^{13}\text{C}$  (qubit B) from a  $^{13}\text{C}$ -labeled chloroform sample diluted in Acetone- $d_6$ . The sample is placed in a superconducting magnet that produces a static magnetic field in  $z$  direction [Fig. 1(a)]. By combining transverse radio-frequency (rf) field with longitudinal field-gradient pulse sequences, we prepare an initial global state of the two spins-1/2 of the form [41],

$$\rho_{AB}^0 = \rho_A^0 \otimes \rho_B^0 + \chi_{AB}, \quad (1)$$

where  $\chi_{AB} = \alpha|01\rangle\langle 10| + \alpha^*|10\rangle\langle 01|$  is a correlation term that satisfies  $\text{Tr}_j[\chi_{AB}] = 0$ , ( $j = A, B$ ). As a result, the initial local states are thermal,  $\rho_j^0 = \exp(-\beta_j H_j)/Z_j$ , with inverse temperature  $\beta_j$  and partition function  $Z_j = \text{Tr}_j[\exp(-\beta_j H_j)]$ .

This condition ensures that the thermodynamic quantities of the local qubits are well defined, even though they are globally correlated. The local spin Hamiltonians are given in a double rotating frame with the nuclear spins resonance frequency by  $H_j = \hbar\nu_0(1 - \sigma_z)/2$ , where  $\sigma_z$  is the usual Pauli operator and  $\nu_0 = 1$  kHz is determined by the offset frequency in the spectrometer observation frame [41]. We denote their respective eigenstates by  $|0\rangle$  and  $|1\rangle$ . To guarantee the positivity of the density operator  $\rho_{AB}^0$ , the correlation strength  $\alpha$  should be bounded by  $|\alpha| \leq \exp[-\hbar\nu_0(\beta_A + \beta_B)/2]/(Z_A Z_B)$  [43]. The duration of the experiment (a few milliseconds) is much shorter than the decoherence time (a few seconds), so that the evolution of the global state can be considered to be unitary to an excellent degree of approximation [14]. The thermal interaction between the two qubits is further realized via the exchange Hamiltonian  $H_{\text{int}} = i(\pi\hbar/2)J(\sigma_A^+\sigma_B^- - \sigma_A^-\sigma_B^+)$ , where  $J = 215.1$  Hz. We implement the corresponding energy conserving evolution operator,  $U_t = \exp(-itH_{\text{int}}/\hbar)$  with  $[U_t, H_A + H_B] = 0$ , by combining free evolution under the scalar coupling between  $^1\text{H}$  and  $^{13}\text{C}$ , and rf-field rotations [Fig. 1(b)].

The experiment is performed in a Varian 500 MHz spectrometer equipped with a double-resonance probe head and a magnetic field-gradient coil. A 50 mg liquid sample of 99%  $^{13}\text{C}$ -labeled  $\text{CHCl}_3$  (Chloroform) is diluted in 0.7 ml of 99.9% deuterated Acetone- $d_6$  and flame sealed in a 5 mm Wildmad LabGlass tube. Intermolecular interactions are negligible due to the high level of dilution and the system may be considered as a set of identical pairs of spins-1/2. The superconducting magnet inside the magnetometer produces a static longitudinal magnetic field  $B_0 \approx 11.75$  T, whose direction is chosen as the positive  $z$  axis. The respective Larmor frequencies of  $^1\text{H}$  and  $^{13}\text{C}$  are about 500 and 125 MHz.

*Quantum fluctuation theorems.*—Because of the nonzero correlations between the two qubits, the global state is not diagonal in the energy representation. Global and local bases are therefore not mutually orthogonal, and the local bases, in which the exchanged heat variable is evaluated, do not contain the entire information about the composite system. As a consequence, the two-projective-measurement scheme cannot account for quantum correlations and quantum coherence. A powerful approach that solves this incompatibility is provided by dynamic Bayesian networks [44,45]. This formalism specifies the local dynamics conditioned on the global states, and hence preserves all the quantum properties of the system [34]. Introducing conditional path trajectories for the two quantum correlated systems and taking the average over the ensemble of all paths generated by the nonequilibrium heat exchange process leads to the integral quantum fluctuation theorem [34],

$$\langle \exp[-(Q_A \Delta\beta + I_0 - I_1 - \Sigma_A - \Sigma_B + \gamma)] \rangle = 1, \quad (2)$$

where  $Q_A$  is the energy change of spin  $A$  and  $\Delta\beta = \beta_A - \beta_B$  the difference of inverse temperatures. In addition,  $I_0$  ( $I_1$ ) is the stochastic quantum mutual information that describes initial (final) correlations between two subsystems, and  $\Sigma_j$  is the stochastic relative entropy characterizing the entropy produced in spin  $j$  (see Ref. [41] for full definitions, including how to define the heat  $Q_A$  directly from the Bayesian network). The last contribution  $\gamma$  originates from the random nature of the conditional dynamics, in analogy to the classical result of Ref. [46]. It vanishes on average, since the global dynamics is unitary and no extra energy is exchanged with an external reservoir [34]. Equation (2) shows that even in the absence of initial correlations,  $I_0 = 0$ , the two-projective-measurement scheme destroys correlations,  $I_1 \neq 0$ , created during the heat exchange.

Expression (2) generalizes the integral fluctuation theorem of Jarzynski and Wójcik,  $\langle \exp(Q_A \Delta\beta) \rangle = 1$ , obtained via the two-projective-measurement scheme [35]. In order to highlight its quantum nature, we write the stochastic quantum mutual informations,  $I_l = J_l + C_l$ , ( $l = 0, 1$ ), as a sum of the stochastic classical mutual information  $J_l$  and of the stochastic quantum relative entropy of coherence  $C_l$ , which is a proper measure of quantum coherence in a given basis [37]. The fluctuation relation (2) thus fully quantifies the presence of quantum correlations between the two subsystems and of quantum coherence in the global and local bases, at all times. Remarkably, contributions from both classical and quantum correlations,  $J_l$  and  $I_l$ , as well as from quantum coherence  $C_l$ , and the relative entropies  $\Sigma_j$ , which do not occur in the two-projective-measurement approach, separately obey an integral fluctuation theorem [34],

$$\langle e^{-I_l} \rangle = \langle e^{-J_l} \rangle = \langle e^{-C_l} \rangle = \langle e^{-\Sigma_j} \rangle = \langle e^{-\gamma} \rangle = 1. \quad (3)$$

A detailed quantum fluctuation relation for heat may be similarly derived for the ratio of the forward heat distribution,  $P_f(Q) = \sum_{\Gamma} \delta(Q - Q_A) \mathcal{P}(\Gamma)$ , and its reverse distribution,  $P_r(Q) = \sum_{\Gamma^*} \delta(Q - Q_A) \mathcal{P}(\Gamma^*)$  [34],

$$\frac{P_f(Q)}{P_r(-Q)} = \frac{\exp(Q\Delta\beta)}{\Psi(Q)}, \quad (4)$$

where the factor  $\Psi(Q)$  depends on the initial correlations between the two qubits, such that the Jarzynski-Wójcik result,  $\Psi_{\text{JW}}(Q) = 1$  is recovered in the absence of initial correlations between the two qubits [35].

*Experimental results.*—In order to analyze the influence of quantum correlations on the second law of thermodynamics, we prepare the two-qubit system in an initial state of the form (1) with inverse spin temperatures  $\beta_A^{-1} = 4.7(3)$  peV [ $\beta_A^{-1} = 4.3(2)$  peV] and  $\beta_B^{-1} = 3.3(3)$  peV [ $\beta_B^{-1} = 3.7(3)$  peV] for  $\alpha = 0$  ( $\alpha \neq 0$ ); the corresponding effective spin temperatures are of the order of 45 nK. We reconstruct the density matrix of the global state using state tomography [32] for a sequence of 22 values of time from  $t = 0$  to  $t = \tau = 2.32$  ms [47]. We determine from these global states the respective local qubit states and all the relevant thermodynamic quantities appearing in the quantum fluctuation relations (2)–(4). The thermal interaction  $H_{\text{int}}$  induces four possible transitions between the eigenstates of the two qubits. This leads to three stochastic values of the heat,  $Q = 0$  (twice) and  $Q = \pm Q_A$ , where  $Q_A = (E_{a_1} - E_{a_0})$  is the energy variation of spin  $A$ , with  $E_{a_0}$  ( $E_{a_1}$ ) the initial (final) energy eigenvalue of  $H_A$ . Contrary to the heat distribution  $P(Q)$ , the three values of  $Q$  do not depend on the (correlated or uncorrelated) initial conditions, and are therefore the same in both cases.

Figures 2(a)–2(d) show the corresponding forward heat distribution  $P_f(Q)$  as well as its (time) reverse  $P_r(Q)$  as a function of time, with [ $\alpha = 0.17(1) + i0.03(1)$ ] and without [ $\alpha = -0.00(1) + i0.0(1)$ ] initial correlations. We observe that the two heat distributions depend explicitly on time and that the forward and reverse distributions are identical in the absence of initial correlations. This follows from the fact that the global spin evolution is invariant under time reversal in that case [35]. Figure 2(e) further exhibits the detailed heat fluctuation theorem (4) for  $t = 1.88$  ms. Without initial correlations, we recover the Jarzynski-Wójcik relation which corresponds to  $\ln[P_f^{\text{JW}}(-Q)/P_r^{\text{JW}}(Q)] = Q\Delta\beta$  (green triangles). For  $\alpha \neq 0$ , the effect of quantum correlations is clearly visible (purple dots), modulating the  $Q$  dependence via the function  $\Psi(Q) \neq 1$ . Quantum correlations therefore modify both the heat distributions and the exponential dependence on the heat variable on the right-hand side of Eq. (4) through the (time-dependent) function  $\Psi(Q)$ .

The function  $\Psi(Q)$  may be determined directly by taking the ratio of the fluctuation relations with and without initial correlations, for fixed  $Q$  and  $\Delta\beta$ . Noting that  $\Psi_{\text{JW}}(Q) = 1$ ,

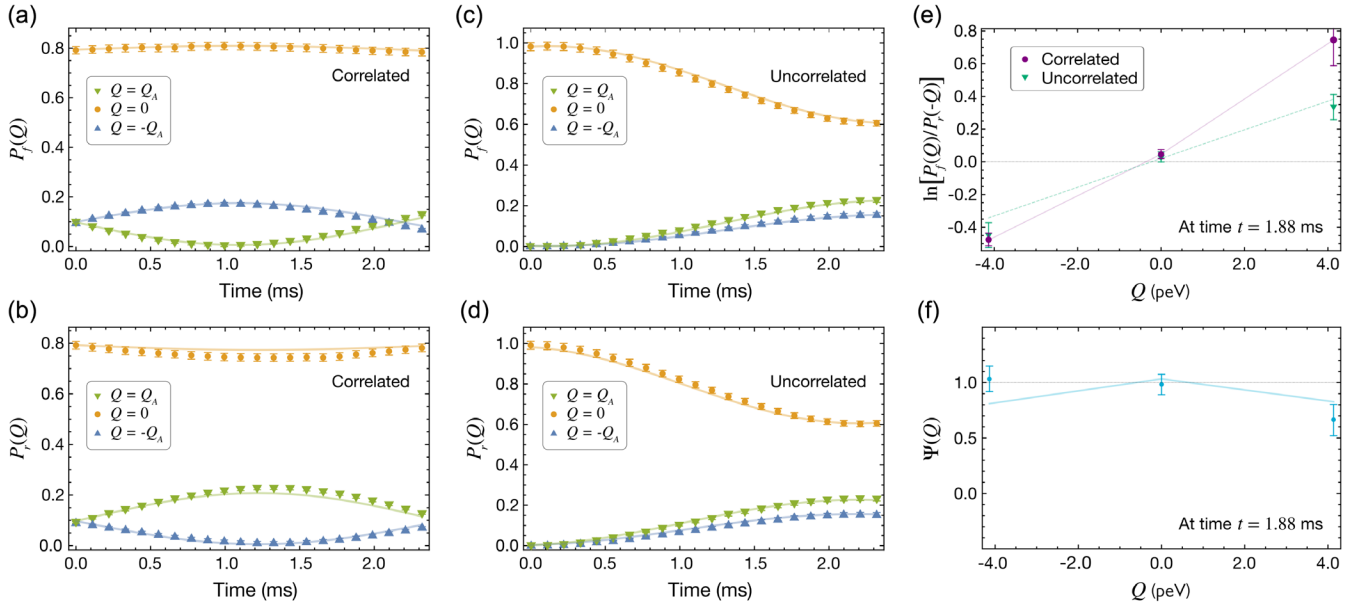


FIG. 2. Detailed quantum fluctuation theorem with and without initial quantum correlations. (a) Forward and (b) reverse heat distributions,  $P_f(Q)$  and  $P_r(-Q)$ , as a function of time in the presence of initial quantum correlations between the two qubits. (c) Forward and (d) reverse heat distributions in the absence of initial correlations between the qubits. Symbols represent data and solid lines are simulations [34]. Error bars are evaluated by a Monte Carlo sampling of the standard deviation. (e) Heat exchange fluctuation theorem, Eq. (4), for  $t = 1.88$  ms, with (purple dots) and without (green triangles) initial correlations. In the absence of initial correlations, the Jarzynski-Wójcik fluctuation relation,  $P_f^{\text{JW}}(Q)/P_r^{\text{JW}}(-Q) = \exp(Q\Delta\beta)$ , is obeyed, while the generalized theorem,  $P_f(Q)/P_r(-Q) = \exp(Q\Delta\beta)/\Psi(Q)$ , that fully accounts for quantum correlations and quantum coherence with  $\Psi(Q) \neq 1$  holds in their presence (solid and dashed lines are guides to the eye). (f) The function  $\Psi(Q)$  may be directly evaluated as the ratio  $\Psi(Q) = [P_f^{\text{JW}}(Q)P_r(-Q)]/[P_r^{\text{JW}}(-Q)P_f(Q)]$  (shown here for  $t = 1.88$  ms).

we find  $\Psi(Q) = [P_f^{\text{JW}}(Q)P_r(-Q)]/[P_r^{\text{JW}}(-Q)P_f(Q)]$ . This function is shown in Fig. 2(f) (symbols) together with theoretical simulations (line), for  $t = 1.88$  ms.

The experimental study of the integral quantum fluctuation relations Eqs. (2)–(3) is represented in Fig. 3. It reveals that not only the sum of all the contributions,  $\sigma = -Q_A\Delta\beta - I_0 + I_1 + \Sigma_A + \Sigma_B - \gamma$ , in the exponent of Eq. (2) satisfies a quantum fluctuation theorem, but that also individual contributions,  $J_l$ ,  $I_l$ ,  $C_l$ ,  $\Sigma_j$  ( $l = 0, 1$  and  $j = A, B$ ), and  $\gamma$ , separately obey such an integral relation. These results are verified at all times and are illustrated for  $t = 1.77$  ms in the figure. Such findings suggest that many versions of the second law of thermodynamics hold independently, both for classical and quantum correlations, as well as for quantum coherence.

As an application, we finally examine the important issue of the energetic cost of creating quantum correlations and quantum coherence [38–40], which has not been studied experimentally so far. Using the concavity of the exponential, the fluctuation relation (2) implies, in the absence of initial correlations ( $I_0 = 0$ ), that the rates of correlation and coherence generation are fundamentally limited by the rate of (dimensionless) energy change,  $\dot{I}_1 \leq \Delta\beta\dot{Q}_A$  and  $\dot{C}_1 \leq \Delta\beta\dot{Q}_A$ . These two inequalities, which bear a striking resemblance to the Clausius inequality

$\beta\dot{Q} \leq \dot{S}$  [1], are confirmed in Fig. 4. Remarkably, the upper bounds are saturated at short times,  $t \leq 0.5$  ms, indicating that the creation of quantum correlation and coherence is thermodynamically optimal in this regime.

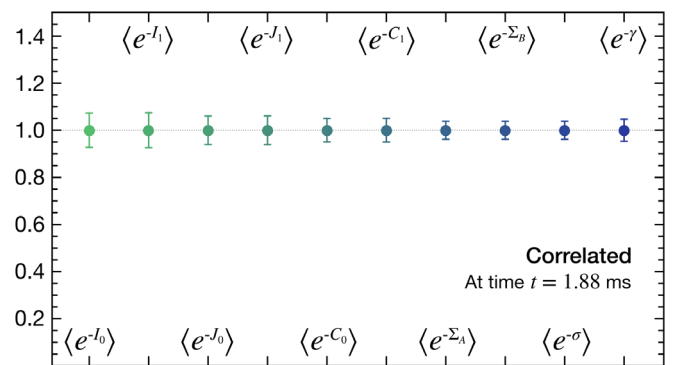


FIG. 3. Integral fluctuation theorems with initial correlations. The individual contributions from classical and quantum correlations,  $J_l$  and  $I_l$ , as well as from quantum coherence  $C_l$ , and the relative entropies  $\Sigma_j$ , for the two qubits ( $l = 0, 1$  and  $j = A, B$ ), and  $\gamma$ , separately verify the quantum integral fluctuation theorem (3). At the same time, the sum of all the quantum contributions  $\sigma = -Q_A\Delta\beta - I_0 + I_1 + \Sigma_A + \Sigma_B - \gamma$  obeys the integral fluctuation relation (2).



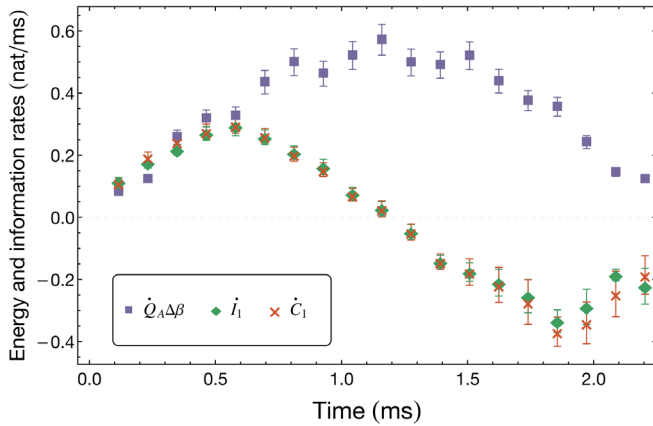


FIG. 4. Energetic cost of creating quantum correlations and coherence. In the absence of initial correlations,  $I_0 = 0$ , the rates of quantum correlation and coherence generation are upper bounded by the rate of (dimensionless) energy change,  $\dot{I}_1 \leq \Delta\beta\dot{Q}_A$  and  $\dot{C}_1 \leq \Delta\beta\dot{Q}_A$ . The derivative  $\dot{X}(t_k)$  at time  $t_k$  is evaluated as the central difference  $X(t_{k+1}) - X(t_{k-1})$ .

We further note that  $\dot{I}_1 \simeq \dot{C}_1$  throughout the process, showing that the rate of classical correlation creation is vanishingly small,  $\dot{J}_1 \simeq 0$ , in our experiment.

**Conclusions.**—We have employed an original dynamic Bayesian network approach to experimentally investigate the quantum thermodynamics of nonequilibrium heat exchange. In contrast to the two-projective-measurement method, this general scheme fully accounts, at all times, for off-diagonal matrix elements in the local energy representation of a system, induced by either quantum correlations or quantum coherence. It thus provides a powerful tool to study generic quantum aspects of thermodynamic processes. We have, in particular, verified for the first time fully quantum, detailed, and integral, fluctuation relations and used these improved formulations of the second law to examine the energetic cost of establishing correlations and coherence. In view of their generality, we expect these results to be useful for the study of the thermodynamic properties of small interacting quantum systems operating far from equilibrium.

We acknowledge financial support from CNPq, CAPES, FAPERJ (Grant No. 203.166/2017), FAPESP (Grant No. 2016/08721-7 and 2018/12813-0), the Ministry of Innovation, Science and Economic Development (Canada), the Government of Ontario, CIFAR, Mike and Ophelia Lazaridis, the São Paulo Research Foundation (Grants No. 2017/07973-5 and No. 2017/50304-7) and from the German Science Foundation (DFG) (Grant No. FOR 2724). R. M. S. gratefully acknowledges support from the Royal Society through the Newton Advanced Fellowship scheme (Grant No. NA140436) and the technical support from the Multiuser Experimental Facilities of Universidade Federal do ABC. This research was performed as part of the Brazilian National Institute of Science and Technology for Quantum Information (INCT-IQ).

- [1] G. Lebon, D. Jou, and J. Casas-Vásquez, *Understanding Non-Equilibrium Thermodynamics* (Springer, Berlin, 2008).
- [2] C. Bustamante, J. Liphardt, and F. Ritort, The nonequilibrium thermodynamics of small systems, *Phys. Today* **58**, No. 7, 43 (2005).
- [3] K. Sekimoto, *Stochastic Energetics* (Springer, Berlin, 2010).
- [4] U. Seifert, Stochastic thermodynamics, fluctuation theorems, and molecular machines, *Rep. Prog. Phys.* **75**, 126001 (2012).
- [5] C. Jarzynski, Equalities and inequalities: Irreversibility and the second law of thermodynamics at the nanoscale, *Annu. Rev. Condens. Matter Phys.* **2**, 329 (2011).
- [6] S. Ciliberto, R. Gomez-Solano, and A. Petrosyan, Fluctuations, linear response, and currents in out-of-equilibrium systems, *Annu. Rev. Condens. Matter Phys.* **4**, 235 (2013).
- [7] M. Esposito, U. Harbola, and S. Mukamel, Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems, *Rev. Mod. Phys.* **81**, 1665 (2009).
- [8] M. Campisi, P. Hänggi, and P. Talkner, Quantum fluctuation relations: Foundations and applications, *Rev. Mod. Phys.* **83**, 771 (2011).
- [9] P. Talkner, E. Lutz, and P. Hänggi, Fluctuation theorems: Work is not an observable, *Phys. Rev. E* **75**, 050102(R) (2007).
- [10] G. Huber, F. Schmidt-Kaler, S. Deffner, and E. Lutz, Employing Trapped Cold Ions to Verify the Quantum Jarzynski Equality, *Phys. Rev. Lett.* **101**, 070403 (2008).
- [11] A. J. Roncaglia, F. Cerisola, and J. P. Paz, Work Measurement as a Generalized Quantum Measurement, *Phys. Rev. Lett.* **113**, 250601 (2014).
- [12] L. Mazzola, G. De Chiara, and M. Paternostro, Measuring the Characteristic Function of the Work Distribution, *Phys. Rev. Lett.* **110**, 230602 (2013).
- [13] R. Dörner, S. R. Clark, L. Heaney, R. Fazio, J. Goold, and V. Vedral, Extracting Quantum Work Statistics and Fluctuation Theorems by Single-Qubit Interferometry, *Phys. Rev. Lett.* **110**, 230601 (2013).
- [14] T. B. Batalhao, A. M. Souza, L. Mazzola, R. Auccaise, R. S. Sarthour, I. S. Oliveira, J. Goold, G. De Chiara, M. Paternostro, and R. M. Serra, Experimental Reconstruction of Work Distribution and Study of Fluctuation Relations in a Closed Quantum System, *Phys. Rev. Lett.* **113**, 140601 (2014).
- [15] S. An, J. Zhang, M. Um, D. Lv, Y. Lu, J. Zhang, Z. Yin, H. T. Quan, and K. Kim, Experimental test of the quantum Jarzynski equality with a trapped-ion system, *Nat. Phys.* **11**, 193 (2015).
- [16] F. Cerisola, Y. Margalit, S. Machluf, A. J. Roncaglia, J. P. Paz, and R. Folman, Using a quantum work meter to test non-equilibrium fluctuation theorems, *Nat. Commun.* **8**, 1241 (2017).
- [17] A. Smith, Y. Lu, S. An, X. Zhang, J. N. Zhang, Z. Gong, H. T. Quan, C. Jarzynski, and K. Kim, Verification of the quantum nonequilibrium work relation in the presence of decoherence, *New J. Phys.* **20**, 013008 (2018).
- [18] Z. Zhang, T. Wang, L. Xiang, Z. Jia, P. Duan, W. Cai, Z. Zhan, Z. Zong, J. Wu, and L. Sun, Experimental demonstration of work fluctuations along a shortcut to adiabaticity with a superconducting Xmon qubit, *New J. Phys.* **20**, 085001 (2018).

- [19] S. Hernandez-Gomez, S. Gherardini, F. Poggiali, F. S. Cataliotti, A. Trombettoni, P. Cappellaro, and N. Fabbri, Experimental test of exchange fluctuation relations in an open quantum system, *Phys. Rev. Research* **2**, 023327 (2020).
- [20] S. Pal, T. S. Mahesh, and B. K. Agarwalla, Experimental verification of quantum heat exchange fluctuation relation, *Phys. Rev. A* **100**, 042119 (2019).
- [21] C. Jarzynski, H. T. Quan, and S. Rahav, Quantum-Classical Correspondence Principle for Work Distributions, *Phys. Rev. X* **5**, 031038 (2015).
- [22] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, Quantum entanglement, *Rev. Mod. Phys.* **81**, 865 (2009).
- [23] A. Streltsov, G. Adesso, and M. B. Plenio, Quantum coherence as a resource, *Rev. Mod. Phys.* **89**, 041003 (2017).
- [24] S. Jevtic, T. Rudolph, D. Jennings, Y. Hirono, S. Nakayama, and M. Muraio, Exchange fluctuation theorem for correlated quantum systems, *Phys. Rev. E* **92**, 042113 (2015).
- [25] Á. M. Alhambra, L. Masanes, J. Oppenheim, and C. Perry, Fluctuating Work: From Quantum Thermodynamical Identities to a Second Law Equality, *Phys. Rev. X* **6**, 041017 (2016).
- [26] J. Åberg, Fully Quantum Fluctuation Theorems, *Phys. Rev. X* **8**, 011019 (2018).
- [27] J. J. Park, S. W. Kim, and V. Vedral, Fluctuation theorem for arbitrary quantum bipartite systems, *Phys. Rev. E* **101**, 052128 (2020).
- [28] J. P. Santos, L. C. Céleri, G. T. Landi, and M. Paternostro, The role of quantum coherence in non-equilibrium entropy production, *npj Quantum Inf.* **5**, 23 (2019).
- [29] G. Francica, J. Goold, and F. Plastina, Role of coherence in the nonequilibrium thermodynamics of quantum systems, *Phys. Rev. E* **99**, 042105 (2019).
- [30] G. Manzano, J. M. Horowitz, and J. M. R. Parrondo, Quantum Fluctuation Theorems for Arbitrary Environments: Adiabatic and Nonadiabatic Entropy Production, *Phys. Rev. X* **8**, 031037 (2018).
- [31] H. Kwon and M. S. Kim, Fluctuation Theorems for a Quantum Channel, *Phys. Rev. X* **9**, 031029 (2019).
- [32] I. Oliveira, R. S. Sarthour, T. Bonagamba, E. Azevedo, and J. C. C. Freitas, *NMR Quantum Information Processing* (Elsevier, Amsterdam, 2007).
- [33] L. M. K. Vandersypen and I. L. Chuang, NMR techniques for quantum control and computation, *Rev. Mod. Phys.* **76**, 1037 (2005).
- [34] K. Micadei, G. T. Landi, and E. Lutz, Quantum Fluctuation Theorems beyond Two-Point Measurements, *Phys. Rev. Lett.* **124**, 090602 (2020).
- [35] C. Jarzynski and D. K. Wójcik, Classical and Quantum Fluctuation Theorems for Heat Exchange, *Phys. Rev. Lett.* **92**, 230602 (2004).
- [36] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England, 2000).
- [37] T. Baumgratz, M. Cramer, and M. B. Plenio, Quantifying Coherence, *Phys. Rev. Lett.* **113**, 140401 (2014).
- [38] M. Huber, M. Perarnau-Llobet, K. V. Hovhannisyán, P. Skrzypczyk, C. Klöckl, N. Brunner, and A. Acín, Thermodynamic cost of creating correlations, *New J. Phys.* **17**, 065008 (2015).
- [39] D. E. Bruschi, M. Perarnau-Llobet, N. Friis, K. V. Hovhannisyán, and M. Huber, The thermodynamics of creating correlations: Limitations and optimal protocols, *Phys. Rev. E* **91**, 032118 (2015).
- [40] A. Misra, U. Singh, S. Bhattacharya, and A. K. Pati, Energy cost of creating quantum coherence, *Phys. Rev. A* **93**, 052335 (2016).
- [41] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.127.180603> for details about the experimental setup, the preparation of the initial state, the tomographic reconstruction of the state, and the theoretical Bayesian network framework, which also includes Ref. [42].
- [42] T. B. Batalhão, A. M. Souza, R. S. Sarthour, I. S. Oliveira, M. Paternostro, E. Lutz, and R. M. Serra, Irreversibility and the Arrow of Time in a Quenched Quantum System, *Phys. Rev. Lett.* **115**, 190601 (2015).
- [43] K. Micadei, J. P. S. Peterson, A. M. Souza, R. S. Sarthour, I. S. Oliveira, G. T. Landi, T. B. Batalhão, R. M. Serra, and E. Lutz, Reversing the direction of heat flow using quantum correlations, *Nat. Commun.* **10**, 2456 (2019).
- [44] R. E. Neapolitan, *Learning Bayesian Networks* (Prentice Hall, Upper Saddle River, 2003).
- [45] A. Darwiche, *Modeling and Reasoning with Bayesian Networks* (Cambridge University Press, Cambridge, England, 2009).
- [46] U. Seifert, Entropy Production along a Stochastic Trajectory and an Integral Fluctuation Theorem, *Phys. Rev. Lett.* **95**, 040602 (2005).
- [47] A less demanding protocol, based on multiple copies of a system and not on full process tomography, has recently been put forward [48]. However, this new scheme has not been experimentally tested yet.
- [48] K. Micadei, G. T. Landi, and E. Lutz, Extracting Bayesian networks from multiple copies of a quantum system, arXiv:2103.14570.