Experimental rectification of entropy production by a Maxwell’s Demon in a quantum system

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Maxwell’s demon explores the role of information in physical processes. Employing information about microscopic degrees of freedom, this “intelligent observer” is capable of compensating entropy production (or extracting work), apparently challenging the second law of thermodynamics. In a modern standpoint, it is regarded as a feedback control mechanism and the limits of thermodynamics are recast incorporating information-to-energy conversion. We derive a trade-off relation between information-theoretic quantities empowering the design of an efficient Maxwell’s demon in a quantum system. The demon is experimentally implemented as a spin-1/2 quantum memory that acquires information, and employs it to control the dynamics of another spin-1/2 system, through a natural interaction. Noise and imperfections in this protocol are investigated by the assessment of its effectiveness. This realization provides experimental evidence that the irreversibility on a nonequilibrium dynamics can be mitigated by assessing microscopic information and applying a feed-forward strategy at the quantum scale.

Connections between thermodynamics and information theory have been producing important insights and useful applications in the past few years, which has turned out to be a dynamic field [1–4]. Its genesis traces back to the famous Maxwell’s demon gedanken experiment [5–9]. In 1867, Maxwell conceived a “neat fingered being”, which has the ability to gather information about the microscopic state of a gas and use this information to transfer fast particles to a hot medium and slow particles to a cold one, engendering an apparent conflict with the second law of thermodynamics. Several approaches and developments concerning this conundrum had been put forward [5–9], but only after more than a century, in 1982, Bennett [10] realized that the apparent contradiction with the second law could be puzzled out by considering the Landauer’s erasure principle [11–14].

Theoretical endeavours to incorporate information into thermodynamics acquire a pragmatic applicability within the recent technological progress, where information just started to be manipulated at the micro- and nanoscale. A modern framework for these endeavors has been provided by explicitly taking into account the change, introduced in the statistical description of the system, due to the assessment of its microscopic information [15]. This outlines an illuminating paradigm for the Maxwell’s demon, where the information-to-energy conversion is governed by fluctuation theorems, which hold for small systems arbitrarily far from equilibrium [16–20]. Generalizations of the second law in the presence of feedback control can be obtained from this framework, establishing bounds for information-based work extraction [21]. Notwithstanding its fundamental relevance, these relations do not provide a clear recipe for building a demon in a laboratory setting. Owing to the challenges associated with a high precision microscopic control, there are only a handful of very recent experiments addressing the information-to-energy conversion at small scales, using Brownian particles [22, 23], single electrons [24–26], and laser pulses [27] regarding the classical scenario, where quantum coherence effects are absent. In the quantum context, there are only two experimental attempts related with information-to-energy conversion. The heat dissipated during a global system-reservoir unitary interaction was investigated in a spin system [28] and single photons in nonthermal states were employed to build a thermodynamics-inspired separability criterion [29].

Here, we contribute to the aforementioned efforts deriving an equality concerning the information-to-energy conversion for a quantum nonunitary feedback process. Such relation involves a trade-off between information-theoretic quantities that provides a recipe to design and implement an efficient Maxwell’s demon in a quantum system where coherence is present. Supported by this trade-off relation and employing Nuclear Magnetic Resonance (NMR) spectroscopy [30–32], we set up an experimental coherent implementation of a measurement-based feedback. Furthermore, we quantify experimentally the effectiveness of this Maxwell’s demon to rectify entropy production, due to quantum fluctuations [33, 34], in a nonequilibrium dynamics.

Theoretical description. Consider the scenario illustrated in Fig. 1. The working system is a small quantum system, initially in the equilibrium state ρ0 at inverse temperature β = (kBT)−1, with kB being the Boltzmann constant. Later on the Maxwell’s demon will also be materialized through a microscopic quantum memory. Sup-


The system starts in the equilibrium state $\rho^{eq}_{0}$ and it is unitarily driven ($\mathcal{U}$) to a nonequilibrium state. Then the demon makes a projective measurement, $M_{l}$, yielding the outcome $l$ with probability $p(l)$. The feedback operation $\mathcal{F}(k)$ is applied with error probability $p(k|l)$. The environment temperature is kept fixed and the whole operation is much faster than the system decoherence time.

pose that the working system is driven away from equilibrium by a fast unitary time-dependent process, $\mathcal{U}$, up to time $\tau_{1}$ (driving the system Hamiltonian from $\mathcal{H}_{0}$ to $\mathcal{H}_{\tau_{1}}$). The purpose of the control mechanism is to rectify the quantum fluctuations introduced by this nonequilibrium dynamics. To this end, the demon acquires information about the system’s state through a complete projective measurement, $\{M_{l}\}$, yielding the outcome $l$ with probability $p(l) = \text{tr}[M_{l}\rho^{eq}_{0}\mathcal{U}^{\dagger}]$. Based on the outcome of this measurement a controlled evolution will be applied. It will be described by unital quantum operations $\mathcal{F}(k)$ ($\mathcal{F}(k)(l) = 1$ for every $k$), which may include a drive on the system’s Hamiltonian from $\mathcal{H}_{\tau_{1}}$ to $\mathcal{H}_{\tau_{2}}$, along the time interval $\tau_{2} - \tau_{1}$ [35]. Furthermore, we consider the possibility of error in the control mechanism, assuming a conditional probability $p(k|l)$ of implementing the feedback process $k$ (associated with the outcome $k$) when $l$ is the actual observed measurement outcome. By a suitable choice of the operations $\{\mathcal{F}(k)\}$, the feedback control mechanism can balance out the entropy production due to the nonequilibrium drive $\mathcal{U}$. A similar protocol might also be employed to information-based work extraction.

Following the scenario presented above, an integral fluctuation relation can be derived [36, 37] as (see Supplemental Material for details):

$$\left\langle e^{-\beta(W - \Delta F(k)) - I(k,l)} \right\rangle = 1,$$

where $W$ is the stochastic work done on the system, $\Delta F(k) = -\beta^{-1} \ln Z_{\tau_{2}}^{k}/Z_{0}$ [with $Z_{k}^{(k)} = \text{tr}(e^{-\beta H_{\tau_{2}}^{(k)}})$ and $Z_{0} = \text{tr}(e^{-\beta H_{0}})$], is the free energy variation for the $k$-th feedback process, $I(k,l) = \ln p(k|l)/p(l)$ is the unaveraged mutual information between the working system and the control mechanism employed [$p(k) = \sum_{l}p(k|l)p(l)$ is the marginal probability distribution of the controlled operation]. The average is computed according to a work distribution probability $P(W)$ that depends on both the measurement and the feedback processes. Equation (1) has the same structure of Sagawa and Ueda’s classical relation [38, 39]. It is also the generalization of the Tasaki quantum identity obtained for unitary control [40], which was previously discussed in Refs. [36, 37]. Jensen’s inequality for convex functions can be used to obtain a lower bound for the mean nonequilibrium entropy production

$$\langle \Sigma \rangle = \beta \left\langle W - \Delta F(k) \right\rangle \geq - \left\langle I(k,l) \right\rangle .$$

If the feedback control is absent, Eq. (2) reduces to the standard Clausius inequality, $\langle \Sigma \rangle \geq 0$. On the other hand, Eq. (2) generalizes the second law, elucidating that the correlations between the system and the demon, expressed by the mutual information $\left\langle I(k,l) \right\rangle$, may be employed to decrease the entropy production beyond the conventional thermodynamic limit. Besides its material importance to the understanding of the underneath gear of the Maxwell’s demon, Eq. (2) does not shed light on how to design an efficient feedback-control protocol. Notice that the right-hand side (rhs) of Eq. (2) is unrelated to the specific form of the feedback operations $\{\mathcal{F}(k)\}$, it is only associated with the feedback error probability $p(k|l)$ and the marginal distribution $p(k)$. Therefore, performance analysis of different types of feedback operations is beyond the scope of the bound in Eq. (2).

We bridge such a gap by deriving an equality for entropy production in the presence of feedback control with experimental relevance for the effective design of a Maxwell’s demon, expressed as (see Supplemental Material):

$$\langle \Sigma \rangle = -I_{\text{gain}} + \left\langle S_{KL} \left( \rho_{\tau_{2}}^{(k,l)} || \rho_{\tau_{2}}^{(k,eq)} \right) \right\rangle + \left\langle \Delta S^{(k,l)} \right\rangle_{\mathcal{F}},$$

with only information-theoretic quantities on the rhs. The information gain $I_{\text{gain}} = S(\rho_{\tau_{1}}) - \sum_{l}p(l)S(\rho_{\tau_{1}}^{(l)})$ quantifies the average information that the demon obtains reading the outcomes of the measurement $\mathcal{M}$ [41–47], with $\rho_{\tau_{1}} = \mathcal{U} \rho^{eq}_{0} \mathcal{U}^{\dagger}$ being the system’s state before the measurement; $\rho_{\tau_{1}}^{(l)}$ the l-th post-measurement state which occurs with probability $p(l)$, and $S(\rho)$ the von Neumann entropy. It is always non-negative for projective measurements [41–44] and it can be interpreted as the reciprocal to the quantity of disturbance impinged on the quantum system due to the measurement operation [45] (see also the Supplemental Material). The Kullback-Leibler (KL) relative entropy, $S_{KL} \left( \rho_{\tau_{2}}^{(k,l)} || \rho_{\tau_{2}}^{(k,eq)} \right) = \text{tr} \left[ \rho_{\tau_{2}}^{(k,l)} \left( \ln \rho_{\tau_{2}}^{(k,l)} - \ln \rho_{\tau_{2}}^{(k,eq)} \right) \right]$, expresses the information divergence between the resulting state of the feedback-controlled process, $\rho_{\tau_{2}}^{(k,l)}$, and the equilibrium state for the final Hamiltonian $\mathcal{H}_{\tau_{2}}^{(k)}$ in the $k$-th feedback process, $\rho_{\tau_{2}}^{(k,eq)} = e^{-\beta \mathcal{H}_{\tau_{2}}^{(k)}}/Z_{\tau_{2}}^{(k)}$. The

Figure 1. Illustration of a Maxwell’s demon operation. The system starts in the equilibrium state $\rho^{eq}_{0}$ and it is unitarily driven ($\mathcal{U}$) to a nonequilibrium state. Then the demon makes a projective measurement, $M_{l}$, yielding the outcome $l$ with probability $p(l)$. The feedback operation $\mathcal{F}(k)$ is applied with error probability $p(k|l)$. The environment temperature is kept fixed and the whole operation is much faster than the system decoherence time.
last term, \( \langle \Delta S^{(k,l)} \rangle_F = \langle S \left( \rho_{1z}^{(k,l)} \right) - S \left( \rho_{1z}^{(l)} \right) \rangle_F \), is the averaged change in von Neumann entropy due to the quantum operation \( F^{(k)} \).

The nonequilibrium entropy production in Eq. (3) is negative iff

\[
\mathcal{I}_{\text{gain}} > \left< S_{KL} \left( \rho_{1z}^{(k,l)} \left| \left| \rho_{1z}^{(k,eq)} \right| \right| \right) \right> + \left< \Delta S^{(k,l)} \right>_F. \quad (4)
\]

This provides a necessary and sufficient condition to implement an effective Maxwell’s demon for the nonunitary protocol considered here. Equation (3) also encompasses the bound \( (\Sigma) \geq -\mathcal{I}_{\text{gain}} \), which is similar to the bounds previously obtained in Refs. [48, 49] considering a different context. In the literature concerning the thermodynamics of information, feedback processes are often regarded as unitary. In this case the last term of the rhs of Eq. (3) does not contribute. Since the postmeasurement state \( \rho_{1z}^{(l)} \) is pure, the average KL relative entropy, \( \left< S_{KL} \left( \rho_{1z}^{(k,l)} \left| \left| \rho_{1z}^{(k,eq)} \right| \right| \right) \right> \), will never be zero for a unitary feedback implemented upon projective measurements (at finite temperature). In a different manner, a nonunitary feedback process can be designed to cancel the term \( \left< S_{KL} \left( \rho_{1z}^{(k,l)} \left| \left| \rho_{1z}^{(k,eq)} \right| \right| \right) \right> \), but in this case the variation of the von Neumann entropy \( \left< \Delta S^{(k,l)} \right>_F \), due to a nonunitary operation, is not null. Along these lines the trade-off concerning these quantities in Eqs. (3) and (4) empower the effective design of a Maxwell’s demon through the performance assessment of different strategies for the controlled operations \( F^{(k)} \).

**Experimental implementation.** We employed a \(^{13}\text{C}\)-labeled CHCl\(_3\) liquid sample and a 500 MHz Varian NMR spectrometer to implement and characterize the aforementioned entropy rectification protocol. The spin 1/2 of the \(^{13}\text{C}\) nucleus is the working system whereas the \(^1\text{H}\) nuclear spin plays the role of a quantum memory for the Maxwell’s demon. Chlorine isotopes’ nuclei can be disregarded providing only mild environmental effects due to the fast relaxation of its energy levels. Details on the experimental setup are provided in the Supplemental Material. Using spatial average techniques the joint initial state, equivalent to \( |0\rangle_H |0\rangle_0^{\text{eq,C}} \), is prepared, where the \(^{13}\text{C}\) is in an equilibrium state of the initial Hamiltonian, \( \mathcal{H}_0^{\text{C}} = \frac{\hbar}{2} \omega_0 \sigma_z^{\text{C}} \) (with \( \frac{\hbar}{2} \omega_0 = 2 \text{ kHz} \)), \( \sigma_{x,y,z} \) being the Pauli matrices, \( |0\rangle_{H,C} \) and \( |1\rangle_{H,C} \) representing the excited and ground state of \( \sigma_z^{\text{H,C}} \), respectively. We consider an initial driving protocol as a sudden quench process, described by a quick change in the Carbon Hamiltonian from \( \mathcal{H}_0^{\text{C}} \) to \( \mathcal{H}_1^{\text{C}} = \frac{\hbar}{2} \omega_1 \sigma_z^{\text{C}} \) (with \( \frac{\hbar}{2} \omega_1 = 3 \text{ kHz} \)). The idea is to change the Hamiltonian so quickly that the state of the system remains unchanged. This state will suddenly become far from equilibrium even including coherence in the energy basis of \( \mathcal{H}_1^{\text{C}} \). The quantum fluctuations, work distribution, and the entropy production in this highly non-adiabatic transformation can be experimentally characterized, in an NMR setting, according the approach presented in Refs. [33, 34]. In the present experiment, this sudden quench is implemented effectively by a short transversal radio-frequency (rf) pulse resonant with the \(^{13}\text{C}\) nuclear spin (with time duration about 9 \( \mu\text{s} \)) represented by the operation \( \mathcal{U} \) (as in Fig. 2(a)).

The feedback mechanism employed is sketched in Fig. 2(a), where the whole feedback operation is much faster than the typical decoherence times, which are on the order of seconds (see Supplemental Material). After the sudden quench \( (\mathcal{H}) \), information is acquired by the demon via the natural \( J \) coupling between \(^{13}\text{C}\) and \(^1\text{H}\) nuclei, \( \frac{\hbar}{2} \pi J \sigma_1^\text{H} \sigma_z^{\text{C}} \) (with \( J = 215.15 \text{ Hz} \)), under a free evolution lasting for about 6.97 ms (equivalent to a CNOT gate). An effective nonselective projective measurement in the energy basis of \( \mathcal{H}_1^{\text{C}} \) is accomplished with an additional longitudinal field gradient, \( \xi_1 \) (applied during 3 ms). It introduces a full dephasing on the \( z \)-component of the memory state. This free evolution followed by dehasing correlates the state of the
working system $^{13}\text{C}$ with the demon’s memory $^{1}\text{H}$ leading to a joint “postmeasurement” state equivalent to $|0\rangle_{H}|0\rangle_{M}M_{0}+|1\rangle_{H}|1\rangle_{M}M_{1}$, where $M_{0}$ and $M_{1}$ are the eigenbasis projectors for $\mathcal{H}_{C}$, with experimentally probed outcome probabilities, $p(l) = 50.0 \pm 0.4\%$ for $l = 0, 1$, as expected for the sudden quench implemented.

Quantum process tomography (QPT) [50] is applied to verify how effective is the demon’s nonselective measurement, with the results displayed Fig. 2(b). The experimentally implemented measurement is very close to the ideal one. In order to investigate the robustness of the feedback process against a control mismatch, we also introduce, in the protocol of Fig. 2(a), a rotation $\mathcal{R}_{C}$ along the $x$-direction on the $^{1}\text{H}$ spin, in such a way that the feedback error probability, $p(0|1) = p(1|0) = \sin^{2}\left(\frac{\phi}{2}\right)$, is changed varying the mismatch angle $\phi$. Figure 2(c) displays the trace distance between the experimental and ideal quantum processes for the demon operation as a function of such an error.

Entropy rectification is achieved by a controlled evolution of the $^{13}\text{C}$ nuclear spin guided by the demon’s memory (encoded in the $^{1}\text{H}$ nuclear spin state). Such conditional evolution is implemented by the operations $\mathcal{T}_{1}$ and $\mathcal{T}_{2}$ represented in Fig. 2(a) and $\mathcal{T}_{2} \mathcal{Y}_{1} = |\phi_{0}\rangle_{H}|\phi_{0}\rangle_{V_{x}}^{(0)} + |\phi_{1}\rangle_{H}|\phi_{1}\rangle_{V_{z}^{(1)}}$, where the mismatched control basis are given by $|\phi_{0}\rangle_{H} = \cos\left(\frac{\phi}{2}\right)|0\rangle_{H} - i\sin\left(\frac{\phi}{2}\right)|1\rangle_{H}$ and $|\phi_{1}\rangle_{H}$ its orthogonal complement; $V_{x}^{(0)} = e^{-i\pi\sigma_{y}^{C}/4}/e^{-i\pi\sigma_{y}^{C}/2}$ and $V_{z}^{(1)} = V_{x}^{(0)}\sigma_{z}^{C}$ are the feedback operations applied on the Carbon nucleus, with $\gamma = 2\arccos\left(1 - e^{-\beta\hbar\omega_{1}}\right)^{-1/2}$. Both controlled operations are put into action by a free evolution under the natural $J$ coupling ($\frac{1}{2} \pi J_{hh}\sigma_{x}^{H}\sigma_{x}^{C}$) combined with individual rotations driven by rf-fields resonant with both Larmor frequencies of $^{13}\text{C}$ and $^{1}\text{H}$ nuclei. We have chosen feedback operations where the system Hamiltonian is not driven, in this case $\mathcal{H}_{2}^{(0)} = \mathcal{H}_{2}^{(1)} = \mathcal{H}_{C} = \mathcal{H}_{C}^{\perp}$. The concluding step for implementing the controlled operations \{$\mathcal{F}(k,l)$\}, is a full dephasing in the eigenbasis of $\mathcal{H}_{2}^{\perp}$. It is supplied by a second longitudinal field gradient, $\xi_{2}$, and local rotations of the Carbon nuclear spin in order to set the dephasing basis.

Performing quantum state tomography (QST) [31] along the experimental implementation of the demon protocol, we can obtain all the information-theoretic quantities in rhs of Eq. (3) (for details see Fig. S2 and the Data Acquisition section in the Supplemental Material). Figure 3(a) displays the entropy production in the feedback controlled operation implemented in our experiment. We achieved negative values showing the realization of entropy rectification, whose effectiveness worsens as the basis mismatch increases. In Figs. 3(b) and 3(c), we note that the bounds based on mutual information, as in Eq. (2), and information gain are not tight in a quantum scenario, as also anticipated by Eq. (3). For the present protocol, it is possible to show that $\langle I^{(k,l)} \rangle \geq I_{\text{gain}}$ (see Supplemental Material). Despite the 4.5% residual error in the trace distance for the zero mismatch case [Fig. 2(e)], the mutual information (between the system
and feedback mechanism) experimentally achieved is very close to its limit, \( \langle I^{(k,l)} \rangle = - \sum p (l) \ln p (l) = \ln 2 \) nats (natural unit of information), as can be observed in Fig. 3(b). As discussed previously the information gain is related to how the system correlates with the memory; hence, it is independent of the control mismatch, which is corroborated by the experimental data in Fig. 3(c).

The \( k \)-th feedback control operation is designed ideally to map the carbon spin into the equilibrium state \( \rho^{(eq)}_{2} \) of the final Hamiltonian \( H^\tau_{F} \) (at inverse temperature \( \beta \)) irrespective of the previous nonequilibrium state \( \rho_{\tau_{1}} \) (produced by the sudden quench). Our aim is to cancel the KL relative entropy, \( S_{KL} (\rho^{(k,l)}_{2} || \rho^{(k,eq)}_{2}) \), which is successfully achieved for the zero basis mismatch, as can be observed in Fig. 3(d). On other hand the full dephasing, in the nonunitary feedback, introduces a finite von Neumann entropy variation \( (\Delta S^{(k,l)})_{P} \), see Figs. 3(d) and 3(e). This variation has no energy cost for the demon, since it is a unital process that does not change the working system mean energy. In the framework of the resource theory of quantum thermodynamics, the full dephasing is regarded as a free operation [51, 52]. When the control mismatch is increased the final state deviates from the thermal state of \( H^\tau_{F} \) and consequently the KL relative entropy also increases as shown in Fig. 3(e).

**Discussion.** Employing an information-to-energy trade-off relation, we designed an entropy rectification protocol based on a Maxwell’s demon. This protocol has been experimentally carried out by a coherent implementation of a measurement-based feedback control on a quantum spin-1/2 system. The demon’s memory is a microscopic quantum ancillary system that acquires information through a natural coupling with the working system. Due to the quantum coherence present in our experiment, we have to execute two dephasing operations in order to perform the Maxwell’s demon. The first dephasing operation is employed to produce a nonselective measurement, whereas the second is essential to accomplish entropy rectification, canceling the \( S_{KL} (\rho^{(k,l)}_{2} || \rho^{(k,eq)}_{2}) \) term in the trade-off relation (3). The present experiment elucidates the role played by different information quantities in the quantum version of the Maxwell’s demon. It also provides evidence that the irreversibility on a quantum nonequilibrium dynamics can be mitigated by assessing microscopic information and applying a feedback strategy. The approach developed here can be applied to general processes regarding information-to-energy conversion, as for instance, information-based work extraction.

A future experimental challenge would be the investigation of feedback protocols based on generalized quantum measurements and the bounds associated with such a scheme. The analysis and the optimization of the energetic cost for information manipulation by the Maxwell’s demon, in the quantum scenario, is also an important topic that deserves further attention. From a broad perspective, understanding the trade-off between information and entropy production at the quantum scale might be important to develop applications of quantum technologies with high efficiency.

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**SUPPLEMENTAL MATERIAL**

This supplemental material provides additional discussions and further (theoretical and experimental) details.

**Work probability distribution.** In the feedback control protocol depicted in Fig. S1, the mean work done on the system is given by the averaged work of each possible history of the feedback process weighted by its corresponding probability

\[
\langle W \rangle = \sum_{k,l} p (k,l) U (\rho^{(k,l)}_{2}) - U (\rho^{eq}_{0}), \tag{S1}
\]

where \( p (k,l) = p (k|l) p (l) \) is the joint probability for the \( l \)-th measurement outcome and \( k \)-th feedback operation, \( U (\rho^{eq}_{0}) = \text{tr} [H_{0} \rho^{eq}_{0}] \) and \( U (\rho^{(k,l)}_{2}) = \text{tr} [H^{(k)}_{F} \mathcal{F}^{(k)} (\rho^{(l)}_{\tau_{1}})] \) are the initial and final internal energy, respectively, \( \rho^{(k,l)}_{2} = \mathcal{F}^{(k)} (\rho^{(l)}_{\tau_{1}}) \) are the possible system’s final states. The operator sum decomposition of the feedback operation is \( \mathcal{F}^{(k)} (\cdot) = \sum \Gamma^{(k)}_{j} (\cdot) \Gamma^{(k)\dagger}_{j} \), whereas the post-measurement state of the \( l \)-th projective measurement is \( \rho^{(l)}_{\tau_{1}} = M^{(l)} \rho^{eq}_{0} M^{\dagger l} / p (l) \). Since the unitary processes considered here do not involve energy exchange with the reservoir, the change in the internal energy is regarded as work. Using the spectral decomposition of both Hamiltonians, \( H_{0} = \sum \varepsilon^{(0)}_{n} \Pi_{n}^{0} \) and \( H^{(k)}_{F} = \sum \varepsilon^{(\tau_{2},k)}_{m} \Pi_{m}^{(\tau_{2},k)} \), one can write Eq. (S1) as

\[
\langle W \rangle = \sum_{m,j,k,l,n} p (k,l) p (m,j,l,n) \Delta^{(k)}_{m,n}, \tag{S2}
\]

with \( p (m,j,k,l,n) \equiv \text{tr} [\Pi_{m}^{(\tau_{2},k)} \Gamma^{(k)}_{j} M_{l} \Pi_{n}^{(0)} \rho^{eq}_{0} U^{\dagger} M_{l} \Gamma^{(k)\dagger}_{j}], \)

\[
p (m,j,k,l,n) \equiv p (k|l) p (m,j,l,n), \quad \Delta^{(k)}_{m,n} = \varepsilon^{(\tau_{2},k)}_{m} - \varepsilon^{(0)}_{n}. \]

We can express the work distribution in the presence of feedback as \( P (W) = \)
the right hand side (rhs) of Eq. (S3) can be simplified
be 
Fluctuation relation in the presence of feedback. For the sake of completeness, we will verify
the validity of the fluctuation relation in Eq. (1) of the main text, which was previously discussed in Refs. [36, 37]. Consider the following average
\[
\langle e^{-\beta(W - \Delta F_{\text{eq}})} \rangle = \sum_{m,j,k,l,n} p(m, j, k, l, n) \frac{e^{-\beta e^0_{m,n}}}{Z_{m,n}^{eq}} p(k) \cdot (S3)
\]
Remembering the definition of \( p(m, j, l, n) \) introduced in the previous section and identifying \( \Pi_{m,n}^{eq} = Z_{m,n}^{-1} e^{-\beta e^0_{m,n}} \) and \( \rho_{\tau_2}^{(k, eq)} = \frac{1}{Z_{\tau_2}} \sum_m e^{-\beta e^m_{\tau_2,k}} \Pi_{m,k,l}^{\tau_2} \), the right hand side (rhs) of Eq. (S3) can be simplified to
\[
\sum_{k,j,l} p(k) \text{ tr} \left( \rho_{\tau_2}^{(k, eq)} \Gamma_j^{(k)} \Gamma_j^{(k)} \right).
\]
Using the completeness of the measurement and the unitarity of the map, \( \mathcal{F}^{(k)}( \mathbb{1} ) = \sum_j \Gamma_j^{(k)} \Gamma_j^{(k)} = \mathbb{1} \), Eq. (S3) turns out to be
\[
\langle e^{-\beta(W - \Delta F_{\text{eq}})} \rangle = \sum_k p(k) \text{ tr} \left( \rho_{\tau_2}^{(k, eq)} \right).
\]
Since \( \text{ tr} (\rho_{\tau_2}^{(k, eq)}) = 1 \) and \( p(k) \) is a normalized distribution, we obtain the fluctuation relation in Eq. (1) of the main text.

Derivation of the trade-off relation. Let us start from KL relative entropy between an arbitrary state, \( \rho \), and the equilibrium state, \( \rho^{eq} \), associated with the Hamiltonian \( \mathcal{H} \) at inverse temperature \( \beta \),
\[
S_{KL}(\rho||\rho^{eq}) = \text{ tr} \left[ \rho \ln \rho - \ln \rho^{eq} \right] = -\text{ tr} \left( \rho \ln \frac{e^{-\beta H}}{Z_{\tau_2}} \right) - S(\rho) = \beta \mathcal{H}(\rho) - \ln Z - S(\rho) = \beta [U(\rho) - F] - S(\rho). \tag{S4}
\]
From the above identity we can write \( \beta U (\rho^{(k,l)}) = S_{KL}(\rho^{(k,l)} || \rho_{\tau_2}^{(k, eq)} ) + \beta F^{(k)} + S(\rho_{\tau_2}^{(k, eq)} ) \) and \( \beta U (\rho_0^{eq}) = \beta F_0 + S(\rho_0^{eq}) \), with \( F^{(k)} = -\beta^{-1} \ln Z^{eq}_{\tau_2} \) and \( F_0 = -\beta^{-1} \ln Z_0 \). These results combined with Eq. (S1) lead to the following expression for the mean nonequilibrium entropy production in the presence of feedback:
\[
\langle \Sigma \rangle = \beta \langle W - \Delta F_{\text{eq}} \rangle - \frac{\beta}{2} H_{\tau_{eq}} + S_{\text{KL}}(\rho_{\tau_2}^{(k, eq)} || \rho_{\tau_2}^{(k, eq)} ) + \sum_{k,l} p(l) S(\rho^{(k,l)}) - S(\rho^{(k,l)}), \tag{S5}
\]
where we have added and subtracted the averaged entropy \( \sum_l p(l) S(\rho^{(k,l)}) \). The rhs of Eq. (S5) can be identified with the information-theoretic quantities (i.e. information gain, KL relative entropy, and the von Neumann entropy variation, respectively) resulting in the trade-off relation in Eq. (3) of the main text.

Information gain. When performing a measurement on a quantum system, the observer acquires information about the system. The description of such a process plays a central role in quantum measurement theory [47]. The information gain was first proposed by Groenewold in 1971 [41] whose aim was to quantify how much information is obtained by a quantum measurement. Groenewold proposed that the information gained by the observer is given by the average uncertainty reduction of the quantum state, and its given by
\[
\mathcal{I}_{\text{gain}} = S(\rho) - \sum_l p(l) S(\rho^{(l)}) \tag{S6}
\]
where \( \rho \) is the state immediately before the measurement and \( \rho^{(l)} \) the post-measurement states, which occur with outcome probability \( p(l) \). A year after its introduction, Lindblad showed that the information gain, as expressed
in Eq. (S6), is non-negative for von Neumann projective measurements [42]. Over a decade later, Ozawa generalized the demonstration of Lindblad showing that the information gain is non-negative for any positive-operator valued measure (POVM) [43]. POVMs and von Neumann measurements are called efficient measurements because each measurement operator is associated with a single outcome (each measurement operator is described by a single Kraus operator). On the other hand, measurements which possess an intrinsic classical uncertainty are called inefficient. Inefficient measurements induce more back-action to the system than quantum mechanics would allow for the same amount of information extracted [47]. Equation (S6) may becomes negative for such inefficient measurements and therefore may not be a good quantifier in this particular case. Recently, Buscemi, Hayashi, and Horodeck [45] generalized the expression for the information gain providing a new framework which gives positive results for any kind of measurement (efficient or inefficient) and recovers Eq. (S6) for efficient measurements. Furthermore, they endorsed the interpretation of the information gain by providing an operational meaning to it. In order to approach this very general class of measurements, in Ref. [45] the measuring apparatus is composed by three ancillary systems apart from the system to be measured (so-called quantum instruments). One ancilla is employed as a purification system and the other two to emulate the desired measurement, similar to the role of the quantum memory in the measurement stage of our feedback protocol. This newly proposed information gain is based on the mutual information of these auxiliary systems, although ultimately it depends only on quantities of the system.

In the present article, we have considered projective measurements (which are experimentally implemented with a very high accuracy assessed by quantum tomography), therefore the Groenewold definition of information gain, in Eq. (S6), is suitable to be applied throughout all of our analyzes. We also note that Eq. (3) of the main text holds for a general projective measurement feedback control based on unitary operations. An interesting point for future investigation is the possibility to derive an information thermodynamics trade-off relation for a feedback mechanism involving inefficient measurements.

For the sake of clarity, we provide below a short demonstration (based on Refs. [44, 47]) that the information gain as defined in Eq. (S6) is non-negative for POVM measurements. Any operator $A$ may be decomposed as $A = PU$, where $P$ is a positive operator and $U$ is a unitary operator. This decomposition is called the polar decomposition of $A$. Using this decomposition we can show that $AA^\dagger = P^2$. Therefore, $A^\dagger A = U^\dagger P^2 U = U^\dagger AA^\dagger U$, which means that the operators $A^\dagger A$ and $AA^\dagger$ possess the same set of eigenvalues (since unitary transformation preserve eigenvalues). For a POVM, $\{M^\dagger_l M_l\}$, the $l$-th post-measurement state, $\rho(l) = M_l \rho M^\dagger_l / p(l)$, is obtained with probability $p(l) = \text{tr} \left( M^\dagger_l M_l \rho \right)$. The measurement operators of the POVM satisfy the completeness relation $\sum_l M^\dagger_l M_l = I$. We can write the state before the measurement, $\rho$, in the following convenient form

$$\rho = \sqrt{p} I \sqrt{p} = \sum_l \sqrt{p} M^\dagger_l M_l \sqrt{p} = \sum_l p(l) \mu(l),$$  

(S7)

where $\mu(l) = \sqrt{p} M^\dagger_l M_l \sqrt{p}/p(l)$. The von Neumann entropy is a concave function, which means $S(\sum_n p(n) \rho(n)) \geq \sum_n p(n) S(\rho(n))$ with $\sum_n p(n) = 1$. From the concavity of the von Neumann entropy we obtain

$$S(\rho) \geq \sum_l p(l) S(\mu(l)).$$  

(S8)

Defining $X_l = \sqrt{p} M_l / \sqrt{p(l)}$ we write $\mu(l) = X_l^\dagger X_l$. So $\mu(l)$ has the same eigenvalues as $\rho(l) = X_l^\dagger X_l = M_l \rho M^\dagger_l / p(l)$. Since the von Neumann entropy is a function of only the eigenvalues of the density operator $S(\mu(l)) = S(\rho(l))$, which implies the non-negativity of the information gain defined in Eq. (S6) for any POVM.

**Experimental set-up.** The liquid sample consist of 50 mg of 99% $^{13}$C-labeled CHCl$_3$ (Chloroform) diluted in 0.7 ml of 99.9% deuterated Acetone-d6, in a flame sealed Wildmad LabGlass 5 mm tube. All experiments are carried out in a Varian 500 MHz Spectrometer employing a double-resonance probe-head equipped with a magnetic field gradient coil. Chloroform sample is very diluted so that the intermolecular interaction can be neglected, and the sample can be regarded as a set of identically prepared pairs of spin-1/2 systems. The sample is placed in the presence of a longitudinal static magnetic field (whose direction is taken to be along the positive $z$ axes) with strong intensity, $B_0 \approx 11.75$ T. The nuclear magnetization of $^1$H and $^{13}$C precess around $B_0$ with Larmor frequencies about 500 MHz and 125 MHz, respectively. Magnetization of the nuclear spins are controlled by time-modulated rf-field pulses in the transverse ($x$ and $y$) direction and longitudinal field gradients.

Spin-lattice relaxation times, measured by the inversion recovery pulse sequence, are $(T_1^H, T_1^C) = (7.42, 11.31)$ s. Transverse relaxations, obtained by the Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence, have characteristic times $(T_2^H, T_2^C) = (1.11, 0.30)$ s. The total experimental running time, to implement the entropy rectification protocol, is about 22.4 ms, which is considerably smaller than the spin-lattice relaxation and therefore decoherence can be disregard. The data for the process tomography, showed in Fig. 2 of the main text, also endorses this consideration, since the experimentally implemented process does not exhibit significant decoherence effects.

The initial state of the nuclear spins is prepared by spatial average techniques [31–34], being $^1$H nucleus pre-


pared in the ground state and the $^{13}\text{C}$ nucleus in a pseudo-thermal state with the populations (in the energy basis of $\mathcal{H}^0$) and corresponding pseudo-temperatures displayed in Tab. SI.

Table SI. Population and pseudo-temperature of the Carbon initial states.

| $p_1^{(0)}$ | $p_2^{(0)}$ | $k_B T$ (peV) |
|------------|------------|--------------|
| 0.96 ± 0.01 | 0.04 ± 0.01 | 2.6 ± 0.2    |
| 0.92 ± 0.01 | 0.08 ± 0.01 | 3.4 ± 0.2    |
| 0.88 ± 0.01 | 0.12 ± 0.01 | 4.2 ± 0.2    |
| 0.84 ± 0.01 | 0.16 ± 0.01 | 4.9 ± 0.2    |
| 0.81 ± 0.01 | 0.19 ± 0.01 | 5.9 ± 0.3    |
| 0.76 ± 0.01 | 0.24 ± 0.01 | 7.0 ± 0.3    |
| 0.73 ± 0.01 | 0.27 ± 0.01 | 8.6 ± 0.4    |
| 0.69 ± 0.01 | 0.31 ± 0.01 | 10.7 ± 0.6   |
| 0.65 ± 0.01 | 0.35 ± 0.01 | 13.8 ± 1.0   |

Data acquisition. Quantum state tomography (QST) is employed to obtain the relevant information quantities in the controlled feedback process. We have performed QST along the protocol implementation of the demon and we have employed an auxiliary circuit as depicted in Figs. S2(a) and S2(b), respectively. QST 1 is used to verify the effective temperature of the initial state. From QST 2 and QST 3 we obtain the information gain, $I_{\text{gain}}$. The mutual information, $I^{(k,l)}$, is obtained from QST 3. The remaining information quantities $\left(S_{KL}\left(\rho_{T_2}^{(k,l)}\right|\rho_{T_2}^{(k,eq)}\right)$ and $\Delta S^{(k,l)}(\tau)$ are obtained from the aforementioned tomographic data combined with QST 4 (obtained from the auxiliary circuit in Fig. S2(b)). Due to the fact that our implementation is based on nonselective measurements, an auxiliary circuit (Fig. S2(b)) is employed to obtain the states $\rho_{T_2}^{(k,l)}$ by QST. This enables us to characterize the information quantities in rhs of Eq. (3) in the main text. The optimized pulse sequence used to implement the Maxwell’s demon is displayed in Fig. S2(c).

The quantum process tomography, as illustrated in Fig. 2(a) of the main text, is carried out by preparing a set of mutually unbiased basis (MUB) states [50], implementing the nonselective projective measurement operation (the full controlled feedback protocol) in QPT 1 (in QPT 2) and then a full quantum state tomography at the end. From this data it is possible to obtain the Choi-Jamiołkowski matrix, $\chi$, of the process. The error in the Maxwell’s demon realization is probed by the process trace distance, $\delta = \frac{1}{2} \text{tr} \left| \chi^{exp} - \chi^{id} \right|$, between the ideal $(id)$ and the experimentally $(exp)$ implemented processes, as displayed in Fig. 2(c) of the main text. Its operational interpretation is related with the bias for the distinguishability between the ideal and experimental processes. The average success probability when distinguish-

Figure S2. Characterization of the Maxwell's demon operation protocol. (a) and (b) Quantum state tomography strategy to obtain the relevant information quantities. The initial states, as displayed, are prepared and the feedback control is applied. This enables the full information characterization of the feedback controlled evolution. QST 1 and QST 2 are single spin-1/2 tomography realized on the Carbon nucleus, whereas QST 3 and QST 4 are joint tomographies implemented in both Hydrogen and Carbon nuclei. (c) Optimized pulse sequence used to implement the measurement-based feedback control operation. Blue (red) circles represent one spin transverse rf-pulses producing rotations on the $x$ ($y$) by the displayed angle. Free evolutions under the natural $J$ coupling $(\frac{1}{2}\pi J\sigma_{z}^{A}\sigma_{z}^{B})$ are represented by two-spins connections (in orange), with the time-length displayed above the connections. The grey regions represent the two longitudinal field gradients.

Information bounds for entropy production. For the projective nonselective measurement implemented in our protocol, the information gain reduces to $I_{\text{gain}} = S(\rho_{T_2})$, in the ideal case. Since the driving process implemented, $\mathcal{U}$, is a unitary sudden quench, the von Neumann entropy after the quench, $S(\rho_{T_2})$, is the same as for the initial equilibrium state, $S(\rho_0)$. This latter entropy is also equal to the classical Shannon entropy $H_{\text{Sh}}(p_0^{(0)}, p_1^{(0)}) = -\sum_{i=0,1} p_i^{(0)} \ln p_i^{(0)}$, of the populations in the initial Hamiltonian ($\mathcal{H}_0$) energy basis (with $p_i^{(0)} = \text{tr} (\Pi_i \rho_0^{(0)})$). So $I_{\text{gain}} = H_{\text{Sh}}(p_0^{(0)}, p_1^{(0)})$. On the other hand, the probability for the $l$-th measurement outcome after the sudden quench is equally weighted in the ideal case, $p(l) = \frac{1}{2}$, for $l = 0, 1$. In the absence of basis mismatch, the correlation generated, by the coherent implementation of the measurement-based feedback, leads ideally to the joint probability distribution $p(k, l)$ of the
controlled operation \((k)\) and measurement outcome \((l)\),
\(p(0,0) = p(1,1) = \frac{1}{2}\) and \(p(0,1) = p(1,0) = 0\). This implies that the marginal
distribution for the control operation is \(p(k) = \sum_l p(k,l) = 1/2\) for \(k = 0, 1\). Accordingly,
\[
\langle I^{(k,l)} \rangle = \sum_{k,l} p(k,l) \ln \frac{p(k,l)}{p(k)p(l)} = H_{Sh}(\frac{1}{2}, \frac{1}{2}) = \ln 2 \text{ nats}
\]
meaning maximum correlation between system and memory. In fact, the measurement-based feedback protocol
was designed to achieve this maximum. Since the Shannon entropy, \(H_{Sh}(p_{0}^{(0)} p_{1}^{(0)})\), is upper bounded by
\(\ln 2\) nats, we conclude that \(I_{\text{gain}} \leq \langle I^{(k,l)} \rangle\), where the inequality is saturated in the limit \(\beta \to 0\) as can be noted in the
experimental data displayed in Fig. 3(b) of the main text.

**Error analysis.** The main sources of error in the experiments are small non-homogeneities of the transverse
rf-field, non-idealities in its time modulation, and non-idealities in the longitudinal field gradient. In order
to estimate the error propagation, we have used a Monte Carlo method, to sampling deviations of the
tomographic data with a Gaussian distribution having widths determined by the variances corresponding to such data.
The standard deviation of the distribution of values for the relevant information quantities is estimated from this
sampling. The variances of the tomographic data are obtained preparing the same state ten times, taking the full
state tomography and comparing it with the theoretical expectation. These variances include random and sys-
tematic errors in both state preparation and data acquisition by QST. The error in each element of the density
matrix estimated from this analysis is about 1\%. All parameters in the experimental implementation, such as
pulses intensity and its time duration, are optimized in order to minimize errors.

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We have considered unital processes in order to obtain the fluctuation relation in Eq. (1). It is worth mentioning that this consideration is not necessary for obtaining the trade-off relation introduced in Eq. (3).

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