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# Generalized Clausius Inequality for Nonequilibrium Quantum Processes

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We show that the nonequilibrium entropy production for a driven quantum system is larger than the Bures length, the geometric distance between its actual state and the corresponding equilibrium state. This universal lower bound generalizes the Clausius inequality to arbitrary nonequilibrium processes beyond linear response. We further derive a fundamental upper bound for the quantum entropy production rate and discuss its connection to the Bremermann-Bekenstein bound.

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All real macroscopic processes are irreversible. In thermodynamics, irreversibility is quantified by means of the entropy  $S$ : For any state transformation, the variation of entropy is written as  $\Delta S = \Delta S_{\text{re}} + \Delta S_{\text{ir}}$ , where  $\Delta S_{\text{re}} = Q/T$  is the entropy change associated with reversible (equilibrium) processes [1]. The Clausius inequality,  $\Delta S_{\text{ir}} \geq 0$ , on the other hand, provides a fundamental characterization of irreversible (nonequilibrium) phenomena by specifying a lower bound for the irreversible entropy change; this lower bound (zero) is trivially independent of how far from equilibrium a process operates. In many cases of interest, however, having a sharper, transformation-dependent lower bound is essential. A case in point is the optimization of the performance of real thermodynamic processes that occur in finite time [2,3]. For classical, near-equilibrium transformations, such a lower bound has been derived using a geometric approach to thermodynamics [4]: The infinitesimal irreversible entropy production is given by the Riemannian distance between initial (equilibrium) and final (nonequilibrium) states,  $dS_{\text{ir}} \geq d\ell^2/2$  [5–8]; this expression is obtained by a second-order expansion around equilibrium and is, therefore, restricted to the linear response regime. The thermodynamic length  $\ell$  measures the number of distinguishable states between initial and final probability distributions,  $p_0$  and  $p_\tau$ , and is identical to Wootters' statistical distance between wave vectors (pure states) in Hilbert space [9]; it is explicitly given by

$$\ell(p_0, p_\tau) = \arccos\left(\int dx \sqrt{p_0(x)p_\tau(x)}\right), \quad (1)$$

and, hence, measures the angle in state space separating the two probability distributions  $p_0$  and  $p_\tau$ .

In this paper, we extend the above results to quantum, far from equilibrium transformations. Specifically, we generalize the familiar Clausius inequality by deriving a universal lower bound for the irreversible entropy production valid for arbitrary quantum processes, using tools from quantum information theory. We, moreover, show that, in contrast to classical nonequilibrium physics, there exists a maximum quantum entropy production rate. Our work is

motivated by recent experiments on driven cold-atom gases, which, for the first time, allow the direct investigation of the nonequilibrium dynamics of isolated many-particle quantum systems beyond the near-equilibrium linear response regime [10,11]. A precise characterization of the quantum nonequilibrium entropy production in this uncharted domain therefore appears necessary. In the following, we start from a recently derived formula for the total work done on a closed quantum system [12] to obtain a microscopic expression for the irreversible entropy production, valid far from equilibrium. We show that the latter is bounded from below by the Bures length [13,14], a quantum generalization for mixed states of Wootters' statistical distance. The Bures length is closely related to the quantum fidelity, a central measure of quantum information theory [15]. We further demonstrate that the quantum entropy production rate  $\sigma$  is bounded from above by a quantity that also depends on the Bures length. This fundamental limit on the entropy variation rate is of purely quantum origin and is connected to the energy-time uncertainty relation. Remarkably, we show that the maximum rate reduces to the Bremermann-Bekenstein bound on information flow [16].

*Quantum nonequilibrium entropy production.*—Let us consider a closed quantum system, initially in a thermal state, whose Hamiltonian  $H_t$  is driven by an external time-dependent parameter during time  $\tau$ . For a slow, reversible transformation, the system remains in thermal equilibrium at all times. By contrast, for a fast change, for instance a parameter quench, the system is driven in a nonequilibrium state. The nonequilibrium entropy variation associated with such a transformation may be defined as [17]

$$\Delta S_{\text{ir}} = \beta \langle W_{\text{ir}} \rangle, \quad (2)$$

where  $\langle W_{\text{ir}} \rangle = \langle W \rangle - \Delta F$  is the difference between the total work  $\langle W \rangle$  done on the system during time  $\tau$  and the free energy difference  $\Delta F$  (the equilibrium work); as usual,  $\beta = 1/(kT)$  denotes the inverse temperature. The irreversible work  $\langle W_{\text{ir}} \rangle$  vanishes for a reversible process and is defined even if the final state of the system is arbitrarily far

from equilibrium. The probability distribution of quantum work is given by the difference  $E_m^\tau - E_n^0$  of final- and initial-system energy eigenvalues, averaged over all initial states [thermal distribution  $p_n^0 = \exp(-\beta E_n^0)/Z_0$ ] and final states (transition probabilities  $p_{m,n}^\tau$ ) [12],

$$\mathcal{P}(W) = \sum_{m,n} \delta(W - (E_m^\tau - E_n^0)) p_{m,n}^\tau p_n^0. \quad (3)$$

An experimental scheme to measure  $\mathcal{P}(W)$  in a modulated cold ion trap has been proposed in Ref. [18]. Equation (3) can be used to show that the entropy production satisfies the fluctuation relation  $\langle \exp(-\Delta S_{\text{ir}}) \rangle = 1$  [12,17]. According to Eq. (3), the mean work is simply  $\langle W \rangle = \sum_{m,n} (E_m^\tau - E_n^0) p_{m,n}^\tau p_n^0$ . By introducing the equilibrium density operator at the final time  $\tau$ ,  $\rho_\tau^{\text{eq}} = \exp(-\beta H_\tau)/Z_\tau$  with eigenvalues  $p_m^\tau$ , we can write

$$\begin{aligned} \langle W \rangle &= 1/\beta \sum_n p_n^0 \ln p_n^0 - 1/\beta \sum_{m,n} p_n^0 p_{m,n}^\tau \ln p_m^\tau \\ &\quad - 1/\beta \ln(Z_\tau/Z_0). \end{aligned} \quad (4)$$

The last term on the right-hand side is equal to  $\Delta F$ , while the first two are  $(1/\beta)$  times the quantum Kullback-Leibler divergence  $S(\rho_\tau \parallel \rho_\tau^{\text{eq}})$ , or quantum relative entropy [19], between the actual density operator of the system  $\rho_\tau$  at time  $\tau$  and the corresponding equilibrium density operator  $\rho_\tau^{\text{eq}}$ . Using Eq. (2), we therefore obtain

$$\Delta S_{\text{ir}} = S(\rho_\tau \parallel \rho_\tau^{\text{eq}}) = \text{tr}\{\rho_\tau \ln \rho_\tau - \rho_\tau \ln \rho_\tau^{\text{eq}}\}. \quad (5)$$

This is an exact expression for the nonequilibrium entropy production and a quantum generalization of recent results presented in Refs. [20,21] (see also Ref. [22]). We note, however, that the relative entropy is not a true metric, as it is not symmetric and does not satisfy the triangle inequality; it therefore cannot be used as a proper quantum distance [23]. Furthermore, Eq. (5) is, in general, difficult to determine explicitly. We next derive a lower bound for the quantum entropy production which we express in terms of the fidelity, one of the most commonly used and well-studied measures in quantum information theory [15].

*Generalized quantum Clausius inequality.*—Inequalities are essential tools of classical and quantum information theory [23]; they allow us to express “impossibilities,” things that cannot happen, and relate hitherto unconnected quantities. An elementary example is Klein’s inequality,  $S(\rho_1 \parallel \rho_2) \geq 0$ , which asserts the non-negativity of the quantum relative entropy [15]. Combined with Eq. (5), it immediately leads to the usual Clausius inequality. We shall establish a generalized Clausius inequality by proving that the irreversible entropy variation is always larger than the Bures length [13,14]. The Bures metric formally quantifies the infinitesimal distance between two density operators as  $\mathcal{L}^2(\rho + \delta\rho, \rho) = \text{tr}\{\delta\rho G\}/2$ , where  $G$  obeys  $\rho G + G\rho = \delta\rho$ . Thermodynamic distances should be physically motivated and, to some degree, unique [4]. Wootters’ statistical distance, being equal to the angle in Hilbert space, is the only Riemannian metric (up to a

constant factor) which is invariant under *all* unitary transformations [9]; it is hence a natural metric on the space of pure states. The Bures metric, on the other hand, is the generalization of Wootters’ metric to mixed states [24]; in this sense it represents a natural, unitarily invariant Riemannian metric on the space of impure density matrices [25]. For any two density operators the finite Bures length  $\mathcal{L}$  is given by

$$\mathcal{L}(\rho_1, \rho_2) = \arccos \sqrt{F(\rho_1, \rho_2)}, \quad (6)$$

where the fidelity  $F$  is defined for an arbitrary pair of mixed quantum states as [26,27]

$$F(\rho_1, \rho_2) = [\text{tr}\{\sqrt{\sqrt{\rho_1}\rho_2\sqrt{\rho_1}}\}]^2. \quad (7)$$

The fidelity is a symmetric, non-negative, unitarily invariant function, which is equal to 1 only when the two states are identical. For pure quantum states,  $\rho_i = |\psi_i\rangle\langle\psi_i|$ , the fidelity reduces to their overlap,  $F(\rho_1, \rho_2) = \text{tr}\{\rho_1\rho_2\} = |\langle\psi_1|\psi_2\rangle|^2$ . It has recently been shown that if  $d(\rho_1, \rho_2)$  is a unitarily invariant norm, then the quantum relative entropy satisfies (Ref. [28], Th. 4)

$$S(\rho_1 \parallel \rho_2) \geq 2 \frac{d^2(\rho_1, \rho_2)}{d^2(e^{1,1}, e^{2,2})}, \quad (8)$$

where  $e^{i,j}$  is the matrix with the  $i, j$  element equal to 1 and all other elements 0. Noting that  $\mathcal{L}(e^{1,1}, e^{2,2}) = \pi/2$ , since the two matrices are orthogonal [ $F(e^{1,1}, e^{2,2}) = 0$ ], we obtain the generalized Clausius inequality,

$$\Delta S_{\text{ir}} \geq \frac{8}{\pi^2} \mathcal{L}^2(\rho_\tau, \rho_\tau^{\text{eq}}). \quad (9)$$

The quantum entropy production  $\Delta S_{\text{ir}}$  is hence bounded from below by the geometric distance between the actual density operator  $\rho_\tau$  at the end of the process and the corresponding equilibrium operator  $\rho_\tau^{\text{eq}}$ , as measured by the Bures length; the latter defines a quantum generalization of the concept of thermodynamic length [5–8] and provides a natural scale to compare  $\Delta S_{\text{ir}}$  with. In other words, inequality (9) quantifies in a precise way the intuitive notion that the irreversible entropy production is larger when a system is driven farther away from equilibrium. Expression (8) shows that  $\Delta S_{\text{ir}}$  is bounded by many distances; however, only the Bures length has a simple physical interpretation. Equation (9) is valid for arbitrary quantum processes, including far-from-equilibrium final states. For infinitesimally close diagonal states, we have  $S(\rho \parallel \rho + d\rho) \simeq 2\mathcal{L}^2(\rho \parallel \rho + d\rho) \simeq d\ell^2(\rho \parallel \rho + d\rho)/2$ . In the limit of classical, quasiequilibrium transformations, Eq. (9) thus reduces to  $dS_{\text{ir}} \geq 2/\pi^2 d\ell^2$ .

A simple illustration of the generalized Clausius inequality (9) is provided by a time-dependent quantum harmonic oscillator, initially at thermal equilibrium,

$$H_t = \frac{p^2}{2m} + \frac{1}{2} m \omega_t^2 x^2, \quad (10)$$

and whose angular frequency is varied from  $\omega_0$  to  $\omega_1$  according to  $\omega_t^2 = \omega_0^2 + (\omega_1^2 - \omega_0^2)t/\tau$  (cf. Fig. 1) [29]. This quantum system is exactly solvable, and both the entropy production and the Bures length can be evaluated explicitly [30]. The distance from equilibrium is controlled here by the ratio  $\omega_1/\omega_0$  between final and initial frequencies. We observe that the relation  $dS_{\text{ir}} \approx d\ell^2/2$  is valid close to equilibrium,  $\omega_1/\omega_0 \approx 1$ , whereas the generalized Clausius inequality (9) holds for all values of  $\omega_1$ . It is worth noting that for very large  $\omega_1$ , initial and final states become maximally distinguishable, i.e., orthogonal, and the Bures length (6) approaches  $\pi/2$ . For the parameters of Fig. 1, this very-far-from-equilibrium regime is reached when the energy of the oscillator is increased by more than a factor of 20. A quantitative estimate of the energy change associated with a given irreversible entropy production will be given below.

**Quantum entropy production rate.**—Nonequilibrium irreversible phenomena are not only characterized by the irreversible entropy change, but also by the rate of entropy production  $\sigma = \Delta S_{\text{ir}}/\tau$ . The entropy rate  $\sigma$  is a central quantity that is associated with the speed of evolution of a nonequilibrium process [1]. In quantum mechanics, the energy of a system imposes a fundamental constraint on its unitary time evolution, as captured, for instance, by the time-energy uncertainty relation  $t \geq \hbar/\Delta E$ : the minimal time it takes for a quantum system to evolve to an orthogonal state is always larger than the inverse of its initial energy spread  $\Delta E$ . A more accurate expression for the “quantum speed limit” has been derived in Ref. [31] for time-independent Hamiltonians. Consider a system initially in state  $\rho_0$  with mean energy  $E_0 = \langle H_0 \rangle$  and energy spread  $\Delta E_0 = (\langle H_0^2 \rangle - \langle H_0 \rangle^2)^{1/2}$ . The minimum time required for the evolution to an arbitrary state  $\rho_\tau$  is then given by

$$\tau_{\min} \approx \max \left\{ \frac{2\hbar \mathcal{L}^2(\rho_\tau, \rho_0)}{\pi E_0}, \frac{\hbar \mathcal{L}(\rho_\tau, \rho_0)}{\Delta E_0} \right\}, \quad (11)$$

where  $\mathcal{L}(\rho_\tau, \rho_0)$  is the Bures length (6) between  $\rho_\tau$  and  $\rho_0$ . The quantum speed limit time is thus entirely determined by

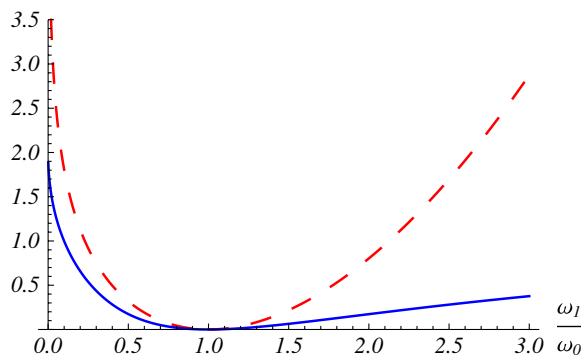


FIG. 1 (color online). Illustration of the generalized Clausius inequality (9) for the quantum harmonic oscillator (10) with linearly varying frequency. We show the entropy production  $\Delta S_{\text{ir}}$  (red, dashed line) and the lower bound  $8/\pi^2 \mathcal{L}^2(\rho_\tau, \rho_\tau^{\text{eq}})$  (blue, solid line) as a function of the final frequency  $\omega_1$  for  $\hbar = \beta = \tau = 1$  and  $\omega_0 = 0.1$ .

the initial energy (mean or variance) and the geometric distance between initial and final states. We will now show that Eq. (11) sets an upper bound to the entropy production rate. We begin by rewriting Eq. (1) in the form  $\Delta S_{\text{ir}} = \beta[\langle H_\tau \rangle - \langle H_0 \rangle - F_\tau + F_0]$ , since  $\Delta S_{\text{ir}} \geq 0$ , and focus on the limit of large excitations,  $\langle H_\tau \rangle \gg \langle H_0 \rangle$ , which is achieved for long enough driving. By applying the triangle inequality and noting, from Eq. (2), that  $\Delta F \leq \langle W \rangle = \langle H_\tau \rangle - \langle H_0 \rangle$ , we obtain

$$\begin{aligned} \Delta S_{\text{ir}} &\leq \beta(\langle H_\tau \rangle + \langle H_0 \rangle + \Delta F) \leq 2\beta(\langle H_\tau \rangle + \langle H_0 \rangle) \\ &\approx 2\beta\langle H_\tau \rangle. \end{aligned} \quad (12)$$

By combining Eqs. (11) and (12), we find that the maximum entropy production rate  $\sigma_{\max} = \Delta S_{\text{ir},\max}/\tau_{\min}$  is

$$\sigma_{\max} \approx 2\beta\langle H_\tau \rangle \min \left\{ \frac{\pi E_0}{2\hbar \mathcal{L}^2(\rho_\tau, \rho_0)}, \frac{\Delta E_0}{\hbar \mathcal{L}(\rho_\tau, \rho_0)} \right\}. \quad (13)$$

The above equation expresses the inherent quantum-mechanical limit to the entropy production rate. Equation (13) simplifies in the limit of far-from-equilibrium transformations, when initial and final states become orthogonal,  $\mathcal{L}(\rho_\tau, \rho_0) \approx \pi/2$ , and of high temperatures,  $E_0 \approx 1/\beta$  ( $\Delta E_0 \approx E_0/\sqrt{N} \leq E_0$ , for  $N$  degrees of freedom), to the Bremermann-Bekenstein bound [16],

$$\sigma \leq \frac{4}{\hbar\pi} \langle H_\tau \rangle. \quad (14)$$

The Bremermann-Bekenstein bound gives the maximum quantum communication rate (capacity) that is possible through a noiseless single channel with signals of finite duration. We stress that the present derivation is solely based on the thermodynamic definition of the entropy production (2) and does not make any reference to information entropy or channels; it is thus free of the caveats of the original derivations, such as the use of the periodic boundary condition approximation [32].

The quantum speed limit time (11), and hence the entropy production rate (13), only holds for slowly driven systems (i.e. quasiequilibrium processes), as Eq. (11) assumes a time-independent Hamiltonian. For arbitrary nonequilibrium transformations, Eq. (11) can be extended using the geometric approach of Ref. [33]. By evaluating the time derivative of the angle between the initial and final states,  $\mathcal{L}(\rho_\tau, \rho_0)$ , we find that the exact quantum speed limit for time-dependent Hamiltonians is

$$\tau_{\min} = \max \left\{ \frac{\hbar \mathcal{L}(\rho_\tau, \rho_0)}{E_\tau}, \frac{\hbar \mathcal{L}(\rho_\tau, \rho_0)}{\Delta E_\tau} \right\}. \quad (15)$$

The minimum time is determined here by the time averaged mean and variance,  $E_\tau = (1/\tau) \int_0^\tau dt \langle H_t \rangle$  and  $\Delta E_\tau = (1/\tau) \int_0^\tau dt (\langle H_t^2 \rangle - \langle H_t \rangle^2)^{1/2}$ , of the energy and not by their initial values [34]. As a consequence, the quantum speed limit time for driven systems can be smaller than for undriven systems when  $E_\tau > E_0$  ( $\Delta E_\tau > \Delta E_0$ ). This is, for instance, the case at zero temperature: According to



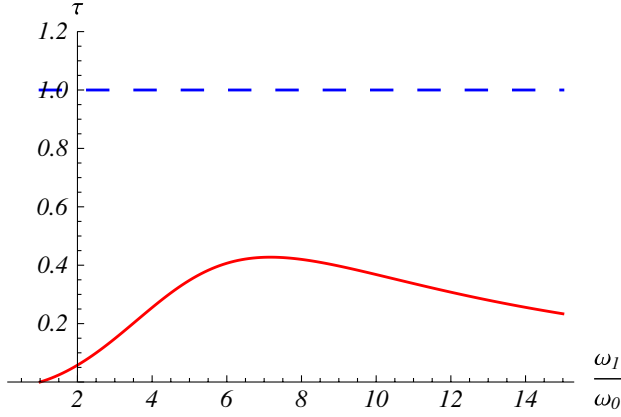


FIG. 2 (color online). Quantum speed limit time  $\tau_{\min}$ , Eq. (13) (red, solid line), and actual driving time  $\tau$  (blue, dashed line) for the linearly parametrized quantum harmonic oscillator (10) with  $\hbar = \tau = 1$ ,  $1/\beta = 0$ , and  $\omega_0 = 0.1$ .

Eq. (11), a quantum system never leaves an initial (non-degenerate) pure state (infinite  $\tau_{\min}$ ) in the absence of driving, while Eq. (13) predicts a finite  $\tau_{\min}$  for a driven Hamiltonian. Figure 2 shows that for the time-dependent oscillator (10) at zero temperature, the actual driving time  $\tau$  can approach the absolute minimum evolution time  $\tau_{\min}$  within a factor of 2, for a simple linear change of its angular frequency.

The general expression for the maximum entropy production rate  $\sigma$  for nonequilibrium quantum processes can eventually be obtained by combining Eqs. (11) and (15); it should be regarded as the extension of the Bremermann-Bekenstein bound (14) to arbitrary distances between initial and final states, arbitrary initial temperature, and arbitrary transformation speed. Written in the form

$$\frac{\langle H_i \rangle}{\Delta S_{\text{ir}}} \geq \frac{2\beta}{\tau} \min \left\{ \frac{E_\tau}{\hbar \mathcal{L}(\rho_\tau, \rho_0)}, \frac{\Delta E_\tau}{\hbar \mathcal{L}(\rho_\tau, \rho_0)} \right\}, \quad (16)$$

it provides an estimate for the minimum energy change occurring with a given entropy variation in a time  $\tau$ .

**Conclusion.**—We have developed a generic geometric characterization of far-from-equilibrium quantum processes based on the distinguishability metric on the space of quantum states. We have first obtained a generalized, more precise form of the Clausius inequality by deriving a lower bound for the irreversible entropy production given by the Bures length between the nonequilibrium density operator of the system and the corresponding equilibrium operator. We have further established the existence of an upper bound for the entropy production rate by employing the notion of quantum speed limit time, which is itself a function of the Bures length. The latter is an extension of the Bremermann-Bekenstein bound to arbitrary nonequilibrium quantum processes.

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