

## Landauer-Büttiker Approach to Strongly Coupled Quantum Thermodynamics: Inside-Outside Duality of Entropy Evolution

Anton Bruch,<sup>1</sup> Caio Lewenkopf,<sup>2</sup> and Felix von Oppen<sup>1</sup>

<sup>1</sup>*Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany*

<sup>2</sup>*Instituto de Física, Universidade Federal Fluminense, 24210-346 Niterói, Brazil*



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We develop a Landauer-Büttiker theory of entropy evolution in time-dependent, strongly coupled electron systems. The formalism naturally avoids the problem of the system-bath distinction by defining the entropy current in the attached leads. This current can then be used to infer changes of the entropy of the system which we refer to as the inside-outside duality. We carry out this program in an adiabatic expansion up to first order beyond the quasistatic limit. When combined with particle and energy currents, as well as the work required to change an external potential, our formalism provides a full thermodynamic description, applicable to arbitrary noninteracting electron systems in contact with reservoirs. This provides a clear understanding of the relation between heat and entropy currents generated by time-dependent potentials and their connection to the occurring dissipation.

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*Introduction.*—Ongoing progress in nanofabrication raises interest in the thermodynamics of nanomachines [1,2], describing the exchange of heat and work with their environment, as well as their efficiencies. The laws of thermodynamics are extremely successful in characterizing machines consisting of a macroscopic number of particles by just a handful of parameters, such as temperature and pressure. How these laws carry over to microscopic systems that consist of only a few particles and exhibit quantum behavior is the central problem of the field of quantum thermodynamics. At small scales, the thermodynamic variables necessarily acquire strong fluctuations [3,4], and the system-bath distinction becomes fuzzy [5–8]. A crucial quantity in this regard is the entropy which links thermodynamics and information, describes irreversibility, and governs the efficiencies of various energy conversion processes [9–15].

Here, we put forward a formalism based on the Landauer-Büttiker scattering approach to describe the entropy evolution generated by (slow) time-dependent potentials in electronic mesoscopic systems strongly coupled to external reservoirs. Our *outside* approach, which focuses on the entropy current in the leads (expressed in terms of scattering states) naturally avoids the system-bath distinction. This distinction has plagued and limited the applicability of previous *inside* approaches [5–8,16–20], which considered the thermodynamic functions of the strongly coupled system directly. While our approach is more general, we find that it reproduces earlier results where available.

In an elementary thermodynamic transformation, an external agent performs work on a system by changing its Hamiltonian, constituting a single stroke of a quantum engine. For electronic nanomachines, this is achieved by

changing the potential in a finite region which is coupled to electronic reservoirs. This type of machine can, for instance, be realized by a quantum dot connected to leads and subject to a time-dependent gate potential. If the gate potential is changed slowly, the coupling to the reservoir ensures thermal equilibrium at all times and the transformation occurs quasistatically. The change of the von Neumann entropy

$$S[\rho] = -\text{Tr}(\rho \ln \rho) \quad (1)$$

associated with the equilibrium state of the system is proportional to the heat  $dQ = TdS$  released into the reservoir at temperature  $T$ .

This should be contrasted with the entropy evolution of a closed quantum system. Its purely unitary time evolution implies that the von Neumann entropy remains unchanged at all times. Here, we want to discuss the entropy evolution of simple electronic nanomachines, which combine fully coherent quantum dynamics with contact to baths and can involve strong coupling between the system and the reservoir. In addressing this problem, quantum effects such as coherences, hybridization, and entanglement are expected to become important. Such electronic nanomachines can be described by the Landauer-Büttiker formalism, which has been very successfully used to understand the conductance [21], electron pumping [22], heat transport and current noise [21,23–26], entanglement creation [27,28], and adiabatic reaction forces [29–32] in a variety of mesoscopic systems.

Viewing the quantum thermodynamics of strongly coupled systems from Landauer-Büttiker theory shifts the emphasis away from the thermodynamic functions of the strongly coupled system (the inside approach) to the

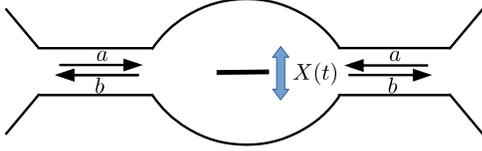


FIG. 1. The scattering potential in the central region, e.g., a quantum dot, slowly changed by an external parameter  $X(t)$ , driving a net heat and entropy current into the leads.

associated currents in the leads (the outside approach). One considers a scattering region connected to ideal leads, as depicted in Fig. 1, where noninteracting electrons propagate freely and under fully coherent quantum dynamics. Relaxation is accounted for by connecting the leads to electronic reservoirs at well-defined temperatures and chemical potentials, which determine the distribution of incident electrons. Conventionally, one calculates energy or particle currents in the leads by accounting for in- and outgoing electrons. By energy and particle conservation, these currents permit one to deduce the change of the energy and the particle number in the scattering region (inside-outside duality). Here, we show that the same approach extends to the entropy, thereby completing the thermodynamic description of strongly coupled systems. Since the von Neumann entropy  $S$  is conserved under coherent unitary dynamics, the change of entropy in the scattering region can also be inferred from the entropy currents carried by the scattered electrons. We find that this method overcomes the limitations of previous approaches arising from the strong system-bath coupling [6–8,33,34].

*Entropy current carried by scattering states.*—We consider a time-dependent scattering region connected to one or multiple ideal leads, in which the electrons propagate in transverse scattering channels, and leave the electronic spin degree of freedom implicit. Electrons in incoming and outgoing channels are described by the annihilation operators  $a$  and  $b$ , related by the scattering matrix  $S$ ,

$$\begin{pmatrix} b_1(\epsilon) \\ \vdots \\ b_N(\epsilon) \end{pmatrix} = \int_{-\infty}^{\infty} \frac{d\epsilon'}{2\pi} S(\epsilon, \epsilon') \begin{pmatrix} a_1(\epsilon') \\ \vdots \\ a_N(\epsilon') \end{pmatrix}. \quad (2)$$

Here, the subscript  $\alpha = 1, \dots, N$  labels the channels and leads. The leads are connected to electronic reservoirs, which determine the distribution of the incoming channels to be  $\langle a_\beta^\dagger(\epsilon) a_\alpha(\epsilon') \rangle = \phi_{\alpha\beta}^{\text{in}}(\epsilon) 2\pi \delta(\epsilon - \epsilon')$  in terms of a diagonal distribution matrix  $\phi_{\alpha\beta}^{\text{in}}(\epsilon) = \delta_{\alpha\beta} f_\alpha(\epsilon)$ , where  $f_\alpha(\epsilon)$  is the Fermi distribution with temperature  $T$  and chemical potential  $\mu_\alpha$ .

The particle current in channel  $\alpha$  through any cross section of the corresponding lead is obtained by accounting for in- and outgoing electrons [21],

$$I_\alpha^N(t) = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \{ \phi_{\alpha\alpha}^{\text{out}}(t, \epsilon) - \phi_{\alpha\alpha}^{\text{in}}(\epsilon) \}, \quad (3)$$

where the one-dimensional density of states  $\rho_\alpha(\epsilon) = [2\pi v_\alpha(\epsilon)]^{-1}$  and the group velocity  $v_\alpha(\epsilon)$  compensate for one another (we set  $\hbar = 1$ ).  $\phi^{\text{out}}(t, \epsilon)$  is given by the Wigner transform

$$\phi_{\alpha\beta}^{\text{out}}(t, \epsilon) = \int_{-\infty}^{\infty} \frac{d\tilde{\epsilon}}{2\pi} e^{-i\tilde{\epsilon}t} \langle b_\beta^\dagger(\epsilon - \tilde{\epsilon}/2) b_\alpha(\epsilon + \tilde{\epsilon}/2) \rangle. \quad (4)$$

Similarly, the energy current  $I_\alpha^E$  in channel  $\alpha$  reads

$$I_\alpha^E(t) = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \epsilon \{ \phi_{\alpha\alpha}^{\text{out}}(t, \epsilon) - \phi_{\alpha\alpha}^{\text{in}}(\epsilon) \}. \quad (5)$$

The heat current  $I_\alpha^Q = I_\alpha^E - \mu_\alpha I_\alpha^N$  carried by the electrons in the leads is a combination of the particle current  $I_\alpha^N$  into the corresponding reservoir with the chemical potential  $\mu_\alpha$  and the energy current  $I_\alpha^E$ . We can express the total heat current in terms of the diagonal elements of the distribution matrix  $\phi^{\text{out}}$ ,

$$I_{\text{tot}}^Q(t) = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} (\epsilon - \mu) \text{tr}_c \{ \phi^{\text{out}}(t, \epsilon) - \phi^{\text{in}}(\epsilon) \}, \quad (6)$$

where the trace runs over the channel and lead space. Here, for simplicity we assume the same chemical potential  $\mu$  in all reservoirs.

To obtain the entropy current, we begin by considering the entropy of a single incoming channel. For a given energy the channel can be either occupied or empty, according to  $f_\alpha(\epsilon)$ , and contributes with

$$\sigma[f_\alpha(\epsilon)] = -f_\alpha(\epsilon) \ln[f_\alpha(\epsilon)] - [1 - f_\alpha(\epsilon)] \ln[1 - f_\alpha(\epsilon)] \quad (7)$$

to the system entropy. By analogy with the particle current, Eq. (3), we write the incoming entropy current as

$$I_\alpha^{\text{S,in}} = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \sigma[f_\alpha(\epsilon)]. \quad (8)$$

Hence, as expected [35], each of the incoming spin-resolved channels carries an entropy current of  $\pi T/6$  towards the scattering region.

Scattering redistributes the electrons between the outgoing channels, thereby modifying the entropy flow into the leads. The scattering-induced correlations between outgoing scattering states [24,26] are encoded in the nondiagonal distribution matrix  $\phi_{\alpha\beta}^{\text{out}}(t, \epsilon)$  for the outgoing electrons. As we show below, the natural extension of Eq. (8) reads

$$I^{\text{S,in(out)}}(t) = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \text{tr}_c \{ \sigma[\phi^{\text{in(out)}}(t, \epsilon)] \}. \quad (9)$$

To motivate Eq. (9), we derive the noninteracting fermionic density matrix for a given distribution matrix  $\bar{\phi}_{\alpha\beta} = \text{Tr}[\rho c_\beta^\dagger c_\alpha]$ . In the scattering setup, the incoming operators describe particles of an equilibrium reservoir and the outgoing operators are linear functions of the incoming ones; see Eq. (2). Hence, all averages can be calculated via Wick's theorem and the single-particle

correlations described by  $\phi$  fully determine all expectation values.

Our derivation exploits the maximum entropy principle that yields the most general density matrix given certain single-particle correlations [36]. (We obtain the same result following the approach of Ref. [37].) The Lagrangian for maximizing the von Neumann entropy under the constraints  $\text{Tr}\rho = 1$  and  $\bar{\phi}_{\alpha\beta} = \text{Tr}[\rho c_{\beta}^{\dagger} c_{\alpha}]$  reads

$$\mathcal{L} = -\text{Tr}[\rho \ln \rho] + \sum_{\alpha\beta} \lambda_{\alpha\beta} (\text{Tr}[\rho c_{\beta}^{\dagger} c_{\alpha}] - \bar{\phi}_{\alpha\beta}) - \gamma (\text{Tr}\rho - 1), \quad (10)$$

where  $\text{Tr}$  denotes the many-particle trace over all possible occupations, and  $\gamma$  and the  $\lambda$  values are Lagrange multipliers. It is convenient to diagonalize the Hermitian matrix  $\bar{\phi}$  and introduce a rotated basis, namely,

$$\bar{\phi} = U \Lambda U^{\dagger} \quad \text{and} \quad c_{\alpha} = \sum_c U_{ac} d_c, \quad (11)$$

where  $U$  is a unitary matrix and  $\Lambda_{\alpha\beta} = \Lambda_{\alpha} \delta_{\alpha\beta}$  is a diagonal matrix containing the real eigenvalues of  $\bar{\phi}$ . In the rotated basis the Lagrangian  $\mathcal{L}$  allows us to maximize the von Neumann entropy with the given constraints. This yields the density matrix

$$\rho = \prod_{\alpha} (1 - \Lambda_{\alpha}) \left( \frac{\Lambda_{\alpha}}{1 - \Lambda_{\alpha}} \right)^{\hat{n}_{\alpha}}, \quad (12)$$

where  $\hat{n}_{\alpha}$  is the occupation of mode  $\alpha$  in the rotated basis. We calculate the entropy  $S$  of this density matrix by summing over all possible occupations in the rotated basis,

$$S = \sum_{\alpha} \sigma[\Lambda_{\alpha}] = \text{tr}(\sigma[\Lambda]), \quad (13)$$

where the sum over the diagonal elements of  $\Lambda$  is included through the single-particle trace  $\text{tr}$ . Finally, rotating back to the original basis,  $\Lambda = U^{\dagger} \bar{\phi} U$ , we find the entropy in terms of the distribution matrix  $\bar{\phi}$ ,

$$S = \text{tr}(\sigma[\bar{\phi}]). \quad (14)$$

For a slowly changing scattering potential, we associate the entropy with the time-dependent distribution matrix  $\phi_{\alpha\beta}(t, \epsilon)$  of the scattering states in Eq. (4), for which the single-particle trace represents an integral over energy and a trace  $\text{tr}_c$  over the channel and lead indices.

Combining the in- and outgoing entropy currents, we write the total entropy current into the leads as

$$I_{\text{tot}}^S(t) = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \text{tr}_c \{ \sigma[\phi^{\text{out}}(t, \epsilon)] - \sigma[\phi^{\text{in}}(\epsilon)] \}. \quad (15)$$

In the case of a static scatterer between two biased reservoirs at zero temperature, the entropy current can be used to quantify the entanglement of outgoing electron-hole pairs created in a tunneling event. Indeed, we verify that an immediate generalization of Eq. (15) reproduces the quantum mutual information between outgoing scattering

channels on the left and right, as obtained in Ref. [27] (see the Supplemental Material [38] for details).

*Entropy current induced by a dynamic scatterer.*—The entropy and heat currents generated by a slowly changing scattering potential  $V[X(t)]$  are obtained by expanding the scattering matrix and the outgoing distribution matrix about the frozen configuration in powers of the velocity  $\dot{X}$  [29–32]. Up to first order, the Wigner transform of the scattering matrix can be expressed in terms of the frozen scattering matrix  $S$  and its first order correction  $A$ ,  $\mathcal{S}(\epsilon, t) = S + \dot{X}A$ . This expansion is well motivated in the regime where  $X(t)$  changes on a characteristic time scale much longer than the electronic dwell time in the scattering region. Accordingly, we write  $\phi^{\text{out}}$  as

$$\phi^{\text{out}} \simeq \hat{I}f + \phi^{\text{out}(1)} + \phi^{\text{out}(2)}, \quad (16)$$

where  $\hat{I}$  is a unit matrix in the channel and lead space and the superscript stands for the order in  $\dot{X}$ . (We omit time and energy labels for better readability.) Similarly, we expand  $\sigma[\phi^{\text{out}}(\epsilon)]$  up to second order about the uncorrelated equilibrium,

$$\begin{aligned} \sigma[\phi^{\text{out}}] &\simeq \hat{I}\sigma[f] + \hat{I} \frac{d\sigma[f]}{df} (\phi^{\text{out}(1)} + \phi^{\text{out}(2)}) \\ &\quad + \frac{1}{2} \hat{I} \frac{d^2\sigma[f]}{df^2} (\phi^{\text{out}(1)})^2. \end{aligned} \quad (17)$$

Note that the second order contribution proportional to  $d^2\sigma[f]/df^2 = (T\partial_{\epsilon f})^{-1}$  is always negative due to the concavity of  $\sigma$ .

By inserting the above expression in Eq. (15), we obtain

$$\begin{aligned} I_{\text{tot}}^S &= \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \text{tr}_c \left\{ \frac{\epsilon - \mu}{T} (\phi^{\text{out}(1)} + \phi^{\text{out}(2)}) \right. \\ &\quad \left. + \frac{1}{2T\partial_{\epsilon f}} (\phi^{\text{out}(1)})^2 \right\}, \end{aligned} \quad (18)$$

where we have used  $\phi^{\text{in}} = \hat{I}f(\epsilon)$ . By the same token, Eqs. (6) and (16) give

$$I_{\text{tot}}^Q = \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} (\epsilon - \mu) \text{tr}_c \{ \phi^{\text{out}(1)} + \phi^{\text{out}(2)} \}. \quad (19)$$

These expressions nicely elucidate the connection between the heat and entropy currents, and the departure from  $dQ = TdS$  beyond the quasistatic limit. At first order in  $\dot{X}$ , corresponding to the quasistatic regime, the entropy current is entirely given by the heat current over temperature  $I_{\text{tot}}^{S(1)} = I_{\text{tot}}^{Q(1)}/T$ ; i.e., the proposed form of the entropy current correctly connects to the quasistatic equilibrium. By contrast, at second order an additional negative correction appears,

$$I_{\text{tot}}^{S(2)} = \frac{I_{\text{tot}}^{Q(2)}}{T} + \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \frac{1}{2T\partial_{\epsilon f}} \text{tr}_c \{ (\phi^{\text{out}(1)})^2 \}. \quad (20)$$

Since  $\text{tr}_c\{(\phi^{\text{out}(1)})^2\}$  contains all of the off-diagonal elements of  $\phi^{\text{out}(1)}$ , it encodes the correlations created by the dynamic scatterer. These correlations determine by how much the entropy current in the leads is smaller than the corresponding heat current over temperature. This net inflow of entropy into the scattering region reflects the local dissipation-induced increase of entropy.

We calculate  $\phi$  explicitly within the gradient expansion [29–31]. Assuming that  $f_\alpha(\epsilon) = f(\epsilon)$ , one writes  $\phi^{\text{out}(1)}$  in terms of the frozen scattering matrix  $S$ ,

$$\phi^{\text{out}(1)}(\epsilon, t) = i\dot{X}\partial_\epsilon f S \partial_X S^\dagger. \quad (21)$$

Inserting  $\phi^{\text{out}(1)}$  into the entropy current equation (20), we obtain the entropy current up to second order,

$$I_{\text{tot}}^S = \frac{I_{\text{tot}}^Q}{T} - \frac{\dot{W}^{(2)}}{T}, \quad (22)$$

with

$$\dot{W}^{(2)} = -\frac{\dot{X}^2}{2} \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \partial_\epsilon f(\epsilon) \text{tr}_c(\partial_X S^\dagger \partial_X S) \geq 0. \quad (23)$$

Remarkably,  $\dot{W}^{(2)} = \gamma\dot{X}^2$  is exactly the dissipated power that the external agent pumps into the system as a result of the time-dependent system Hamiltonian.  $\dot{W}^{(2)}$  was derived in Refs. [29–31] in terms of the friction coefficient  $\gamma$  of the backaction force that needs to be overcome by the external agent. Thus, from our *outside perspective*, dissipation leads to an inflow of entropy into the scattering region in addition to the heat-current contribution.

We are now ready to discuss the inside-outside duality of entropy evolution: We utilize the acquired knowledge about the entropy current (the outside perspective) to draw conclusions about the evolution of the entropy  $s$  of the strongly coupled subsystem located in the scattering region (the inside perspective). The direct calculation of the thermodynamic functions of such a subsystem has proven problematic in the past due to difficulties in taking proper account of the coupling Hamiltonian and the presence of strong hybridization [6–8,33]. These problems are naturally avoided within the Landauer-Büttiker formalism. Since this formalism considers fully coherent unitary dynamics in both the leads and the scattering region, the von Neumann entropy associated with the scattering states is conserved in a scattering event. Hence, an additional inflow of entropy is reflected in an increased entropy  $s$  stored in the scattering region. As a result, the entropy is source-free,

$$\frac{ds}{dt} + I_{\text{tot}}^S = 0. \quad (24)$$

We can use this continuity equation and Eq. (22) to infer the evolution of  $s$ . Invoking energy and particle conservation, we identify  $\dot{Q} = -I_{\text{tot}}^Q$  as the heat leaving the scattering region from the inside perspective. Thus, the entropy evolution can be expressed in terms of the thermodynamic functions of the (strongly) coupled subsystem as

$$\frac{ds}{dt} = \frac{\dot{Q}}{T} + \frac{\dot{W}^{(2)}}{T}. \quad (25)$$

Therefore, dissipation leads to a local increase of entropy, which is provided by the scattered electrons. This constitutes the inside-outside duality of entropy evolution.

Integrated over a full cyclic transformation of  $X$ , the entropy current needs to vanish, as it derives from a source-free thermodynamic state function; see Eq. (24). Averaged over a cycle, Eq. (22) thus implies that all extra energy pumped into the scattering region  $\dot{W}^{(2)}$  eventually has to be released as heat into the leads,

$$\overline{I_{\text{tot}}^{Q(2)}} = \overline{\dot{W}^{(2)}}. \quad (26)$$

Equation (25) bears some similarity with the entropy-balance equation of standard nonequilibrium thermodynamics [41]. However, we emphasize that we do not make any assumptions beyond Landauer-Büttiker theory and a slow time dependence of the scatterer. Specifically, unlike standard nonequilibrium thermodynamics, our approach does not require local equilibrium in the scattering region. It is also worthwhile to point out that Eqs. (24) and (25) can be interpreted as general definitions of the entropy  $s$  of the strongly coupled system. We will show next that this reproduces earlier results for the resonant level model, which were based on less general definitions.

*Application to the resonant level model.*—To emphasize the advantage of the outside approach over calculating the thermodynamic functions of a subsystem directly, we connect here to the thermodynamics of the resonant level model derived earlier from an *inside perspective*. This model consists of a single localized electronic level  $H_D = \epsilon_d(t)d^\dagger d$ , which can be changed in time by an external agent. It is coupled to a free electron metal  $H_B = \sum_k \epsilon_k c_k^\dagger c_k$  via a coupling Hamiltonian  $H_V = \sum_k (V_k d^\dagger c_k + \text{H.c.})$  and was intensively studied in the past [6,8], with the difficulties in Ref. [33] pointed out and overcome in Ref. [7].

The inside approach demands a splitting of the coupling Hamiltonian  $H_V$  between the effective system and the bath, which strongly limits its applicability to the resonant level model in the wideband limit of energy-independent hybridization [7,8]. By contrast, the outside approach developed here yields the strong coupling thermodynamics for arbitrary noninteracting electron systems and, furthermore, reproduces the results for the resonant level. Deriving the distribution matrix  $\phi$  for this model explicitly, we show order by order that both the heat current  $I^Q$  in Eq. (19) and the entropy current  $I_{\text{tot}}^S$  in Eq. (18) exactly reproduce the absorbed heat  $\dot{Q} = -I_{\text{tot}}^Q$  and the entropy change  $\dot{s} = -I_{\text{tot}}^S$  from the inside perspective [7] (see the Supplemental Material [38]). Thereby, we also explicitly confirm the inside-outside duality of entropy evolution: The dissipated power  $\dot{W}^{(2)}$  was shown to lead to a local increase of entropy for the resonant level in Ref. [7], and we demonstrate here that this is reflected in an additional inflow of entropy  $I_{\text{tot}}^S$

carried by the scattering states, leaving the entropy source-free [Eq. (24)].

*Conclusion.*—We developed a Landauer-Büttiker approach to entropy evolution in strongly coupled fermionic systems, which considers a fully coherent quantum dynamics in combination with coupling to macroscopic equilibrium baths. This formalism naturally avoids the system-bath distinction and is applicable to arbitrary non-interacting electron systems. We showed that the entropy current generated by a dynamic scatterer depends on the correlations between different scattering channels, which are generated in the scattering event. At quasistatic order, the entropy current is just the heat current over temperature, while at next order the dissipation induced by the finite velocity transformation yields a net inflow of entropy into the scattering region. This inflow reflects the dissipation-induced local increase of entropy constituting the inside-outside duality of entropy evolution.

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