

Clausius Inequality and Optimality of Quasistatic Transformations for Nonequilibrium Stationary States

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Nonequilibrium stationary states of thermodynamic systems dissipate a positive amount of energy per unit of time. If we consider transformations of such states that are realized by letting the driving depend on time, the amount of energy dissipated in an unbounded time window then becomes infinite. Following the general proposal by Oono and Paniconi and using results of the macroscopic fluctuation theory, we give a natural definition of a renormalized work performed along any given transformation. We then show that the renormalized work satisfies a Clausius inequality and prove that equality is achieved for very slow transformations, that is, in the quasistatic limit. We finally connect the renormalized work to the quasipotential of the macroscopic fluctuation theory, which gives the probability of fluctuations in the stationary nonequilibrium ensemble.

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A main goal of nonequilibrium thermodynamics is to construct analogues of thermodynamic potentials for nonequilibrium stationary states. These potentials should describe the typical macroscopic behavior of the system as well as the asymptotic probability of fluctuations. As it has been shown in Ref. [1], this program can be implemented without the explicit knowledge of the stationary ensemble and requires as input the macroscopic dynamical behavior of systems that can be characterized by the transport coefficients. This theory, now known as macroscopic fluctuation theory, is based on an extension of Einstein equilibrium fluctuation theory to stationary nonequilibrium states combined with a dynamical point of view. It has been very powerful in studying concrete microscopic models but can be used also as a phenomenological theory. It has led to several new interesting predictions [2–7].

From a thermodynamic viewpoint, the analysis of transformations from one state to another one is most relevant. This issue has been addressed by several authors in different contexts. For instance, following the basic papers [8–10], the case of Hamiltonian systems with finitely many degrees of freedom has recently been discussed in Refs. [11,12], while the case of Langevin dynamics is considered in Ref. [13].

We here consider thermodynamic transformations for driven diffusive systems in the framework of the macroscopic fluctuation theory. With respect to the authors mentioned above, the main difference is that we deal with systems with infinitely many degrees of freedom, and the spatial structure becomes relevant. For simplicity of notation, we restrict to the case of a single conservation law, e.g., the conservation of mass. We thus consider an open

system in contact with boundary reservoirs, characterized by their chemical potential λ , and under the action of an external field E . We denote by $\Lambda \subset \mathbb{R}^d$ the bounded region occupied by the system, by x the macroscopic space coordinates, and by t the macroscopic time. With respect to our previous work [1,3,4], we here consider the case in which λ and E depend explicitly on the time t , driving the system from a nonequilibrium state to another one. The macroscopic dynamics is given by the hydrodynamic equation for the density that satisfies the following general assumptions based on the notion of local equilibrium. For stochastic lattice gases, these assumptions can be proven rigorously and the macroscopic transport coefficient can be characterized in terms of the underlying microscopic dynamics [14].

The macroscopic state is completely described by the local density $u(t, x)$ and the associated current $j(t, x)$. In the sequel, we drop the dependence on the space coordinate x from the notation. The macroscopic evolution is given by the continuity equation

$$\partial_t u(t) + \nabla \cdot j(t) = 0, \quad (1)$$

together with the constitutive equation $j(t) = J(t, u(t))$ expressing the local current in function of the local density. For driven diffusive systems, the constitutive equation takes the form

$$J(t, \rho) = -D(\rho)\nabla\rho + \chi(\rho)E(t), \quad (2)$$

where the diffusion coefficient $D(\rho)$ and the mobility $\chi(\rho)$ are positive matrices. In the case of time-independent driving, the right hand side does not depend explicitly on time, and we denote the current simply by $J(\rho)$.

The transport coefficients in Eq. (2) satisfy the local Einstein relation $D(\rho) = \chi(\rho)f''(\rho)$, where f is the equilibrium free energy per unit of volume. The interaction with the external reservoirs specifies the boundary conditions for the evolution defined by Eqs. (1) and (2). Recalling that $\lambda(t)$ is the chemical potential of the reservoirs, this boundary condition reads $f'(u(t, x)) = \lambda(t, x)$, $x \in \partial\Lambda$. We shall also assume that when λ and E do not depend on time, there is a unique and globally attractive stationary solution for the flow defined by Eqs. (1) and (2) that is denoted by $\bar{\rho} = \bar{\rho}_{\lambda, E}$. In particular, $\bar{\rho}_{\lambda, E}$ is the typical density profile in the stationary nonequilibrium state corresponding to time-independent chemical potential λ and external field E .

We now fix time-dependent paths $\lambda(t)$ of the chemical potential and $E(t)$ of the driving field. Given a density profile ρ , we denote by $(u(t), j(t))$, $t \geq 0$, the solution of Eqs. (1) and (2) with initial condition ρ . Let $W_{[0, T]} = W_{[0, T]}(\lambda, E, \rho)$ be the energy exchanged between the system and the external driving in the time interval $[0, T]$, that is,

$$W_{[0, T]} = \int_0^T dt \left\{ \int_{\Lambda} dx j(t) \cdot E(t) - \int_{\partial\Lambda} d\sigma \lambda(t) j(t) \cdot \hat{n} \right\}, \quad (3)$$

where \hat{n} is the outer normal to $\partial\Lambda$ and $d\sigma$ is the surface measure on $\partial\Lambda$. The first term on the right hand side is the energy provided by the external field, whereas the second term is the energy provided by the reservoirs.

In view of the boundary conditions and the Einstein relation, by using the divergence theorem in Eq. (3), we deduce that

$$W_{[0, T]} = F(u(T)) - F(\rho) + \int_0^T dt \int_{\Lambda} dx j(t) \cdot \chi(u(t))^{-1} j(t), \quad (4)$$

where F is the equilibrium free-energy functional,

$$F(\rho) = \int_{\Lambda} dx f(\rho(x)). \quad (5)$$

Consider two stationary states corresponding to (time-independent) (λ_0, E_0) and (λ_1, E_1) and denote by $\bar{\rho}_0 = \bar{\rho}_{\lambda_0, E_0}$ and $\bar{\rho}_1 = \bar{\rho}_{\lambda_1, E_1}$ the associated density profiles. Such states can be either equilibrium or nonequilibrium states. We can drive the system from the initial state $\bar{\rho}_0$ at time $t = 0$ to the final state $\bar{\rho}_1$ at time $t = +\infty$ by considering a time-dependent forcing $[\lambda(t), E(t)]$ satisfying $(\lambda(0), E(0)) = (\lambda_0, E_0)$ and $(\lambda(+\infty), E(+\infty)) = (\lambda_1, E_1)$. As the second term on the right hand side of Eq. (4) is positive, by letting $W = W_{[0, +\infty]}$ be the total energy exchanged in the transformation, we deduce the Clausius inequality

$$W \geq \Delta F = F(\bar{\rho}_1) - F(\bar{\rho}_0). \quad (6)$$

When the initial and final states are equilibrium states, e.g., the external field E vanishes and the chemical potential λ is constant, the inequality (6) is a standard

formulation of the second law of thermodynamics. Moreover, by considering a sequence of transformations in which the variation of the driving becomes very slow, it is not difficult to show that equality in Eq. (6) is achieved in the quasistatic limit; we refer to Ref. [15] for the details. On the other hand, for nonequilibrium states the inequality (6) does not carry any information. Indeed, as nonequilibrium states support a nonvanishing current, in the limit $T \rightarrow +\infty$, the second term on the right hand side of Eq. (4) becomes infinite so that the left hand side of Eq. (6) is infinite while the right hand side is bounded. By interpreting the ideas in Ref. [16], further developed in Refs. [10, 17], we next define a renormalized work for which a significant Clausius inequality can also be obtained for nonequilibrium stationary states.

To this aim, we recall the quasipotential, which is the key notion of the macroscopic fluctuation theory. Consider a system with time-independent driving and let $[\hat{u}(t), \hat{j}(t)]$, $t \in [T_1, T_2]$ be a pair density current satisfying the continuity equation $\partial_t \hat{u} + \nabla \cdot \hat{j} = 0$. According to the basic principles of the macroscopic fluctuation theory [1, 3, 4], the probability of observing this path is given, up to a prefactor, by $\exp\{-\varepsilon^{-d} \beta I_{[T_1, T_2]}(\hat{u}, \hat{j})\}$ where ε is the scaling parameter, i.e., the ratio between the microscopic length scale (say the typical intermolecular distance) and the macroscopic one, $\beta = 1/\kappa T$ (here T is the temperature and κ is Boltzmann's constant), and the action functional I has the form

$$I_{[T_1, T_2]}(\hat{u}, \hat{j}) = \frac{1}{4} \int_{T_1}^{T_2} dt \int_{\Lambda} dx [\hat{j}(t) - J(\hat{u}(t))] \cdot \chi(\hat{u}(t))^{-1} \times [\hat{j}(t) - J(\hat{u}(t))]. \quad (7)$$

In particular, if (\hat{u}, \hat{j}) solves Eqs. (1) and (2), then $I_{[T_1, T_2]}(\hat{u}, \hat{j}) = 0$. The above statement therefore implies that the typical behavior of the system is described by the hydrodynamic equations. The quasipotential is the functional on the set of density profiles defined by the variational problem

$$V(\rho) = \inf\{I_{(-\infty, 0]}(\hat{u}, \hat{j}), \hat{u}(0) = \rho\}, \quad (8)$$

where the infimum is carried out over all the trajectories satisfying the prescribed boundary condition. Namely, $V(\rho)$ is the minimal action to bring the system from the typical density profile $\bar{\rho}$ to the fluctuation ρ . The probability of a density profile ρ in the stationary nonequilibrium ensemble is then given, up to a prefactor, by $\exp\{-\varepsilon^{-d} \beta V(\rho)\}$. In particular, the minimizer of V is the typical density profile $\bar{\rho}$. For equilibrium states it can be shown [4] that V coincides, apart an affine transformation, with the free-energy functional (5). Moreover, as shown in Ref. [1], the functional V solves the stationary Hamilton-Jacobi equation

$$\int_{\Lambda} dx \nabla \frac{\delta V}{\delta \rho} \cdot \chi(\rho) \nabla \frac{\delta V}{\delta \rho} - \int_{\Lambda} dx \frac{\delta V}{\delta \rho} \nabla \cdot J(\rho) = 0, \quad (9)$$

where $\delta V/\delta\rho$ vanishes at the boundary $\partial\Lambda$ and ρ satisfies the boundary condition $f'(\rho(x)) = \lambda(x)$, $x \in \partial\Lambda$.

In the case of time-independent driving, the current $J(\rho)$ in Eq. (2) can be decomposed as $J(\rho) = J_S(\rho) + J_A(\rho)$, where $J_S(\rho) = -\chi(\rho)\nabla\frac{\delta V}{\delta\rho}$ and $J_A(\rho) = J(\rho) - J_S(\rho)$. In view of the stationary Hamilton-Jacobi Eq. (9), the above decomposition is orthogonal in the sense that for each density profile ρ

$$\int_{\Lambda} dx J_S(\rho) \cdot \chi(\rho)^{-1} J_A(\rho) = 0. \quad (10)$$

We shall refer to $J_S(\rho)$ as the symmetric current and to $J_A(\rho)$ as the antisymmetric current. This terminology refers to symmetric and antisymmetric parts of the microscopic dynamics [1,3]. We remark that J_S is proportional to the thermodynamic force and the above decomposition depends on the external driving.

Since the quasipotential V is minimal in the stationary profile, the above definitions imply that $J_S(\bar{\rho}) = 0$; namely, the stationary current is purely antisymmetric. In particular, $J_A(\bar{\rho})$ is the typical current in the stationary nonequilibrium ensemble associated, and it is therefore experimentally accessible. In view of the general formula (4) for the total work, the amount of energy per unit of time needed to maintain the system in the stationary profile $\bar{\rho}$ is

$$\int_{\Lambda} dx J_A(\bar{\rho}) \cdot \chi(\bar{\rho})^{-1} J_A(\bar{\rho}). \quad (11)$$

We shall next consider time-dependent driving and define a renormalized work by subtracting from the total work the energy needed to maintain the system out of equilibrium. We fix, therefore, $T > 0$, a density profile ρ , and space-time-dependent chemical potentials $\lambda(t)$ and external field $E(t)$, $t \in [0, T]$. Let $[u(t), j(t)]$ be the corresponding solution of Eqs. (1) and (2) with initial condition ρ . Recalling Eq. (11), we define the renormalized work $W_{[0,T]}^{\text{ren}} = W_{[0,T]}^{\text{ren}}(\lambda, E, \rho)$ performed by the reservoirs and the external field in the time interval $[0, T]$ as

$$W_{[0,T]}^{\text{ren}} = W_{[0,T]} - \int_0^T dt \int_{\Lambda} dx J_A(t, u(t)) \cdot \chi(u(t))^{-1} J_A(t, u(t)). \quad (12)$$

In this formula, $W_{[0,T]} = W_{[0,T]}(\lambda, E, \rho)$ is given in Eq. (3),

$$J(t, u) = J_S(t, u) + J_A(t, u),$$

$$J_S(t, u) = -\chi(u)\nabla\frac{\delta V_{\lambda(t), E(t)}(u)}{\delta u}$$

in which u is a generic density profile, $J(t, u)$ is given by Eq. (2), and $V_{\lambda(t), E(t)}$ is the quasipotential relative to the state $[\lambda(t), E(t)]$ with frozen t . Observe that the definition of the renormalized work involves the antisymmetric current $J_A(t)$ computed not at density profile $\bar{\rho}_{\lambda(t), E(t)}$ but at

the solution $u(t)$ of the time-dependent hydrodynamic equation. That is, at time t we subtract the power the system would have dissipated if its actual state $u(t)$ had been the stationary profile corresponding to $[\lambda(t), E(t)]$. This choice, which is certainly reasonable for slow transformations, leads to a Clausius inequality. Indeed, by using Eq. (4) and the orthogonality between the symmetric and the antisymmetric part of the current,

$$W_{[0,T]}^{\text{ren}}(\lambda, E, \rho) = F(u(T)) - F(\rho) + \int_0^T dt \int_{\Lambda} dx J_S(t, u(t)) \cdot \chi(u(t))^{-1} J_S(t, u(t)).$$

Consider a density profile ρ and a space-time-dependent chemical potential and external field $[\lambda(t), E(t)]$, $t \geq 0$, converging to (λ_1, E_1) as $t \rightarrow +\infty$. Let $\bar{\rho}_1 = \bar{\rho}_{\lambda_1, E_1}$ be the stationary profile associated to (λ_1, E_1) and $[u(t), j(t)]$, $t \geq 0$, be the solution of Eqs. (1) and (2) with initial condition ρ . Since $u(T)$ converges to $\bar{\rho}_1$, the symmetric part of the current, $J_S(u(T))$, relaxes as $T \rightarrow +\infty$ to $J_S(\bar{\rho}_1) = 0$. Under suitable assumptions on the transformation, the last integral in the previous formula is convergent as $T \rightarrow +\infty$. By letting $W^{\text{ren}} = \lim_{T \rightarrow \infty} W_{[0,T]}^{\text{ren}}$, we thus get

$$W^{\text{ren}}(\lambda, E, \rho) = F(\bar{\rho}_1) - F(\rho) + \int_0^{\infty} dt \int_{\Lambda} dx J_S(t, u(t)) \cdot \chi(u(t))^{-1} J_S(t, u(t)), \quad (13)$$

where F is the equilibrium free-energy functional (6). In particular,

$$W^{\text{ren}}(\lambda, E, \rho) \geq F(\bar{\rho}_1) - F(\rho), \quad (14)$$

which is a meaningful version of the Clausius inequality for nonequilibrium states. Furthermore, by considering a sequence of transformations $[\lambda(t), E(t)]$, which vary on a time scale $1/\delta$, we realize that the integrand on the second term in the right hand side of Eq. (13) is of order δ^2 while the integral essentially extends, due to the finite relaxation time of the system, over an interval of order δ^{-1} . Therefore, in quasistatic limit $\delta \rightarrow 0$ equality in Eq. (14) is achieved. We refer to Ref. [15] for more details.

For special transformations, the integral in Eq. (13), which represents the excess work over a quasistatic transformation, can be related to the quasipotential. Consider at time $t = 0$ a stationary nonequilibrium profile $\bar{\rho}_0$ corresponding to some driving (λ_0, E_0) . The system is put in contact with new reservoirs at chemical potential λ_1 and a new external field E_1 . For $t > 0$, the system evolves according to the hydrodynamic Eqs. (1) and (2) with initial condition $\bar{\rho}_0$, time-independent boundary condition λ_1 , and external field E_1 . In particular, as $t \rightarrow +\infty$ the system relaxes to $\bar{\rho}_1$, the stationary density profile corresponding to (λ_1, E_1) . A simple calculation shows that along such a path

$$\int_0^\infty dt \int_\Lambda dx J_S(t, u(t)) \cdot \chi(u(t))^{-1} J_S(t, u(t)) \\ = V_{\lambda_1, E_1}(\bar{\rho}_0) - V_{\lambda_1, E_1}(\bar{\rho}_1) = V_{\lambda_1, E_1}(\bar{\rho}_0). \quad (15)$$

The quasipotential $V_{\lambda_1, E_1}(\bar{\rho}_0)$ thus represents the excess work, with respect to a quasistatic transformation, along the path that solves Eqs. (1) and (2) with initial condition $\bar{\rho}_0$ and time-independent driving (λ_1, E_1) .

To connect the above result with classical equilibrium thermodynamics, consider an equilibrium state with vanishing external field and constant chemical potential λ_0 and let $\bar{\rho}_0$ be the corresponding homogeneous density, i.e., $\lambda_0 = f'(\bar{\rho}_0)$. The system is put in contact with a new environment with chemical potential λ_1 . In this case, recalling that f is the free energy per unit of volume and that the temperature of the system is the same of the environment, the availability per unit of volume is defined (see Ref. [18] Ch. 7) pippard) by $a = f(\bar{\rho}_0) - \lambda_1 \bar{\rho}_0$. The function a , which depends on state of the system and the environment, can be used to compute the maximal useful work that can be extracted from the system in the given environment. More precisely, by letting $\bar{\rho}_1$ be such that $f'(\bar{\rho}_1) = \lambda_1$, then $-\Delta a = f(\bar{\rho}_0) - f(\bar{\rho}_1) - \lambda_1(\bar{\rho}_0 - \bar{\rho}_1) \geq 0$ is the maximal useful work per unit of volume that can be extracted from the system in the given environment. By computing the quasipotential for equilibrium states (see Ref. [4]), we get $V_{\lambda_1, 0}(\bar{\rho}_0) = -|\Lambda| \Delta a$. Therefore, while a definition of thermodynamic potentials, that is, functionals of the state of the system, does not appear possible in nonequilibrium thermodynamics, the quasipotential is the natural extension of the availability.

In terms of the underlying microscopic ensembles, as discussed in Ref. [15], the quasipotential $V_{\lambda_1, E_1}(\bar{\rho}_0)$ can be obtained by computing the relative entropy of the ensemble associated to (λ_0, E_0) with respect to the one associated to (λ_1, E_1) . By consideration of a Markovian model for such underlying dynamics, it is also possible to give a microscopic definition of the exchanged work that in the hydrodynamic scaling limit converges to Eq. (3). The corresponding fluctuations can be deduced from those of the empirical current [3].

The definition of renormalized work we have introduced is natural and ensures, as we have discussed, both its finiteness and the validity of a Clausius inequality. From an operational point of view, the quasipotential, a generically nonlocal quantity, can be obtained from the measurement of the density correlation functions. In fact, V is the Legendre transform of the generating functional of density correlation functions [1]. On the other hand, the identity $W^{\text{ren}} = \Delta F$, which is achieved for quasistatic transformations, requires the knowledge of the total current in the intermediate stationary states that can be directly measured.

One may ask whether there exist, with respect to Eq. (12), alternative renormalizations of the total work. For instance,

in the recent work of Ref. [19], Maes and Netocny considered the topic of a renormalized Clausius inequality in the context of a single Brownian particle in a time-dependent environment. To compare the approach in Ref. [19] to the present one, consider N independent diffusions in the thermodynamic limit $N \rightarrow \infty$. Each diffusion solves the Langevin equation $\dot{X} = E(t, X) + \sqrt{2}\dot{w}$, where E is a time-dependent vector field and \dot{w} denotes white noise. The corresponding stationary measure with E frozen at time t is denoted by $\exp\{-v(t, x)\}$. The scheme discussed here can be now applied, the hydrodynamic equations are given by Eqs. (1) and (2) with $D = 1$ and $\chi(\rho) = \rho$. Our renormalized work is given by Eq. (12) with $J_A(t, \rho) = \rho[E(t, x) + \nabla v(t, x)]$. The renormalization introduced in Ref. [19] is instead obtained by introducing a potential field such that the corresponding stationary state has minimal entropy production. Namely, they write $E = f - \nabla U$ and subtract from the energy exchanged the space-time integral of $|J_t^\varphi|^2/\rho$ where $J_t^\varphi = \rho(f - \nabla\varphi) - \nabla\rho$ and $\varphi = \varphi(t, x; \rho)$ is chosen so that $\nabla \cdot J_t^\varphi = 0$. While the two renormalization schemes are different, both satisfy the Clausius inequality (14) with $F(\rho) = \int dx \rho \log \rho$. Observe that in this case of independent particles, our renormalization is local whereas the dependence of J_t^φ on ρ is nonlocal. It is not clear to us how the approach in Ref. [19] can be generalized to cover the case of interacting particles in the hydrodynamic scaling limit.

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