

Entropy production of diffusion in spatially periodic deterministic systemsJ. R. Dorfman,^{1,*} P. Gaspard,^{2,†} and T. Gilbert^{3,‡}¹*Department of Physics and Institute for Physical Science and Technology, University of Maryland, College Park, Maryland 20742*²*Center for Nonlinear Phenomena and Complex Systems, Université Libre de Bruxelles, Code Postal 231, Campus Plaine, B-1050 Brussels, Belgium*³*Department of Chemical Physics, The Weizmann Institute of Science, Rehovot 76100, Israel*

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This paper presents an *ab initio* derivation of the expression given by irreversible thermodynamics for the rate of entropy production for different classes of diffusive processes. The first class is Lorentz gases, where noninteracting particles move on a spatially periodic lattice, and collide elastically with fixed scatterers. The second class is periodic systems, where N particles interact with each other, and one of them is a tracer particle that diffuses among the cells of the lattice. We assume that, in either case, the dynamics of the system are deterministic and hyperbolic, with positive Lyapunov exponents. This work extends methods originally developed for a chaotic two-dimensional model of diffusion, the multi-baker map, to higher-dimensional, continuous-time dynamical systems appropriate for systems with one or more moving particles. Here we express the rate of entropy production in terms of hydrodynamic measures that are determined by the fractal properties of microscopic hydrodynamic modes that describe the slowest decay of the system to an equilibrium state.

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I. INTRODUCTION

In 1902, Gibbs described a mechanism by which the entropy could increase toward its equilibrium value in mechanical systems that obey Newton's equations [1]. Gibbs' mechanism is based on the assumption that the microscopic dynamics is mixing. The mixing would allow coarse-grained probabilities to reach their equilibrium values after a long time, a result that has taken on rigorous meaning in light of the modern definition of mixing [2]. The second ingredient of Gibbs' mechanism is the assumption that the entropy of a physical system should be defined as a quantity that is now referred to as the coarse-grained entropy. The use of the coarse-grained entropy could be justified by the fact that, if the entropy should be given according to Boltzmann by the logarithm of the number of complexities of a system, then it can only be defined by introducing cells of nonvanishing size in systems described by continuous coordinates [3].

The aim of the present paper is to apply the program set up by Gibbs to hyperbolic, deterministic dynamical systems sustaining a transport process of diffusion. We assume that the systems obey Liouville's theorem, namely, that phase space volumes are preserved by the dynamics, which is a major assumption used by Gibbs. Specifically, the systems we consider are either periodic Lorentz gases, in which moving particles diffuse through a lattice interacting only with fixed scatterers, or are periodic repetitions of interacting N -particle systems such that a tagged particle is followed as it undergoes diffusion among the unit cells. The present work is an extension to continuous time, and interacting systems, of previous works [4–8] concerned with the multi-baker map as a chaotic model of diffusion. In the previous work,

we used the fact that an initial nonequilibrium distribution function, in the multi-baker "phase space," rapidly develops a fractal structure due to the chaotic nature of the dynamics. This structure is such that variations of the distribution function on arbitrarily fine scales develop as the system evolves in time. The final stages of the approach to equilibrium are then controlled by the decay of fractal, microscopic hydrodynamic modes of the system, in this case diffusive modes, which decay with time as $\exp(-Dk^2t)$, where k is a wave number characterizing a particular mode associated with a macroscopic density variation over a distance of order k^{-1} , D is the diffusion coefficient, and t is the time. For the multi-baker system it is possible to express the rate of entropy production in this final stage in terms of measures of sets that are determined by the nonequilibrium phase-space distribution in the set, in particular, by the values of the fractal hydrodynamic modes in the set.

In this paper we show that it is possible to apply the same methods to calculate the rate of entropy production for diffusive flows in periodic Lorentz gases and for tracer diffusion in periodic, interacting N -particle systems as long as the microscopic dynamics is deterministic, mixing, and chaotic. Our method is based on the explicit construction of the microscopic fractal hydrodynamic modes of diffusion, which characterize the long-time relaxation of the system towards thermodynamic equilibrium. Our main result is that we obtain by this method exactly the expression for the rate of entropy production as given by irreversible thermodynamics for these systems [9]. The source of this agreement can be traced to the role played by the fractal hydrodynamic modes, both for requiring a coarse graining of the phase space to properly incorporate the effects of their fractal properties on entropy production in the system, as well as for describing the slowest decay of the system as it relaxes towards equilibrium.

The plan of the paper is the following. The mathematical methods needed to describe spatially periodic systems are

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provided in Sec. II. Then the nonequilibrium distribution is defined in Sec. III. Once the distribution function has been constructed, we can identify the microscopic hydrodynamic modes of diffusion, and they are then constructed in Sec. IV. Here we use the properties of these modes to introduce the notion of a hydrodynamic measure of a set in phase space, and we also identify the sets that are used in the coarse graining of the phase space. These sets are not arbitrary, but must have some specific properties in order to be useful for the calculation of the rate of entropy production, which is carried out in Sec. V. In Sec. VI we conclude with a discussion of the method and results obtained here, and with an outline of directions for future work.

II. SPATIALLY PERIODIC SYSTEMS

A. The phase space

We consider a deterministic dynamical system of phase-space dimension M , which is spatially periodic in the form of a d -dimensional lattice \mathcal{L} . We will label the positions of the periodic cells \mathcal{M} on the lattice by the vector $\vec{l} \in \mathcal{L}$, and the phase-space coordinates within an elementary cell by $X \in \mathcal{M}$. The lattice \mathcal{L} is isomorphic to \mathbb{Z}^d . The total phase space of the system is the direct product $\mathcal{M} \otimes \mathcal{L}$ of dimension M . The time displacement operator over a time interval t , acting on points (\vec{l}, X) , is denoted by Φ^t , which is also called the *flow*. On the other hand, the time displacement operator acting on points X inside the basic unit cell is denoted by ϕ^t .

Examples of such systems are the following.

1. The two-dimensional Lorentz gas

In this system a point particle moves in free flight and undergoes elastic collisions on hard disks forming a triangular lattice. The phase space is defined by the spatial and velocity coordinates (\vec{r}, \vec{v}) of the moving particle. The spatial coordinates vary over the plane excluding the area occupied by the disks: $\vec{r} \in \mathcal{Q}$ with $\dim \mathcal{Q} = 2$. The velocity coordinates form another two-dimensional vector $\vec{v} \in \mathbb{R}^2$. Energy is conserved during the motion so that each energy shell is preserved by the dynamics. In each energy shell, we can thus reduce the coordinates to the two positions $\vec{r} = (x, y) \in \mathcal{Q}$ and the velocity angle $\varphi \in [0, 2\pi[$. The position space decomposes into a triangular lattice of hexagonal elementary cells, each containing a single disk: $\mathcal{Q} = \cup_{\vec{l} \in \mathcal{L}} \mathcal{T}^{\vec{l}} \mathcal{C}$, where $\mathcal{T}^{\vec{l}}$ denotes the translation by the lattice vector \vec{l} , and \mathcal{C} is the elementary cell of the position space \mathcal{Q} . The elementary cell of the phase space thus has the coordinates $X = (x, y, \varphi) \in \mathcal{M} = \mathcal{C} \otimes [0, 2\pi[$. In the two-dimensional Lorentz gas, the phase space has the dimension $\dim \mathcal{M} = 3$ while the lattice has the dimension $d = \dim \mathcal{L} = 2$. The flow of the hard-disk Lorentz gas preserves the Lebesgue measure $dX = dx dy d\varphi$. We notice that the horizon of the hard-disk Lorentz gas must be finite in order for the diffusion coefficient to be finite [10], which we assume in the following.

Similar considerations apply to the Lorentz gas in which a point particle moves in a periodic lattice of attractive Yukawa potentials. In this system the diffusion coefficient is positive

and finite for high enough energy [11]. Here, too, the phase space has three dimensions and is the union of the constant energy surfaces for each cell of the lattice.

2. The three-dimensional Lorentz gas

This Lorentz gas is the direct generalization of the two-dimensional one. The spatial coordinates vary over the space with the exclusion of the volume occupied by the spheres: $\vec{r} \in \mathcal{Q}$ with $\dim \mathcal{Q} = 3$. The velocity coordinates form another three-dimensional vector $\vec{v} \in \mathbb{R}^3$. In each energy shell, we can reduce the coordinates to the three positions $\vec{r} = (x, y, z) \in \mathcal{Q}$ and the two velocity spherical angles $\theta \in [0, \pi]$ and $\varphi \in [0, 2\pi[$. The position space decomposes into a lattice of elementary cells containing one or several disks: $\mathcal{Q} = \cup_{\vec{l} \in \mathcal{L}} \mathcal{T}^{\vec{l}} \mathcal{C}$. The elementary cell of the phase space has thus the coordinates $X = (x, y, z, \cos \theta, \varphi) \in \mathcal{M} = \mathcal{C} \otimes [-1, +1] \otimes [0, 2\pi[$, where, again, \mathcal{C} is the elementary cell of the position space \mathcal{Q} . In the three-dimensional Lorentz gas, the phase space has the dimension $\dim \mathcal{M} = 5$ while the lattice has the dimension $d = \dim \mathcal{L} = 3$. The flow of the hard-disk Lorentz gas preserves the Lebesgue measure $dX = dx dy dz d\cos \theta d\varphi$. Here, also, we suppose that the horizon is finite in order for the diffusion coefficient to be finite.

3. Diffusion of a tracer in a system on a torus

The molecular dynamics simulation of the diffusion of a tracer particle moving in a fluid can be performed by considering a finite number of particles modeling the fluid and the tracer particle, all of them moving with interactions, in a domain delimited by periodic boundary conditions. The total number of particles is equal to N . The center of mass can be taken at rest. The vector \vec{l} can be used to locate the position of the cell containing the tracer particle as it moves on the checkerboard lattice made of infinitely many images of the system, which tiles the d -dimensional space of the system. Then the diffusion coefficient of the tracer particle can be computed by adding the appropriate lattice vector to \vec{l} each time the tracer particle crosses a boundary. The energy and total momentum are to be conserved so that here X denotes the phase-space coordinates of an elementary cell of the phase space, which is of dimension $M = \dim \mathcal{M} = 2dN - 2d - 1$ after elimination of the d degrees of freedom of the center of mass, while the lattice is of dimension $d = \dim \mathcal{L} = 2$ or 3.

In summary, we will denote by X the coordinates on an energy-momentum shell of a microcanonical ensemble for the periodic system. We suppose that the flow Φ^t preserves the Lebesgue measure dX . Moreover, we assume that the diffusion coefficient of the system is finite, the mean drift vanishes, and the microscopic dynamics is chaotic and mixing. In the sequel \vec{l} will denote the lattice position vector and \mathcal{T} will denote the translation operator on the lattice. Subsets of the unit cell \mathcal{M} will be denoted by capital roman letters. The notation A will be used for a set belonging to the elementary phase-space cell at the origin $\vec{l} = 0$ of the lattice. The notation $A_{\vec{l}} = \mathcal{T}^{\vec{l}} A$ will be used when we want to refer to a set in a specific lattice cell at position \vec{l} .

B. Lattice Fourier transforms

We will also need to define lattice Fourier transforms [12]. We will need the preliminary result.

Definition 1. Consider a function $G(\vec{l}, X)$, which is a function of the lattice coordinate \vec{l} , and the unit cell coordinate X . This function can then be expressed in terms of a lattice Fourier transform as

$$G(\vec{l}, X) = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} e^{i\vec{k} \cdot \vec{l}} \tilde{G}(\vec{k}, X), \quad (1)$$

where \mathcal{B} denotes the first Brillouin zone of the reciprocal lattice, $|\mathcal{B}|$ is its volume, and \tilde{G} is the lattice Fourier transform of G .

It is important to note the following.

Lemma 1. If G is only a function of the lattice vector \vec{l} , then \tilde{G} does not depend upon the unit cell coordinate X , but only upon \vec{k} .

III. THE NONEQUILIBRIUM DISTRIBUTION

We consider a periodic deterministic dynamical system with a finite diffusion coefficient.

The nonequilibrium measure

We now construct the statistical ensemble that we will use for the rest of this paper. We assume that the coordinates of the ensemble are distributed on the lattice in such a way that the distribution can be described by an initial ensemble density $\rho(\vec{l}, X, 0)$, where ρ denotes the number of systems per unit phase-space volume. We take this density to be close to that of total equilibrium and write it in the form

$$\rho(\vec{l}, X, 0) = \rho_{\text{eq}} [1 + R(\vec{l}, X, 0)], \quad (2)$$

where the equilibrium distribution ρ_{eq} is, for Lorentz gases, uniform with respect to the cells \vec{l} , and with respect to the phase variables X in agreement with the assumption that the Lebesgue measure dX is preserved by the flow Φ^t . For the case of tracer diffusion, ρ_{eq} is the equilibrium microcanonical distribution for the N particles in a cell on the lattice. The initial deviation from equilibrium in the cell located at \vec{l} is denoted by $R(\vec{l}, X, 0)$, which we assume to be Lebesgue integrable when weighted with the equilibrium distribution. Using the lattice Fourier transform, Eq. (1), we can express this deviation in the form

$$R(\vec{l}, X, 0) = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} e^{i\vec{k} \cdot \vec{l}} \tilde{R}(\vec{k}, X). \quad (3)$$

The phase-space density (2) leads us to the definition of the nonequilibrium measure of a set $A_{\vec{l}}$ belonging to the phase-space cell $\mathcal{T}^{\vec{l}} \mathcal{M}$ corresponding to the lattice vector \vec{l} .

Definition 2. The equilibrium measure $\nu(A_{\vec{l}})$ of a set $A_{\vec{l}}$ is defined by

$$\nu(A_{\vec{l}}) \equiv \int_{A_{\vec{l}}} dX \rho_{\text{eq}}, \quad (4)$$

and the nonequilibrium measure, $\mu_t(A_{\vec{l}})$, of the same set is defined by

$$\mu_t(A_{\vec{l}}) \equiv \int_{A_{\vec{l}}} dX \rho(\vec{l}, X, t) = \nu(A_{\vec{l}}) + \delta\mu_t(A_{\vec{l}}), \quad (5)$$

where

$$\delta\mu_t(A_{\vec{l}}) = \int_{A_{\vec{l}}} dX \rho_{\text{eq}} R(\vec{l}, X, t). \quad (6)$$

We will simplify matters a bit by assuming that the initial deviation from equilibrium $R(\vec{l}, X, 0)$ depends only upon the cell \vec{l} , but not on the initial phase X of the system within the cell. In this case the Fourier transform $\tilde{R}(\vec{k}, X)$ does not depend upon X either, and will henceforth be denoted by $F_{\vec{k}}$.

The time-dependent distribution function $\rho(\vec{l}, X, t)$ is the solution of Liouville's equation and is given by

$$\begin{aligned} \rho(\vec{l}, X, t) &= \rho_{\text{eq}} [1 + R(\vec{l}, X, t)], \\ &= \rho_{\text{eq}} \left\{ 1 + \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \tilde{L}[\Phi^{-t}(\vec{l}, X, 0)]} \right\}. \end{aligned} \quad (7)$$

Here \tilde{L} denotes the projection on the lattice coordinate, thus $\tilde{L}[\Phi^{-t}(\vec{l}, X, 0)]$ is the lattice vector of the cell in which a moving particle would be located at time $-t$ if it were in cell \vec{l} at time $t=0$ with phase X . We can express the time-dependent deviation from total equilibrium in the form

$$R(\vec{l}, X, t) = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot [\vec{l} + \vec{d}(X, t)]}, \quad (8)$$

where the *backward* displacement of the lattice vector \vec{l} over a time interval t is defined by

$$\vec{d}(X, t) = \tilde{L}[\Phi^{-t}(\vec{l}, X, 0)] - \vec{l}. \quad (9)$$

We will make heavy use of the fact that $\vec{d}(X, t)$ depends upon the time interval $(-t, 0)$ and upon the phase point X , at the initial time, but not upon the initial cell \vec{l} . In other words, the periodicity of the lattice and the dynamics produce a ‘‘winding number’’ $\vec{d}(X, t)$ that does not depend upon the cell in which the trajectory is located at the initial time. Here we remark that the long time limit of the displacement vector $\vec{d}(X, t)$ may be a wildly varying function of the phase coordinate X . Thus, we expect that the decaying modes of the time-dependent distribution (7) are singular functions of the phase coordinates.

We define the time-dependent density of the tracer particle by integrating the phase-space density over the coordinates X of an elementary phase space cell of the lattice [13]:

$$n(\vec{l}, t) \equiv \int_{\mathcal{M}} dX \rho(\vec{l}, X, t) = \mu_t(\mathcal{M}_{\vec{l}}). \quad (10)$$

Using Eq. (8), we obtain the density as

$$n(\vec{l}, t) = \nu(\mathcal{M}) \left[1 + \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} \times \frac{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}}} \right]. \quad (11)$$

We consider times t that are long compared to the mean time between collisions of the moving particles, but that are short compared to the time needed for the system to relax to total equilibrium. For such times, we expect the time-dependent deviation from total equilibrium to decay exponentially with a rate $-s_k$ given by the van Hove relation [14]

$$s_k \equiv \lim_{t \rightarrow \infty} \frac{1}{t} \ln \langle e^{i\vec{k} \cdot \vec{d}(X, t)} \rangle_{\mathcal{M}},$$

$$= \lim_{t \rightarrow \infty} \frac{1}{t} \ln \frac{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}}}, \quad (12)$$

which gives the decay rate of a hydrodynamic mode of diffusion of wave number \vec{k} . An expansion in powers of the wave number gives

$$s_k = -D\vec{k}^2 + O(\vec{k}^4) \quad (13)$$

with diffusion coefficient D . Here we have assumed that the diffusive motion of the tracer particle is invariant under space inversion so that all the odd powers of the wave number vanish. We notice that the existence of the successive terms of the expansion in powers of the wave number depends on the existence of the super-Burnett and higher diffusion coefficients [15], which has been recently proved for the hard-disk periodic Lorentz gas with a finite horizon [16].

The definition (12) shows that

$$\frac{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}}} = C(\vec{k}, t) e^{s_k t}, \quad (14)$$

where $C(\vec{k}, t)$ is a function of the wave number \vec{k} with a subexponential dependence on time, i.e.,

$$\lim_{t \rightarrow \infty} \frac{1}{t} \ln C(\vec{k}, t) = 0. \quad (15)$$

Accordingly, the tracer density can be written as

$$n(\vec{l}, t) = \nu(\mathcal{M}) \left[1 + \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} C(\vec{k}, t) e^{s_k t} \right]. \quad (16)$$

Notice that $n(\vec{l}, t)$ obeys a form of the diffusion equation appropriate for our lattice system, given by

$$\frac{\partial n(\vec{l}, t)}{\partial t} = \frac{\nu(\mathcal{M})}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} C(\vec{k}, t) s_k e^{s_k t} + \dots$$

$$= -D \frac{\nu(\mathcal{M})}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} \vec{k}^2 e^{i\vec{k} \cdot \vec{l}} C(\vec{k}, t) e^{s_k t} + \dots, \quad (17)$$

which—in the scaling limit where the size of the unit cell becomes small, and for large times and small wave numbers—is the diffusion equation

$$\frac{\partial n(\vec{l}, t)}{\partial t} \simeq D \frac{\partial^2 n(\vec{l}, t)}{\partial \vec{l}^2}. \quad (18)$$

IV. THE HYDRODYNAMIC MODES

A. The hydrodynamic measures

Spatially periodic deviations from total equilibrium characterized by the wave number \vec{k} relax exponentially at the rate given by van Hove's relation [14]. Here our purpose is to determine the nonequilibrium state corresponding to this mode of exponential relaxation. This state can be defined as a measure, which we call a hydrodynamic measure, associated with the hydrodynamic mode of diffusion. It is the microscopic analog of the solutions $\exp(i\vec{k} \cdot \vec{l} - Dk^2 t)$ of wave number \vec{k} for the macroscopic diffusion equation (18).

We introduce the hydrodynamic measures by considering the deviations from the equilibrium measure for a set $A_{\vec{l}}$:

$$\delta\mu_t(A_{\vec{l}}) = \int_{A_{\vec{l}}} dX \rho_{\text{eq}} R(\vec{l}, X, t),$$

$$= \int_{A_{\vec{l}}} dX \rho_{\text{eq}} \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot [\vec{l} + \vec{d}(X, t)]}$$

$$= \frac{\nu(\mathcal{M})}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} \frac{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}}}$$

$$\times \frac{\int_{A_{\vec{l}}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}, \quad (19)$$

where we have used the property that the backward displacement $\vec{d}(X, t)$ is independent of the initial lattice vector \vec{l} so

that the integral over the set $A_{\vec{l}}$ is equal to the integral over set A of the elementary cell at the origin of the lattice. In the last line, we have factorized the exponential decay according to Eq. (14), which is independent of set A , from a further factor, which depends on set A but that is expected to have a well-defined limit for $t \rightarrow \infty$, because both its numerator and denominator are expected to decay exponentially as $\exp(s_k t)$. This observation is the motivation for the following definition.

Definition 3. The hydrodynamic measure $\chi_{\vec{k}}(A, t)$ is defined by

$$\chi_{\vec{k}}(A, t) \equiv \nu(\mathcal{M}) \frac{\int_A dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}{\int_{\mathcal{M}} dX \rho_{\text{eq}} e^{i\vec{k} \cdot \vec{d}(X, t)}}. \quad (20)$$

We emphasize that the hydrodynamic measures are independent of the cell location \vec{l} . We notice that the hydrodynamic measures are complex measures because of the lattice Fourier transform.

One important property of the hydrodynamic measures is that the total hydrodynamic measure of a unit cell is constant in time, as follows from Definition 3. That is,

Lemma 1. For the set $A = \mathcal{M}$, i.e., the phase-space region associated with an entire unit cell, the hydrodynamic measure is

$$\chi_{\vec{k}}(\mathcal{M}, t) = \nu(\mathcal{M}). \quad (21)$$

Another observation follows.

Lemma 2. If we make a \vec{k} -expansion of the hydrodynamic measure of the form

$$\chi_{\vec{k}}(A, t) = \nu(A) + i\vec{k} \cdot \vec{T}(A, t) + \vec{k}\vec{k} : \vec{g}(A, t) + \dots, \quad (22)$$

it follows from Eq. (21) that $\vec{T}(\mathcal{M}, t) = 0$ and that $\vec{g}(\mathcal{M}, t) = 0$, etc.

Properties like these have already been used in the various symmetric, multi-baker models [4–7]. For a system with vanishing mean drift $\langle \vec{d}(X, t) \rangle_{\mathcal{M}} = 0$, the two first coefficients of the \vec{k} expansion of the hydrodynamic measures can be expressed as

$$\vec{T}(A, t) = \nu(A) \langle \vec{d}(X, t) \rangle_A \quad (23)$$

$$\vec{g}(A, t) = \frac{1}{2} \nu(A) [\langle \vec{d}(X, t) \vec{d}(X, t) \rangle_{\mathcal{M}} - \langle \vec{d}(X, t) \rangle_A \langle \vec{d}(X, t) \rangle_A] \quad (24)$$

with the definition

$$\langle \rangle_A \equiv \frac{\int_A dX \rho_{\text{eq}}(\cdot)}{\int_A dX \rho_{\text{eq}}}. \quad (25)$$

Thanks to the hydrodynamic measures, Eq. (20) and Eq. (14), we finally derive from Eq. (19) an expression for the measure of a set A in cell $\mathcal{M}_{\vec{l}}$:

$$\begin{aligned} \mu_t(A_{\vec{l}}) &= \nu(A) + \delta\mu_t(A_{\vec{l}}), \\ &= \nu(A) + \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} C(\vec{k}, t) e^{s_k t} \chi_{\vec{k}}(A, t). \end{aligned} \quad (26)$$

B. Conservation of measure and de Rham-type equation

Since the time evolution is a measure-preserving Liouville operator, the measure of any set A remains constant as the set follows the motion of the system in phase space. Therefore, we may express this conservation of measure as

$$\mu_{t+\tau}(A) = \mu_t(\Phi^{-\tau} A). \quad (27)$$

Here τ denotes some time interval, and $\Phi^{-\tau} A$ is the pre-image of set A under the flow, obtained by following the backward evolution of the points of A over a time interval τ . This simple result has some important consequences, among them a de Rham-type equation for the hydrodynamic measures.

We assume that sets A are sufficiently small that all the points in them will flow through the same sequence of cells over some large time interval ($-\mathcal{T} \leq \tau \leq \mathcal{T}$). In such a case, the set of points $\Phi^{-\tau} A$ are all in the *same* cell with location denoted by $\vec{l} + \vec{d}(X_A, \tau)$, which is determined by the backward evolution of an arbitrary phase point X_A in set A . Using Eq. (26), we can express the application of Eq. (27) to a set $A_{\vec{l}}$ as

$$\begin{aligned} &\frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot \vec{l}} C(\vec{k}, t + \tau) e^{(t+\tau)s_k} \chi_{\vec{k}}(A, t + \tau) \\ &= \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} d\vec{k} F_{\vec{k}} e^{i\vec{k} \cdot [\vec{l} + \vec{d}(X_A, \tau)]} C(\vec{k}, t) e^{s_k t} \chi_{\vec{k}}(\Phi^{-\tau} A, t). \end{aligned} \quad (28)$$

Since this equation must be true for all cells \vec{l} for all $F_{\vec{k}}$, and for all sets A satisfying the above condition, the only way it can be satisfied is if the integrands are equal almost everywhere. Equating the integrands leads to the equation

$$C(\vec{k}, t + \tau) e^{s_k \tau} \chi_{\vec{k}}(A, t + \tau) = e^{i\vec{k} \cdot \vec{d}(X_A, \tau)} C(\vec{k}, t) \chi_{\vec{k}}(\Phi^{-\tau} A, t). \quad (29)$$

Since the hydrodynamic measures do not depend on the lattice vector, the pre-image $\Phi^{-\tau} A$ under the full flow over the lattice can be reduced to the pre-image $\phi^{-\tau} A$ under the flow defined with periodic boundary conditions inside the elementary cell \mathcal{M} at $\vec{l} = 0$.

Now we assume that the dynamics is hyperbolic in order to assert that the hydrodynamic measures $\chi_{\vec{k}}(A, t)$ reach asymptotic forms exponentially rapidly, that is, on a time scale of the order of the inverse of the positive Lyapunov exponent for the system. In this case, we can replace the hydrodynamic measures in Eq. (29) by their asymptotic forms, denoted by $\chi_{\vec{k}}(A)$, in the long time limit $t \rightarrow \infty$. Moreover, we have the property that

$$\lim_{t \rightarrow \infty} \frac{C(\vec{k}, t + \tau)}{C(\vec{k}, t)} = 1 \quad (30)$$

as a consequence of the subexponential behavior (15) of the functions $C(\vec{k}, t)$.

In the long time limit $t \rightarrow \infty$, combining Eqs. (29) and (30) we obtain the following.

Lemma 3.3. The hydrodynamic measures satisfy a de Rham-type equation:

$$e^{s_k \tau} \chi_{\vec{k}}(A) = e^{i\vec{k} \cdot \vec{d}(X_{\phi^{\tau} A}, \tau)} \chi_{\vec{k}}(\phi^{-\tau} A). \quad (31)$$

Explicit solutions of this equation have been found for multi-baker maps [8,17,18]. For the hard-disk Lorentz gas, these solutions lead to the cumulative functions constructed in Ref. [19] with one-dimensional sets A and in Ref. [12] with two-dimensional sets A . An alternative form of Eq. (31) is

$$e^{s_k \tau} \chi_{\vec{k}}(\phi^{\tau} A) = e^{i\vec{k} \cdot \vec{d}(X_{\phi^{\tau} A}, \tau)} \chi_{\vec{k}}(A). \quad (32)$$

Equation (32) has an expansion in powers of the wave number \vec{k} that will be useful in the calculation of the rate of entropy production. In obtaining these expansions we will make use of the \vec{k} expansions of the $\chi_{\vec{k}}$ functions given in Eq. (22). The wave number expansion of Eq. (32) leads to the following equation for terms of order \vec{k} :

$$\vec{k} \cdot \vec{T}(\phi^{\tau} A) = \vec{k} \cdot \vec{T}(A) + \nu(A) \vec{k} \cdot \vec{d}(X_{\phi^{\tau} A}, \tau). \quad (33)$$

C. Partition of phase space and sum rules

We consider a partition $\{A_j\}$ of the elementary cell \mathcal{M} at $\vec{l}=0$ of the phase space into disjoint sets A_j :

$$\cup_{A_j \subset \mathcal{M}} A_j = \mathcal{M}, \quad A_i \cap A_j = \emptyset, \forall i, j, i \neq j. \quad (34)$$

We notice that the images $\phi^{\tau} A_j$ also form a partition of the elementary cell of phase space:

$$\mathcal{M} = \cup_{A_j \subset \mathcal{M}} \phi^{\tau} A_j. \quad (35)$$

We can apply the de Rham-type equation (32) to one set A_j of the partition (34) and sum both members of Eq. (32) over all the sets $A_j \in \mathcal{M}$ to obtain

$$e^{s_k \tau} \sum_j \chi_{\vec{k}}(\phi^{\tau} A_j) = \sum_j e^{i\vec{k} \cdot \vec{d}_j} \chi_{\vec{k}}(A_j), \quad (36)$$

with the notation

$$\vec{d}_j \equiv \vec{d}(X_{\phi^{\tau} A_j}, \tau). \quad (37)$$

Since the sets $\phi^{\tau} A_j$ form a partition of \mathcal{M} into disjoint sets, we infer from Eq. (21) that

$$\sum_j \chi_{\vec{k}}(\phi^{\tau} A_j) = \chi_{\vec{k}}(\mathcal{M}) = \nu(\mathcal{M}), \quad (38)$$

so that Eq. (36) becomes

$$\nu(\mathcal{M}) e^{s_k \tau} = \sum_j e^{i\vec{k} \cdot \vec{d}_j} \chi_{\vec{k}}(A_j). \quad (39)$$

Now, we perform a wave number expansion of both members of Eq. (39) using Eqs. (13) and (22). Using the properties

$$\sum_j \nu(A_j) = \nu(\mathcal{M}), \quad (40)$$

$$\sum_j \vec{T}(A_j) = \vec{T}(\mathcal{M}) = 0, \quad (41)$$

$$\sum_j \vec{g}(A_j) = \vec{g}(\mathcal{M}) = 0, \quad (42)$$

the identification of the terms that are of the first and second powers of the wave number \vec{k} gives us the following two sum rules:

$$\sum_j \vec{d}_j \nu(A_j) = 0, \quad (43)$$

$$\sum_j [\vec{d}_j \vec{T}(A_j) + \vec{T}(A_j) \vec{d}_j + \vec{d}_j \vec{d}_j \nu(A_j)] = 2 D \tau \nu(\mathcal{M}) \vec{l}. \quad (44)$$

Equation (44) is fundamental for the following development because it constitutes a sum rule relating the diffusion coefficient to the first coefficients $\vec{T}(A_j)$ of the wave number expansion of the hydrodynamic measures, which is linear in the wave number \vec{k} . The measures $\vec{T}(A_j)$ have been interpreted elsewhere as the stationary nonequilibrium measures associated with a gradient of concentration of tracer particles across the system. In the case of the multi-baker maps, $\vec{T}(A_j)$ is given by the difference of the Takagi function at both ends of the one-dimensional sets A_j [7]. The sum rule (44) thus relates the diffusion coefficient to the generalization of the Takagi function for the present system.

V. ENTROPY PRODUCTION

A. Definitions

In this section we are going to calculate of the rate of irreversible entropy production over a time τ , assuming that $t \gg \tau$. For our calculation of the rate of entropy production in a unit cell of the periodic lattice, we use a partition of the total phase space into the small disjoint sets A_j defined in the paragraph above Eq. (28). We suppose that the partition is invariant under the spatial translations $T^{\vec{l}}$. The phase-space cell located at the lattice vector \vec{l} is decomposed by this partition as

$$\mathcal{M}_{\vec{l}} = \cup_{A_j \subset \mathcal{M}_{\vec{l}}} A_j. \quad (45)$$

This partition can be seen as a translationally invariant grid extending over the whole phase space.

We begin by defining the entropy of the lattice cell $\mathcal{M}_{\vec{l}}$ at time t as the coarse-grained entropy of this cell with respect to the partition (45):

$$S_t(\mathcal{M}_{\vec{l}}|\{A_j\}) \equiv - \sum_{A_j \subset \mathcal{M}_{\vec{l}}} \mu_t(A_j) \ln \frac{\mu_t(A_j)}{\nu(A_j)} + S_{\text{eq}}(\mathcal{M}_{\vec{l}}|\{A_j\}), \quad (46)$$

where we have set Boltzmann's constant equal to unity, $k_B = 1$. The first term on the right-hand side of Eq. (46) is the nonequilibrium relative entropy with respect to the equilibrium entropy for this partition. The equilibrium entropy is given by

$$S_{\text{eq}}(\mathcal{M}_{\vec{l}}|\{A_j\}) = - \sum_{A_j \subset \mathcal{M}_{\vec{l}}} \nu(A_j) \ln \frac{\nu(A_j)}{c}, \quad (47)$$

where c is a constant that fixes the absolute value of the equilibrium entropy. The time variation of the entropy over a time interval τ is, of course, only due to the change in the relative entropy, and is defined as the difference

$$\begin{aligned} \Delta^\tau S(\mathcal{M}_{\vec{l}}) &\equiv S_t(\mathcal{M}_{\vec{l}}|\{A_j\}) - S_{t-\tau}(\mathcal{M}_{\vec{l}}|\{A_j\}), \\ &= S_t(\mathcal{M}_{\vec{l}}|\{A_j\}) - S_t(\Phi^\tau \mathcal{M}_{\vec{l}}|\{\Phi^\tau A_j\}). \end{aligned} \quad (48)$$

On the other hand, the *entropy flow* is defined as the difference between the entropy that enters the cell $\mathcal{M}_{\vec{l}}$ and the entropy that exits that cell:

$$\begin{aligned} \Delta_e^\tau S(\mathcal{M}_{\vec{l}}) &\equiv S_{t-\tau}(\Phi^{-\tau} \mathcal{M}_{\vec{l}}|\{A_j\}) - S_{t-\tau}(\mathcal{M}_{\vec{l}}|\{A_j\}), \\ &= S_t(\mathcal{M}_{\vec{l}}|\{\Phi^\tau A_j\}) - S_t(\Phi^\tau \mathcal{M}_{\vec{l}}|\{\Phi^\tau A_j\}). \end{aligned} \quad (49)$$

Accordingly, the *entropy production* over a time τ , assuming that $t \gg \tau$, is defined as

$$\begin{aligned} \Delta_i^\tau S(\mathcal{M}_{\vec{l}}) &\equiv \Delta^\tau S(\mathcal{M}_{\vec{l}}) - \Delta_e^\tau S(\mathcal{M}_{\vec{l}}), \\ &= S_t(\mathcal{M}_{\vec{l}}|\{A_j\}) - S_t(\mathcal{M}_{\vec{l}}|\{\Phi^\tau A_j\}). \end{aligned} \quad (50)$$

B. Calculation of the entropy production

Equation (50) gives the expression of the entropy production as the difference between the entropy with respect to the original partition into sets A_j and the entropy with respect to a partition into sets that are the images $\Phi^\tau A_j$ of sets A_j after time τ . We notice that each set $\Phi^\tau A_j$ belongs to a single unit cell $\mathcal{M}_{\vec{l}}$ by a previous assumption. Moreover, since the partition is invariant under translation from cell to cell, the partition $\{\Phi^\tau A_j\}$ is identical to the partition $\{\phi^\tau A_j\}$ obtained by using the flow on the torus.

Written out in full, this entropy production is

$$\begin{aligned} \Delta_i^\tau S(\mathcal{M}_{\vec{l}}) &= - \sum_{A_j \subset \mathcal{M}_{\vec{l}}} \mu_t(A_j) \ln \frac{\mu_t(A_j)}{\nu(A_j)} \\ &\quad + \sum_{\phi^\tau A_j \subset \mathcal{M}_{\vec{l}}} \mu_t(\phi^\tau A_j) \ln \frac{\mu_t(\phi^\tau A_j)}{\nu(A_j)}, \end{aligned} \quad (51)$$

where we have used $\nu(\phi^\tau A_j) = \nu(A_j)$. Next, we expand in powers of the deviations of the measures from their equilibrium values and find

$$\begin{aligned} \Delta_i^\tau S(\mathcal{M}_{\vec{l}}) &= \frac{1}{2} \sum_{\phi^\tau A_j \subset \mathcal{M}_{\vec{l}}} \frac{[\delta \mu_t(\phi^\tau A_j)]^2}{\nu(A_j)} \\ &\quad - \frac{1}{2} \sum_{A_j \subset \mathcal{M}_{\vec{l}}} \frac{[\delta \mu_t(A_j)]^2}{\nu(A_j)} + O(\delta \mu_t^3). \end{aligned} \quad (52)$$

We now use the explicit forms for the measures $\delta \mu_t(A)$ given by the second term on the right-hand side of Eq. (26). After some algebra and the use of the conservation of measures, as well as the summation formulas (41) and (42), we find that the right-hand side of Eq. (52) becomes

$$\begin{aligned} \Delta_i^\tau S(\mathcal{M}_{\vec{l}}) &= \frac{1}{2} \frac{1}{|\mathcal{B}|^2} \int_{\mathcal{B}} d\vec{k}_1 F_{\vec{k}_1} \int_{\mathcal{B}} d\vec{k}_2 F_{\vec{k}_2} e^{i\vec{l} \cdot (\vec{k}_1 + \vec{k}_2)} \\ &\quad \times C(\vec{k}_1, t) C(\vec{k}_2, t) \\ &\quad \times e^{(s_{k_1} + s_{k_2})t} \sum_j \frac{1}{\nu(A_j)} \vec{k}_1 \vec{k}_2 : [\vec{T}(A_j) \vec{T}(A_j) \\ &\quad - \vec{T}(\phi^\tau A_j) \vec{T}(\phi^\tau A_j)]. \end{aligned} \quad (53)$$

Here, the summation is over the sets A_j , that form a partition of the unit cell $\mathcal{M}_{\vec{l}}$. Now we use the identity (33) and the sum rule (44) to obtain our central result:

$$\begin{aligned} \Delta_i^\tau S(\mathcal{M}_{\vec{l}}) &= -D\tau \frac{\nu(\mathcal{M})}{|\mathcal{B}|^2} \int_{\mathcal{B}} d\vec{k}_1 F_{\vec{k}_1} \int_{\mathcal{B}} d\vec{k}_2 F_{\vec{k}_2} \vec{k}_1 \cdot \vec{k}_2 \\ &\quad \times e^{i\vec{l} \cdot (\vec{k}_1 + \vec{k}_2)} C(\vec{k}_1, t) C(\vec{k}_2, t) e^{(s_{k_1} + s_{k_2})t} \\ &\simeq D\tau \frac{1}{n_{\text{eq}}} \left[\frac{\partial n(\vec{l}, t)}{\partial \vec{l}} \right]^2, \end{aligned} \quad (54)$$

since the tracer density is expressed according to Eq. (16) and $n_{\text{eq}} = \nu(\mathcal{M})$.

Here we have used the isotropy of the motion in order to eliminate correlations between the displacements in orthogonal directions. This results in the factor $\vec{k}_1 \cdot \vec{k}_2$ appearing in the integrand in Eq. (54). We have also implied a scaling limit in order to write the last line of this equation.

VI. CONCLUSIONS

In this paper, the irreversible entropy production has been derived from statistical mechanics for a process of diffusion in periodic, deterministic dynamical systems. The derivation starts from Gibbs' coarse-grained entropy, and assumes that the system is spatially periodic, and that the dynamics satisfies Liouville's theorem, is chaotic, and mixing. We chose a coarse-graining partition of phase space that has the property that any two trajectories starting in the same set of the partition will remain close together over some specified time interval. The central quantities appearing in our derivation

are the hydrodynamic measures. They define, at the microscopic level, the hydrodynamic modes of diffusion, which are exponentially damped at a rate given by the van Hove dispersion relation for diffusion. These hydrodynamic measures describe the approach to the thermodynamic equilibrium under the diffusion process. In deterministic systems, the hydrodynamic measures turn out to be singular. Indeed, the quantity $\tilde{T}(A)$ is a measure describing a nonequilibrium stationary state corresponding to a gradient of concentration across the system and, in the multi-baker, the cumulative function of this measure is known to be the continuous but nondifferentiable Takagi function [17]. On the other hand, it has been shown elsewhere that the hydrodynamic measures are singular in periodic Lorentz gases [19]. It should be emphasized here that the hyperbolicity of the system is used to argue that the time-dependent hydrodynamic measures, defined by Eq. (20), approach their asymptotic forms, with variations on arbitrarily fine scales in phase space, on time scales determined by the positive Lyapunov exponents, which are very short compared to the time scales necessary for the relaxation of the density distribution to equilibrium. Of course, not all systems with macroscopic diffusion and a positive rate of entropy production are chaotic, e.g., wind-tree models with nonoverlapping trees [20]. In such systems the mechanism responsible for positive entropy production is expected to develop on algebraic rather than exponential time scales, as is the case for chaotic systems.

Moreover, if the hydrodynamic measures were regular, expression (54) for the entropy production would vanish, as shown in the case of the multi-baker map [4,7]. Accordingly, the singular character of the hydrodynamic measures plays an essential role in the positiveness of the entropy production expected from irreversible thermodynamics. A further point we want to emphasize is that the present derivation naturally leads to a positive entropy production in agreement with the second law of thermodynamics. Finally, we point out that our derivation of the expression given by irreversible thermodynamics for the rate of entropy production applies not only to Lorentz gases, but also to tracer diffusion taking place in a spatially periodic N particle system, where the mechanism for tracer diffusion is provided by the particles interacting with each other. Without this interaction, the motion of the tracer particle would be ballistic, and the mean square displacement would grow quadratically with time.

The next step to be taken in the development of this approach to the theory of entropy production in the relaxation of fluid systems to thermal equilibrium is to generalize the method given here, for tracer diffusion in a periodic N particle system, to viscous and heat flows. We shall report on the derivation of entropy production for these other transport processes in future publications. Finally, one would like to

remove the restriction to periodic systems, and to consider the entropy production for a general, isolated N particle system relaxing to thermal equilibrium. This remains open for future work.

It is necessary to mention that our approach to entropy production in fluids has been criticized by Rondoni and Cohen in a series of papers [21]. This is not the place to provide a detailed response to their criticisms, which we will do in separate publications. However, it is appropriate here to mention two issues that are of some importance for our response to their comments. (1) Rondoni and Cohen find it troublesome that the time needed for our method to be applicable depends on the nature of the partition chosen for the calculation of the relative entropy and its change with time. While this observation is indeed correct, the consequences are not problematic. The essential point to note is that in order for a nonzero rate of irreversible entropy production to result from our method, we require a partition that is coarser than the scale of variation of the nonequilibrium distribution function. As we have tried to make clear, the nonequilibrium distribution function develops a fractal structure on a time scale set by the magnitude of the largest positive Lyapunov exponent. For the systems described here, this is a time scale that is very short compared to the time scale over which the system relaxes to equilibrium. In fact, this time scale is on the order of the mean free time between collisions, a scale sufficiently short that more traditional methods for computing the rate of entropy production, based upon the Boltzmann equation, for example, also do not apply. (2) Rondoni and Cohen have also objected to our use of multi-baker maps and Lorentz gases as examples of systems to which nonequilibrium thermodynamics might be applied. While one might object to their view of the utility of simple model systems for the development of physical intuition, our calculation of the rate of entropy production for tracer diffusion in an N particle system shows that the method can be applied to much more general systems than multi-baker maps and Lorentz gases. Our future work will be devoted to seeing exactly how general this method might be.

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