Up-hill diffusion, creation of density gradients: Entropy measure for systems with topological constraints

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It is always some constraint that yields any nontrivial structure from statistical averages. As epitomized by the Boltzmann distribution, the energy conservation is often the principal constraint acting on mechanical systems. Here we investigate a different type: the topological constraint imposed on "space." Such a constraint emerges from the null space of the Poisson operator linking an energy gradient to phase space velocity and appears as an adiabatic invariant altering the preserved phase space volume at the core of statistical mechanics. The correct measure of entropy, built on the distorted invariant measure, behaves consistently with the second law of thermodynamics. The opposite behavior (decreasing entropy and negative entropy production) arises in arbitrary coordinates. An ensemble of rotating rigid bodies is worked out. The theory is then applied to up-hill diffusion in a magnetosphere.

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I. INTRODUCTION

There are plenty of examples that seemingly violate the principle of entropy maximization. So-called up-hill diffusion, creating density gradients, is often observed in multiphase fluids and solids undergoing spinodal decomposition [1,2], in metallic alloys [3], nanoporous materials [4], and magmas [5]. By separating the different components of the mixture, Helmholtz free energy achieves a local minimum, characterized by nonuniform concentrations, that is stable against fluctuations [2]. With a completely different mechanism, astronomical plasmas accumulate within the magnetic fields of stars and planets through the process of inward diffusion [6–9] and generate an heterogeneous density profile. That the determining factor is not the energy constraint is made apparent by the experimental observation of non-neutral plasma particles climbing up the potential hill [8], as well as by numerical calculations concerning their thermal equilibrium [10]. Here the *topological constraints* [11–16] affecting the phase space are the underlying principle. Once the constraints are broken, the quasistationary state is destroyed and the systems progressively approach thermal death. Accretion of galaxies under the action of gravitation [17,18], ferromagnetism mediated by the magnetic field [19,20], spontaneous creation of planetary magnetospheres through electromagnetic interaction [6–8], vortical structures in magnetofluids preserving helicities [21], living organisms harvesting "negentropy" [22], and self-organization of data flows in information theory [23] are some of the most paradigmatic examples of such ordered structures that originate from the topological invariants affecting the relevant "phase space."

In the present paper, we study the nonequilibrium statistical mechanics of Hamiltonian systems subjected to the aforementioned topological constraints. To formulate statistical mechanics, the phase space must be "homogeneous" so that the equilibrium state becomes a uniform distribution, as the entropy principle predicts [24]. However, a topological constraint introduces inhomogeneity to the phase space by limiting the accessible region. Then we have to formulate an alternative "proper phase space" in which the homogeneity recovers. The statistical processes in the proper phase space may appear very differently when projected into the original constrained phase space [12]. In the context of the present study, the degeneracy of the Poisson bracket represents such a topological constraint (the adiabatic invariants are the physical origin of the constraint) [11,21]. Then the proper phase space is the Casimir leaf [13–15], which is a symplectic submanifold. When the Casimir leaf is inhomogeneously embedded in the original phase space, the distorted metric on the leaf (viewed from the original phase space of the degenerate Poisson bracket) gives rise to nontrivial structures. In particular, we show that the entropy defined on the invariant measure of the proper phase space behaves consistently with the second law of thermodynamics. Due to the noncovariant nature of differential entropy [25,26], the time evolution of the uncertainty measured in arbitrary coordinates may "flip" and appear as an entropy decrease in the Cartesian perspective. It is the Jacobian of the coordinate change that yields the ordered structure, while the probability distribution is flattened in the proper variables.

The theory, which finds its roots in the phenomenological observation that particle density in planetary magnetopsheres tends to be homogenized in the magnetic coordinates [27,28], shows that the proper phase space upon which statistical mechanics can be formulated differs from the *a priori* variables used to represent a general physical system. These findings may pave the way for a new and rigorous understanding of the statistical mechanics governing constrained systems.

II. TOPOLOGICALLY CONSTRAINED HAMILTONIAN MECHANICS

We start with a short review of the Hamiltonian formalism. Hamiltonian mechanics is the result of interaction between matter (energy or Hamiltonian function H) and space (Poisson operator $\mathcal{J}$) according to the equation

$$\boldsymbol{v} = \mathcal{J}\nabla H, \tag{1}$$

where $\boldsymbol{v} = \dot{\boldsymbol{x}}$ is the flow velocity in n-dimensional phase space. Equation (1) admits two typologies of constants of motion: those that can be ascribed to the specific form of the Hamiltonian function, i.e., to the properties of matter, and the so-called Casimir invariants that originate from the eigenvectors with 0-eigenvalue (the null space or kernel) of the Poisson operator, i.e., from the properties of space. This second kind of invariants, which limits the accessible regions of phase space as a result of the constraining environment, is at the core of the theory developed in the present work. Specifically, due to antisymmetry $\mathcal{J}^T = -\mathcal{J}$, whenever the operator $\mathcal{J}$ has a kernel $\boldsymbol{\xi}$ such that $\mathcal{J}\boldsymbol{\xi} = \mathbf{0}$, the system is subjected to *topological constraints*:

$$\boldsymbol{\xi} \cdot \boldsymbol{v} = 0. \tag{2}$$

Equation (2) can be thought of as the formal definition of topological constraint. We remark that the above result holds for any Hamiltonian and even if $\mathcal{J}$ does not satisfy the Jacobi identity (see Ref. [29]). However, because of Darboux's theorem [13–15], the Jacobi identity ensures that the kernel is integrable, i.e., that a Casmir invariant exists:

$$\boldsymbol{\xi} = \lambda \nabla C, \tag{3}$$

where, for now, we assume that the rank of $\mathcal{J}$ is $n-1$ (see Ref. [30]), and the two functions λ and C are the integration factor and Casimir invariant ($\dot{C} = \lambda^{-1}\boldsymbol{\xi} \cdot \boldsymbol{v} = 0$), respectively.

III. NONCOVARIANT NATURE OF DIFFERENTIAL ENTROPY

It is now useful to make some considerations on the noncovariant nature of differential entropy. Extension of Shannon's discrete entropy to continuous probability distributions is a delicate process [25,26]. Indeed, the quantity

$$\tilde{S} = -\int_V p(\boldsymbol{x}) \log p(\boldsymbol{x})\, dV \tag{4}$$

is not, in general, the entropy of the continuous probability distribution $p(\boldsymbol{x})$ on the volume element $dV = dx^1 \wedge \cdots \wedge dx^n$. The reason is that $\tilde{S}$ is not covariant, i.e., its value changes depending on the chosen coordinate system, and (4) tacitly assumes that dV is an invariant measure. Unfortunately, this is not always the case, and (4) has to be amended with Jaynes' functional:

$$S^J = -\int_V p(\boldsymbol{x}) \log \left[\frac{p(\boldsymbol{x})}{\mathfrak{J}(\boldsymbol{x})}\right] dV, \tag{5}$$

where the Jacobian $\mathfrak{J}(\boldsymbol{x})$ compensates the coordinate dependence of the logarithm. In the Hamiltonian picture, one can always find a time-independent function $\mathfrak{J}(\boldsymbol{x})$ nullifying the Lie derivative of $\mathfrak{J}dV$ with respect to the dynamical flow $\boldsymbol{v}$, i.e., such that $\mathfrak{L}_{\boldsymbol{v}}\mathfrak{J}(\boldsymbol{x})dV = 0$. The obtained $\mathfrak{J}$ with (5) will then give the desired covariant form of entropy. It is useful to recast (5) as

$$\Sigma = -\int_{V_I} P(\boldsymbol{y}) \log P(\boldsymbol{y})\, dV_I. \tag{6}$$

Here P is the probability distribution of $\boldsymbol{y}$ and dV_I is the invariant measure of the system $dV_I = dy^1 \wedge \cdots \wedge dy^n = \mathfrak{J}dV$ satisfying $\mathfrak{L}_{\boldsymbol{u}} dV_I = 0$, with $\boldsymbol{u} = \dot{\boldsymbol{y}}$.

IV. ENSEMBLE OF ROTATING RIGID BODIES

We are now ready to test the theory with a simple 3D example. In three dimensions Eq. (1) can always be cast in the form $\boldsymbol{v} = \boldsymbol{w} \times \nabla H$, where $\boldsymbol{w}$ is a properly chosen vector (see Ref. [31]). The Euler's rotation equation for the motion of a rigid body with angular momentum $\boldsymbol{x}$ and moments of inertia I_x, I_y, and I_z can be obtained by setting $H = (x^2/I_x + y^2/I_y + z^2/I_z)/2$ and $\boldsymbol{w} = \boldsymbol{x}$. The kernel $\boldsymbol{\xi}$ associated to this operator, i.e., the topological constraint (2) affecting the phase space of a rigid body, is soon identified to be $\boldsymbol{\xi} = \boldsymbol{x}$. Indeed, $\boldsymbol{\xi} \cdot \boldsymbol{v} = \boldsymbol{x} \cdot \boldsymbol{x} \times \nabla H = 0$. At the same time, one can verify that the Jacobi identity (see Ref. [32]) is satisfied $\boldsymbol{x} \cdot \nabla \times \boldsymbol{x} = 0$, making the system Hamiltonian. The Jacobi identity also guarantees that the kernel is integrable [remember (3)] to give the integration factor $\lambda = 1$ and the Casimir invariant $C = \boldsymbol{x}^2/2$, so that $\boldsymbol{w} = \nabla C$. Furthermore, the invariant measure turns out to be $dV_I = dx \wedge dy \wedge dz$, as follows from $\nabla \cdot \boldsymbol{v} = 0$. Since this is the original statistical measure, one can directly apply (4) to define the entropy of an ensemble of such rigid bodies. However, suppose that we consider a slightly more complicated rotation pattern, such as

$$\boldsymbol{v} = \lambda(\boldsymbol{x}) \nabla \frac{\boldsymbol{x}^2}{2} \times \nabla H, \tag{7}$$

where, for example, $\lambda = e^{z^2/2}$. Since $\dot{z} \propto \lambda$, high values of z will be less probable, and (7) may represent the anisotropic rotation of a rigid body that tends to spin around the axis with angular momenta x,y. Equation (7) still satisfies the Jacobi identity and thus represents an Hamiltonian system with the same Casimir element C. However, the invariant measure becomes

$$dV_I = e^{-z^2/2}\, dx \wedge dy \wedge dz = dC \wedge d\chi \wedge dz, \tag{8}$$

where we introduced new coordinates (C,χ,z), with $\chi = e^{-z^2/2} \arctan(y/x)$. Separating the constant of motion C, the new 2D canonical equations are

$$\boldsymbol{u} = \begin{bmatrix} \dot{\chi} \\ \dot{z} \end{bmatrix} = \begin{bmatrix} -H_z \\ H_\chi \end{bmatrix}. \tag{9}$$

One can verify that (9) is divergence free.

In order to study the statistical mechanics of the new system, we now consider an ensemble of objects obeying (9) and let them interact by adding to the Hamiltonian an interaction potential ϕ. Its ensemble average must go zero $\langle\phi\rangle = 0$, since the total energy of the system has to be preserved. In addition, and this is the key point of the paper, there are grounds for the ergodic hypothesis in the novel coordinates (C,χ,z) [and not in the original variables (x,y,z)] because of the invariant measure (8). In other words, it is licit to exchange ensemble averages with time averages *only* on (8):

$$\begin{aligned} 0 = \langle d\phi \rangle &= \langle \phi_\chi \rangle d\chi + \langle \phi_z \rangle dz \\ &= \bar{\phi}_\chi d\chi + \bar{\phi}_z dz = \bar{\Gamma}_\chi(t) d\chi + \bar{\Gamma}_z(t) dz, \end{aligned} \tag{10}$$

with Γ_χ and Γ_z Gaussian white noises and where the bar indicates long-time averaging. In (10) first we substituted ensemble averages with time averages, and then represented the various components with random processes of zero time average. We remark that this would not have been possible in the original coordinates (x,y,z), as they are not measure preserving. Neglecting the constant C, the equations

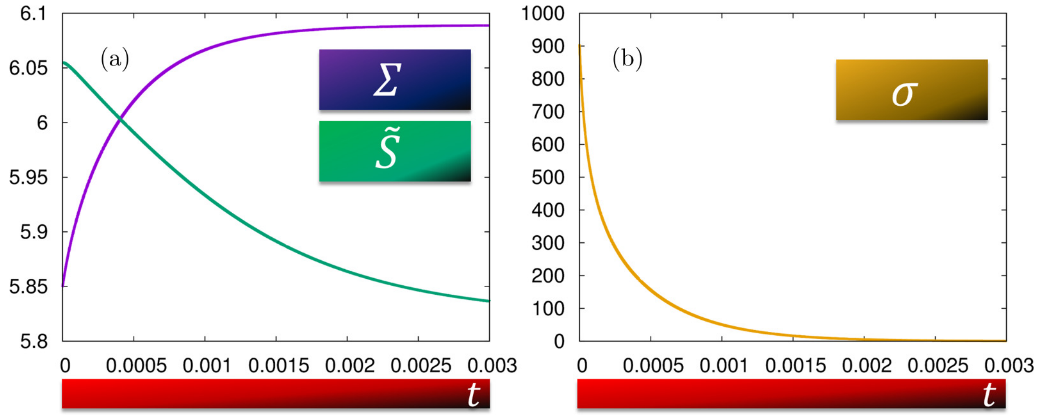

FIG. 1. (a) Σ (increasing line) and $\tilde{S}$ (decreasing line) as a function of time t. (b) σ as a function of time t. Arbitrary units are used. Initial condition is a flat distribution f on dV.

accounting for the interaction become

$$\begin{bmatrix}\dot{\chi}\\ \dot{z}\end{bmatrix} = \begin{bmatrix}-H_z - \Gamma_z\\ H_\chi + \Gamma_\chi\end{bmatrix}. \tag{11}$$

Note that, while the Hamiltonian is no more a constant, C is still a Casimir invariant: the rigid bodies will explore the surface of phase space defined by C.

The next step is to build the Fokker-Planck equation associated to (11). We refer the reader to Refs. [9,34] for a detailed description of the procedure. The result is

$$\frac{\partial P}{\partial t} = H_z \frac{\partial P}{\partial \chi} - H_\chi \frac{\partial P}{\partial z} + \frac{1}{2} D_\chi \frac{\partial^2 P}{\partial \chi^2} + \frac{1}{2} D_z \frac{\partial^2 P}{\partial z^2}. \tag{12}$$

Here P is the probability distribution on (χ,z), and D_χ, D_z are the diffusion coefficients associated with the white noises. Finally, we seek for an explicit expression of the entropy production rate σ of the system. Define the Fokker-Planck velocity $\mathbf{Z}$ to be the vector field such that (12) is written as $\partial_t P = -\nabla \cdot (\mathbf{Z} P)$. Then, recalling (6):

$$\frac{d\Sigma}{dt} = \int_{V_I} \{P \nabla \cdot \mathbf{Z} + \nabla \cdot [P \log(P) \mathbf{Z}]\}\, dV_I. \tag{13}$$

The first term represents the ensemble average of the Fokker-Planck velocity divergence, while the second factor can be cast as a surface integral representing entropy flow out L. It is straightforward to deduce that

$$\sigma = \langle \nabla \cdot \mathbf{Z} \rangle, \tag{14a}$$

$$L = -\int_{V_I} \nabla \cdot [P \log(P) \mathbf{Z}]\, dV_I. \tag{14b}$$

Substituting the expression of $\mathbf{Z}$ in (14a), we obtain

$$\sigma = -\frac{1}{2} D_\chi \left\langle \frac{\partial^2 \log P}{\partial \chi^2} \right\rangle - \frac{1}{2} D_z \left\langle \frac{\partial^2 \log P}{\partial z^2} \right\rangle. \tag{15}$$

In Fig. 1 we report the results of the numerical simulation of (12). The entropy Σ, defined on the invariant measure (8) of the system behaves consistently with the second law of thermodynamics and the associated entropy production σ is positive. On the other hand, the wrong measure of entropy $\tilde{S} = -\int f \log f\, dV = \Sigma + \langle \lambda \rangle$, defined by the distribution function f on the original phase space $dV = dx \wedge dy \wedge dz$, decreases. Furthermore, diffusion flattens the distribution P, and since preservation of particle number requires $P\, dV_I = f\, dV$, $f = P/\lambda$ creates an ordered structure by approaching $f \propto \lambda^{-1}$.

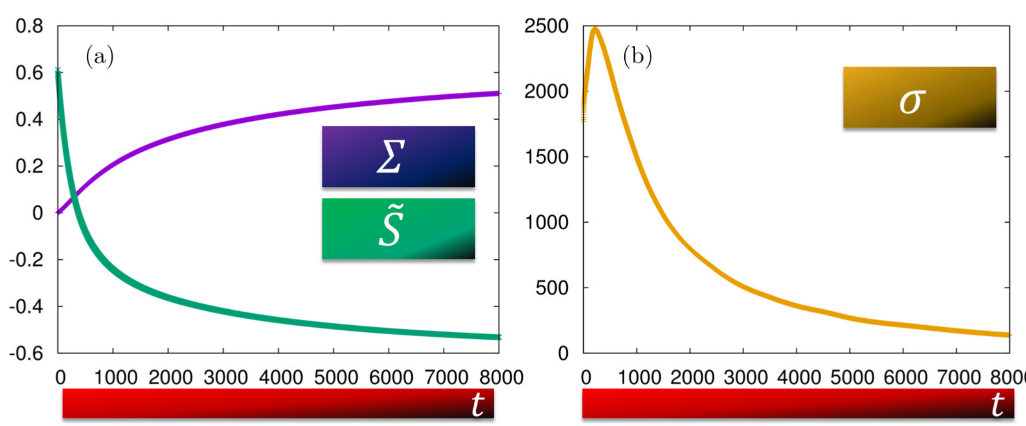

FIG. 2. (a) Σ (increasing line) and $\tilde{S}$ (decreasing line) as a function of time t. (b) σ as a function of time t. Arbitrary units are used. Initial condition is a Maxwell-Boltzmann distribution.

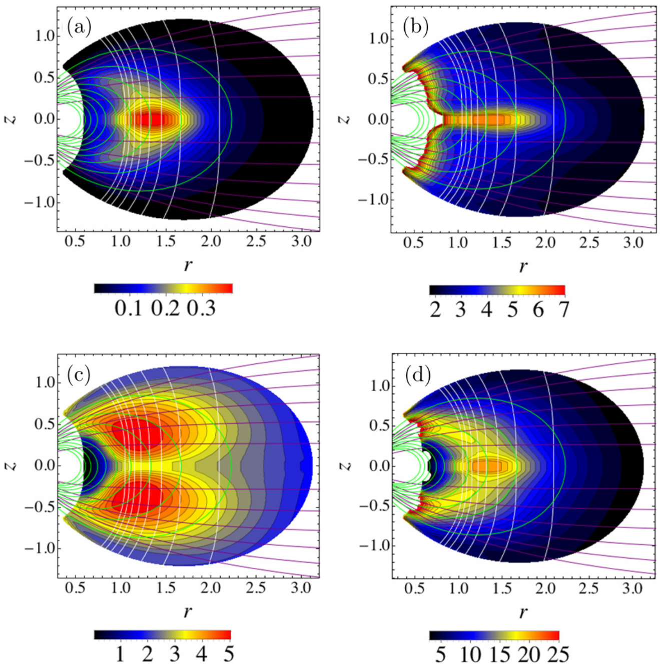

FIG. 3. Self-organized plasma after entropy maximization. (a) Spatial profile of particle density (a.u.). (b) Temperature anisotropy $T_\perp/T_\parallel$. (c) Parallel temperature $T_\parallel(eV)$. (d) Perpendicular temperature $T_\perp(eV)$. White (vertical) lines, green (circular) lines, and purple (spreading from the left to the right) lines represent contours of B, ψ, and l.

V. SELF-ORGANIZED CONFINEMENT IN MAGNETOSPHERE

Let us show how the theory can be applied to the study of a real self-organizing system: a magnetosphere. In astronomical plasmas, charged particles are trapped by planetary magnetospheres as they spiral around the magnetic field $\boldsymbol{B} = \nabla\psi \times \nabla\theta$, where $\psi = \psi(r,z)$ is the flux function and θ the toroidal angle of a cylindrical coordinate system (r,z,θ). This dynamics (cyclotron motion) is characterized by preservation of the magnetic moment $\mu = mv_\perp^2/2B = \text{const}$, where m is the particle mass, $v_\perp$ the particle velocity perpendicular to magnetic field lines, and $B = |\boldsymbol{B}|$. Because of the topological constraint μ, it turns out [9,35] that the invariant measure of magnetized particles is $dV_I = d\mu \wedge dv_\parallel \wedge dl \wedge d\psi \wedge d\theta = Bd\mu \wedge dv_\parallel \wedge dx \wedge dy \wedge dz = BdV$, where l and $v_\parallel$ are length and velocity along $\boldsymbol{B}$, respectively. The electromagnetic interaction diffuses the constrained particles on the statistical measure dV_I and maximizes the associated entropy Σ. Due to the inhomogeneous Jacobian B, the process will appear as creating density gradients and temperature anisotropy in the Cartesian perspective, while the entropy $\tilde{S}$ defined on dV is minimized. This scenario is exemplified in Figs. 2 and 3 obtained by simulation of the Fokker-Planck equation derived in Refs. [9,35].

VI. SUMMARY AND CONCLUSIONS

Hamiltonian mechanics, formulated in terms of an energy function and a Poisson operator, encompasses a broad class of physical systems that often seem to deviate from the laws of thermodynamics. In the present paper we have shown that the key to a proper construction of statistical mechanics resides in the kernel of the Poisson operator, which dictates topological constraints. Such constraints are intrinsically different from the usual energy conservation yielding the Boltzmann distribution, and they act independently to limit the accessible phase space. As a consequence, the invariant measure required to introduce a consistent notion of entropy is distorted with respect to the original reference frame exploited to represent the physical system. The probability distribution will be flattened and the entropy will be maximized on the properly constructed invariant measure, while the Jacobian of the coordinate change will cause an entropy "flip" in the original coordinates. Here, due to the noncovariance of differential entropy, the system will seemingly violate the second law of thermodynamics and an order structure will arise.

The theory has been tested with an ensemble of rotating rigid bodies and then applied to explain the nonequilibrium statistical mechanics of up-hill diffusion in magnetospheric plasmas.

Finally, the same arguments will remain valid whenever the invariant measure carrying the ergodic hypothesis is connected by an inhomogeneous Jacobian to the coordinates used to observe the physical system.

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[29] This identity is satisfied by any Poisson operator and is the essential feature of the algebraic structure of Hamiltonian systems. Defining the Poisson bracket of two functions f and g as $\{f,g\} = \nabla f \cdot \mathcal{J}\nabla g$, the identity for three functions f, g, and h reads $\{f,\{g,h\}\} + \{g,\{h,f\}\} + \{h,\{f,g\}\} = 0$.
[30] If the rank of $\mathcal{J}$ is $n-k$ and the Jacobi identity is satisfied, there will be k Casimir invariants. Again, this is a consequence of Darboux's theorem [15].
[31] If $\boldsymbol{w} = \boldsymbol{B}/B^2$ with $\boldsymbol{B}$ the magnetic field, and setting $H = \phi$, with ϕ the electric potential, one obtains the equations of motion for a magnetized particle performing $\boldsymbol{E} \times \boldsymbol{B}$ drift. In a vacuum, $\boldsymbol{B} = \nabla\xi$ for some potential ξ. In this case, ξ is a Casimir invariant.
[32] In three dimensions the Jacobi identity reads as $\boldsymbol{w} \cdot \nabla \times \boldsymbol{w} = 0$. When satisfied, the constraint $\boldsymbol{w} \cdot \boldsymbol{v} = 0$ is integrable [33], and thus two scalars λ and C can be found such that $\boldsymbol{w} = \lambda\nabla C$. Furthermore, $\mathfrak{I} = \lambda^{-1}$ defines an invariant measure since $\nabla \cdot (\mathfrak{I}\boldsymbol{v}) = 0$, with the result that, in three dimensions, the Jacobi identity, Casimir invariant, and invariant measure imply each other provided that $\boldsymbol{v} = \boldsymbol{w} \times \nabla H$.
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