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Shape-dependent thermodynamics and nonlocal hydrodynamics in a non-Gibbsian steady state of a drift-diffusion system

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Shape-dependent thermodynamics and nonlocal hydrodynamics are argued to occur in dissipative steady states of driven diffusive systems. These predictions are confirmed by numerical simulations. Unlike power-law correlations, these phenomena cannot be explained by a hypothesis of ''criticality.'' Instead, they require the effective Hamiltonian of the system to contain very long-range potentials, making the invariant probability measures formally ''non-Gibbsian.'' [S1063-651X(98)50606-X]

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Power-law decay of correlations are generic in dissipative steady states of open, driven systems with conservation laws [1,2]. In equilibrium systems such power laws can be due to two different mechanisms: the interaction potentials in the system Hamiltonian can themselves be long-range power laws (e.g., dipolar) or else the potentials can be short-ranged but the system may be at a critical point [3]. The latter circumstance has prompted the view that dissipative, nonequilibrium systems are attracted without any tuning of parameters to a critical state, or exhibit so-called ''self-organized criticality'' (SOC) [4]. We shall show here that such an interpretation, taken literally, is not true in an important class of such systems. As we shall explain, an *effective Hamiltonian* may be introduced to characterize the nonequilibrium statistics. We then exhibit phenomena that can be explained *only* by the presence there of effective power-law potentials of very long range. Such an explanation for power-law correlations was earlier proposed in *driven diffusive systems* [5] and, more recently, in a shaken *granular flow* [6]. Because the underlying dynamics are local in these systems, this mechanism may be justly termed ''self-organized long-range interactions'' (SOLRI). We shall demonstrate in the class of systems considered two closely related phenomena: *shape-dependent thermodynamics* and *nonlocal hydrodynamics*. These phenomena could not occur if the effective Hamiltonian were short-ranged but critical. In fact, the induced potentials must have such an extreme long-range, many-body character that the nonequilibrium measures are formally *non-Gibbsian*. (For an excellent review of the relevant notions, see [7].) The indicators of the non-Gibbsian nature, shape-dependent thermodynamics and nonlocal hydrodynamics, have great importance in themselves. They are expected to occur in physical drift-diffusion systems, e.g., electrolytes and semiconductors.

Correlations of power-law type are predicted in such systems by linear fluctuating hydrodynamics [1,2]. In a DDS with one species of (unit-charged) particle at constant mean density ρ, in contact with a heat bath at temperature T and in an applied electric field $\mathbf{E}$, the equation in Fourier space for density fluctuations $\delta\hat{\rho}(\mathbf{k},t)$ is

$$\delta\dot{\hat{\rho}}(\mathbf{k},t)=[i\mathbf{c}_{\mathbf{E}}\cdot\mathbf{k}-\mathbf{k}\cdot\mathbf{D}_{\mathbf{E}}\cdot\mathbf{k}]\delta\hat{\rho}(\mathbf{k},t)-i\mathbf{k}\cdot\delta\hat{\mathbf{J}}(\mathbf{k},t). \quad (1)$$

Here $\mathbf{c}_{\mathbf{E}}(\rho)=d\mathbf{j}_{\mathbf{E}}/d\rho(\rho)$ is the drift velocity, a density derivative of the conduction current. The latter is given, at low density ρ and small field-strength E, by Ohm's law, $\mathbf{j}_{\mathbf{E}}=\boldsymbol{\sigma}\cdot\mathbf{E}=\rho\boldsymbol{\mu}\cdot\mathbf{E}$, with $\boldsymbol{\sigma}$ the conductivity tensor and $\boldsymbol{\mu}$ the

mobility tensor. $\mathbf{D_E}(\rho)$ is the diffusion tensor in the conducting state, while $\delta\hat{\mathbf{J}}(\mathbf{k},t)$ is the Fourier component of the current noise with zero mean and covariance $\langle\delta\hat{\mathbf{J}}(\mathbf{k},t)\delta\hat{\mathbf{J}}(\mathbf{k}',t')\rangle=2\mathbf{L}^s_{\mathbf{E}}(\rho)\delta^3(\mathbf{k}+\mathbf{k}')\delta(t-t')$. In the limit $E\to 0$, $\mathbf{L_E}(\rho)$ coincides with the Onsager matrix $\mathbf{L}=\boldsymbol{\sigma}k_BT$. It is easy to solve Eq. (1) for the static structure function $\hat{S}_{\mathbf{E}}(\mathbf{k})\equiv\lim_{t\to\infty}\langle\delta\hat{\rho}(\mathbf{k},t)\delta\hat{\rho}(-\mathbf{k},t)\rangle$:

$$\hat{S}_{\mathbf{E}}(\mathbf{k})=\frac{\mathbf{k}\cdot\mathbf{L_E}\cdot\mathbf{k}}{\mathbf{k}\cdot\mathbf{D_E}\cdot\mathbf{k}}. \tag{2}$$

(See [1].) Inverse Fourier transforming gives a long-range density-density correlation $S_{\mathbf{E}}(\mathbf{r})\sim r^{-d}$ in dimension d, when $\mathbf{L_E}$ and $\mathbf{D_E}$ are not proportional. This will generally be true for $E\neq 0$. Such power-law correlations have been verified in numerical simulations of simple *driven lattice gas* (DLG) models [9,10]. Correlations of this type are expected more generally in locally driven or damped conservative systems without detailed balance. For example, r^{-d} correlations of similiar origin have recently been predicted and verified computationally in the homogeneous cooling state of rapid granular flows [11]. Our results have implications for all such systems.

We illustrate our points with the DLG models [9,12]. These were originally introduced as models of solid electrolytes, or superionic conductors [13,14]. However, unlike physical drift-diffusion systems of charged particles, these models have only short-ranged dynamical interactions and are thus perfect for our theoretical objectives. We shall comment later how the results carry over to realistic DDS with Coulombic interactions. The particles of the DLG model live on a cubic lattice $\mathbf{Z}^d$. Assuming hard-core exclusion, the occupancy $\eta_{\mathbf{x},t}\in\{0,1\}$ for each site $\mathbf{x}\in\mathbf{Z}^d$ and time $t\geqslant 0$. The evolution of the configuration is via a Kawasaki exchange dynamics, specified by the rate $c_{\mathbf{E}}(\mathbf{x},\mathbf{y};\eta)$ for exchange of occupancy of nearest neighbor sites $\mathbf{x},\mathbf{y}$ in the configuration η, i.e., for the transition $\eta\to\eta^{\mathbf{xy}}$. In addition to assuming that rates are functions of occupancies at sites within a finite range of $\{\mathbf{x},\mathbf{y}\}$, the main assumption is *local* detailed balance:

$$c_{\mathbf{E}}(\mathbf{x},\mathbf{y};\eta)=c_{\mathbf{E}}(\mathbf{x},\mathbf{y};\eta^{\mathbf{xy}})\exp\{-\beta[H(\eta^{\mathbf{xy}})-H(\eta)+\mathbf{E}\cdot(\mathbf{x}-\mathbf{y})(\eta_{\mathbf{x}}-\eta_{\mathbf{y}})]\}, \tag{3}$$

for some short-ranged lattice-gas Hamiltonian $H(\eta)$, e.g., an Ising model. The condition (3) encourages particles to hop in the direction of the electric field $\mathbf{E}$ and, in infinite volume, sets up an irreversible steady state with a mean current. In fact, these models have space-ergodic, homogeneous, and time-invariant measures $\mu_{\rho,\mathbf{E},\beta}$ for each density $\rho\in[0,1]$, expected to be unique at small β. The question arises whether these measures in infinite volume are ''Gibbsian'' for an effective Hamiltonian

$$H_{\text{eff}}(\eta)=\sum_{A\subset\mathbf{Z}^d}\Phi_A(\eta) \tag{4}$$

with some set of many-body potentials Φ_A depending on spins $\eta_{\mathbf{x}}$ at sites $\mathbf{x}\in A\subset\mathbf{Z}^d$. If it exists, this will generally not be the same as the short-ranged Hamiltonian $H(\eta)$ used in defining the dynamics. It turns out that almost *any* reasonable measure (with local densities) is ''Gibbsian'' if one permits extremely long-ranged, many-body potentials and, indeed, there are a myriad of physically inequivalent such Hamiltonians [e.g., see [15], Theorem V.2.2(a)]. To guarantee uniqueness and other standard properties of usual Gibbs measures, the condition of *absolute summability* is required,

$$\sum_{A\ni\mathbf{x}}\|\Phi_A\|_\infty<\infty \tag{5}$$

for all $\mathbf{x}\in\mathbf{Z}^d$, where $\|\Phi_A\|_\infty\equiv\sup_\eta|\Phi_A(\eta)|$. Following rather common practice [7], we shall agree here to call only measures with the latter property ''Gibbsian.'' It is a rigorous theorem of Asselah [16] (or Appendix B of [8]) that the following alternative holds: either *all* space-ergodic, invariant measures of the DLG are Gibbsian with absolutely summable potential or else *none* of them are. It may also be proved that in the domain of analyticity, no absolutely summable power-law potential can produce a correlation $\propto r^{-d}$ (Theorem 1 of [3]), such as is observed in the DLG. Thus, under the first alternative, one is led to conclude that the Gibbs measure must be critical to account for the observed correlation decay. Alternative number one for our purposes may thus be termed SOC. On the other hand, in alternative number 2 the potentials are nonsummable (=long-ranged), so that this case corresponds to the SOLRI scenario. To support the latter, we make some key comparisons with long-ranged systems.

An analogy was already remarked a few years ago between the DDS and dipolar systems [5]. Of course, dipole spin-spin correlations are also $\propto r^{-d}$ even at high temperature. This is consistent with our point of view, because the dipole potential just misses being absolutely summable (and is thus ''non-Gibbsian'' according to our criterion). An important consequence of this nonsummability was recognized early [17], namely, that the thermodynamics of dipolar systems is *shape dependent*. This situation arises because the dipole potential energy sums are only conditionally convergent and hence may lead to different values depending upon the order of summation, i.e. the shape, at least at nonzero field [18]. In uniformly magnetized (=high field), ellipsoidally-shaped samples of dipolar materials the shape dependence of thermodynamic free-energy functions is simply parameterized [19], in good agreement with experiment [20].

It was argued in [8] that a similiar shape dependence occurs in DDS. The thermodynamic functions of interest are the ''pressure'' $p_{\mathbf{E}}$ and the ''Helmholtz free-energy'' $f_{\mathbf{E}}$. The former is defined by the thermodynamic limit

$$p_{\mathbf{E}}(\mu,\beta|\rho_*)=\lim_{\Lambda\to\mathbf{Z}^d}\frac{1}{\beta|\Lambda|}\log\langle e^{\beta\mu N_\Lambda}\rangle_{\rho_*,\beta,\mathbf{E}}, \tag{6}$$

where Λ is a sequence of lattice volumes converging to $\mathbf{Z}^d$, $N_\Lambda(\eta)$ is the number of particles within Λ for the configuration η, and the average $\langle\cdot\rangle_{\rho_*,\beta,\mathbf{E}}$ is with respect to the invariant measure $\mu_{\rho_*,\beta,\mathbf{E}}$ of the DLG for reference density ρ_*. The ''Helmholtz free energy'' $f_{\mathbf{E}}$ is then introduced by the Legendre transform $f_{\mathbf{E}}(\rho,\beta|\rho_*)=\sup_\mu[\mu\rho-p_{\mathbf{E}}(\mu,\beta|\rho_*)]$. We include the ρ_* as a reminder of the reference density and the $\mathbf{E}$ to indicate the strength of the applied electric field. Since the measures here are for irre-

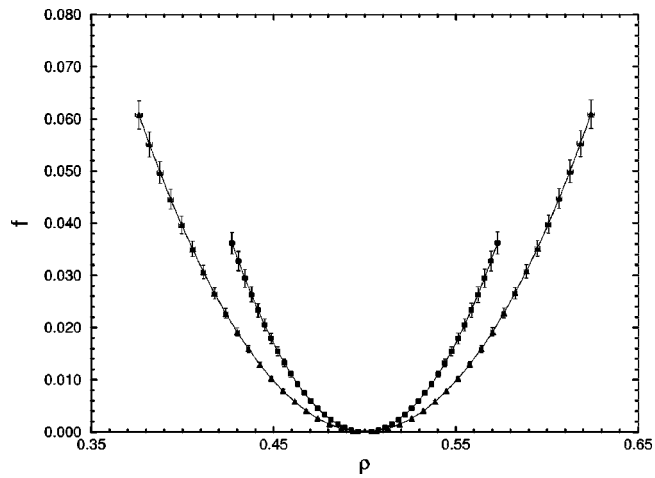

FIG. 1. Free energy as a function of density for $E=10$, $\beta=0.2$, $\rho_*=0.5$ for aspect ratio 3:1 (triangles) and 1:3 (squares).

versible steady states with Ohmic dissipation, these are not usual free energies. They coincide with the equilibrium free-energies in the limit $E\to 0$. The physical interpretation of $f_{\mathbf{E}}(\rho,\beta|\rho_*)$ is as an ''excess dissipation function,'' i.e., as the total energy dissipated per volume by an external field to change the density to ρ from its reference value ρ_* (in addition to the Ohmic dissipation intrinsic to the reference state) [8]. The argument for shape dependence is that the ''susceptibility'' (essentially, the isothermal compressibility) may be written both in terms of the free-energy, $\chi_{\mathbf{E}}(\rho,\beta)=[\beta\partial^2 f_{\mathbf{E}}/\partial\rho^2(\rho,\beta)]^{-1}$, and also in terms of the structure-function, via the limit $\chi_{\mathbf{E}}=\lim_{k\to 0}\hat{S}_{\mathbf{E}}(\mathbf{k})$. However, the latter limit is indeterminate when the structure function has the form in Eq. (2) and depends upon the wave number vector direction $\hat{\mathbf{k}}$ along which the limit is taken. Thus, the free-energy itself must be shape dependent, by the same argument as for dipole systems.

To test this prediction we have performed a Monte Carlo simulation of the DLG on a periodic square $S\times S$ lattice with Ising Hamiltonian $H=-\frac{1}{2}\Sigma_{\langle\mathbf{x},\mathbf{y}\rangle}\eta_{\mathbf{x}}\eta_{\mathbf{y}}$ where $\langle\mathbf{x},\mathbf{y}\rangle$ denotes nearest neighbor sites, for which the (inverse) critical temperature is $\beta_c\approx 0.31$ [12]. To stay well within the single phase (high temperature) regime, we used $\beta=0.2$, $E=10.0$, and reference density $\rho_*=0.5$. We have determined the thermodynamic functions for rectangular subblocks Λ of the $S\times S$ system, in which various aspect ratios of the sides of the rectangles were chosen. The pressure was evaluated by a double limit. First the infinite volume limit was obtained by a linear extrapolation in $1/S\to 0$ on the Monte Carlo average $\langle e^{\beta\mu N_\Lambda}\rangle_S$ in the steady state with $S=64$, 128, 256, 512. The thermodynamic limit in Eq. (6) for the pressure was then evaluated by a second linear extrapolation in the inverse volume $1/|\Lambda|$ of the subblock going to zero. The largest subblock edge in this second extrapolation had a length of 18. The Legendre transform to the free-energy was then carried out. The results are shown in Fig. 1 for subblocks with aspect ratios of 3:1 and 1:3 for sidelengths parallel and perpendicular to the field, respectively. Error bars reflect both statistical deviations in independent runs and the double extrapolation procedure. The two functions are clearly distinct. We see that the DLG, considered as a model of a current-carrying electrochemical cell, has well-defined free-energies but the results depend upon the shape of the cell.

This shape dependence leads, however, to a rather serious puzzle about the hydrodynamic behavior of the DDS. The general problem is to describe how an initial smooth density profile relaxes to a constant density in the driven steady state. In [8] a nonlinear hydrodynamic equation was derived by the formal method of nonequilibrium distributions to describe this irreversible process. Density fields varying on a length-scale of the order of ϵ^{-1} compared with the lattice distance were formally shown to evolve by a *drift-diffusion equation*,

$$\dot{\rho}(\mathbf{r},t)=-\nabla\cdot\left[\epsilon^{-1}\mathbf{j}_{\mathbf{E}}(\rho)-\beta\mathbf{L}_{\mathbf{E}}(\rho)\cdot\nabla\left(\frac{\delta\mathcal{F}_{\mathbf{E}}}{\delta\rho}\right)\right], \tag{7}$$

over times of order ϵ^{-2}. This equation has the ''Onsager form,'' with $\mathbf{j}_{\mathbf{E}}(\rho)$ the conduction current, $\mathbf{L}_{\mathbf{E}}(\rho)$ the Onsager coefficient matrix, and $\mathcal{F}_{\mathbf{E}}[\rho]$ the free-energy functional. Explicit analytical formulas were given in [8] for each of these quantities, e.g. a Green-Kubo formula for the Onsager matrix. These formulas are exact even at high field strengths E, although they may be difficult to evaluate concretely. The free-energy functional in [8] was *nominally* given by the local expression $\mathcal{F}_{\mathbf{E}}[\rho]=\int d\mathbf{r}f_{\mathbf{E}}[\rho(\mathbf{r})]$. However, as observed there, such a form is indeterminate. Since the free-energy depends upon the limiting shape, which value is to be used?

We can now resolve this issue. The free-energy functional actually shown in [8] to be relevant to hydrodynamics is given by a Legendre transform $\mathcal{F}_{\mathbf{E}}[\rho]=\int d\mathbf{r}\rho(\mathbf{r})\mu(\mathbf{r})-\mathcal{P}_{\mathbf{E}}[\mu]$ of the pressure functional

$$\mathcal{P}_{\mathbf{E}}[\mu]=\frac{1}{\beta}\lim_{\epsilon\to 0}\epsilon^d\log\left\langle\exp\left[\sum_{\mathbf{x}}\beta\mu(\epsilon\mathbf{x})\eta_{\mathbf{x}}\right]\right\rangle_{\rho_*,\beta,\mathbf{E}}. \tag{8}$$

Simple computation then gives $\delta\mathcal{P}_{\mathbf{E}}/\delta\mu(\mathbf{r})|_{\mu=0}=\rho_*$ and

$$\left.\frac{\delta^2\mathcal{P}_{\mathbf{E}}}{\delta\mu(\mathbf{r})\delta\mu(\mathbf{r}')}\right|_{\mu=0}=\beta\lim_{\epsilon\to 0}\epsilon^{-d}[\langle\eta_{[\epsilon^{-1}\mathbf{r}]}\eta_{[\epsilon^{-1}\mathbf{r}']}\rangle_{\rho_*,\mathbf{E}}-\rho_*^2]$$
$$\equiv\beta S_{\mathbf{E}}(\mathbf{r}-\mathbf{r}'). \tag{9}$$

It follows that $\mathcal{P}_{\mathbf{E}}[\mu]=\int d\mathbf{r}\rho_*\mu(\mathbf{r})+\beta/2\int d\mathbf{r}\int d\mathbf{r}'S_{\mathbf{E}}(\mathbf{r}-\mathbf{r}')\mu(\mathbf{r})\mu(\mathbf{r}')+O(\mu^3)$, and the Legendre transform yields

$$\mathcal{F}_{\mathbf{E}}[\rho]=\frac{1}{2\beta}\int d\mathbf{r}\int d\mathbf{r}'S_{\mathbf{E}}^{-1}(\mathbf{r}-\mathbf{r}')\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')+O(\delta\rho^3), \tag{10}$$

where $\delta\rho(\mathbf{r})\equiv\rho(\mathbf{r})-\rho_*$ and $S_{\mathbf{E}}^{-1}$ is the operator inverse of $S_{\mathbf{E}}$. In other words, $\widehat{S_{\mathbf{E}}^{-1}}(\mathbf{k})=\mathbf{k}\cdot\mathbf{D}_E\cdot\mathbf{k}/\mathbf{k}\cdot\mathbf{L}_E\cdot\mathbf{k}$. Hence, the inverse kernel $S_{\mathbf{E}}^{-1}(\mathbf{r})$ is $\propto r^{-d}$ for large r, too. We see that the hydrodynamic equation of the DDS at finite field strengths E thus must have an explicit, severely *nonlocal* form. [H. Spohn has emphasized to us that Eq. (7) linearized about the homogeneous state of density ρ_* will still be local if Eq. (2) holds, since then $\mathbf{L}_{\mathbf{E}}(\rho_*):\nabla\nabla\int d\mathbf{r}'S_{\mathbf{E}}^{-1}(\mathbf{r}-\mathbf{r}')\delta\rho(\mathbf{r}')=\mathbf{D}_{\mathbf{E}}(\rho_*):\nabla\nabla\delta\rho(\mathbf{r})$.] However, with the free-energy functional in Eq. (10) replacing the local expression, all results of [8] remain valid: the H-theorem, the fluctuation-dissipation theorem, etc.

Such nonlocal hydrodynamic behavior should also be present in equilibrium systems with long-ranged interactions, e.g., dipolar hard sphere systems or ferrofluids. In one such system, the Kawasaki lattice gas with a long-ranged Kac pair-potential, there is a rigorous result [21] that a hydrodynamic equation of the same form as Eq. (7) holds, with a similar nonlocal expression for the free-energy as in Eq. (10), simply replacing $S_{\mathbf{E}}^{-1}(\mathbf{r})$ by the kernel $J(\mathbf{r})$ of the Kac potential. We see again a very striking and fruitful analogy between dissipative, driven systems and equilibrium systems with long-range interactions.

Our results verify that the SOLRI scenario holds in our model, and not SOC. For Gibbsian measures with summable potentials there is no shape-dependence of thermodynamics, such as we observe here, even at the critical point ([15], Theorem I.2.5). Boundary conditions play a role *below* the critical point in the phase coexistence region in determining which of multiple phases will occur, but even then the free energies are independent of the phase. The ordinary thermodynamic limit, with no shape-dependence, remains valid directly at the critical point. Likewise, the dynamics of short-ranged systems in the coexistence region is expected to be described by the Cahn-Hilliard dynamics, which is local. It is actually a little perplexing how to interpret the SOC point of view that nonequilibrium steady states are always ''critical,'' when these are observed themselves to undergo continuous phase transitions at sharp values of temperature and/or density. Such transitions occur both in the DLG [12] and in granular flow [6], not to mention dipole systems [22]. Away from the transition point both DDS and dipole systems have a finite correlation length ξ which characterizes the crossover from a critical power law $\propto r^{-(d-2+\eta)}$ at intermediate range $r \ll \xi$ into the asymptotic power-law $\propto r^{-d}$ at long-range $r \gg \xi$ [12,22].

It is most interesting to consider the implications of our model calculation for real systems. We expect that the main conclusions concerning shape dependence and nonlocality will hold for physical drift-diffusion systems, such as fluid or solid electrolytes, semiconductors, and, at a more mesoscopic level, colloidal suspensions [23]. In real charged-particle systems there is the added complication of a dynamics which is itself long-ranged, via Coulombic interactions. However, these are expected to be Debye-screened and effectively short ranged. The drift-diffusion equations were justified long ago for nonequilibrium processes in electrolyte solutions at low density and small fields within Debye-Hückel theory [24]. The equations are of the same form as those we have considered, simply generalized to multiple ionic species. The effects of the Coulomb interactions are calculable and can all be incorporated into an effective Onsager matrix, with cross-species terms due to ionic-cloud distortion and electrophoresis. The effects considered in our work correspond to contributions to the invariant measures and thermodynamic potentials in at least the E^2 power of the field strength. It would be interesting to make a theoretical estimate of the order of magnitude of such effects in electrolyte solutions. Perhaps the most accessible predictions are the long-ranged correlations themselves, which could be observed in light-scattering experiments similiar to those carried out on simple fluids subject to a temperature gradient (see [25] for a recent review). Also of possible practical interest are the implications of a nonlocal hydrodynamics for granular flow.

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