Thermodynamic formalism in the thermodynamic limit: Diffusive systems with static disorder

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The chaotic properties of *diffusive* systems with *static disorder* can be calculated from a free-energy-type function, the Ruelle pressure, $\psi(\beta)$ depending on an inverse temperaturelike variable, β. For a typical system of physical interest, we show that, in the thermodynamic limit, the Ruelle pressure has a singularity and two branches (a high and low temperature ''phase''), corresponding to transitions between different localized states, with an extended state possible at the transition point. More generally, for all systems with static disorder in any number of dimensions, the Ruelle pressure depends sensitively on rare atypical fluctuations in the static disorder, and is independent of the global structure of the disorder that determines the transport coefficients. [S1063-651X(96)50408-3]

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A challenging question in the physics of fluids out of equilibrium is to understand how irreversible and complex macroscopic behavior stems from simple reversible microscopic evolution laws. In the past few years, dynamical systems theory has successfully been used to relate macroscopic transport coefficients such as diffusion coefficients to fundamental dynamical quantities such as Lyapunov exponents and Kolmogorov-Sinai entropies [1]. A unifying method for deriving these and related quantities from a single function is the thermodynamic formalism of Ruelle, Sinai, and Bowen [2,3]. In this formalism the Ruelle or topological pressure $\psi(\beta)$ is defined in terms of the long time limit of a ''free energy'' per unit time, obtained from a dynamical partition function, and β is an inverse temperaturelike variable. The thermodynamic formalism has been proven to be a powerful tool to calculate Lyapunov exponents and other dynamical properties for the many body systems of interest in statistical mechanics [3,5–7]. It can be studied by typical statistical mechanics methods, such as molecular dynamics [8] and kinetic theory methods [5,9]. Moreover, nonanalyticities in β have been interpreted, in chaos theory, as dynamical phase transitions [3,4]. In the many particle systems of interest to statistical mechanics, similar nonanalyticities and phase transitions occur. Demonstration of their existence and their interpretation as transitions between different localized states is the subject of this paper.

The goal of this paper is to investigate the large system-size behavior of the Ruelle pressure for diffusive systems with static disorder. Here we restrict ourselves to a simple model, in which independent particles move diffusively in a random static environment characterized by scatterers distributed in space (or on a lattice), namely the Lorentz (lattice) gas (LLG) [5]. But the same conclusions apply to more general models of random walks in random environments [10]. For these models it was possible to calculate $\psi(\beta,\rho)$ near $\beta=1$ using a mean field theory [5,10], yielding Lyapunov exponents, escape rates, etc. for open systems that depend on the density of scatterers ρ [5] and even on the configuration of scatterers [10]. These quantities can be related to nonequilibrium properties such as transport coefficients. Using a method described in [5], one can also determine these chaos quantities from computer simulations averaging over different configurations of scatterers. For one dimensional systems, the results are in reasonably good agreement with the mean field predictions, and the more advanced methods of kinetic theory can be used to calculate corrections beyond the Boltzmann or mean field approximation.

Simulation results for the Ruelle pressure have been obtained for one-dimensional *finite* LLG systems with up to 10^4 sites at different β values $(0\leqslant\beta\leqslant 2)$. We present an explanation of the simulation results based upon a theoretical argument that for large LLG systems in a given configuration, the Ruelle pressure for $\beta<1$ (respectively $\beta>1$) is determined by the largest cluster of scatterers (empty sites). In the limit of large system size L this pressure is extremely sensitive to rare spatial fluctuations in the distribution of randomness, and localization of trajectories in the ''most'' or ''least'' chaotic regions makes its value no longer representative for the whole disorder in the system.

This is the case for all β, except in a small region around $\beta=1$ whose size we have estimated, and which shrinks to a point in the limit of infinite systems. At this point the system is in an extended state, where trajectories explore the whole system. For all other values of β, $\psi(\beta)$ tends to a value $\psi_{\pm}(\beta)$ which is *independent* of the *density* of scatterers. Here the $(\pm)$ subscript refers to the case $\beta>1$, respectively $\beta<1$. The two branches are identical to the Ruelle pressure of a system with *all* sites occupied $(-)$ or empty $(+)$, which corresponds to localization, respectively, on clusters of scatterers or in large empty regions.

All this implies that in the thermodynamic limit the Ruelle pressure away from the singular β values carries no dynamical information specific to transport properties, which do depend on the structure and correlations of the quenched randomness.

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This peculiar behavior of the thermodynamic formalism should be a general feature of diffusive systems with static disorder, both for discrete and continuous systems. Moreover we show that the approach of the Ruelle pressure to its limiting value is extremely slow, of $O((\ln L)^{-\alpha})$, where α depends on the model and on the sign of $1-\beta$.

We consider a Lorentz lattice gas on a d-dimensional cubic lattice. A fraction $\rho=N/L^d$ of all L^d sites, chosen at random, are impurity sites, occupied by a scatterer. A particle moves at times $t=0,1,2,\ldots$ ballistically from site to site with velocity equal to a nearest-neighbor lattice vector per unit time until it hits an impurity site, where the moving particle is transmitted, reflected or deflected with probabilities p,q,s normalized as $p+q+2(d-1)s=1$. The dynamical state $x_t=\{\mathbf{r}_t,\mathbf{c}_t\}$ of the moving particle at time t is given by its position $\mathbf{r}_t$ and velocity $\mathbf{c}_t$. We consider both the case of absorbing boundary conditions (open systems) and periodic boundary conditions (closed systems).

To study the chaotic properties of LLG's we use [5] the thermodynamic formalism, where a partition function is introduced as a sum over all points Ω in a dynamic phase space

$$Z_L(\beta,t|x_0)=\sum_{\Omega}\,[P(\Omega,t|x_0)]^{\beta}. \tag{1}$$

A point Ω consists of a possible trajectory of t time steps, i.e., $\Omega=\{x_1,x_2,\ldots,x_t\}$, and $P(\Omega,t|x_0)$ is the probability that the system follows a trajectory Ω starting from x_0 at $t=0$. The temperaturelike parameter β has no direct physical interpretation, but it allows us to scan the structure of the probability distribution P. The corresponding *Ruelle* or *topological pressure* is defined as

$$\psi_L(\beta)=\lim_{t\to\infty}\frac{1}{t}\langle \ln Z_L(\beta,t|x_0)\rangle_{\hat\rho}, \tag{2}$$

which is independent of x_0. The average denoted by $\langle\cdots\rangle_{\hat\rho}$ is performed over *all* configurations of scatterers.

The probability P can be expressed in terms of the transition matrix $w(x|y)$, i.e.,

$$P(\Omega,t|x_0)=\prod_{n=1}^{t}\,w(x_n|x_{n-1}), \tag{3}$$

where $\Sigma_x w(x|y)\leqslant 1$ for open and closed systems.

It has been demonstrated [5] that the Ruelle pressure is related to the largest eigenvalue $\Lambda_L(\beta)$ of the matrix $w_\beta(x|y)\equiv[w(x|y)]^\beta$ by $\psi_L(\beta)=\langle \ln\Lambda_L(\beta)\rangle_{\hat\rho}$, where $w_\beta(x|y)$ is a large random matrix with a rank of order L, whose elements depend upon the realized configuration of scatterers. Numerically, the Ruelle pressure can be computed by determining $\Lambda_L(\beta)$ for a number of randomly generated configurations and averaging $\ln\Lambda_L(\beta)$ over the different realizations.

We outline the main arguments for the one-dimensional (1D) case. Elsewhere [11] we will generalize these results to higher dimensions and present more rigorous derivations. We begin by pointing out that at full coverage of the lattice by scatterers, the LLG reduces to a persistent random walk. For this case the largest eigenvalue is known to be [5]

$$\Lambda_L^{PRW}(\beta)=(a+b)\left[1-\frac{a}{2b}k^2\right]+O(k^3), \tag{4}$$

where $a\equiv p^\beta$, $b\equiv q^\beta$ with $p+q=1$, and $k=0$ for closed systems and $k=\pi/L$ for open systems.

To demonstrate that $\psi_L(\beta,\rho)$ tends to $\psi_\pm(\beta)$ in the thermodynamic limit ($L\to\infty$), we construct upper and lower bounds which approach each other as $L\to\infty$. We start by observing that $Z_L(\beta)$ in (1) is a sum of positive terms, in which $\Sigma_x[w(x|y)]^\beta\leqslant W(\beta)$ with $W(\beta)\equiv 1$ for $\beta>1$ and $W(\beta)\equiv a+b$ for $\beta<1$, both for *closed* and *open* systems. Repeated application of this inequality yields the L-independent upper bound for the topological pressure $\psi_-(\beta)=\ln(a+b)$ for $\beta<1$ and $\psi_+(\beta)=0$ for $\beta>1$.

Next we construct the lower bounds by considering a fixed configuration of scatterers, starting with the case $\beta<1$. We observe that for each configuration of scatterers there exists a largest cluster of scatterers, the length of which is M. (If there are several clusters of maximal size, we choose one of them arbitrarily.) If we restrict the sum in (1) to any subset of trajectories, we get a lower bound for $Z_L(\beta,t)$ as all terms are positive. We choose to keep only those terms for which the particle remains inside the largest cluster. The sum over this set of trajectories is precisely the partition function for a persistent random walk on a lattice of size M with open boundaries. Thus we have the inequalities

$$\langle \ln Z_M^{PRW}(\beta,t)\rangle_{\hat\rho}\leqslant\langle \ln Z_L(\beta,t)\rangle_{\hat\rho}\leqslant t\ln(a+b), \tag{5}$$

where the left-hand term will be determined using (4).

To obtain our lower bound on the Ruelle pressure we need the probability distribution for the largest cluster size, M, which can be obtained from the literature. For our purposes a qualitative estimate will suffice, and more precise estimates will be given in Ref. [11]. The average value of M is roughly determined by the relation $L\rho^{\langle M\rangle}(1-\rho)^2\sim 1$. Indeed the cluster can be located at L different sites, has to be limited by two empty sites, and contains M scatterers. Consequently $M\sim\ln L$ typically, so that $\langle 1/M^2\rangle_{\hat\rho}\sim(\ln L)^{-2}$ and vanishes in the thermodynamic limit. Thus for $\beta<1$ the Ruelle pressure approaches the branch $\psi_\infty(\beta,\rho)=\psi_-(\beta)=\ln(a+b)$. A lower bound for the partition function for $\beta>1$ is obtained by keeping only those trajectories in (1) which remain in the largest region free of scatterers, i.e., the largest hole, whose size is again denoted by M. In fact for a given initial state there is only one trajectory which remains confined to the largest hole, once the particle finds it. This is the trajectory where the particle is continually reflected back into the hole by the scatterers at either end. Its contribution to the partition function is $q^{\beta n_B}$ where n_B is the number of backscatterings of the particle at the boundary sites of the hole. For large t, $n_B\sim t/M$ so that $Z_L(\beta,t|x_0)\geqslant q^{\beta t/M}$ and the lower bound for the pressure is

$$\psi_L(\beta,\rho)\geqslant\beta\langle 1/M\rangle_{\hat\rho}\ln q. \tag{6}$$

The distribution for the largest hole is simply related to that for the largest cluster and we find that $\langle 1/M\rangle_{\hat\rho}\sim O((\ln L)^{-1})$ which vanishes as $L\to\infty$. Consequently we find that for LLG models, in the thermodynamic limit, $\psi_\infty(\beta,\rho)=\psi_+(\beta)=0$, for $\beta>1$.

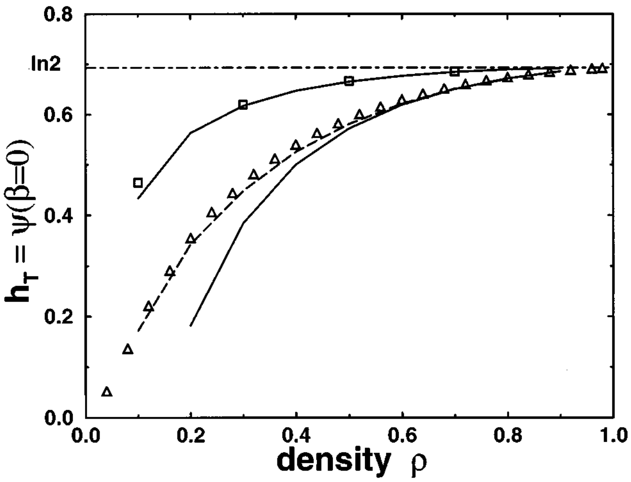

FIG. 1. Ruelle pressure as a function of the density ρ for $L=100$ (△) or $L=10{,}000$ (□). Lower bounds from a numerical determination of the largest cluster size distribution (solid lines) and from an estimate based on effective high density clusters (dashed line).

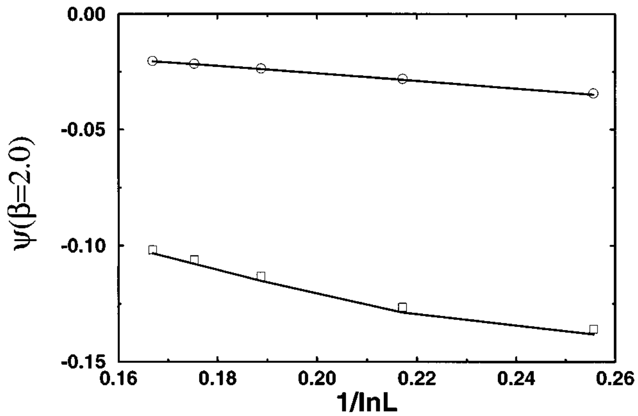

FIG. 2. Ruelle pressure as a function of the system size in the case of strong backscattering ($p=0.2$) for $\rho=0.2$ (○) or $\rho=0.8$ (□). Lower bounds (solid lines) were obtained using a numerical determination of the largest cluster size distribution.

For the 1D LLG we have tested the above predictions against numerical measurements of the largest eigenvalue $\Lambda_L(\beta)$ typically averaged over 10^4 configurations of scatterers. For $\beta=0$, Fig. 1 compares numerical measurements of the topological entropy $h_T = \psi_L(0,\rho)$ (open symbols) with the lower bound in Eq. (5) (continuous lines) for system sizes $L=100$ (triangles) and $L=10^4$ (squares). All points remain below the upper bound of ln2. For $L=100$ we checked that the estimates based upon the largest cluster are indeed lower bounds. As the system size becomes larger it becomes more likely to find a very large cluster dominating the sum in (1). Indeed for $L=10^4$ measurements of $\psi_L(0,\rho)$ and predictions for the lower bounds essentially coincide, except at small densities. The prediction plotted here uses the distribution for the size of the largest cluster as evaluated numerically from random configurations. Similar results were obtained when using theoretical estimates. It should be noted that as the Ruelle pressure converges to its limiting value only logarithmically with system size, the final limit is not accessible numerically. By defining effective ''high density'' clusters with a density above average [12] the lower bounds can be improved to provide a very good estimate for h_T, also at $L=100$, as is shown by the dashed line in Fig. 1.

Simulation results at $\beta=2$ are also in agreement with the lower bound based on the largest hole. For strong backscattering ($p=0.2$), it is not only a lower bound but also a good estimate for the Ruelle pressure for all system sizes (see Fig. 2). This is not the case for weak backscattering ($p=0.8$), where it is less likely that the particle will remain in the largest hole. In all cases the numerical results are found between the upper and lower bounds.

We conclude that in the large system limit, for β away from singular points, properties calculated from the thermodynamic formalism are totally independent of the properties of the disordered lattice such as the density of impurity sites. Nevertheless the thermodynamic formalism appears to be of considerable value even for these systems in a small region about $\beta=1$. For instance, without the thermodynamic formalism, a calculation of the Lyapunov exponents requires not only the largest eigenvalue $\Lambda_L(1)$ of the matrix $w(x|y)$, but also the corresponding left and right eigenvectors. The question remains as to the size of the region about $\beta=1$ where the dominant orbits extend over the entire system. The scaling of the most relevant physical properties of the system by $1/L^2$ suggests replacing β in the thermodynamic formalism by the scaled variable $(1-\beta)L^2$. By comparing our lower bounds on $\Lambda_L(\beta)$ for a given L with the corresponding mean field estimates obtained in [5]—which give a good estimate for the contributions of the extended orbits—we find that the region about $\beta=1$ where extended orbits dominate the pressure is at most of order $1/(\ln L)^\alpha$ where $\alpha=2$ for $\beta<1$ and $\alpha=1$ for $\beta>1$ [11].

In summary, we have shown analytically that the Ruelle pressure $\psi(\beta)$ in LLG's has a singularity at $\beta=1$ in the thermodynamic limit. The preceding results for the Ruelle pressure can be generalized to hopping models on bond and site disordered lattices [10,11]. In general there exists a finite set of singularities. For β values away from these points, the dominant orbits are localized on the largest cluster with a well-defined periodic structure (e.g., compact clusters of the same type of ''impurity'' bonds or sites [10,11], or of alternating ''impurity'' and ''regular'' bonds or sites [10]). At the singular points the trajectories are extended over the whole system ($\beta=1$) or over larger clusters with a well-defined degree of disorder, which is lower than that of the whole system. The different localized states may be considered as ''phases,'' separated by singularities where the states are fully or partially extended.

It is worth pointing out the striking similarity between the dominance of large clusters or holes for the LLG models in determining the pressure, and the nonexponential decay of the density in diffusive systems with fixed random traps, which is also due to the rare occurrence of large areas without traps [13]. Other related phenomena are Griffiths singularities [14] and Lifshitz tails [15].

The above results can be generalized to d-dimensional lattices, and the same conclusions about the Ruelle pressure apply in the thermodynamic limit. In fact we conjecture that in the thermodynamic limit all diffusive systems with static disorder have orbits that are localized for $\beta<1$ on the largest cluster with the most ''chaotic'' scattering and for $\beta>1$ on clusters with the least ''chaotic'' scattering.

For continuous Lorentz gases this would imply that the orbits for $\beta>1$ are localized in the largest empty regions. For $\beta<1$ the situation is less simple. In the case of randomly distributed hard nonoverlapping spherical scatterers of equal radius the escape rate formalism [1] leads to similar results as obtained here for the LLG [11]. The essential difference between periodic and random arrangements of scatterers is that in the thermodynamic limit the dominant orbits always remain extended in the former case but are always localized on *rare* spatial density fluctuations in the latter, and do not explore the typical spatial arrangements of the scatterers.

Finally, based on our experience with these models of static disorder, we conjecture that models of fluids where all particles move will have a more interesting Ruelle pressure for $\beta<1$, but a trivial Ruelle pressure for $\beta>1$, since that region will be dominated by rare trajectories where the particles do not collide.

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