Lyapunov Spectrum and the Conjugate Pairing Rule for a Thermostatted Random Lorentz Gas: Kinetic Theory

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We calculate all four nonzero Lyapunov exponents for a three-dimensional, dilute, random Lorentz gas by combining dynamical systems and Boltzmann equation methods. In the presence of an external field and a Gaussian thermostat the Lyapunov exponents, calculated up to second order in the applied field, satisfy a conjugate pairing rule. Agreement of the results obtained here with those of computer simulations of Dellago and Posch [following Letter, Phys. Rev. Lett. **78**, 211 (1997)]. [S0031-9007(96)02152-7]

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The application of methods from dynamical systems theory to the study of transport phenomena in fluids has recently become an area of active study. Formal theories based upon the assumption that, microscopically, fluids can be considered to be hyperbolic dynamical systems with nonzero Lyapunov exponents have led to very interesting connections between transport coefficients and dynamical quantities such as Lyapunov exponents, and Kolmogorov-Sinai (KS) entropies. For example, the viscosities and diffusion coefficients for fluids can be expressed in terms of the sum of all Lyapunov exponents, when the fluids are in a steady state produced by the application of a thermostatted external force on the particles [1,2]. When the *conjugate pairing rule* applies, the transport coefficients can be obtained from the determination of the values of a single conjugate pair of Lyapunov exponents, rather than having to determine all of them. This rule, which was discovered by Evans, Morriss, and Cohen [3], states that all of the nonzero Lyapunov exponents can be ordered in pairs λ_i, λ_i' in such a way that the sum $\lambda_i + \lambda_i'$ is independent of the index *i*. This generalizes the pairing rule for symplectic systems, where the sum is always zero. A general derivation of this rule has been given by Dettmann and Morriss [4] for Hamiltonian systems with smooth potentials, which are coupled to a thermostat that maintains a constant kinetic energy in the system. It applies to the case of diffusion studied here, with the small exception that we treat hard sphere particles. However, up to the present work there have not been any explicit analytical calculations of the spectrum of Lyapunov exponents for systems usually treated in computer simulations with more than one pair of exponents. A new complication, which did not arise in the two-dimensional case studied earlier [5], stems from the fact that the time evolution operator needed for the computation of the Lyapunov exponents involves time ordered products of noncommuting matrices. Thus it had not yet been possible to make detailed comparisons of numerical with theoretical results for the exponents of systems with more than one pair of exponents.

In this paper we present an analytical calculation of the four Lyapunov exponents for a dilute, three-dimensional Lorentz gas with randomly placed hard sphere scatterers, both for an equilibrium system and for a system where the moving particle is placed in a thermostatted applied field. In both cases our results for each of the exponents agree very well with computer simulations of Dellago and Posch [6] and satisfy the conjugate pairing rule to second order in the electric field $\vec{E}$.

Recently a systematic analytic method was developed that can be used to calculate the sums of the positive and negative Lyapunov exponents, respectively, for dilute systems from an extended Lorentz-Boltzmann equation [5] where a radius of curvature appears as one of the variables. In this paper we use a different method, which combines the ordinary Lorentz-Boltzmann equation with dynamical systems methods, to compute both the sum of the positive Lyapunov exponents, i.e., the KS entropy and the sum of the negative exponents for equilibrium and nonequilibrium steady state systems. Further, we obtain all four of the individual exponents by combining kinetic theory methods with results from the theory for eigenvalues of products of random matrices. In the steady state case we compute the exponents to second order in the applied field. In a subsequent paper we will present more formal methods based upon the arguments given here and elsewhere [5,7] which allow a systematic generalization of these results to higher densities and to larger fields.

We consider the motion of a particle in a random, fixed array of hard sphere scatterers. The number density of the scatterers will be denoted by *n*, the radius of each scatterer by *a*, and we require $na^3 \ll 1$. The position and velocity of the particle are denoted by $\vec{r}$ and $\vec{v}$, respectively, and the particle has mass *m* and charge *q* which couples to a constant external electric field $\vec{E}$. To avoid an infinite increase in energy an isokinetic constraint is applied, which keeps the kinetic energy and, of course, the speed

v of the moving particle constant at all times. The equations of motion are $\dot{\vec{r}} = \vec{v}$; $\dot{\vec{v}} = q\vec{E}/m - \alpha\vec{v}$; $\alpha = q\vec{E} \cdot \vec{v}/mv^2$. Here $-m\alpha\vec{v}$ is the force that maintains a constant kinetic energy for the moving particle. We take the direction of the field to be along the z axis, and set $\vec{E} = (0, 0, E)$. When the particle hits a scatterer, there is a discontinuous change from a precollision velocity $\vec{v}$ to a post-collision velocity $\vec{v}' = \vec{v} - 2(\vec{v} \cdot \hat{n})\hat{n}$, where $\hat{n}$ is the outward normal of the spherical scatterer at the point of impact.

As shown by Sinai [8] the positive (negative) Lyapunov exponents can be related to a radius of curvature matrix ρ of an expanding (contracting) trajectory bundle. That is, we consider the trajectory of the moving particle $[\vec{r}(t), \vec{v}(t)]$, which we call the reference trajectory, and a bundle of trajectories which are infinitesimally close to the reference trajectory. We measure the spreading or contraction of this trajectory bundle with time t by considering a plane $\Sigma(t)$ in space which is perpendicular to the reference trajectory at time t, and then measuring both the spatial and the velocity distances from the reference trajectory to the intersection of the adjacent trajectories with this plane. We denote the spatial difference between the reference trajectory and another one in the bundle by $\delta\vec{r}_\perp(t)$ and the velocity separation of these two trajectories by $\delta\vec{v}_\perp(t)$, defined as the component perpendicular to $\vec{v}(t)$ of the velocity difference of the two trajectories at the points of intersection of the trajectories with the plane $\Sigma(t)$. By construction, both $\delta\vec{r}_\perp(t)$ and $\delta\vec{v}_\perp(t)$ lie in $\Sigma(t)$. Then, in this plane, the radius of curvature matrix $\boldsymbol{\rho}$ is defined by the relation $\delta\vec{v}_\perp(t) = v\boldsymbol{\rho}(t)^{-1} \cdot \delta\vec{r}_\perp(t)$. By means of this construction, $\boldsymbol{\rho}$ is a 2×2 matrix with nonzero eigenvalues. The eigenvalues will be positive (negative) on the expanding (contracting) trajectory bundles.

We first consider the equation of motion for $\boldsymbol{\rho}$ and then relate this matrix to the Lyapunov exponents. The equation of motion for $\boldsymbol{\rho}$ during a free flight can be derived by using the equations of motion given above, and the geometry of the particle trajectories in the thermostatted field. It is of the form

$$\frac{d}{dt}\boldsymbol{\rho} = v\mathbf{I} + \epsilon\cos\theta\boldsymbol{\rho} + \frac{\epsilon^2}{v}\sin^2\theta\boldsymbol{\rho} \cdot \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \cdot \boldsymbol{\rho}\,. \tag{1}$$

Here $\epsilon = qE/mv$ and θ is the angle between $\vec{v}$ and $\vec{E}$, so introducing an arbitrary azimuthal angle φ, one has $\vec{v}(t) = v(\sin\theta(t)\cos\varphi(t), \sin\theta(t)\sin\varphi(t), \cos\theta(t))$, with $\dot{\theta} = -\epsilon\sin\theta$ and $\dot{\varphi} = 0$. The matrix $\boldsymbol{\rho}$ is described in a time dependent coordinate system with its basis vectors in the plane $\Sigma(t)$, such that the first one is orthogonal to $\vec{E}$. Because of the scattering of the moving particle by the hard spheres, there is a discontinuous change in $\boldsymbol{\rho}$ at each collision. On the expanding manifold the value of $\boldsymbol{\rho}$ immediately after a scattering event can be written in terms of a scattering geometry and the precollision matrix $\boldsymbol{\rho}'$ as

$$\boldsymbol{\rho}^+ = [(2/a)\cos\phi\mathbf{P} + \mathbf{I}/\boldsymbol{\rho}']^{-1},$$
$$\mathbf{P} = \begin{pmatrix} 1 + \tan^2\phi\cos^2\psi & \tan^2\phi\cos\psi\sin\psi \\ \tan^2\phi\cos\psi\sin\psi & 1 + \tan^2\phi\sin^2\psi \end{pmatrix}, \tag{2}$$

where ϕ is the angle between the outward normal $\hat{n}$ on the sphere and $\vec{v}$, and ψ is the angle between $\hat{n}$ and the plane through $\vec{v}$ and $\vec{E}$.

The separation of the trajectories in space at time t, $\delta\vec{r}_\perp(t)$ can be written in terms of the initial separation $\delta\vec{r}_\perp(0)$ as

$$\delta\vec{r}_\perp(t) = T\exp\left[v\int_0^t \frac{\mathbf{I}}{\boldsymbol{\rho}(t')}\,dt'\right] \cdot \delta\vec{r}_\perp(0)$$
$$\equiv \mathbf{U}(t) \cdot \delta\vec{r}_\perp(0)\,, \tag{3}$$

where T is the time ordering operator. The sum of the positive Lyapunov exponents is defined as the limit value for $t \to \infty$ of the growth rate $1/t\ln S(t)$ of an area element $S(t)$ in the plane $\Sigma(t)$ [8]. $S(t)$ is given by $\det\mathbf{U}(\mathbf{t})S(0)$, where $S(0)$ is an arbitrary initial surface element. Writing U as $\prod_{i=1}^N \exp[v/\boldsymbol{\rho}(t_i)\Delta t] + O((\Delta t)^2)$ where $t = N\Delta t$ and using the fact that the determinant of a product of matrices is equal to the product of the determinants, one finds that $\lambda_1^+ + \lambda_2^+ = \lim_{t\to\infty}(\log\det\mathbf{U})/t = \lim_{t\to\infty}[\mathrm{Tr}(\ln\mathbf{U})]/t = \lim_{t\to\infty}[v\int_0^t dt'\mathrm{Tr}\,\boldsymbol{\rho}^{-1}(t')]/t$.

By using the fact that reversing the direction of time and the velocities of a contracting trajectory bundle leads to an expanding bundle, we can apply these results to obtain the sum of the negative Lyapunov exponents also.

A calculation of the sum of the positive exponents for a dilute random Lorentz gas using kinetic theory arguments proceeds as follows: We first note that immediately after a collision, the matrix $\boldsymbol{\rho}$ is, to leading order in n, given by $[(2/a)\cos\phi\mathbf{P}]^{-1}$ since the additional term appearing on the right hand side of Eq. (2) will give contributions on the order of na^2 compared to the other terms which are of order a^{-1}. Thus, at low density, the matrix $\boldsymbol{\rho}$ at any time depends only upon the parameters ϕ and ψ, of the most immediate past collision and not upon the parameters of any collision prior to that. One can then calculate the time integral of the trace of $\boldsymbol{\rho}^{-1}$ by following the collision history of the moving particle and breaking up the integration time into a set of intervals, each starting at the instant after a collision and ending at the time immediately before the next collision. The length of these time intervals will be distributed according to the distribution of free flight times. Next, one assumes that the motion of the particle is ergodic, so that the time averages can be replaced by an ensemble average over a suitable distribution of velocity directions, collision parameters, and free flight times. We assume that the system is in a spatially homogeneous nonequilibrium steady state produced by the field and the thermostat. The equilibrium case is easily obtained by setting the field strength equal to zero. We then write

$$\lambda_1^+ + \lambda_2^+ = \frac{v\nu}{4\pi^2}\int_0^{\pi} d\theta \sin\theta \int_0^{2\pi} d\varphi \times \int_0^{\infty} \nu\, dt\, \exp[-\nu t] \int_0^t dt' \int_0^{\pi/2} d\phi \times \int_0^{2\pi} d\psi\, \sin\phi \cos\phi\, f(\theta'(\phi,\psi)) \mathrm{Tr}\boldsymbol{\rho}^{-1}(\theta,\phi,\psi,t'). \quad (4)$$

Here ν is the collision frequency, or inverse mean free time, given by $\nu = na^2 v\pi$, and $\nu \exp -[\nu t]$ is the low density form of the distribution of free flight times, which is independent of the field strength since the particle's speed is constant. The quantity $na^2 v \sin\phi \cos\phi \sin\theta f(\theta'(\phi,\psi)) d\phi d\psi d\theta$ is the number of particles scattered per unit time to angles between θ and $\theta + d\theta$ with scattering angles between ϕ and $\phi + d\phi$ and ψ and $\psi + d\psi$. Here θ' is the precollision value of the angle that the velocity makes with the z axis, such that after collision this angle is θ, given by $\cos\theta' = \cos\theta - 2\cos\phi[\cos\theta\cos\phi + \sin\theta\sin\phi\cos(\psi - \varphi)]$, where φ is an azimuthal angle for the collision with the scatterer at the initial time. Further $f(\theta)$ is the stationary angular distribution function for directions of the velocity of the moving particle, obtained by solving the Lorentz-Boltzmann equation for a spatially homogeneous steady state; i.e., $f(\theta) = 1 + 2\epsilon\cos\theta/\nu + O(\epsilon^2)$. Normalization factors have also been included in Eq. (4). The trace of $\boldsymbol{\rho}^{-1}$ is determined up to second order in ϵ by solving Eq. (1) as a power series in ϵ with the initial condition $\boldsymbol{\rho} = (2\cos\phi/a\mathbf{P})^{-1}$. These calculations may be simplified by working in a representation where $\boldsymbol{\rho}$ is diagonal just after collision. All that then remains is to carry out the required integrals in Eq. (4).

The sum of the negative Lyapunov exponents can be computed in a similar way. Here one uses the fact that negative Lyapunov exponents are determined by trajectory bundles that contract exponentially with time. The equation for the radius of curvature matrix for the contracting trajectory bundle is obtained from Eq. (1) by reversing the time direction ($t \to -t$) and reversing the direction of the velocity ($\theta \to \pi - \theta$). One can then follow the motion of the contracting bundle from scatterer 1 to scatterer 2 (see Fig. 1) and require that the radius of curvature matrix just before the collision with 2 is very close (with corrections of relative order na^3) to $[2\cos\phi/a\mathbf{P}]^{-1}$ where $\mathbf{P}$ is given by Eq. (2) with ϕ, ψ the scattering angles at the collision with 2. This is required if the trajectory bundle continues to be contracting after the collision with 2.

We denote the velocity direction after the collision with 1 by θ again, and the velocity direction just before the collision with 2 by θ_f. The relation between these two angles is $\cos\theta_f = \cos\theta + \epsilon t \sin^2\theta + O(\epsilon^2)$, where t is the time between the two collisions. The sum of the negative Lyapunov exponents can then be obtained by changing the sign on the right hand side of Eq. (4)—to account for the fact that the trajectory bundle is contracting—and using the solution to the equation for the contracting radius of curvature matrix, with the above mentioned condition immediately before the collision with 2, in the integrand. In this way one finds that

$$\frac{a}{v}\sum_i \lambda_i^{\pm} = \mp 2\tilde{n}\left(\ln\frac{\tilde{n}}{2} + C\right) - \left(\frac{2}{3} \mp \frac{1}{18}\right)\frac{\tilde{\epsilon}^2}{\tilde{n}}. \quad (5)$$

C is Euler's constant, $\tilde{n} = \pi n a^3$, and $\tilde{\epsilon} = a\epsilon/v$. The total sum of the negative and positive exponents agrees with the general results of Evans and Hoover for the relation between the phase space contraction to a steady state attractor and the entropy production $\sum_i \lambda_i = \langle \frac{d\dot{\Gamma}}{d\Gamma}\rangle = -2\langle\alpha\rangle$ [1,2], where $\Gamma = (\vec{r}, \vec{v})$ is a vector in phase space. The average of the friction coefficient α is related to the diffusion coefficient D by $\langle\alpha\rangle = 2D\epsilon^2/v^2$ for small fields, and one can immediately check that this relation is satisfied by our results. Further, Pesin's theorem [9] allows us to identify the sum of the positive Lyapunov exponents with the KS entropy of the system. The zeroth order term in ϵ is the equilibrium KS entropy. The term of order $\tilde{n}\ln\tilde{n}$ agrees with the value of the Kolmogorov-Sinai entropy for a periodic Lorentz gas at low density given by Chernov [10], while our results also give the order $\tilde{n}$ terms as well as the KS entropy for the system in small fields. The agreement with simulations is excellent for small densities and electric fields as can be seen from the results of Dellago and Posch in the accompanying Letter [6]. It is worth pointing out that the computer results provide an accurate check of the order $\tilde{n}$ terms in the equilibrium KS entropy.

To calculate the individual exponents we must combine the results for the sums of the two positive or two negative exponents with a calculation of the largest magnitude using results from the theory of products of random matrices [11]. This latter method can be applied without major difficulties because for low densities, the radius of curvature matrix depends only on the previous collision and not on the collisions that preceded it.

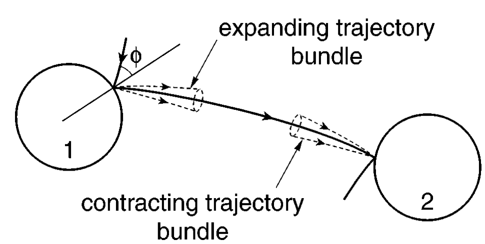

FIG. 1. A particle scattered by sphere 1 with scattering angle ϕ collides with sphere 2 after a free flight. The expanding trajectory bundle, related to the positive Lyapunov exponent, is indicated with dashed lines at scatterer 1. The converging bundle, relevant for the calculation of the negative Lyapunov exponents, is shown as dashed lines at sphere 2.

Therefore the operator $\mathbf{U}(t)$ becomes a product of random matrices $\mathbf{U}_i$ where $\mathbf{U}_i = \exp(v \int_{t_i}^{t_{i+1}} \frac{1}{\boldsymbol{\rho}(t')} dt')$ is the free propagator between collision i and $i + 1$. Each $\mathbf{U_i}$ is determined by the value of $\boldsymbol{\rho}$ after the ith collision (i.e., by the scattering angles ϕ and ψ, which, as before, are sampled from a random distribution) and by the random time intervals $t_{i+1} - t_i$ between collisions. From the theory for products of random matrices [11] it is known that the largest Lyapunov exponent is equal to $\lambda_{\max} = \nu\langle \ln\Lambda\rangle_\beta$, with $\Lambda = |\mathbf{U_i} \cdot \hat{r}|$, where $\hat{r} = (\cos\beta, \sin\beta)$ is an arbitrary unit vector. The subscript β indicates an average over β, which has to be performed in addition to the average over the random matrix parameter. In our case one can show that Λ has a rotationally invariant distribution up to order ϵ to lowest order in the density, i.e., $P(\beta) = \frac{1}{2\pi} + \cos 2\beta O(\epsilon^2)$. The term to order ϵ^2 in $P(\beta)$ does not contribute to the Lyapunov exponents.

The calculation of the individual Lyapunov exponents is now a straightforward extension of the method used above. For the calculation of the largest positive exponent λ_1^+, the required average of $\ln|\mathbf{U_i} \cdot \hat{\mathbf{r}}|$ is taken over the distribution of free flight times, the collision parameters ϕ, ψ, and over β. Again, one expands both the distribution function f and the logarithm of the matrix product in powers of ϵ and computes λ_1^+ as a series in ϵ. The calculation of the negative Lyapunov exponent with the largest magnitude λ_1^- is similar to the calculation of the sum of the negative exponents computed earlier. One considers the contracting bundle along the trajectory from collision 1 to 2 in Fig. 1. The result for the four Lyapunov exponents can now be given as

$$\lambda_1^{\pm} = \mp \frac{v}{a}\,\tilde{n}\left(\ln\frac{\tilde{n}}{2} + C - \ln 2 + 1/2\right) - \left(\frac{1}{3} \mp \frac{1}{36}\right)\frac{v\tilde{\epsilon}^2}{a\tilde{n}}, \tag{6}$$

$$\lambda_2^{\pm} = \mp \frac{v}{a}\,\tilde{n}\left(\ln\frac{\tilde{n}}{2} + C + \ln 2 - 1/2\right) - \left(\frac{1}{3} \mp \frac{1}{36}\right)\frac{v\tilde{\epsilon}^2}{a\tilde{n}}.$$

Therefore the Lyapunov exponents fulfill the conjugate pairing rule, $\lambda_i^+ + \lambda_i^- = -\langle\alpha\rangle$ independent of the index i. The field dependent corrections to the two different equilibrium values of the positive (negative) exponents are the same.

The quantitative agreement of (6) with the simulations for both the equilibrium and the steady state systems is again very satisfactory [6].

We conclude with a few remarks. We have shown that it is possible to compute the Lyapunov spectrum for a simple random system using analytical methods, based upon the Boltzmann transport equation. The results obtained here are in excellent agreement with computer simulations and provide a concrete example of the validity of the conjugate pairing rule for thermostatted systems. The success of this calculation suggests that it might very well be possible to obtain analytical results for Lorentz gases at higher densities and at higher field strengths and for more complicated systems where all of the particles move.

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