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Diffusion and long-time tails in the overlapping Lorentz gas

Jonathan Machta and Steven M. Moore
Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01003
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A number of new features of the overlapping Lorentz gas are obtained using a method based on describing the system as a random walk on a disordered lattice. The velocity correlation function is shown to have a contribution which decays like $t^{-[2+1/(d-1)]}$. This term is the dominant long-time tail for $d>3$ and above the percolation threshold. New values are obtained for the exponents describing the vanishing of the diffusion coefficient and the intermediate-time tail near threshold.

This Rapid Communication presents a qualitative study of the self-diffusion process in a simple dynamical system. The system in question is the overlapping Lorentz gas consisting of a single point particle moving with unit speed according to classical mechanics in a random array of overlapping spherical (or hyperspherical) scatterers. Previous studies of the overlapping Lorentz gas have relied on kinetic theory,[1-5] mode-coupling theory,[6,7] and computer simulation.[8-10] The present work takes quite a different approach and approximates the motion of the particle as a random walk on a disordered lattice. This approach was developed by Machta and Zwanzig[11] for the Lorentz gas with circular scatterers on a triangular lattice. Similar methods have been applied recently to study transport in a variety of disordered materials.[12,13] Using this approach we find new features in the transport properties of the overlapping Lorentz gas.

Lorentz gases are important models in the study of nonequilibrium transport phenomena[1]—they are simple enough that significant progress can be made by studying them analytically and yet complex enough to display behavior which is qualitatively like real systems, particularly classical fluids. When the density of scatterers n is sufficiently low, the particle moves diffusively through the system. Like fluid systems, the diffusion coefficient has a nonanalytic dependence on the density[2] at $n=0$ and the velocity correlation function has an asymptotic power-law decay or long-time tail.[3] As the density of scatterers in the overlapping Lorentz gas is increased a critical density is reached at which the void space in which the particle moves no longer percolates and diffusion ceases. Self-consistent kinetic theories[4,5] and self-consistent mode-coupling theories[6] have been developed which predict a percolation threshold and exponents for the vanishing of the diffusion coefficient as the threshold is approached. These self-consistent theories have also been used to study the decay of the velocity correlation function (VCF) at high density. Below the percolation threshold they predict an ultimate $t^{-(1+d/2)}$ long-time tail and, as the threshold is approached, an increasingly long penultimate decay or intermediate-time tail before the onset of the asymptotic long-time tail. By matching the ultimate and penultimate long-time tails, the amplitude of the long-time tail near the threshold has also been predicted.[14]

In this paper we derive a number of new results for the overlapping Lorentz gas. We show that a trapping mechanism, discussed elsewhere by Machta,[15] leads to a new contribution to the long-time behavior of the VCF which decays like $t^{-[2+1/(d-1)]}$. Our other findings concern the transport properties of the system near the percolation threshold. These results agree qualitatively with the self-consistent theories and with a real-space renormalization-group treatment,[16] but disagree in the values of the exponents.

The basic idea of our approach is a coarse graining in which the particle motion is described as a random walk on a disordered lattice. The sites of the lattice are voids or chambers formed between the scatterers, and the bonds are the relatively narrow channels connecting neighboring chambers. If the connecting channels are narrow, the particle collides many times with the walls of a single chamber before moving to the next chamber and, hence, loses memory of the conditions of its entrance to the chamber. We therefore treat the dynamics as a Markovian random walk.

The partitioning of the void space between the scatterers into sites and bonds of a random lattice is naturally accomplished through a Voronoi tesselation (Wigner-Seitz cells) constructed around the centers of the scatterers. This construction is described and applied to a random array of spheres by Kerstein.[17] In the discussion which follows we use the language of three dimensions; the arguments are generalized to other dimensions by appropriate reinterpretation of "area," "volume," "surface," and "triangle." The Voronoi tesselation partitions space into polyhedra whose faces are surfaces perpendicular to and bisecting the lines joining the centers of the scatterers. Only those surfaces which are closest to the centers of scatterers are kept. The bonds of the random lattice are defined by the edges of the tesselation and the sites by the vertices.

The centers themselves are the vertices of packed polyhedra with triangular faces, three centers determining each triangle. Given spheres of radius R located at these centers, the intersection of each triangle with the spherical surfaces defines a minimal area in void space perpendicular to each bond which we call the "port," whose area is denoted by $A_{i,j}$ (port between site i and j). The ports and spherical surfaces of the scatterers surrounding a site define the boundary of the "chamber" associated with the site j, whose volume is denoted by V_j.

A bond is either blocked or unblocked. The latter occurs if the area $A_{i,j}$ of the port is nonzero.[13] The moving particle can pass directly between two sites only if they are connected by an unblocked bond. These considerations are illustrated in Fig. 1, which shows a portion of a random array of circles and the Voronoi tesselation about their centers.

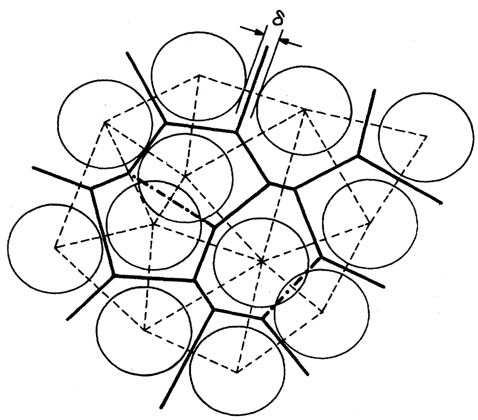

FIG. 1. Sketch of a small region of the configuration space showing scatterers (circles), bonds constructed via the Voronoi tesselation and a typical channel width, δ. Open bonds are indicated by heavy solid lines, while blocked bonds are indicated by heavy broken lines.

The next step in the analysis consists of assigning transition rates to the unblocked bonds of the lattice. The technique for doing this was discussed in Ref. 11 and, using a projection operator formalism, in Ref. 18. To determine the rate $W_{i,j}$ for hopping from site j to site i, we appeal to the ergodic hypothesis. The probability of leaving a chamber through a given port in time dt if the particle is distributed within the chamber according to the ergodic measure is the ratio of the phase space available for leaving the chamber in time dt to the total phase space in the chamber. Thus we identify the transition rate, for open bonds, as

$$W_{ij} = S(d) A_{ij} / V_j \ , \tag{1}$$

where $S(d) = \Gamma(d/2)/2\pi^{1/2}\Gamma[(d+1)/2]$ is a geometric factor which depends on the dimension d. For blocked bonds the transition rate vanishes.

If the moving particle bounces many times in a chamber before moving to the next chamber there will be no correlation between entering and leaving a chamber and the random walk taken by the moving particle is described by the Markovian master equation,

$$dP_i(t)/dt = \textstyle\sum_j [W_{i,j}P_j(t) - W_{j,i}P_i(t)] \ , \tag{2}$$

where $P_i(t)$ is the probability that the moving particle is in chamber i at time t. We conjecture that the non-Markovian aspects of a more accurate stochastic description are irrelevant to the determination of the exponents studied here and take Eq. (2) as a starting point for the analysis. It was shown in Ref. 11 that Eq. (2) gives very accurate results for the two-dimensional Lorentz gas on a triangular lattice near close packing.

The transport properties which are emphasized in this paper depend only on the small-W behavior of the transition-rate probability density, $\rho(W)$. Small values of W result from small port areas whose likelihood we now calculate. To understand the geometry of the Lorentz gas at various scatterer densities we may either vary the density of the scatters N/V holding the radius fixed, or we may hold the density fixed and vary the radius. For the following argument we take the latter approach. The triangle (whose vertices are scatterer centers) defining the port ij can be parametrized by the minimum radius Δ_{ij} for which the port area A_{ij} vanishes. If the actual scatterers have a radius R which is slightly less than Δ_{ij}, port ij will be small and nearly triangular with a linear size characterized by $\delta_{ij} = (\Delta_{ij} - R)$ and area proportional to δ_{ij}^2. For arbitrary d,

$$A_{ij} \sim (\delta_{ij})^{d-1} \ . \tag{3}$$

Let $G(X)$ be the cumulative probability that $\Delta_{ij} < X$ and let $F_R(X)$ be the cumulative probability that $\delta_{ij} < X$. The relation between these two distributions is then

$$1 - F(\delta) = [1 - G(\delta + R)]/p(R) \ , \tag{4}$$

where $p(R)$ is the fraction of unblocked bonds,

$$p(R) = [1 - G(R)] \ . \tag{5}$$

Thus, for small δ,

$$F_R(\delta) \sim \delta[G'(R)/p(R)] \ . \tag{6}$$

Since triangles of all sizes are possible in the tesselation, both $G'(R)$ and $p(R)$ are finite for any R. This means that the probabilty density for δ is finite at $\delta = 0$, a result announced[12] previously for the geometrically identical "Swiss-cheese" model of random media. Finally, since finite chamber volumes can coexist with arbitrarily small ports, the small W singularity in $\rho(W)$ is controlled by the small-δ behavior of $F_R(\delta)$ through Eqs. (1), (3), and (6),

$$\rho(W) \sim \frac{dF}{d\delta}\frac{d\delta}{dA}\frac{dA}{dW} \sim W^{\alpha-1}, \quad W \to 0 \ , \tag{7a}$$

with

$$\alpha = 1/(d-1) \ . \tag{7b}$$

Similarly, near the percolation threshold,

$$p - p_c \approx G'(R_c)(R_c - R) = [R_c G'(R_c)/dn_c](n_c - n) \ , \tag{8}$$

where $n = NR^d/V$ is the reduced density conventionally used to characterize Lorentz gases. From Eq. (8) we see that, for exponent relations, the distance to the percolation threshold may be measured either in terms of the bond probability p or the reduced density n.

The small-W singularity in $\rho(W)$ determines a number of new features of transport in the overlapping Lorentz gas. To obtain these features we apply known results from percolation theory and the theory of disordered random walks. The first new result is a contribution to the long-time behavior of the VCF due to trapping in cul-de-sacs. Some fraction of the sites are connected by only one open bond to the other sites in the network. The velocity entering such a cul-de-sac is anticorrelated with the velocity for leaving, resulting in a contribution to the VCF which decays like $-\exp(-Wt)$, where W is the transition rate for leaving the cul-de-sac. To leading order, each cul-de-sac contributes additively to the VCF and integrating over the small-W singularity in $\rho(W)$ yields a power-law decay of the VCF. The general problem of random walks with cul-de-sacs was studied in Ref. 15, where it was found that the new contribution to the VCF decays as $t^{-(2+\alpha)}$. In that paper it was shown that this mechanism is distinct from the mode-coupling[6,7,19] or ring-event mechanism,[1,3] which leads to a $t^{-(d/2+1)}$ decay of the VCF for $n < n_c$ and no long-time tail for $n > n_c$. The dominant mechanism thus depends on the dimension and the density.

We define exponents l_+ and l_- describing the asymptotic decay of the velocity correlation function $\phi(t)$ above and below the threshold, respectively,

$$\phi(t) \sim \begin{cases} t^{-l_+}, & n > n_c \\ t^{-l_-}, & n < n_c \ . \end{cases} \tag{9}$$

Applying the results of Ref. 7 and Ref. 15 with α given in Eq. (7b) we have

$$l_- = \begin{cases} 2, & d=2 \\ 2+1/(d-1), & d \geqslant 3 \ , \end{cases} \tag{10a}$$

and

$$l_+ = 2+1/(d-1) \ . \tag{10b}$$

Numerical values of these and other exponents are listed in Table I and compared there to other theories. Since the formation of a cul-de-sac requires at least $d+1$ scatterers and since the moving particle bounces many times from each scatterer forming the cul-de-sac before leaving, this new long-time tail first appears in a density expansion of the VCF at order n^{d+1} and would be difficult to predict from a kinetic theory calculation.

Next we consider features of the overlapping Lorentz gas which appear near the percolation threshold. Here we apply results from the theory of percolation on a regular lattice with a distribution of bond strengths. This theory was developed by Kogut and Straley,[20] Ben-Mizrahi and Bergman,[21] and Straley.[22] Although these three references disagree with one another, we believe the arguments presented most recently by Straley[22] are correct. He finds that the conductivity exponent as a function of α,[23] $t(\alpha)$, is given by the greater of $(d-2)\nu+1/\alpha$ and t, where t is the conductivity exponent for ordinary percolation and ν is the correlation length exponent. The exponent τ characterizes the disappearance of the diffusion constant at the percolation threshold: $D \sim (n_c - n)^\tau$. From the Einstein relation we have that $\tau = t(\alpha)$. Use of Ref. 22 for $t(\alpha)$ and Eq. (7b) for α yields

$$\tau = \begin{cases} t, & d=2 \\ (d-2)\nu+(d-1), & d \geqslant 3 \ . \end{cases} \tag{11}$$

The singular distribution of weak bonds is irrelevant in two dimensions but has a marked effect in three dimensions and higher.

Gefen, Aharony, and Alexander[24] have shown that near the percolation threshold there is anomalous diffusion on length scales much greater than the mean-bond length but less than the correlation length. They find anomalous diffusion,

$$\langle r^2(t) \rangle \approx t^{(2\nu-\beta)/(2\nu+t-\beta)} \ , \tag{12}$$

in the range $1 < \langle r^2(t) \rangle < \xi$, where $\langle r^2(t) \rangle$ is the mean-squared displacement of the moving particle and β is the exponent for the probability of being on the infinite cluster. We use their results to obtain an intermediate-time power-law decay of the VCF near the percolation threshold. The only modification which must be made in the present case is to replace t by τ. Since the velocity correlation function $\phi(t)$ is one half the second derivative of the mean-squared displacement we obtain an intermediate-time tail with exponent f given by

$$f = 1+\tau/(2\nu+\tau-\beta) \ , \tag{13}$$

where the exponent f is defined as the asymptotic behavior of the VCF exactly at n_c,

$$\phi(t) \sim t^{-f}, \quad n = n_c \ . \tag{14}$$

The exponent relation, Eq. (13), was derived previously by Keyes.[14]

Numerical values of τ and f are given in Table I for two and three dimensions and compared with self-consistent theories. In evaluating these expressions we take the most recent lattice percolation values for t, ν, and β for $d=2$, (Ref. 25) and $d=3$ (Ref. 26) from the literature. There is strong evidence that the geometrical properties of continuum percolation characterized by β and ν are in the same universality class as lattice percolation.[27,28]

As one approaches n_c, the power law t^{-f} persists to longer and longer times before crossing over to t^{-l_+} or t^{-l_-}. In computer experiments near the percolation threshold it is likely that only the intermediate-time regime can be observed. Gotze, Leutheusser, and Yip[6] show that this leads to an apparent exponent which diminishes from l_- to f as the threshold is approached from below. Computer simulations of the two-dimensional overlapping Lorentz gas

TABLE I. Values of the exponents for $d=2$ and 3 as compared to the self-consistent theories of Refs. 4 and 6. The self-consistent theories for τ, f, and l_+ are independent of dimension. For $d=2$ and 3, self-consistent values of l_- agree with the present work. Required lattice values of t, ν, and β were taken from recent values in the literature for $d=2$ (Ref. 25) and 3 (Ref. 26).

	Present work		Self-consistent theories	
	$d=2$	$d=3$	Ref. 4	Ref. 6
t	1.29	2.2	...	...
τ	1.29	2.89	$\frac{1}{2}$	1
l_-	2	$\frac{5}{2}$	Same as present work	Same as present work
f	1.34	1.69	$\frac{4}{3}$	$\frac{3}{2}$
l_+	3	$\frac{5}{2}$	∞	∞

by Alder and Alley[10] show this effect, and extrapolation of their density-dependent exponent to n_c leads to the value $f = 1.26$, in good agreement with our results and those of Ref. 4.

The results of our method, summarized in Table I, differ from those of previous studies using self-consistent mode coupling and kinetic theories. The reason for this difference is twofold. First of all, self-consistent theories and equivalent effective medium theories[29] of disordered media break down near the percolation threshold.[30] The second reason, which becomes increasingly important in higher dimensions, is that these theories do not incorporate the singularity in the distribution of transition rates.

In conclusion, we have studied the overlapping Lorentz gas using a method based upon treating the dynamics as a random walk on a disordered lattice. An important property of this random walk is that it has a random distribution of transition rates with a singularity at small transition rates. This singularity leads to the new contribution to the velocity correlation function and the deviation of our exponents from those of conventional percolation theory. Halperin, Feng, and Sen[12] argue that these deviations are a generic feature of transport properties in continuum systems with a percolation threshold, and the overlapping Lorentz gas serves as a particularly simple model in which to test these ideas.

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