

Density-Dependent Diffusion in the Periodic Lorentz Gas

R. Klages^{1,3} and Christoph Dellago²

Received November 22, 1999; final June 13, 2000

We study the deterministic diffusion coefficient of the two-dimensional periodic Lorentz gas as a function of the density of scatterers. Based on computer simulations, and by applying straightforward analytical arguments, we systematically improve the Machta–Zwanzig random walk approximation [*Phys. Rev. Lett.* **50**:1959 (1983)] by including microscopic correlations. We furthermore, show that, on a fine scale, the diffusion coefficient is a non-trivial function of the density. On a coarse scale and for lower densities, the diffusion coefficient exhibits a Boltzmann-like behavior, whereas for very high densities it crosses over to a regime which can be understood qualitatively by the Machta–Zwanzig approximation.

KEY WORDS: Deterministic diffusion; periodic Lorentz gas; computer simulations; random walk; chaotic scattering; Boltzmann approximation.

1. INTRODUCTION

One of the central themes in the theory of chaotic transport is the problem of deterministic diffusion. Over the past several years, deterministic diffusion coefficients have been computed for a variety of low-dimensional model systems, in particular for the periodic Lorentz gas.⁽¹⁾ This model mimics classical diffusion in a crystal, but is as well isomorphic to a periodic system of two hard disks per unit cell. Motivated especially by the rigorous mathematical analysis of Bunimovich and Sinai,⁽²⁾ a great deal of

¹ Center for Nonlinear Phenomena and Complex Systems, Université Libre de Bruxelles, Campus Plaine CP 231, Blvd du Triomphe, B-1050 Brussels, Belgium.

² Department of Chemistry, University of Rochester, Rochester, New York 14627. E-mail: dellago@chem.rochester.edu

³ Present address: Max Planck Institute for Physics of Complex Systems, Noethnitzer Str. 38, D-01187 Dresden, Germany. E-mail: rklages@mipiks-dresden.mpg.de

work on the dynamical and transport properties of this model has been published.⁽³⁻⁶⁾ In particular, its diffusion coefficient has been computed by a variety of methods.⁽⁷⁻¹³⁾

In this paper we focus on the behavior of the diffusion coefficient in the two-dimensional periodic Lorentz gas under variation of the density of scatterers. In Section 2 we define the model, briefly sketch the simple analytical approximation obtained by Machta and Zwanzig⁽⁷⁾ and compare it to detailed numerical results obtained from computer simulations. In Section 3 we explain how the Machta–Zwanzig argument can be corrected systematically by taking microscopic correlations into account. In Section 4 we compare our numerical diffusion coefficient to a simple Boltzmann approximation and argue that, on a coarse scale, there is a dynamical crossover for the diffusion coefficient as a function of the density. More detailed computer simulations show that the diffusion coefficient exhibits a non-trivial fine structure as a function of the density. In Section 5 we summarize our results and relate them to analogous findings in low-dimensional maps.

2. DIFFUSION AS A SIMPLE MARKOV PROCESS

The geometry of the periodic Lorentz gas is depicted in Fig. 1a: A point particle of mass m moves with constant velocity v in an array of circular hard scatterers of radius R arranged on a triangular lattice. Upon collisions with the scatterers the particle is reflected elastically. In the following we use units for which $v = 1$, $m = 1$, and $R = 1$. The lattice spacing of the disks is then $2 + w$, where w is the smallest inter disk distance. The gap size w is related to the number density n of the disks by

$$n = 2/[\sqrt{3}(2 + w)^2] \quad (1)$$

The gap size w , or the number density of the disks n , respectively, is the only control parameter. At close packing $w = 0$ the moving particle is trapped in a single triangular region formed between three disks, see Fig. 1a. For $0 < w < w_\infty = 4\sqrt{3} - 2 = 0.3094$, the particle can move across the entire lattice, but it cannot move collision-free for an infinite time. For $w > w_\infty$ the particle can move arbitrarily far between two collisions. Here, w_∞ denotes the gap size at which the particle first sees such an “infinite horizon.” Bunimovich and Sinai proved that for $0 < w < w_\infty$ the system is ergodic, and that the diffusion coefficient exists, while it diverges for $w > w_\infty$.⁽²⁾

In ref. 7 Machta and Zwanzig have derived a simple analytical approximation for the diffusion coefficient D . The basic idea is that diffusion

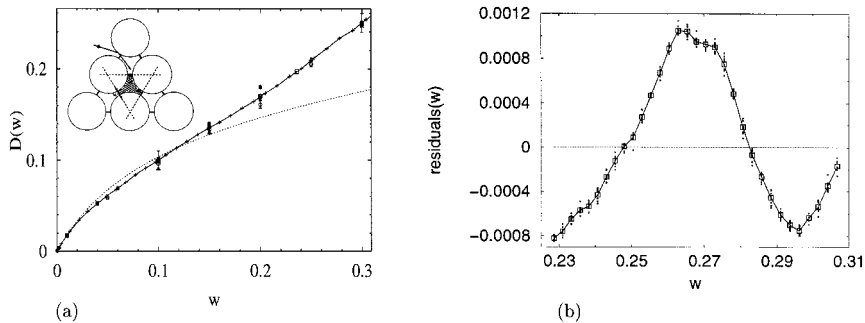


Fig. 1. (a) Geometry of the periodic Lorentz gas and its diffusion coefficient D as a function of the gap size w . The dotted line in the diagram represents the Machta-Zwanzig random walk model Eq. (2), the different symbols refer to single data points obtained in the literature from various methods (see text). The crosses connected with lines are our results from computer simulations. All of our data points in this plot have error bars smaller than the symbols. Error bars of the other data points have been included as far as available. (b) Blowup of the region of large w of (a) in terms of the residuals, that is, the deviations of the diffusion coefficients D from a linear fit.

can be treated as a Markovian hopping process between the triangular trapping regions indicated in Fig. 1a. For this purpose, they have calculated the average rate τ^{-1} at which a particle leaves such a trap. According to a simple phase space argument, this rate is determined by the fraction of phase space available for leaving the trap divided by the total phase space volume of the trap. Furthermore, for random walks on two-dimensional isotropic lattices the diffusion coefficient is $D = l^2/(4\tau)$, where $l = (2 + w)/\sqrt{3}$ is the distance between the centers of the traps. This leads to the Machta-Zwanzig random walk approximation of the diffusion coefficient

$$D_{\text{MZ}} = \frac{w(2 + w)^2}{\pi[\sqrt{3}(2 + w)^2 - 2\pi]} \quad (2)$$

which is shown as the dotted line in the diagram of Fig. 1a. Included in this figure are single data points for the diffusion coefficient obtained by various authors using different methods: The filled circles are numerical results of ref. 7, the stars are from ref. 8. These data points have been obtained by employing the Green-Kubo formula, where D is determined from an integral over the velocity autocorrelation function. The squares have been computed in ref. 11 by periodic orbit expansions, the empty circles and triangles are from ref. 13, where they have been computed by applying the escape rate formalism, and via the fractal dimension of the repeller of the

Lorentz gas, respectively. The crosses connected with lines represent our new data which we have obtained from computer simulations by means of the Einstein formula

$$D = \lim_{t \rightarrow \infty} \frac{\langle \Delta r^2(t) \rangle}{4t}, \quad (3)$$

where the brackets indicate an ensemble average after time t . We calculated $\langle \Delta r^2(t) \rangle$ by averaging over long trajectories. Depending on the density n each trajectory contained from 6×10^8 to 3×10^9 collisions. After a short transient $\langle \Delta r^2(t) \rangle$ grows linearly with a slope of $4D$. We determined the diffusion coefficient D by fitting a straight line to $\langle \Delta r^2(t) \rangle$ in the linear regime. Though in principle equivalent with the Green–Kubo formalism, the Einstein approach is numerically more efficient in the Lorentz gas.

Figure 1(a) demonstrates that the Machta–Zwanzig approximation Eq. (2) is only valid in the limit of small gap sizes $w \rightarrow 0$, where according to Eq. (2) the diffusion coefficient should go linearly in w , as has been pointed out already in ref. 7, and as has been proven rigorously in ref. 14. For larger values of w and up to approximately $w < 0.1$ Eq. (2) then overestimates the exact diffusion coefficient, whereas for $w > 0.1$ it clearly underestimates diffusion. In the following section, we will discuss the physical reason for these deviations of the Machta–Zwanzig approximation from the numerically exact values over the entire density regime, and how it can be corrected.⁴ Note that for $w \rightarrow w_\infty$ there is no evidence for any singularity in the diffusion coefficient reminiscent of critical behavior. In ref. 9, this feature has been understood based on the analysis of velocity autocorrelation functions.

To learn more about the detailed dependence of D on the gap size w , we performed further computer simulations in the region of large w . The results are presented in Fig. 1b. Here, the respective residuals of D have been plotted, that is, the deviations of the diffusion coefficients from a linear fit in w over the whole region as shown in the figure. At each value of w ten independent runs of more than 2×10^9 collisions each have been carried out yielding the diffusion coefficients depicted by the dots. The squares correspond to the averages over the ten runs, where the size of the squares indicates the size of the numerical error. The numerical results clearly demonstrate that, on a fine scale, the diffusion coefficient is a very non-trivial function of w as a parameter. We will come back to these irregularities and discuss their possible microscopic origin in Section 4.

⁴ It has been pointed out to us that, along the lines suggested in ref. 14, analytical corrections to the Machta–Zwanzig diffusion coefficient have been obtained by other people.⁽¹⁵⁾

3. CORRECTION OF THE MACHTA-ZWANZIG APPROXIMATION

3.1. Collisionless Flights across a Trap

As Machta and Zwanzig remarked themselves, for larger w there is a non-vanishing probability p_{cf} for the particle to move collision-free across a trap. In Fig. 2a, we have calculated this probability from computer simulations as well as in a simple analytical approximation, which is based on straightforward applying the Machta-Zwanzig phase space argument described in Section 2.⁽¹⁶⁾ Note that collisionless flights occur only for $w > 2(\sin(\pi/3) + 1)/\sqrt{3} - 2 \simeq 0.1547$. If we rely on the Machta-Zwanzig picture of diffusion as a hopping process with frequency τ^{-1} over distances l , we can correct Eq. (2) in the following way: If a particle moves collision-free across a trap, it travels within the time τ over a *larger* distance than assumed in Eq. (2). For this larger distance we take the distance between a center of a trap and the center of its next nearest neighbor which is $l_2 = \sqrt{3}l$. These processes would thus yield a larger diffusion coefficient $D_{l_2} = l_2^2/(4\tau) = 3D_{MZ}$, where D_{MZ} is the diffusion coefficient of Eq. (2). We now define the corrected Machta-Zwanzig diffusion coefficient D_{cf} by weighting the contribution of collisionless flights via the probability p_{cf} of Fig. 2a,

$$\begin{aligned} D_{cf} &= [1 - p_{cf}] D_{MZ} + p_{cf} 3D_{MZ} \\ &= [1 + 2p_{cf}] D_{MZ} \end{aligned} \quad (4)$$

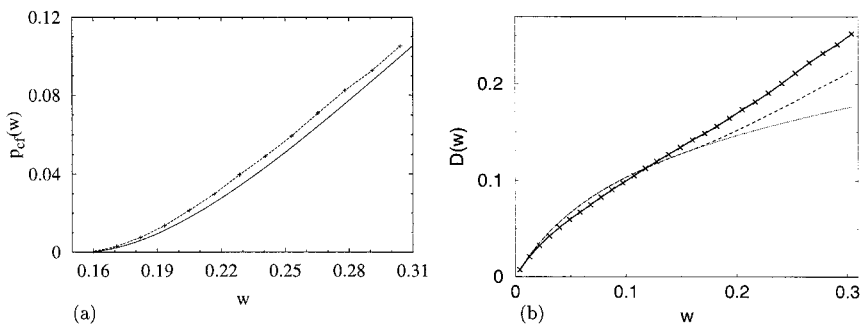


Fig. 2. Correction of the Machta-Zwanzig approximation by collisionless flights: (a) probability p_{cf} of collisionless flights across a trap, numerical results (dotted line with crosses), and a straightforward analytical approximation (bold line) (b) diffusion coefficient D , numerical results (bold line with crosses) in comparison to the Machta-Zwanzig approximation Eq. (2) (dotted line), and compared to the correction of Eq. (4) by including p_{cf} (dashed line).

The result is plotted in Fig. 2b. For $w > 0.1547$, the revised formula Eq. (4) improves the original Machta–Zwanzig approximation considerably, however, it is still much smaller than the numerically exact results.

3.2. Probability of Backscattering

A second significant contribution to the correction of Eq. (2) is determined by the backscattering probability p_{bs} , which is the probability of the moving particle to leave the trap through the same gap where it entered. We computed p_{bs} numerically by repeatedly injecting the particle through a specific gap and observing through which gap it left the trap. The particles are initially situated at one entrance of a trap, and they are uniformly distributed in the respective phase space of this entrance, which consists of the position of a particle on the entrance line, $-1 \geq 2x/w \geq 1$, and of the sine of the angle between the velocity direction and an axis perpendicular to the entrance, $-1 \geq \sin \alpha \geq 1$. Alternatively, p_{bs} can be determined from a single long trajectory. The backscattering probability obtained from our simulations is shown in Fig. 3a.

The Markovian approximation of Machta and Zwanzig Eq. (2) corresponds to a backscattering probability of $1/3$. However, for small values of w the numerically computed probability is significantly larger than $1/3$, whereas for larger w it is considerably smaller. The reduced probability for backward scattering at larger w is in part due to collisionless flights across the trap. It is remarkable that the backscattering probability is different from $1/3$ even for gap sizes where the average number of collisions between inter-trap hops is large: At $w = 0.055$, where the backscattering probability reaches its maximum value of $p_{\text{bs}} = 0.38$, more than 17 collisions occur before the particle hops to the next trap, and for $w = 0.02$, where p_{bs} is still close to 0.36, the number of collisions is greater than 50. These collision rates are indeed in good agreement with the Machta–Zwanzig approach yielding 19 for $w = 0.055$ and 52 for $w = 0.02$. However, based on a lower bound of about three collisions per trap residence time for all w below the infinite horizon Machta and Zwanzig concluded that the diffusion coefficient approximation Eq. (2) should be accurate for all these w . This appeared to be confirmed in respective computer simulations.⁽⁷⁾ Figure 3a demonstrates that even 50 collisions are not sufficient to obtain randomization. This accounts for the deviations between our numerical results and the Machta–Zwanzig approximation visible in Fig. 1. Based on the work in ref. 2, related conclusions on the validity of the Machta–Zwanzig approximation have been drawn in ref. 14.

Due to such memory effects, the detailed functional form of p_{bs} appears to be quite intricate as well: Below the maximum at $w = 0.055$

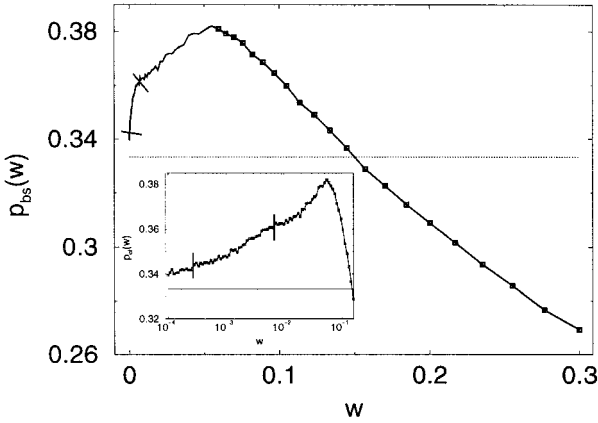
there are at least three regions where p_{bs} decreases approximately linearly in w with different values of the slope. As the numerical results indicate, the function must eventually drop extremely sharply to $p_{\text{bs}}(0) = 1/3$. Details of these regions are shown in the magnification included in the figure. The vertical lines separating different regions correspond to the respective lines separating regions of different slope in the main figure. A very close look reveals that the fine structure of all these different regions appears to be quite similar, however, we note that this structure is essentially within the range of our numerical errors.

We furthermore remark that a plot of the initial conditions in the $(x, \sin \alpha)$ -plane leading to backscattering for a given gap size w yields a fractal set of these coordinates.⁽¹⁶⁾ The modifications of such a fractal structure by varying w must be related to the changes seen in the function p_{bs} of Fig. 3a. This may explain why, even on the coarse scale of Fig. 3a, p_{bs} is not a simple function of w . Moreover, the detailed changes of such a fractal set may be reflected in the respective detailed changes of p_{bs} on the fine scale, as illustrated in the magnification.

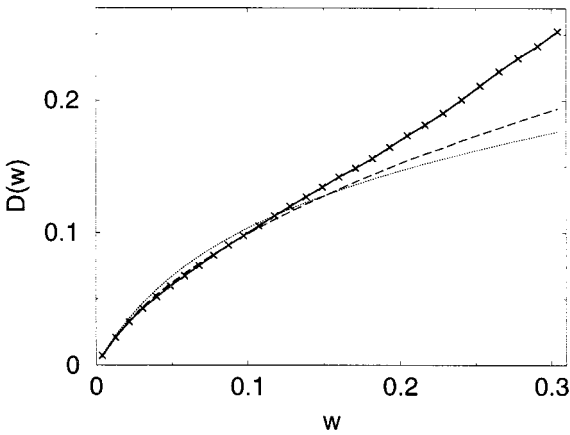
Having the probability of backscattering p_{bs} we can now perform a second correction of the original Machta–Zwanzig diffusion coefficient Eq. (2). For this purpose, we again assume that diffusion can be treated as a hopping process with a frequency τ^{-1} over distances l . We now inquire to which traps the particle can move by performing two jumps within a total time interval of 2τ . There are only two possibilities: either the particle suffers backscattering, that is, it goes back to its original trap and does not contribute to any actual displacement within 2τ , or it moves over a distance l_2 to the left or to the right of its original trap. Thus, the corresponding diffusion coefficient reads

$$D_{\text{bs}} = \frac{[1 - p_{\text{bs}}] l_2^2}{8\tau} = [1 - p_{\text{bs}}] 3/2 D_{\text{MZ}} \quad (5)$$

D_{bs} is plotted in comparison to our Einstein formula results, and to the original Machta–Zwanzig Eq. (2), in Fig. 3b. For smaller w , the corrected diffusion coefficient approximates the numerically exact values quite well. Thus, we conclude that the existence of backscattering is basically responsible for the Machta–Zwanzig argument overestimating diffusion for small values of w . For larger w Eq. (5) again improves the original Machta–Zwanzig approximation, like the previous approximation Eq. (4), however, like this it yields much smaller results than the correct values.



(a)



(b)

Fig. 3. Correction of the Machta-Zwanzig approximation by backscattering: (a) backscattering probability p_{bs} as a function of the gap size w . For larger w the single data points are plotted by symbols and are connected with lines, for smaller w only the lines are shown. The dotted line corresponds to the value of $1/3$ of equal probability for *any* gap, as it is assumed in the Machta-Zwanzig approximation. The inset is a half-logarithmic blowup of the initial region for small w . The bars included in the figure refer approximately to the regions of different slope in the main figure. (b) numerical results for the diffusion coefficient (bold line with crosses) in comparison to the Machta-Zwanzig approximation Eq. (2) (dotted line), and the correction via p_{bs} Eq. (4) (dashed line).

3.3. Combining Collisionless Flights, Backscattering, and a Symbolic Dynamics

So far we have identified two microscopic scattering mechanisms leading to deviations from the Machta–Zwanzig approximation: collisionless flights across the trap and backscattering. One might think of combining these two processes to obtain a single expression for the diffusion coefficient. This can be performed by simply replacing the Machta–Zwanzig diffusion coefficient D_{MZ} in Eq. (5) by D_{cf} of Eq. (4) yielding

$$D_1 = 3/2[1 - p_{\text{bs}}][1 + 2p_{\text{cf}}] D_{\text{MZ}} \quad (6)$$

This function, denoted as a first order approximation, is shown in Fig. 4a in comparison to the numerically exact results and to the Machta–Zwanzig Eq. (2). For larger w , this combined approximation is much closer to the numerical results than the original Machta–Zwanzig formulation, however, there still remains a notable quantitative difference.

In the same figure, corrections of higher order based on the idea of Eq. (6) are depicted. In the following we just outline the basic concept. The higher-order corrections of the diffusion coefficient are obtained by numerically computing the probabilities of higher-order backscattering, and by building them into a respectively generalized version of Eq. (5). This generalized expression of the backscattering diffusion coefficient D_{bs} is then combined with the respective collisionless flight-diffusion coefficient D_{cf} of Eq. (4) in the same way as before. The probabilities of higher-order backscattering have been computed on the basis of a simple symbolic dynamics, as it can be defined in case of simple backscattering: We followed a long trajectory of a particle in the Lorentz gas. For each visited trap we labeled the entrances through which the particle entered with z , the exit to the left of this entrance with l , and the one to the right with r . Thus, a trajectory in the Lorentz gas can be mapped to a sequence of symbols z , l , and r . Note that the symbolic dynamics we are using here is different to the one applied in other work in that we are labeling the three gaps of a trap, whereas in previous work the single disks accessible after a collision have been chosen, which required an alphabet of 12 symbols.^(17, 3) $p(z) = p_{\text{bs}}$ is then the backscattering probability depicted in Fig. 3a, whereas, due to symmetry, $p(l) = p(r) = (1 - p(z))/2$ corresponds to forward scattering. Combining these probabilities with the correction by collisionless flights yields Eq. (6) as a first order approximation.

Higher-order correlations can be calculated by taking into account the probabilities of longer symbol sequences.⁽¹⁶⁾ For example, the second order approximation involves probabilities corresponding to nine symbol sequences each consisting of two symbols, $p(zz)$, $p(zl)$, $p(zr)$, $p(lz)$, $p(ll)$, $p(lr)$,

$p(rz)$, $p(rl)$, $p(rr)$. In complete analogy to Eq. (6), a respective second-order approximation of the diffusion coefficient is then computed on the basis of these numerical probabilities, and by associating to them the respective distances traveled within a time interval of 3τ . In the third-order approximation the probabilities correspond to sequences of three symbols, for example, three times backscattering within a time interval of 4τ corresponding to $p(zzz)$, etc., and lead to a respective third order diffusion coefficient. In general, the k th order approximation involves the probability of sequences of k symbols. Figure 4a shows that by including such correlations the respective higher-order approximations of the diffusion coefficient converge, and globally move closer to the numerical exact results.

We therefore studied the effect of increased (or decreased) backscattering with the help of a lattice gas computer simulation. In such a simulation the Lorentz gas is mapped to a honeycomb lattice where the sites of the lattice represent the traps. The moving particle hops from site to site with frequency τ^{-1} , which is identical to the exact hopping frequency used in Machta-Zwanzig theory. We first describe the first order approximation: At each step the particle hops back to the site where it came from with probability p_{bs} or to one of the other sites with probability $(1 - p_{bs})/2$. The backscattering probability p_{bs} used in the lattice gas simulations is the one numerically obtained from simulations in the Lorentz gas. Also in the lattice gas simulations we determine the diffusion coefficient from the Einstein

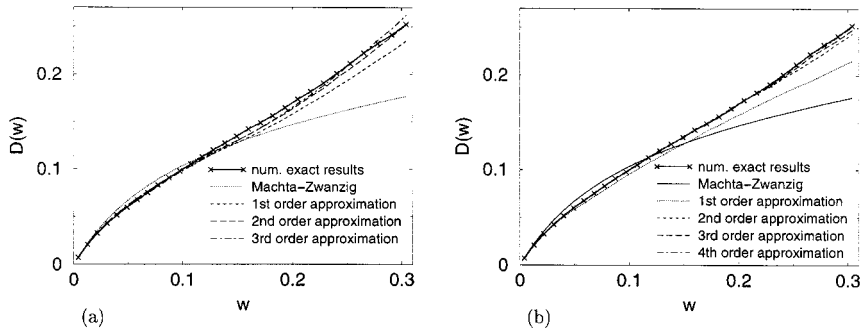


Fig. 4. Systematic approximations of the exact diffusion coefficient in higher order: (a) combined approach of employing the numerical probabilities of collisionless flights and of backscattering within a hierarchy of analytical expressions of the diffusion coefficient. The first-order approximation is given by Eq. (6) by using the probabilities p_{cf} and p_{bs} of Figs. 2a and 3a. The second and third order are computed as described in the text and include higher-order backscattering events. (b) diffusion coefficients obtained from lattice gas simulation by using probabilities of correlated hopping sequences as calculated in Lorentz gas simulations (see text).

formula Eq. (3). The results of these simulations are shown in Fig. 4b by a dotted line. Taking into account these first order correlations brings the diffusion coefficient closer to the correct value, but there still is a considerable deviation. This indicates the importance of higher order correlations, which can be obtained by correlating more than two hops between neighboring traps. To determine the diffusion coefficient for such multiple hopping events by lattice gas simulations we used the probabilities $p(lrz)$ calculated up to fourth order from long trajectories in the Lorentz gas. The results of these calculations are shown in Fig. 4b. As can be seen in the figure, the diffusion coefficient obtained from this scheme converges very quickly to the numerically exact results, in particular for small w . For larger w , the convergence is somewhat slower, however, the fourth order approximation can be hardly distinguished from the numerically exact results on the scale of Fig. 4b. For growing order the diffusion coefficient converges to the correct results exactly.

4. BOLTZMANN APPROXIMATION FOR THE DIFFUSION COEFFICIENT

In the previous section, we have discussed how the Machta–Zwanzig theory can be improved systematically. In case when the scatterers are distributed randomly in the plane without overlap, an alternative approach for understanding diffusion is provided by kinetic theory. In refs. 18 and 19, for low densities the diffusion coefficient has been computed from the Boltzmann equation to

$$D_{\text{Bo}}(n) = \frac{3}{8} l_c(n) v \quad (7)$$

where $l_c = 1/(2Rn)$ is the collision length of the moving particle. At higher number densities n the excluded volume of the scatterers becomes important, and the revised shorter collision length, which can also be obtained exactly from the Machta–Zwanzig argument,⁽⁷⁾ yields

$$D_{\text{Bo}}(n) = \frac{3}{16} \left(\frac{1}{n} - \pi \right) \quad (8)$$

where we have changed to units with $v = 1$ and $R = 1$. We now investigate whether this Boltzmann approximation is somewhat helpful to understand diffusion in the periodic case. Here, n is related to the gap size w by Eq. (1). Note that for $w = 0$, where the particle is trapped, the corresponding density is $n < \infty$ and the collision length is still finite. Thus, Eq. (8) implies a diffusion coefficient of $D_{\text{Bo}}(w = 0) > 0$, that is, this approximation inevitably

leads to an offset between $D_{\text{Bo}}(w=0)$ and the exact result of $D(w=0)=0$. Therefore, it can immediately be concluded that D_{Bo} is not correct at the highest densities.

In Fig. 5, Eq. (8) is shown in comparison to the numerically exact results and to the Machta–Zwanzig approximation Eq. (2). For this purpose, the diffusion coefficient has been plotted as a function of $1/n$, and an offset of $\Delta D = 0.025$ has been subtracted from Eq. (8). According to this equation, $D_{\text{Bo}}(n)$ should go linearly in $1/n$ with a slope of $3/16$, and the offset ΔD is the only fit parameter. Indeed, in Fig. 5 we observe that, on a sufficiently coarse scale and over a wide range of smaller densities, D matches surprisingly well to this functional form. In particular, the value of the slope appears to be almost exact. Only for larger densities D deviates from a linearity in $1/n$, where the Machta–Zwanzig approximation seems to describe the functional form at least qualitatively quite well.

We remark that for the random Lorentz gas the Boltzmann equation has been rigorously derived in ref. 20. However, for the periodic Lorentz gas it is not at all obvious that a Boltzmann approximation can be used, since here a diffusion coefficient exists only in the regime of very high densities. On the other hand, there stands our observation that Eq. (8) yields the correct linearity of the diffusion coefficient in $1/n$ for smaller densities with even the almost exact value of the slope. In trying to understand this usefulness of the Boltzmann equation for the periodic case we first remark that, except excluded volume effects, there are no further Enskog density corrections in a Lorentz gas as related to screening of particles, or to many-particle collisions.⁽²¹⁾ On the other hand, the existence of an offset at large densities may be related to the fact that the Boltzmann approximation represents only the first term in a series expansion in n , which is known as the density expansion of kinetic theory. For the random Lorentz gas, such a density expansion has been carried out explicitly in refs. 18 and 19. It has been found that there exist higher-order terms being logarithmic in the density corresponding to so-called ring collisions, which diminish the Boltzmann diffusion coefficient quantitatively. However, these logarithmic corrections are difficult to see in the functional form of the diffusion coefficient. It is not known to us whether a density expansion has been performed for the periodic Lorentz gas. But the offset we find might be associated to the existence of such higher-order corrections in the density, and the apparent Boltzmann-like linear behavior may be related to the observation that in similar systems higher-order corrections do not change the functional form of the diffusion coefficient in a drastic way.

Based on this, we may come back to the non-trivial fine structure of the diffusion coefficient depicted in Fig. 1b. We believe that these fluctuations can be understood as a signature of long-range correlations in

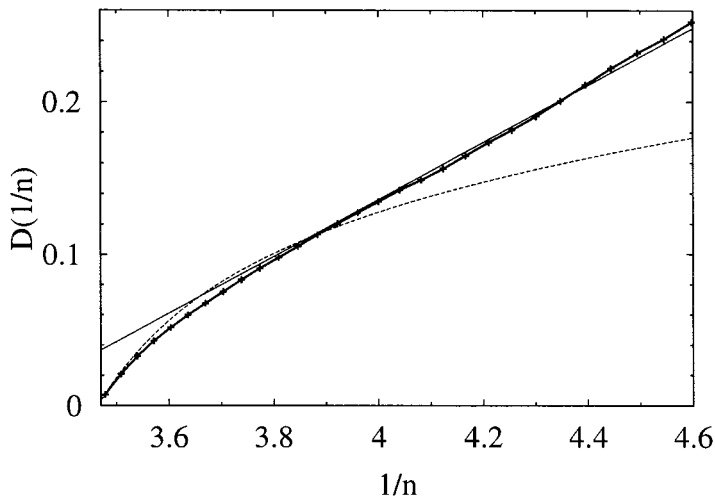


Fig. 5. Diffusion coefficient in the periodic Lorentz gas with respect to the inverse of the number density of the scatterers n . The Boltzmann approximation Eq. (8) is shown after subtracting an offset of $\Delta D = 0.025$ (thin bold line). It is compared to the numerically exact results (thick bold line with crosses), and to the Machta-Zwanzig approximation Eq. (2).

microscopic scattering events. A certain subclass of such processes are the ring collisions mentioned above leading to a series of logarithmic corrections, and divergences, in the density expansion of the diffusion coefficient. It might be conjectured that the existence of these divergences in the density expansion of kinetic theory is due to the problem of approximating a diffusion coefficient, which is in fact a function as complicated as the one shown in the inset of Fig. 1b, in form of a simple series expansion, as has already been remarked in ref. 3.

5. CONCLUSIONS

We have performed a detailed comparison between computer simulation results and the analytical approximation of Machta and Zwanzig for the density-dependent diffusion coefficient in the two-dimensional periodic Lorentz gas. In particular, we have discussed how their approximation can be corrected systematically over the full diffusive regime by including correlations in the hopping mechanism. Our corrections are conceptually very simple. Nevertheless, the physical mechanisms of free flights and backscattering, on which they rely, show up in other transport processes like diffusion-controlled chemical reaction,⁽²²⁾ and also in more complicated models (see the discussion in refs. 23 and 24). We furthermore

found that on a coarse scale there exists a dynamical crossover from a function linear in $1/n$ for lower densities, as qualitatively described by the Boltzmann diffusion coefficient, to a behavior at high densities, which can qualitatively be understood by the Machta–Zwanzig argument. We remark that these two approaches to understand the diffusion coefficient, that is, improving the Machta–Zwanzig argument, and applying a Boltzmann approximation, rather complement than contradict each other: The complicated microscopic scattering processes responsible for the limited validity of the Machta–Zwanzig approximation apparently just superpose in a way such that the resulting diffusion coefficient is approximately linearly in $1/n$ on a coarse scale. A similar relation between nonlinear corrections and a linear response has been discussed in refs. 25 and 26 with respect to the existence of Ohm's law in a periodic Lorentz gas with an external field. That this picture is in detail too simple is demonstrated by our finding of deviations from such a linear behavior on a very fine scale.

As has been discussed explicitly in ref. 3, fundamental physical and mathematical properties of the Lorentz gas are shared by simple one- and two-dimensional chaotic maps. In this respect, we remark that a dynamical crossover between different asymptotic laws for a parameter-dependent diffusion coefficient has already been found in periodic one-dimensional chaotic maps.^(23, 24) As in our discussion above, for these maps two different random walk models have been used to describe the corresponding asymptotic behavior: For small parameters, the respective random walk basically depends on the hopping probability of a particle leaving a cell, thus corresponding to the Machta–Zwanzig approximation, whereas for larger parameters the respective random walk scales with the distance a particle travels per time step, thus corresponding to the Boltzmann approximation, where diffusion is proportional to the collision length of the moving particle. We furthermore note that the diffusion coefficients in this kind of maps have been found to be fractal with respect to variation of the control parameter, and that specific correlated microscopic scattering events could be identified as being responsible for this fractal structure.^(27, 23, 28) The same phenomena have been encountered in related two-dimensional multi-baker maps.⁽²⁹⁾ Whether the diffusion coefficient of the periodic Lorentz gas is as well such a non-differentiable function of the density remains an open question.

ACKNOWLEDGMENTS

This article is dedicated to G. Nicolis on occasion of his 60th birthday. R.K. wants to express his gratitude to G. Nicolis for many scientific discussions and advice, and for his continuing support over the last two years.

In addition, R.K. is indebted to P. Gaspard for his interest in this problem, and for many valuable discussions. Helpful comments by L. A. Bunimovich, E. Barkai, H. van Beijeren, and S. Hess are gratefully acknowledged. R.K. thanks the European Commission for financial support within its Training and Mobility Program.

REFERENCES

1. H. A. Lorentz, *Proc. Roy. Acad. Amst.* **7**:438, 585, 684 (1905).
2. L. A. Bunimovich and Ya. G. Sinai, *Commun. Math. Phys.* **78**:247, 479 (1980).
3. P. Gaspard, *Chaos, Scattering, and Statistical Mechanics* (Cambridge University Press, Cambridge, 1998).
4. *Microscopic Simulations of Complex Hydrodynamic Phenomena*, Vol. 292 of *NATO ASI Series B: Physics*, M. Mareschal and B. L. Holian, eds. (Plenum Press, New York, 1992).
5. *Chaos and Irreversibility*, Vol. 8 of *Chaos*, T. Tél, P. Gaspard, and G. Nicolis, eds. (American Institute of Physics, College Park, 1998).
6. J. R. Dorfman, *An Introduction to Chaos in Nonequilibrium Statistical Mechanics* (Cambridge University Press, Cambridge, 1999).
7. J. Machta and R. Zwanzig, *Phys. Rev. Lett.* **50**:1959 (1983).
8. A. Baranyai, D. J. Evans, and E. G. D. Cohen, *J. Stat. Phys.* **70**:1085 (1993).
9. H. Matsuoka and R. Martin, *J. Stat. Phys.* **88**:81 (1997).
10. P. Cvitanovic, P. Gaspard, and T. Schreiber, *Chaos* **2**:85 (1992).
11. G. Morriss and L. Rondoni, *J. Stat. Phys.* **75**:553 (1994).
12. P. Gaspard and F. Baras, pp. 301–322 of ref. 4.
13. P. Gaspard and F. Baras, *Phys. Rev. E* **51**:5332 (1995).
14. L. A. Bunimovich, *Progr. Phys.* **10**:127 (1985); in *Statistical Physics and Dynamical Systems* (Birkhäuser, Boston, 1985); *Soviet J. of Theor. and Exp. Physics* **89**:842 (1985).
15. L. A. Bunimovich (private communication).
16. R. Klages and C. Dellago, preprint chao-dyn/9910038 (1999).
17. P. Cvitanović, lecture notes, available on www.nbi.dk/ChaosBook/.
18. A. Weijland and J. van Leeuwen, *Physica* **38**:35 (1968).
19. C. Bruin, *Physica* **72**:261 (1974).
20. C. Boldrighini, L. A. Bunimovich, and Ya. G. Sinai, *J. Stat. Phys.* **32**:477 (1983).
21. S. Chapman and T. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, Cambridge, 1970).
22. I. Claus and P. Gaspard, *Microscopic Chaos and Reaction-Diffusion Processes in the Periodic Lorentz Gas*, to appear in *J. Stat. Phys.*
23. R. Klages, *Deterministic Diffusion in One-Dimensional Chaotic Dynamical Systems* (Wissenschaft & Technik-Verlag, Berlin, 1996).
24. R. Klages and J. R. Dorfman, *Phys. Rev. E* **55**:R1247 (1997).
25. E. G. D.Cohen and L. Rondoni, *Chaos* **8**:357 (1998).
26. N. Chernov, C. Eyink, J. L. Lebowitz, and Ya. G. Sinai, *Comm. Math. Phys.* **154**:569 (1993).
27. R. Klages and J. R. Dorfman, *Phys. Rev. Lett.* **74**:387 (1995).
28. R. Klages and J. R. Dorfman, *Phys. Rev. E* **59**:5361 (1999).
29. P. Gaspard and R. Klages, *Chaos* **8**:409 (1998).