Normal diffusion in crystal structures and higher-dimensional billiard models with gaps

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We show, both heuristically and numerically, that three-dimensional periodic Lorentz gases—clouds of particles scattering off crystalline arrays of hard spheres—often exhibit normal diffusion, even when there are *gaps* through which particles can travel without ever colliding, i.e., when the system has an infinite horizon. This is the case provided that these gaps are not "too big", as measured by their dimension. The results are illustrated with simulations of a simple three-dimensional model having different types of diffusive regime, and are then extended to higher-dimensional billiard models, which include hard-sphere fluids.

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The Lorentz gas is a classical model of transport processes, in which a cloud of non-interacting point particles (modelling electrons) undergo free motion between elastic collisions with fixed hard spheres (atoms) [1]. It has been much studied as a model system for which one can carry out in detail the programme of statistical physics: to relate the known microscopic dynamics to the macroscopic behavior of the system, which in this case is diffusive [2, 3].

When the scatterers are arranged in a periodic crystal structure, the dynamics of this *billiard model* can be reduced to a single unit cell [3]. The curved shape of the scatterers implies that nearby trajectories separate exponentially fast, so that the system is hyperbolic (chaotic) and ergodic [4].

In two dimensions, it has been shown that the cloud of particles in the periodic Lorentz gas undergoes normal diffusion, provided that the geometrical *finite horizon* condition is satisfied: particles cannot travel arbitrarily far without colliding with a scatterer [4, 5]. By *normal diffusion*, we mean that the distribution of particle positions behaves like solutions of the diffusion equation; in particular, the mean-squared displacement (variance) grows asymptotically linearly in time: $\langle r(t)^2 \rangle \sim 2dDt$ when $t \to \infty$, where $r(t)$ is the displacement of a particle at time t from its initial position, d is the spatial dimension, $\langle \cdot \rangle$ denotes a mean over initial conditions, and the diffusion coefficient D gives the asymptotic growth rate.

When the horizon is infinite, however, particles can undergo arbitrarily long free flights along *corridors* in the structure. It was long argued [6–8] and has recently been proved [9], that there is then weak superdiffusive behavior, with $\langle r(t)^2 \rangle \sim t \log t$, so that the diffusion coefficient no longer exists.

For *higher-dimensional* periodic Lorentz gases, rigorous results on ergodic properties [10] and diffusive properties [11] have been obtained; recent progress in their analysis has been made [12, 13], including in the limit of small scatterers [14]. In particular, higher-dimensional Lorentz gases are believed to exhibit normal diffusion when the horizon is finite [11].

Nonetheless, the study of billiard models in higher dimensions, especially three dimensions, has received surprisingly little attention from the physics community, despite their interest as simple models of transport in three-dimensional crystals. This can be attributed to increased simulation times and the difficulty of visualisation in higher dimensions, but also to an apparent general belief that the diffusive properties of higher-dimensional systems should be analogous to those in the 2D case. Hypercubic Lorentz gases (with infinite horizon) in $d \leq 7$ dimensions were studied in [15], but no strong conclusions about diffusive properties could be drawn.

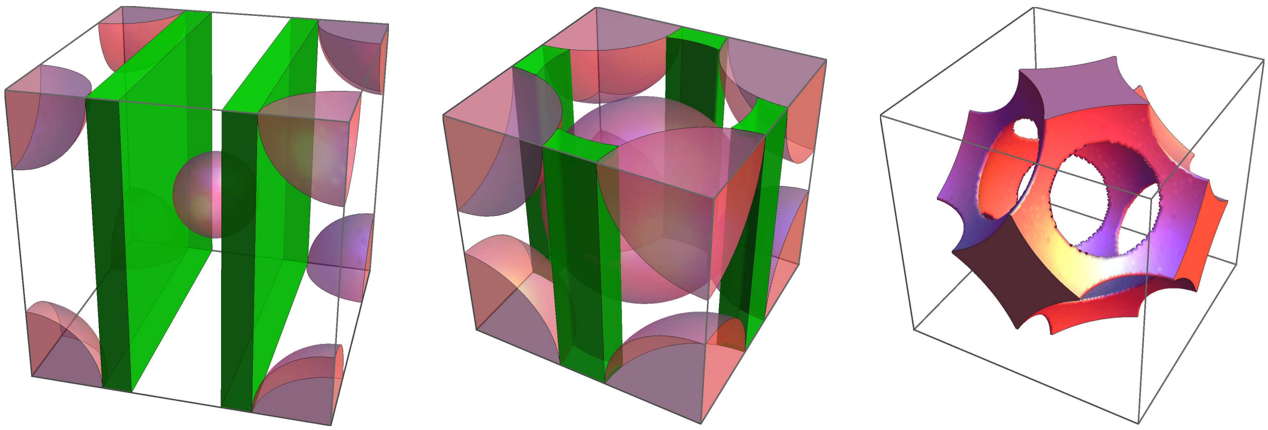
In particular, it was believed that a finite horizon was necessary for a system to show normal diffusion, with weak superdiffusion occurring for an infinite horizon [11, 16]. While periodic Lorentz gases with finite horizon are known to *exist* in any dimension [17], constructing such a model turns out to be a difficult task, even in three dimensions. Furthermore, crystals of spheres arranged in any Bravais lattice (and in many other crystal structures) have been shown to always have small gaps which prevent a finite horizon [17–19].

In this Letter, we show, using heuristic arguments and careful numerical simulations, that in fact periodic Lorentz gases in three and higher dimensions with infinite horizon—that is, with *gaps*, or holes, in the structure—can exhibit *normal* diffusion. The key observation is that the gaps in configuration space, which are higher-dimensional analogs of the corridors in 2D, can be of different dimensions. Structures with gaps of the highest possible dimension exhibit weak superdiffusion, as in the 2D infinite horizon case, whereas lower-dimensional gaps give normal diffusion. Nonetheless, higher moments of the displacement distribution are affected by the small proportion of arbitarily long trajectories in the structure.

To test the analytical arguments, we perform careful numerical simulations of a 3D periodic Lorentz gas model with spheres of two radii, which can be varied to obtain different types of diffusive regime. In particular, a a finite-horizon regime may be obtained by allowing the spheres to overlap; otherwise, gaps of different dimensions can be found. Here, results will be given for representative cases in each regime; a detailed analysis of the model will be given elsewhere.

Finally, we extend the analytical arguments to higher-dimensional billiards, which includes the class of hard-sphere fluids [20], thus providing an approach to understanding the diffusive behavior of such systems in terms of the geometry of their configuration space.

Model and gaps in configuration space:- We begin by introducing a simple two-parameter 3D periodic Lorentz gas model, with which the different types of diffusive regime can



(a) (b) (c)

FIG. 1: (Color online) Spherical scatterers (light color; purple online) and gaps (dark color; green online) in the 3D periodic Lorentz model discussed in the text: (a) vertical planar gaps for $a = 0.25$ and $b = 0.15$; and (b) vertical cylindrical gaps for $a = 0.4$ and $b = 0.4$ (a body-centred cubic structure). The gaps are shown in a single unit cell, but in fact extend throughout the whole of space. (c) When $a = 0.55$ and $b = 0.4$, the scatterers overlap, leaving an infinite, connected available space for the particles, which is depicted; for clarity, the spheres are omitted. In this case, the horizon is finite—there are no gaps in the structure.

be explored. The model consists of a cubic lattice of spheres of radius a, with an additional spherical scatterer of radius b at the centre of each cubic unit cell, themselves forming another (interpenetrating) cubic lattice. The side length of the cubic unit cell is taken equal to 1. By varying the radii of the spheres, a range of models with different properties can be obtained; a "phase diagram" showing the possibilities and a detailed study of its properties will be presented elsewhere. This is a 3D version of the 2D model studied in refs. [21, 22].

When $b = 0$, we obtain a simple cubic lattice. In this case, we can insert planes parallel to the lattice directions which do not intersect any obstacles—we call these planar *gaps*. This remains the case for small enough b, as shown in Fig. 1(a). For $b \geq 1 - 2a$, however, *all of the planar gaps are blocked*. There are still gaps of infinite extent in the structure, but they are now *cylindrical gaps*, as shown in Fig. 1(b). These are infinitely long tubes which do not intersect any scatterer, given by the product of a line with an area; the latter is the projection of the gap along the axis of the cylinder.

By tuning a and b appropiately, we can also obtain a model with *finite horizon*. To do so, we allow the scatterers to overlap, since otherwise constructing an explicit model is difficult. Each adjacent pair of a-spheres overlap when $a > \frac{1}{2}$; choosing a large enough value of the radius b of the central sphere then allows us to block all gaps in the structure, thereby obtaining a finite-horizon model, as we will show elsewhere. Note that unlike in the 2D case, in 3D the free space in between the overlapping scatterers forms an infinite connected network. Physically, this overlapping model can correspond to a sphere of non-zero radius colliding with disjoint scatterers. Note that the known rigorous results on higher-dimensional Lorentz gases assume that the scatterers are disjoint, and thus do not directly apply to this model [12].

Distribution of free paths:- Several approaches to determining the diffusive properties of systems with infinite horizon involve calculating the tail of the free-path length distribution, that is, the proportion of trajectories, starting from random initial conditions in a unit cell, which have a free-path length T before colliding which is longer than a large time t [6, 23, 24]; we denote this by $\mathbb{P}(T > t)$.

Consider straight trajectories emanating in all directions $\mathbf{v}$ from a given initial condition $\mathbf{x}_0$ which lies inside a gap G. Since energy is conserved at collisions, all particles can be taken to have speed 1. The possible positions $\mathbf{x}_t$ of the trajectories at time t then lie on a sphere S_t of radius t and surface area $S(t) = 4\pi t^2$, centred on $\mathbf{x}_0$. The proportion $\mathbb{P}(T > t)$ of trajectories which have not collided during time t is given by the ratio $\mathbb{P}(T > t) := A(t)/S(t)$, where $A(t)$ is the area of the intersection $I_t := G \cap S_t$ of the gap G with the sphere S_t.

If G is a planar gap, then the intersection I_t is approximately the product of a circle of radius t with an interval (which is asymptotically flat as $t \to \infty$) with the same width w as the gap. Thus $A(t) \simeq 2\pi wt$, giving the asymptotic behavior $\mathbb{P}(T > t) \sim C/t$ when $t \to \infty$, where C is a constant. This result was previously found for a simple cubic lattice [6, 11]; a detailed calculation is given in [24].

When G is a cylindrical gap, however, its intersection I_t with the sphere S_t is asymptotically the cross-sectional area A of the cylinder, giving the asymptotics $\mathbb{P}(T > t) \sim C/t^2$.

The tail $\mathbb{P}(T > t)$ of the free-path distribution is strongly related to the system's diffusive properties. Friedman & Martin [6] proposed that the asymptotic decay rate of the velocity autocorrelation function $C(t) := \langle \mathbf{v}(0) \cdot \mathbf{v}(t) \rangle$ is the same as that of $\mathbb{P}(T > t)$, since $C(t)$ is dominated by trajectories which do not collide up to time t. The finite-time diffusion coefficient $D(t) := \frac{d}{dt} \langle r(t)^2 \rangle$ is given by $D(t) = \frac{1}{d} \int_0^t C(s) ds$, so that $D(t)$ converges, to the diffusion coefficient D, only if the autocor-

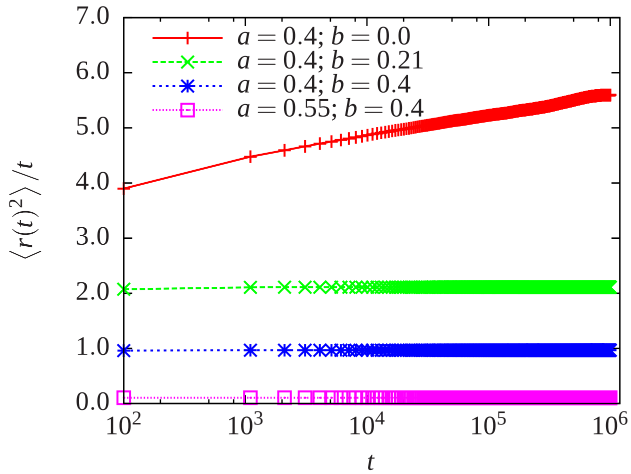


FIG. 2: (Color online) Linear–log plot of $\langle r(t)^2 \rangle / t$ vs. t in different diffusive regimes: finite horizon ($a = 0.55$; $b = 0.4$); cylindrical gaps in a body-centred cubic lattice ($a = b = 0.4$); single large cylindrical gap ($a = 0.4$; $b = 0.21$); simple cubic lattice ($a = 0.4$; $b = 0.0$); and body-centred cubic lattice ($a = b = 0.1$) with planar gaps. Means are taken over up to 4.1×10^7 initial conditions; error bars are of the order of the symbol size. Linear growth (weak superdiffusion) occurs only when there are planar gaps.

relation $C(t)$ decays faster than $1/t$ [3].

Thus we expect that a 3D periodic Lorentz gas should exhibit normal diffusion when $\mathbb{P}(T > t)$ decays faster than $1/t$, as is the case with cylindrical gaps (and when the horizon is finite), but weak superdiffusion when it decays like $1/t$. This is also in agreement with an equivalent condition on the moment of the free path distribution between collisions [8].

Numerical results:- To test the above hypotheses, we perform careful numerical simulations of the model in its different regimes, calculating the time evolution of the mean squared displacement $\langle r(t)^2 \rangle$ in each case. We use a stringent test to distinguish normal from weakly anomalous diffusion: $\langle r(t)^2 \rangle / t$ is plotted as a function of $\log t$ [21, 25]. Normal diffusion corresponds to an asymptotically flat graph, since the logarithmic correction is absent, and the diffusion coefficient is proportional to the asymptotic height of the graph. Weak superdiffusive $t \log t$ behavior for the mean-squared displacement, on the other hand, gives asymptotic linear growth [25].

Numerical results are shown in Fig. 2. We see that the arguments given in the previous section are confirmed: diffusion is normal, with $\langle r(t)^2 \rangle \sim t$, when the horizon is finite, and is weakly superdiffusive, with $\langle r(t)^2 \rangle \sim t \log t$, when there is a planar gap. Furthermore, the numerics clearly show that *diffusion is normal* also in the case that there are only cylindrical gaps. This is the case even when the cylindrical gaps are "large", for example when $a = 0.4$ and $b = 0.21$, when the gaps depicted in 1(b) merge to form a single cylindrical gap, still without any planar gaps in the structure. Thus we conclude that the heuristic arguments correctly predict the type of diffusion which occurs in these systems.

Holes in higher-dimensional billiards:- Fluids of hard spheres are isomorphic to higher-dimensional chaotic billiard models, although with cylindrical instead of spherical scatterers [20]. By extending the above arguments, we can hope to obtain information on correlation decay and diffusive properties for general higher-dimensional chaotic billiards by analysing the gaps in their configuration space.

To define these higher-dimensional gaps, we consider initial positions in a configuration space of dimension d, from which infinitely long non-colliding trajectories emanate along certain directions. We call a connected set of such initial positions for which these non-colliding trajectories point in the same direction(s) a *gap* in configuration space. Note that it is possible for a given set of initial conditions to have such trajectories pointing in different, unconnected directions—this is the case, for example, in Fig. 1(b), where there are also cylindrical gaps in a horizontal direction (not shown). In such cases, we consider each such set of different directions as a distinct gap. For a related discussion of higher-dimensional gaps in the context of sphere packings see ref. [18].

As shown above for the 3D case, the key geometrical property determining the diffusive behavior of a system is the dimension of its gaps. We define the *dimension* of a gap G to be the dimension g of the largest affine subspace which lies completely within the gap, i.e., which does not intersect any scatterer. In a system with a d-dimensional configuration space, there can be gaps with any dimension between 1 and $d-1$, or no gaps at all (finite horizon).

To calculate the tail $\mathbb{P}(T > t)$ of the free-path distribution due to such gaps, we take coordinates $\mathbf{x} := (x_1, \ldots, x_d)$ in the d-dimensional configuration space, with the initial position at the origin. The sphere S_t is then given by $\sum_{i=1}^{d} x_i^2 = t^2$. Consider a gap G, of dimension g. Inside the gap, there is a largest subspace, also of dimension g, i.e., it has g freely-varying coordinates. By rotating the coordinates, this subspace can be written as $x_1 = x_2 = \cdots = x_c = 0$, where $c := d - g$ is the *codimension* of the gap, giving the number of coordinates in the subspace which are fixed. The intersection $I_t = G \cap S_t$ of the gap with the sphere is thus given by $\sum_{i=c+1}^{d} v_i^2 = t^2$. This is a g-dimensional sphere, with surface area $K_g t^{g-1}$, where K_g is a dimension-dependent constant. The tail of the free-path distribution is given by the ratio of the area of intersection I_t to the area of the sphere S_t, giving the asymptotics

$$\mathbb{P}(T > t) \sim Z_c \frac{K_g t^{g-1}}{K_d t^{d-1}} = K t^{-(d-g)} = K t^{-c}, \qquad (1)$$

where Z_c is the c-dimensional cross-sectional area of the gap in the directions orthogonal to the affine subspace, and K is an overall constant.

We thus see that the decay is faster for gaps of smaller dimension (larger codimension), but it is always eventually dominated by the contribution of trajectories lying along the gaps. The dominant contribution to the tail of the free-path distribution, and hence to the diffusive properties, thus comes from the gap of largest dimension.

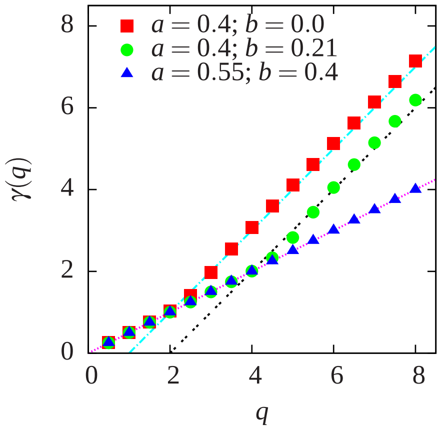


FIG. 3: Growth rate $\gamma(q)$ of higher moments $\langle r^q(t) \rangle$ as a function of q; geometries are as in Fig. 2. For $a = 0.4$ and $b = 0.21$, the means were calculated over 2.4×10^8 initial conditions, up to a time $t = 10000$, to capture the weak effect of the cylindrical gaps. The straight lines show the expected gaussian behavior ($q/2$) and behavior for large q with planar ($q-1$) and cylindrical ($q-2$) gaps.

We thus conjecture that d-dimensional chaotic, periodic billiard models can generically be expected to exhibit normal diffusion, at the level of the mean-squared displacement, provided that the largest-dimensional gap is of dimension less than $d-1$, that is, if its codimension is larger than 1.

Higher moments:- A more sensitive probe of diffusive properties is given by the growth rates $\gamma(q)$ of the qth moments of the displacement distribution, $\langle r^q(t) \rangle \sim t^{\gamma(q)}$, as a function of the real parameter q [23, 26]. If the decay rate of $\mathbb{P}(T > t) \sim t^{-c}$, then long trajectories dominate $\langle r^q(t) \rangle$ for large q, giving $\gamma(q) = q - c$, while the low moments show diffusive gaussian behavior, with $\gamma(q) = q/2$ [23]. A crossover between the two behaviors thus occurs at $q = 2c$. If the horizon is finite, then there are no long free flights, and gaussian behavior is expected for all q. The numerical calculation of higher moments is difficult, due to the weak effect of free flights [25]. Nonetheless, by taking means over a very large number of initial conditions, it is possible to see the effect of the different types of gaps for our 3D Lorentz gas model: as shown in Fig. 3, they are in agreement with the above argument. Thus, higher moments can distinguish the subtle effects of different types of gaps.

In conclusion, we have shown that the diffusive properties of periodic three-dimensional Lorentz gases, and by extension of higher-dimensional periodic billiard models, depend on the highest dimension of gap in the configuration space. By introducing a simple 3D model in which each type of diffusive regime occurs, we showed that if there is a finite horizon or cylindrical gaps, then the diffusion is normal, whereas planar gaps give weak superdiffusion. Nonetheless, higher moments distinguish between different types of gaps. The concept of infinite horizon is thus no longer sufficiently precise for higher-dimensional systems, and must be replaced by maximal gap dimension. In future work we will extend our numerical investigations to higher-dimensional models.

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