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Diffusion of oriented particles in porous media

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ABSTRACT

Diffusion of particles in porous media often shows subdiffusive behavior. Here, we analyze the dynamics of particles exhibiting an orientation. The features we focus on are geometrical restrictions and the dynamical consequences of the interactions between the local surrounding structure and the particle orientation. This interaction can lead to particles getting temporarily stuck in parts of the structure. Modeling this interaction by a particular random walk dynamics on fractal structures we find that the random walk dimension is not affected while the diffusion constant shows a variety of interesting and surprising features.

1. Introduction

The diffusion of particles driven by thermal noise is a phenomenon widely present in nature. The distance $r(t)$ particles can move from their starting point in a given time depends on several features of the system. If for instance the pathways of the particles are partly obstructed their progress will be slower. Such obstructions can be found in a variety of systems and are often of completely different origins. Typical examples are the slowed-down diffusion process in biological systems [1–3], or processes in chemistry [4], or physics [5,6]. Other examples include processes occurring in porous media like sandstone or in applications like filter systems [7].

Especially the diffusion in porous materials shows interesting features depending on the structure of the porosity. Already in the 1980s, it was found that some porous materials exhibit self-similarities over a certain range of length scales [8,6,9]. For such materials the mean squared displacement $\langle r^2(t)\rangle$ (MSD) shows anomalous diffusion behavior on certain time scales, i.e. one finds

$$\langle r^2(t)\rangle = Dt^{\gamma}, \tag{1}$$

where D is the diffusion coefficient and $0 < \gamma < 2$ is the (anomalous) diffusion exponent. In the case of diffusion in porous materials subdiffusion is observed with $\gamma < 1$. In order to analyze such behavior further, anomalous diffusion was investigated on fractal structures, where the self-similarity has no cut-off at larger length scales and thus at long time scales the behavior described by (1) persists. For such processes the exponents γ were then determined to high precision.

The new twist we introduce here is that the diffusing particles possess an orientation, i.e. they are no longer isotropic ‘point’ particles. Due to the resulting spatial extension (orientation) the interaction of particles with their surrounding cannot be neglected, which leads to a changed dynamics. Thus, a new move class has to be developed in order to capture these features.

Such oriented particles play an important role in a variety of systems, we just mention as examples electronic dipoles, liquid crystals [10–13], fibers, and ferromagnetic nanoparticles [14–17]. They are also of great technological importance for instance in liquid crystal displays.

Considering diffusion of such oriented particles in porous materials (or in biological tissue) immediately raises a number of questions: How do the oriented particles move within the porous structure? Does the transport of these particles follow the same laws as point particles do? If there are differences, are these only local effects or can they be observed also in the overall behavior? Do we obtain different exponents for the MSD or do we even get a new functional dependence for the MSD?

The answers to these questions are crucial for our understanding of diffusion processes in complex structures, as biological systems, and in their applications, as for instance the design of filter systems [18].

For the analysis of transport processes of oriented particles different continuum approaches have been used. These are theories

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like the Ericksen–Leslie theory [19,20] or 'mesoscopic continuum physics' [21–23]. These theories introduce either additional fields or variables to take into account the properties of the material. In order to embed the orientation of the particles for instance in the case of the Ericksen–Leslie theory a director field is introduced describing the mean orientation of the liquid crystals. The mesoscopic continuum physics approach enlarges the domain, i.e. it describes the properties on a higher-dimensional space – for example for liquid crystals on $\mathbb{R}^3 \times \mathbb{S}^2$.

While it is certainly a highly interesting attempt to include the self-similarity features of the material into the above approaches for instance by using fractal derivatives in the continuum equations [24–28], we here choose a different route based on simulating the diffusion processes. This approach has been widely used [5,6,29,9,30–33] to investigate diffusion of 'point' like particles within fractals and porous materials.

In the analysis presented here our focus is on capturing the interaction of the oriented particles with the walls of the porous material. We envision the oriented particles to be ellipsoids, where the long axis is a little longer than the size of the smallest pores and channels. Then, particles with an orientation perpendicular to a channel will not enter it, and if such a particle is nonetheless inside such a channel it will get stuck temporarily. The latter case can happen, if the orientation of the particle is changed while being inside a channel (for instance by a thermal fluctuation), or if the fluid in which the particles are dispersed pushes a particle into a channel. Note that external fields might allow to influence (at least the probability of) the orientation. We have implemented these features by designing a special move class for the random walks modeling the diffusion process.

In order to capture the self-similarity feature of the porous materials we performed random walks on special two-dimensional fractals, the Sierpinski carpets (SC) [34,35,29,9,33,32], a simple and often used model for porous structures. Below we will first introduce the details of our model, thereafter we describe the data acquisition followed by the data analysis, and finally the results are presented.

2. Model

SCs are constructed by recursively applying a generator pattern to itself. A generator pattern consists of $n \times n$ sub-squares of which m are labeled black and the rest white. The latter ones are removed and the generator is scaled up such that the sub-squares have the size of the original generator. Then every scaled-up black sub-squares is replaced by a copy of the generator. If this iteration procedure is repeated *ad infinitum* the limiting object is our fractal structure where each black square (site) is assigned a tuple (x, y) of integer coordinates. The fractal dimension d_f of a SC is given as $d_f = \log m / \log n$. It is a measure to characterize the property of self-similarity of the fractal.

The (typical) generator patterns used in this Letter and the corresponding d_f are given in Fig. 1. Here, all patterns have the size 7×7.

Note that sites with only one neighbor are called (narrow) pore in the following. Furthermore, we introduce the term (narrow) channel/passage for a group of sites, where each site has only two neighbors.

Generator A (Fig. 1(a)) is a symmetric pattern in x- and y-direction where the particles can move in the middle of the structure independently of their orientation. However, if they want to leave this area they have to pass through narrow passages where they are likely to get stuck. Note that in both directions there is an even number of sticking sites, where particles cannot move if they have the wrong orientation. Generator B (Fig. 1(b)) is an asymmetric generator, but close to the structure of generator A,

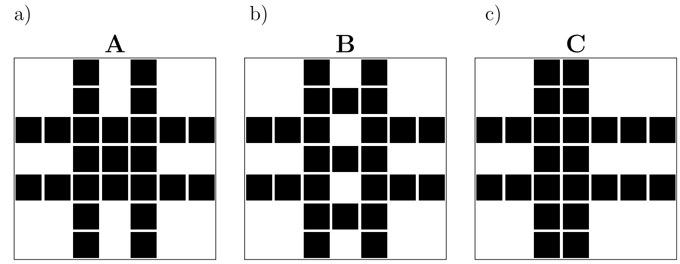

Fig. 1. These Sierpinski carpet generator patterns are used for the recent investigations. The patterns A & B have the fractal dimension $d_f = 1.694$, whereas pattern C has $d_f = 1.633$. The random walk dimension d_w are $d_w^A = 2.254$, $d_w^B = 2.289$, and $d_w^C = 2.265$.

where the middle part is split such that there are only narrow passages. Here, the movement of the particles should depend stronger on their orientation, as there are less possibilities to get stuck in x- than in y-direction. The last generator C (Fig. 1(c)) is an asymmetric pattern where the focus is laid on the different effects of narrow passages versus wide passages: there are two small passages in x-direction and one wide channel (twice the size of the x-direction) in y-direction.

The particles moving on the fractal structures described above are considered to be independent of each other. Apart from its coordinate tuple (x, y) each particle is in addition characterized by its orientation. In order to simplify the description the orientation can only take values x or y, i.e. particles are either oriented horizontal in the x-direction or vertical in the y-direction.

The move class of an oriented particle is then as follows: First, in each time step, the orientation of a particle is chosen randomly with probability p_y to be oriented in y-direction and with $p_x = 1 - p_y$ to be oriented in x-direction. This represents possible influences for instance by external fields.

Then for each particle position (x, y) it will be checked, whether there is a neighboring site in the orientation direction of the particle, e.g. for a particle oriented in x-direction either a black square at $(x - 1, y)$ or $(x + 1, y)$ has to exist. If there is no neighbor, no move can be done, which represents a particle that is temporarily stuck in a narrow pore/channel.

If a neighboring site exists, we choose one of the four possible neighbors with equal probability (blind ant algorithm [36]). If the chosen position (x_{new}, y_{new}) does not exist, again no move can be done. This reflects that the particle can only move within the pore space. If (x_{new}, y_{new}) exists, it will be checked whether (x_{new}, y_{new}) has at least one neighboring site in the orientation direction of the moving particle. If there is a neighbor, the move is allowed as the passage is wide enough so that the particle can move into it. Otherwise, the particle can only move to this site with $p_p \leqslant 1$. This is motivated by having a surrounding fluid that pushes particles into narrow pores/channels. Combing the rules of the move class above, one can determine for each site (x, y) the resulting probabilities of a particle to move or to stay in one time step. Four typical situations of the resulting probabilities of this move class are depicted exemplarily in Fig. 2. Note that the orientation is chosen in each time step according to p_y and that the sum over all movement probabilities plus the probability to stay equals 1. In Fig. 2(a) the particle can move in one of four directions or stay on its current position. Due to the orientation of the particle it can be pushed into the pore/channel (upwards) with probability $p_p/4$ or it stays with $(1 - p_p)/4$. In Fig. 2(b) the particle has four possible choices: either it moves up- or downwards with probability $1/4$, it is pushed into the pore/channel with $p_p/4$ or it stays at its current position with $1/2 - p_p/4$. The last two cases represent situations where a particle is within a pore/channel. In Fig. 2(c) the particle can leave the pore/channel with $1/4$