The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Normal and anomalous Knudsen diffusion in 2D and 3D channel pores

Stephan Zschiegner,[1,2] *Stefanie Russ*,[3] *Armin Bunde*,[2]
Marc-Olivier Coppens,[4] *Jörg Kärger*[1]

[1]Universität Leipzig, [2]Universität Giessen, [3]FU Berlin, [4]RPI Troy (NY) USA
Email corresponding author: stephan.zschiegner@physik.uni-giessen.de

1. Introduction

In general, diffusion of gas particles depends on the collisions between the gas molecules as well as on the collisions of the gas with the pore walls. Of particular interest for many real gases is the so-called Knudsen regime, where the interaction of the molecules with the pore walls plays the crucial role and the intramolecular collisions can be neglected. We implement pores with different roughness, by considering the first four iterations of a generalized fractal Koch curve. For these model pores we have performed detailed investigations of diffusion coefficients using a cube-based algorithm.

We know, the diffusion properties can be mapped onto Levy walks [1] to predict the diffusion properties. In 2D linear channel pores we observe anomalous diffusion [2, 3], which can also be induced by using different reflection laws in smooth or rough 3D pores. Normal diffusion is found in convoluted 2D pores and in all 3D pores when applying diffuse reflection.

2. Channel pores in 2D

We generate pores of different roughness as shown in [4], by employing the first iterations of a generalized fractal Koch curve. The diffusivity decreases with increasing surface roughness [5, 6]; in linear 2D pores, anomalously fast diffusion (superdiffusion) is observed [2,3]. Here we show that the logarithmic time dependence of the diffusion vanishes when the pore axis varies along the pore length (see Fig. 1). In a first example the elevation of the pore is shifted up or down according to a Random Walk. As a second example we use a pore that is alternating up and down by one pore diameter. In both examples the very long jumps that determine the asymptotical diffusion behaviour are suppressed, which leads to normal diffusion.

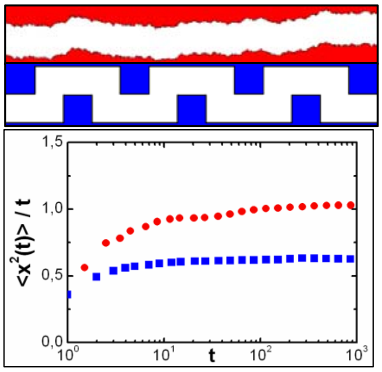

Fig. 1: Diffusion in non-linear 2D channel pores where no very long jumps are possible. For pores with an elevation that varies like a random walk (red) and alternating pores (blue) the scaled mean square displacement $<x^2(t)>/t$ is asymptotically constant.

3. Normal and anomalous diffusion in 3D

In 3D pores two angles determine the jump length. Jumps parallel to the pore axis generated by Lambert' law of reflection, where the probability of a particle to be reflected into the solid angle $d\Omega$ is $P(\vartheta,\varphi) \sim \cos\vartheta d\Omega$, become negligibly rare. This leads to normal diffusion (Fig. 2, middle).

Varying the applied law of particle reflection can change this diffusion behaviour in linear pores (Fig. 2). A homogeneous angular distribution ($P(\vartheta,\varphi) \sim d\Omega$) results in superdiffusion in smooth pores. This effect weakens with increasing roughness since many trajectories parallel to the pore axis are blocked (compare Fig. 1 in [3]). Pronounced vertically reflected particles ($P(\vartheta,\varphi) \sim \cos\vartheta d\vartheta$) diffuse anomalously fast only in rough pores, where long trajectories start at pore wall elements perpendicular to the pore axis. Since these wall elements are absent in smooth pores, normal diffusion is observed in this case.

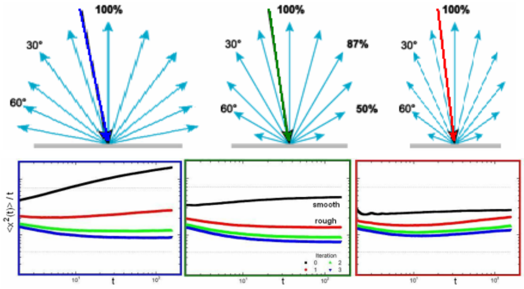

Fig. 2: Different reflection laws in 3D influence the diffusion behavior. A homogeneous angular distribution results in anomalous fast diffusion in smooth pores (left), while pronounced vertically reflected particles superdiffuse in rough pores (right). Diffuse reflection (Lambert's law, middle) yields normal diffusion in all pores.

The different reflection laws can be used to generate a geometry-induced bias in 2D and 3D. When the applied pore structure has an arrow-shaped geometry (Fig. 3), homogeneously reflected particles move in the arrow direction, while pronounced perpendicularly reflected particles move into the opposite direction. Diffuse reflection implies no bias at all.

Fig. 3: Different reflection laws in 3D influence the diffusion behavior. Homogeneous and pronounced perpendicularly reflected particles induce a bias in opposite directions. Diffuse reflection (Lambert's law) implies no bias.

4. Conclusion

We have simulated diffusion coefficients in two- and three-dimensional systems of channel pores. In 2D, anomalously fast diffusion arises for linear channels, while particles diffuse normally for random or alternating 2D pores where long jumps are suppressed. In 3D, particles can superdiffuse either in smooth or in rough pores when using different laws of reflection. For diffuse reflection following Lambert's law normal diffusion is observed. Furthermore the non-diffuse reflection laws in 2D and 3D lead to a bias in arrow-shaped pores.

References

[1] J. Klafter, M.F. Shlesinger, G. Zumofen, Phys. Today **49** (1996) 33.
[2] P. Levitz, Europhys. Lett. **39** (1997) 593.
[3] S. Russ, S. Zschiegner, A. Bunde, J. Kärger, Phys. Rev. E **72** (2005) 030101.
[4] S. Zschiegner, S. Russ, A. Bunde, J. Kärger, Diff. Fund. **4** (2007) 13.1.
[5] K. Malek, M.-O. Coppens, Phys. Rev. Lett. **87** (2001) 125505.
[6] S. Zschiegner, S.Russ, A. Bunde, J. Kärger, Europhys. Lett. **78** (2007) 20001.