INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS A: MATHEMATICAL AND GENERAL

J. Phys. A: Math. Gen. **38** (2005) 3743–3749 doi:10.1088/0305-4470/38/17/003

Modelling light transport in dry foams by a coarse-grained persistent random walk

M F Miri[1] **and H Stark**[2]

[1] Institute for Advanced Studies in Basic Sciences, Zanjan 45195-159, Iran
[2] Universität Konstanz, Fachbereich Physik, D-78457 Konstanz, Germany

Received 3 August 2004, in final form 2 March 2005
Published 13 April 2005
Online at stacks.iop.org/JPhysA/38/3743

Abstract

A two-dimensional persistence random walk model with two states is considered. In the immobile state, the particle chooses a new direction according to some probability function, while in the mobile state it persists on its direction and performs a ballistic motion. Starting from a master equation, we are able to calculate the diffusion constant for this model. The model was developed to explain Monte Carlo simulations of photon diffusion in two-dimensional dry foams. While it gives the leading term in the diffusion constant D, it fails to reproduce the details in D which depend on the amount of disorder in foams. This might show that diffusing photons probe local structural correlations in foams.

PACS numbers: 05.40.Fb, 82.70.Rr

1. Introduction

A multitude of different physical problems have been subsumed under the concept of a random walk. This is exemplified by such diverse fields as the transport in disordered media [1, 2], crystallographic statistics [3, 4] and the dynamics of stock prices [5]. Among the numerous models, the persistent random walk is possibly the simplest one to incorporate a form of momentum in addition to random motion [6]. In its basic realization on a one-dimensional lattice, a persistent random walker possesses constant probabilities for either taking a step in the same direction as the immediately preceding one or for reversing its motion. First introduced by Fürth as a model for diffusion in a number of biological problems [7], and shortly after by Taylor in the analysis of turbulent diffusion [8], the persistent random walk model is now generalized to study, e.g., polymers [9], diffusion in solids [10], dispersal of spores [11], cell movement [12] and general transport mechanisms [13–15].

The persistent random walk can be regarded as a particular case of the general concept of the multistate random walk [3]. This concept first arose in the original two-state model of Lennard-Jones [3, 16] where the random walker switches between an immobile and a mobile diffusive state. As such, the two-state model is applicable in the processes of electrophoresis and chromatography. Similar ideas are developed in the context of atomic diffusion in crystals [17].

Here we introduce a special persistent random walk in two dimensions which can be characterized by two different states. In the immobile state, the walker chooses a new direction in space during each time step; in the mobile state it persists on its present direction and performs a ballistic motion. The description has some similarities with a study on the diffusion of particles in a dense periodic Lorentz gas [18]. The random walk model investigated in this paper was developed within our work on diffusive light transport in cellular structures such as foams [19, 20] which is well established by experiments [21]. Our goal was to find a coarse-grained and analytically solvable model to explain features of our Monte Carlo simulations on the diffusive spreading of photons in disordered Voronoi foams [20]. While the model gives the leading factor in the diffusion constant D, it does not reproduce the details of the simulations. This helps us to obtain further insight into photon diffusion in cellular structures, as we will explain below.

The paper is organized as follows. In section 2 we first briefly review our work on the diffusive transport of photons in disordered Voronoi foams and then introduce the coarse-grained random walk model. The diffusion constant is a key parameter in diffusive transport that can be measured experimentally. Section 3 explains in detail how it is extracted from the current model. Finally, in section 4 we discuss our result and conclude.

2. Model

Two-dimensional cellular structures such as foams consist of air bubbles separated by liquid films. In relatively dry foams, the thickness of the films is much smaller than the cell diameter and three of them always meet in vertices [22]. Since the cells are much larger than the wavelength of light, one can employ geometrical optics and follow a light beam or photon as it is reflected by the liquid films with a probability r called the intensity reflectance. This naturally leads to a persistent random walk of the photons in space, since the new direction chosen by the photon in the $(n+1)$th step depends on the direction of the nth step. We started to implement this scenario in an ordered honeycomb structure as the simplest model for a two-dimensional foam [19]. As an idealization, we assumed that the intensity reflectance does not depend on the angle of incidence and that the films or edges have a uniform thickness so that the transmitted light ray does not change its direction. However, the honeycomb structure is highly idealistic. Therefore, we constructed disordered Voronoi foams to include the topological and geometrical disorder of real foams in our studies [20]. The main result of our Monte Carlo simulations is summarized in the empirical formula for the diffusion constant $D(r)$,

$$D(r) = \frac{1-r}{2r} \langle l \rangle c(1 - b_1 + b_2 r), \tag{1}$$

where $\langle l \rangle$ stands for the mean edge length in the Voronoi foam. The main behaviour is governed by the first factor on the right-hand side, which is also found in the honeycomb structure. However, there is a small but systematic deviation from it described by the last factor with $0 < b_1 < 0.1$ and $b_2 \approx 0.1$. Both constants show a clear dependence of the disorder in the Voronoi foam.

With the following coarse-grained random walk model, we attempt to justify the empirical equation (1). We note that a photon reflected by a liquid film does not leave the cell and therefore does not contribute to diffusion. Hence, in our coarse-grained approach we assume that the photon or random walker merely changes its direction of motion in space but keeps its current position which is equal to the position of the cell in the foam. On the other hand, at each transmission through a liquid film, the photon persists on its current direction and changes its location to a neighbouring cell.

To rationalize this model we introduce the probability $P_n(x, y|\theta)\,\mathrm{d}x\,\mathrm{d}y$ that the photon after its nth step arrives in the area $\mathrm{d}x\,\mathrm{d}y$ at position $\mathbf{x} = (x, y)$ where the direction of this step is given by the angle θ relative to the x-axis. Then the following master equation expresses the evolution of $P_n(x, y|\theta)$:

$$P_{n+1}(x, y|\theta) = r \int_{-\pi}^{\pi} P_n(x, y|\gamma) R(\theta - \gamma)\,\mathrm{d}\gamma + t P_n(x - \bar{L}\cos\theta, y - \bar{L}\sin\theta|\theta). \tag{2}$$

The first term on the right-hand side mimics the 'reflection' of the photon with a probability r. While according to the coarse-grained model it stays at the position (x, y) during the step $n+1$, it changes its direction by an angle $\theta - \gamma$ according to the probability function $R(\theta - \gamma)$. On the other hand, the second term describes the transmission with a probability $t = 1 - r$. The photon performs a ballistic motion with step length $\bar{L}$ travelling along the direction θ from position $(x - \bar{L}\cos\theta, y - \bar{L}\sin\theta)$ to (x, y). In the following, we explore consequences of this random walk model.

3. Diffusion constant

The diffusion constant follows from the evaluation of the second moment of $P_n(x, y|\theta)$ with respect to the spatial coordinates x and y. The probability distribution as an exact solution of the master equation (2) is hard to obtain. However, there is a more direct method for the evaluation of the first and second moments which employs the characteristic function associated with $P_n(x, y|\theta)$. It is the Fourier transform of the probability distribution with respect to $\mathbf{x} = (x, y)$ and the Fourier series expansion in the cyclic variable θ [13],

$$P_n(\boldsymbol{\omega}|m) = P_n(\omega, \alpha|m) = \int_{-\pi}^{\pi} \mathrm{e}^{\mathrm{i}m\theta} \int\int \mathrm{e}^{\mathrm{i}\boldsymbol{\omega}\cdot\mathbf{x}} P_n(x, y|\theta)\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}\theta, \tag{3}$$

where ω and α are the polar representation of the vector $\boldsymbol{\omega} = (\omega_x, \omega_y)$. Now, any moment of $P_n(x, y|\theta)$ can be generated from $P_n(\omega, \alpha|m)$ by appropriate derivatives:

$$\langle x^{k_1} y^{k_2}\rangle_n = (-\mathrm{i})^{k_1+k_2} \left.\frac{\partial^{k_1+k_2} P_n(\boldsymbol{\omega}|m=0)}{\partial\omega_x^{k_1}\,\partial\omega_y^{k_2}}\right|_{\boldsymbol{\omega}=\mathbf{0}}. \tag{4}$$

So if we introduce the Taylor expansion

$$P_n(\omega, \alpha|m) \approx Q_{0,n}(\alpha|m) + \mathrm{i}\omega\bar{L}\,Q_{1,n}(\alpha|m) - \frac{\omega^2\bar{L}^2}{2} Q_{2,n}(\alpha|m) + \cdots, \tag{5}$$

the first and second moments of $P_n(x, y|\theta)$ are

$$\begin{aligned} \langle x\rangle_n &= \bar{L}\,Q_{1,n}(0|0) \qquad & \langle y\rangle_n &= \bar{L}\,Q_{1,n}\left(\frac{\pi}{2}\middle|0\right), \\ \langle x^2\rangle_n &= \bar{L}^2 Q_{2,n}(0|0) \qquad & \langle y^2\rangle_n &= \bar{L}^2 Q_{2,n}\left(\frac{\pi}{2}\middle|0\right). \end{aligned} \tag{6}$$

Fourier transforming equation (2) one obtains

$$P_{n+1}(\omega, \alpha|m) = t \sum_{k=-\infty}^{\infty} \mathrm{i}^k\,\mathrm{e}^{-\mathrm{i}k\alpha} J_k(\omega\bar{L}) P_n(\omega, \alpha|k+m) + r R(m) P_n(\omega, \alpha|m), \tag{7}$$

where we have used the convolution theorem for both the spatial $[(x, y)]$ and the cyclic (θ) coordinates. We also introduced the Fourier representation of the kth-order Bessel function

$$J_k(z) = \frac{1}{2\pi\,\mathrm{i}^k}\int_{-\pi}^{\pi} \mathrm{e}^{\mathrm{i}z\cos\theta}\,\mathrm{e}^{-\mathrm{i}k\theta}\,\mathrm{d}\theta \tag{8}$$

and

$$R(m) = \int_{-\pi}^{\pi} \mathrm{e}^{\mathrm{i}m\theta} R(\theta)\,\mathrm{d}\theta. \tag{9}$$

Since we are only interested in the Taylor coefficients $Q_{1,n}(\alpha|m)$ and $Q_{2,n}(\alpha|m)$, we insert equation (5) into equation (7). Using the Taylor expansion of the relevant Bessel functions $J_k(z)$ $(|k| \leqslant 2)$ and $J_k(0) = \delta_{0,k}$ [23] and collecting all terms with the same power in ω, results in the following recursion relations for the $Q_{i,n}(\alpha|m)$:

$$\begin{aligned}
Q_{0,n+1}(\alpha|m) &= [t + rR(m)]Q_{0,n}(\alpha|m),\\
Q_{1,n+1}(\alpha|m) &= [t + rR(m)]Q_{1,n}(\alpha|m) + t\left[\frac{\mathrm{e}^{-\mathrm{i}\alpha}}{2}Q_{0,n}(\alpha|m+1) + \frac{\mathrm{e}^{\mathrm{i}\alpha}}{2}Q_{0,n}(\alpha|m-1)\right],\\
Q_{2,n+1}(\alpha|m) &= [t + rR(m)]Q_{2,n}(\alpha|m) + t[\mathrm{e}^{-\mathrm{i}\alpha}Q_{1,n}(\alpha|m+1) + \mathrm{e}^{\mathrm{i}\alpha}Q_{1,n}(\alpha|m-1)]\\
&\quad + t\left[\frac{1}{2}Q_{0,n}(\alpha|m) + \frac{\mathrm{e}^{-2\mathrm{i}\alpha}}{4}Q_{0,n}(\alpha|m+2) + \frac{\mathrm{e}^{2\mathrm{i}\alpha}}{4}Q_{0,n}(\alpha|m-2)\right].
\end{aligned} \tag{10}$$

We solve this set of coupled linear difference equations using the method of the z-transform [24]. The z-transform $F(z)$ of a function F_n of a discrete variable $n = 0, 1, 2, \ldots$ is defined by

$$F(z) = \sum_{n=0}^{\infty} F_n z^n. \tag{11}$$

One then derives the z-transform of F_{n+1} simply as $F(z)/z - F_{n=0}/z$. Note the similarities of this rule with the Laplace transform of the time derivative of a continuous function [23]. We are mainly interested in identifying diffusive behaviour for large n. Since the z-transform of $F_n = Dn$ is $Dz/(1-z)^2$, i.e., $z = 1$ is a pole of order two, one can extract the diffusion constant from $F(z)$ by evaluating

$$D = \lim\nolimits_{z\to 1} F(z)(1-z)^2. \tag{12}$$

The z-transform of equations (10) leads to a set of algebraic equations which immediately gives

$$\begin{aligned}
Q_0(z,\alpha|m) &= \frac{Q_{0,n=0}(\alpha|m)}{1 - z[t + rR(m)]},\\
Q_1(z,\alpha|m) &= \frac{Q_{1,n=0}(\alpha|m)}{1 - z[t + rR(m)]} + \frac{t\,\mathrm{e}^{-\mathrm{i}\alpha}}{2}\frac{zQ_{0,n=0}(\alpha|m+1)}{\{1 - z[t + rR(m)]\}\{1 - z[t + rR(m+1)]\}}\\
&\quad + \frac{t\,\mathrm{e}^{\mathrm{i}\alpha}}{2}\frac{zQ_{0,n=0}(\alpha|m-1)}{\{1 - z[t + rR(m)]\}\{1 - z[t + rR(m-1)]\}},\\
Q_2(z,\alpha|m) &= \frac{Q_{2,n=0}(\alpha|m)}{1 - z[t + rR(m)]} + t\,\mathrm{e}^{-\mathrm{i}\alpha}\frac{zQ_{1,n=0}(\alpha|m+1)}{\{1 - z[t + rR(m)]\}\{1 - z[t + rR(m+1)]\}}\\
&\quad + t\,\mathrm{e}^{\mathrm{i}\alpha}\frac{zQ_{1,n=0}(\alpha|m-1)}{\{1 - z[t + rR(m)]\}\{1 - z[t + rR(m-1)]\}} + \frac{t}{2}\frac{zQ_{0,n=0}(\alpha|m)}{\{1 - z[t + rR(m)]\}^2}\\
&\quad \times\left\{1 + \frac{zt}{1 - z[t + rR(m+1)]} + \frac{zt}{1 - z[t + rR(m-1)]}\right\}
\end{aligned}$$

$$
\begin{aligned}
&+\frac{t\,\mathrm{e}^{-2\mathrm{i}\alpha}}{4}\frac{zQ_{0,n=0}(\alpha|m+2)}{\{1-z[t+rR(m)]\}\{1-z[t+rR(m+2)]\}}\\
&\times\left\{1+\frac{2zt}{1-z[t+rR(m+1)]}\right\}\\
&+\frac{t\,\mathrm{e}^{2\mathrm{i}\alpha}}{4}\frac{zQ_{0,n=0}(\alpha|m-2)}{\{1-z[t+rR(m)]\}\{1-z[t+rR(m-2)]\}}\\
&\times\left\{1+\frac{2zt}{1-z[t+rR(m-1)]}\right\}. \qquad (13)
\end{aligned}
$$

These expressions contain the sum of several terms whose inverse z-transform are readily calculated.

If we assume an initial probability distribution $P_0(x, y|\theta)$ which is isotropic in both the spatial (x, y) and angular (θ) variables, we immediately obtain

$$\langle x\rangle_n = \langle y\rangle_n = 0 \qquad (14)$$

as it should be. Otherwise, for an arbitrary $P_0(x, y|\theta)$, the relevant function $Q_{1,n}(\alpha|0)$ (see equations (6)) contains terms which are either constant or behave as c^n with $c < 1$. They are associated with the randomization of the initial distribution of the random walkers but are not essential for large n.

The behaviour of the mean-square displacements is associated with $Q_{2,n}(\alpha|0)$ (see equations (6)). We checked that for large n or in the long-time limit it is purely diffusive, i.e.,

$$\langle x^2\rangle_n = 2D_x\tau \qquad \text{and} \qquad \langle y^2\rangle_n = 2D_y\tau, \qquad (15)$$

where we introduced the time $\tau = n\bar{L}/c$ which passes when the random walker makes n steps at a speed c. Using equation (12), we extract the diffusion constants from $Q_2(z, \alpha|0)$ and obtain

$$D_x = D_y = \frac{1-r}{2r}\bar{L}c(1-a+ar) \qquad (16)$$

with

$$a = \frac{R(m=1)R(m=-1)-1}{[1-R(m=1)][1-R(m=-1)]}. \qquad (17)$$

We have assumed a fixed step length in the above derivation. However, since cells in a disordered Voronoi foam are of various sizes (see, e.g., [20]), one might generalize our model by allowing a distribution of step lengths. Nevertheless, we do not expect this narrow distribution to change the functional form of $D_x(r)$ and $D_y(r)$ in equation (16).

4. Discussions and conclusions

The diffusion constant of equation (16) has the same structure as equation (1) which we deduced from our Monte Carlo simulations on Voronoi foams. At first sight, this looks like a promising result. Let us first check equation (16) for consistency. If the random walker during its immobile state does not change its direction at all, then $R(\theta) = \delta(\theta)$, where $\delta(\theta)$ stands for the delta function. One therefore finds $R(m = \pm1) = 1$ or $a \to -\infty$ which means that the diffusion constant diverges as it should do for a ballistic motion. On the other hand, $R(\theta) = \delta(\theta - \pi)$ gives $R(m = \pm1) = -1$ and $a = 0$ which results in the diffusion constant of the one-dimensional persistent random walk, as expected, with the step length $\bar{L}$ [3].

A distribution function which is symmetric with respect to $\theta = 0$ fulfils $R(m) = R(-m)$ and we can write the constant a as $a = -[1 + R(m = 1)]/[1 - R(m = 1)]$ where

$|R(m = 1)| \leqslant 1$ since the probability distribution $R(\theta)$ is normalized to one. This means that a is zero for $R(m = 1) = -1$ and then decreases continuously to $a = -1$ at $R(m = 1) = 0$ until it diverges at $R(m = 1) = 1$. So a is small only, as in our Voronoi-foam study (see equation (1)), when $R(\theta)$ is peaked around $\theta = \pi$. Then the diffusion constant is mainly determined by the first factor in equation (16). For our Voronoi foams, we find that reflection is roughly governed by the probability function $R(\theta) = \frac{1}{4}\left|\sin\frac{\theta}{2}\right|$ which leads to $R(m = 1) = -1/3$. This gives $a = -1/2$ and results in an extra factor of $(3 - r)/2$ in the diffusion constant. Moreover the sign of a is always negative just contrary to the sign of the constants b_1 and b_2 in equation (1). So, whereas our model contains the leading $(1-r)/(2r)$ term in the diffusion constant, it cannot explain the additional factor $1-b_1+b_2r$ in equation (1).

We tried to extend our model by introducing a distribution of step lengths or by giving each photon a helicity, as was done in [19] in the case of a honeycomb foam. Still, we were not successful in explaining the details of photon diffusion in Voronoi foams (as summarized in equation (1)) beyond the $(1 - r)/(2r)$ term in the diffusion constant. We therefore have to conclude that our coarse-grained random walk model does not describe all characteristic details of the diffusion of photons in Voronoi foams. This gives hints that the law of geometrical optics and the explicit structure of the Voronoi foam creates a special type of random walk for the photons which is hard to access by an effective model. For example, the fact that the change of the direction of the photon depends on the orientation of the reflecting edge seems to be important. Furthermore, due to local correlations of the cells in the Voronoi foam, successive reflections are correlated with regard to step lengths and reflection angles which is not contained in our model.

In conclusion, by studying a coarse-grained random-walk model we find hints that geometrically induced correlations between photon steps are important for the diffusive propagation of photons in a Voronoi foam. To include these correlations into an effective model is, however, non-trivial and we are currently working on it.

Acknowledgments

We would like to thank H Larralde for fruitful discussions at the early stages of this work. HS acknowledges financial support from the Deutsche Forschungsgemeinschaft under Grant No. Sta 352/5-1. MFM thanks the International Graduate College at the University of Konstanz for financial support.

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