Diffusive transport of light in a two-dimensional disordered packing of disks: Analytical approach to transport mean free path

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We study photon diffusion in a two-dimensional random packing of monodisperse disks as a simple model of granular media and wet foams. We assume that the intensity reflectance of disks is a constant r. We present an *analytic* expression for the transport mean free path l^* in terms of the velocity of light in the disks and host medium, radius R and packing fraction of the disks, and the intensity reflectance. For glass beads immersed in air or water, we estimate transport mean free paths about half the experimental ones. For air bubbles immersed in water, l^*/R is a *linear* function of $1/\varepsilon$, where ε is the liquid volume fraction of the model wet foam. This throws light on the empirical law of Vera *et al.* [Appl. Opt. **40**, 4210 (2001)] and promotes more realistic models.

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I. INTRODUCTION

There is a good reason to study wave propagation in turbid or random media: Multiply scattered waves can *probe* temporal changes in physical systems [1–3]. Thus, light transport through fog [4], milky liquids, nematic liquid crystals [5], granular media [6–9], foams [10–16], and human tissue [17]; propagation of elastic waves in the Earth's crust [18,19]; acoustic waves in the fluidized or sedimenting suspensions [20]; etc., have attracted much attention.

In a turbid medium, light undergoes many scattering events before leaving the sample and the transport of light energy is diffusive [3]. Therefore, the photon can be considered as a random walker. The transport mean free path l^*, over which the photon direction becomes randomized, depends on the structural details of the opaque medium. Experimental techniques like diffuse-transmission spectroscopy (DTS) [21] and diffusing-wave spectroscopy (DWS) [22] can be used to measure l^*. In DTS, the average fraction T of incident light transmitted through a slab of thickness L is measured. The transport mean free path is then deduced from $T \propto l^*/L$. Utilizing the temporal intensity fluctuations in the speckle field of the multiply scattered light, DWS determines l^* and the mean-squared displacement of the scattering sites due to time evolution, thermal motion, or flow.

A plethora of light-scattering experiments show that light transport reaches its diffusive limit in granular media [6–9] and foams [10–16], which means that photons perform a random walk. However, the mechanisms underlying this random walk are not elucidated. A wet foam is composed of spherical gas bubbles dispersed in liquid. A relatively dry foam consists of polyhedral cells separated by thin liquid films. Three of them meet in the so-called Plateau borders which then define tetrahedral vertices [23]. In their studies of foams with the liquid volume fraction ε in the range $0.008 < \varepsilon < 0.3$, Vera, Saint-Jalmes, and Durian [11] observed the empirical law

$$l^* \approx 2R\left(\frac{0.14}{\varepsilon} + 1.5\right), \tag{1}$$

where R is the average bubble radius. Recent studies of scattering from Plateau borders [11,24], vertices [25], and films [26–31] or transport effects such as total internal reflection of photons inside the Plateau borders [12,32] have not yet clarified the empirical law of Vera *et al.* For granular media, systematic measurements of the transport mean free path l^* as a function of the refractive indices of grains and the host medium (air, water, etc.), grain size, and packing fraction have not been performed. Menon and Durian [6] determined $l^* \approx 15R$ for glass spheres of radius $R = 47.5$ μm dispersed in air. For glass beads dispersed in water, Leutz and Rička [8] found $l^* \approx 14R - 16R$ for $80\ \mu\text{m} \leqslant R \leqslant 200\ \mu\text{m}$. Their samples had a packing fraction $\phi \approx 0.64$. Crassous [9] performed numerical simulations to find l^* as a function of refractive indices of the grain and host medium, but only for packing fraction $\phi \approx 0.64$.

It is instructive to consider *simple* or even toy models of granular media and wet foams, which allow an *analytic* access to the transport mean free path l^*. Apparently, such models pave the way for a deeper understanding of fascinating DWS experiments. In this paper, we consider two-dimensional packing of monosize disks. The disks are much larger than the wavelength of light; thus, one can employ ray optics to follow a light beam or photon as it is reflected by the disks with a probability r called the intensity reflectance. We assume that the intensity reflectance is constant and the velocity of light inside and outside the disks is c/n_{in} and c/n_{out}, respectively. We show that the photon's random walk based on the above rules is a persistent random walk [26,33,34]. Writing a master equation to describe the photon transport, we find in Sec. III A the transport mean free path as

$$l^* = \frac{\pi R}{4} \frac{\left(\frac{3}{2r} - 1\right)\left[\frac{r}{1-r} + \left(\frac{1-\phi}{\phi}\right)^2\right]}{\left(\frac{\phi}{n_{in}} + \frac{1-\phi}{n_{out}}\right)\left(n_{in}\frac{r}{1-r} + n_{out}\frac{1-\phi}{\phi}\right)}, \tag{2}$$

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where ϕ and R denote the packing fraction and radius of disks, respectively. We further study our model by numerical simulation of the photon's random walk. We observe the overall agreement between our numerical and analytical estimates of the transport mean free path.

For glass beads immersed in air or water, we find transport mean free paths about half the experimental ones [6,8]. For air bubbles immersed in the water, we use Eq. (2) to derive l^* as a function of the liquid volume fraction $\varepsilon = 1 - \phi$. $r \approx 0.20$ is estimated as a weighted average of Fresnel's intensity reflectance. We find that in the range $0.08 < \varepsilon < 0.15$, our analytical result agrees well with the relation $l^* \approx R(0.11/\varepsilon + 2.37)$. In other words, we find that l^*/R is a *linear* function of $1/\varepsilon$. Using the hybrid lattice gas model for two-dimensional foams and Fresnel's intensity reflectance, Sun and Hutzler performed a numerical simulation of photon transport and found $l^* \approx R(0.26/\varepsilon + 4.90)$ [24]. Quite remarkably, our *analytic* estimate of the transport mean free path throws light on the empirical law of Vera *et al.* and the numerical simulation of Sun and Hutzler.

Our article is organized as follows. In Sec. II we introduce the two-dimensional packing of disks as a simple model for a granular medium or a wet foam. Photon transport in a random packing of disks using constant intensity reflectance is discussed in Sec. III. Discussions, conclusions, and an outlook are presented in Sec. IV.

II. MODEL

As a *simple* model for a two-dimensional disordered granular medium, wet foam, and bubbly liquid, we choose the random packing of circular disks. All nonoverlapping disks have the same radius R and cover a fraction ϕ of the plane. To address the photon transport in such a medium, we have made the following assumptions: (i) Disks or grains, are much larger than the wavelength of light; thus, one can employ ray optics to follow a light beam or photon as it is reflected by the disks with a probability r called the intensity reflectance. (ii) r is a constant, with no dependence on the incidence angle. (iii) Although disks of refractive index n_{in} are immersed in a medium of refractive index n_{out}, the incident and transmitted rays have the same direction. In other words, we assume that the angle of refraction equals the angle of incidence. (iv) The velocity of light inside and outside the disks is c/n_{in} and c/n_{out}, respectively.

Our first assumption is inspired by the experiments [6–9]. Our second and third assumptions do not agree with Fresnel's formulas and Snell's law, respectively. Consequently, our model does not consider the total internal reflection of rays. However, we deliberately adopt a step-by-step approach to photon transport in granular media and will consider more realistic models later.

As already mentioned, we model single-photon paths in a packing of disks as a random walk with rules motivated by ray optics; i.e., an incoming light beam is reflected from a disk surface with a probability r or it traverses the disk surface with a probability $t = 1 - r$. This naturally leads to a persistent random walk of the photons [26], where the walker remembers its direction from the previous step [33,34]. Persistent random walks are employed in biological problems [35], turbulent diffusion [36], polymers [37], Landauer diffusion coefficient for a one-dimensional solid [38], and general transport mechanisms [39,40]. More recent applications are reviewed in [41]. In the following section, we adopt the approach of [28,39] to study persistent random walk of the photons in a granular medium.

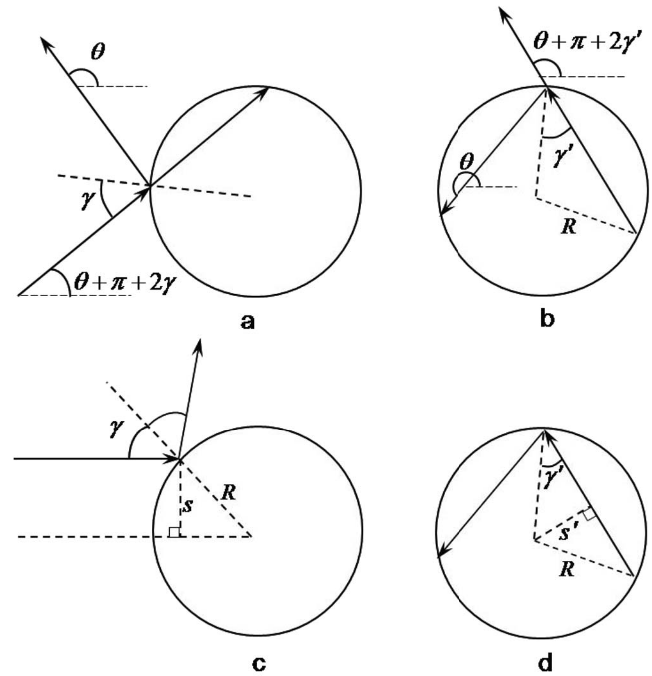

FIG. 1. (a) Path of a photon moving in the host medium and hitting a disk with an incidence angle γ. (b) Path of a photon moving in a disk and hitting its surface with an incidence angle γ'. The step length inside the disk is $2R\cos\gamma'$, where R is radius of the disk. (c) A photon impinging on a disk with an impact parameter s. Note $s = R\sin\gamma$. (d) The distance s' is related to the incidence angle γ' by $s' = R\sin\gamma'$.

III. PHOTON TRANSPORT IN A TWO-DIMENSIONAL PACKING OF DISKS

A. Analytical treatment

The photon random walk in a packing of disks consists of steps inside and outside the disks. We denote the average length of steps inside and outside the grains by $\bar{L}_{in}$ and $\bar{L}_{out}$, respectively. We characterize each step by an angle relative to the x axis. As Fig. 1(a) demonstrates, on hitting a disk with an incidence angle γ, a photon moving in the host medium along the direction $\theta + \pi + 2\gamma$ will be either reflected to the direction θ or enter the disk. The probability distribution of the random variable γ $(0 < \gamma < \pi/2)$ is $F(\gamma) = \cos\gamma$; see the Appendix. Similarly, a photon moving in a disk along the direction $\theta + \pi + 2\gamma'$ and hitting its surface with an angle γ' will be either reflected to the direction θ or enter the host medium; see Fig. 1(b). Quite remarkably, the probability distributions of the incidence angles γ and γ' are the same; see the Appendix. We can therefore conclude that the diffusions of the photons inside and outside the grains are not inherently different. As will be shown in the following, we write a master equation to describe the photon diffusion inside (outside) the grains, utilizing the step length $\bar{L}_{in}$ ($\bar{L}_{out}$) and velocity c/n_{in} (c/n_{out}), and extract the diffusion constant D_{in} (D_{out}). According to the two-state Lennard-Jones model [34], the diffusion constant of photons in the granular medium is

$$D_m = f_{in}D_{in} + f_{out}D_{out}, \tag{3}$$

where f_{in} ($f_{out}=1-f_{in}$) is the fraction of time that the photons spend inside (outside) the disks.

We introduce the probability $P_n(x,y|\theta)dxdy$ that the photon after its nth step of length $\bar{L}$ along the direction θ arrives in the area $dxdy$ at position $\mathbf{x}=(x,y)$. Then the following master equation expresses the evolution of $P_n(x,y|\theta)$:

$$P_{n+1}(x,y|\theta) = \frac{1}{2}r\int_{-\pi/2}^{\pi/2} P_n(x-\bar{L}\cos\theta, y-\bar{L}\sin\theta|\theta+\pi+2\gamma) \times F(\gamma)d\gamma + tP_n(x-\bar{L}\cos\theta, y-\bar{L}\sin\theta|\theta). \tag{4}$$

The first term on the right-hand side describes the reflection of the photon with a probability r. The photon which has arrived at position $(x-\bar{L}\cos\theta, y-\bar{L}\sin\theta)$ along the direction $\theta+\pi+2\gamma$ changes its direction by an angle $\pi+2\gamma$ according to the probability function $F(\gamma)$ [42]. The second term describes the transmission with a probability $t=1-r$. The photon performs a ballistic motion with step length $\bar{L}$ along direction θ from position $(x-\bar{L}\cos\theta, y-\bar{L}\sin\theta)$ to (x,y).

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 (4) is hard to obtain. However, there is a more direct method for the evaluation of the moments which employs the characteristic function $\mathbf{P}_n(\omega_x,\omega_y|m)$ associated with $P_n(x,y|\theta)$ [34]:

$$\langle x^{k_1}y^{k_2}\rangle_n \equiv \int\int\int x^{k_1}y^{k_2}P_n(x,y|\theta)dx\,dy\,d\theta = (-i)^{k_1+k_2}\frac{\partial^{k_1+k_2}\mathbf{P}_n(\vec{\omega}|m=0)}{\partial\omega_x^{k_1}\partial\omega_y^{k_2}}\bigg|_{\vec{\omega}=0}, \tag{5}$$

where k_1 and k_2 are either zero or positive integers,

$$\mathbf{P}_n(\vec{\omega}|m) \equiv \int_{-\pi}^{\pi} e^{im\theta}\int\int e^{i\vec{\omega}\cdot\mathbf{x}}P_n(x,y|\theta)dx\,dy\,d\theta, \tag{6}$$

and $\vec{\omega}=(\omega_x,\omega_y)$. We are interested in the first and second moments of $P_n(x,y|\theta)$; thus, we focus on the Taylor expansion

$$\mathbf{P}_n(\omega,\alpha|m) \approx Q_{0,n}(\alpha|m) + i\omega\bar{L}Q_{1,n}(\alpha|m) - \frac{\omega^2\bar{L}^2}{2}Q_{2,n}(\alpha|m) + \cdots, \tag{7}$$

where ω and α are the polar representation of the vector $\vec{\omega}=(\omega_x,\omega_y)$. From Eqs. (5) and (7) it follows that

$$\langle x\rangle_n = \bar{L}Q_{1,n}(0|0),$$

$$\langle y\rangle_n = \bar{L}Q_{1,n}\left(\frac{\pi}{2}|0\right),$$

$$\langle x^2\rangle_n = \bar{L}^2Q_{2,n}(0|0),$$

$$\langle y^2\rangle_n = \bar{L}^2Q_{2,n}\left(\frac{\pi}{2}|0\right). \tag{8}$$

Fourier transforming Eq. (4), we obtain

$$\mathbf{P}_{n+1}(\omega,\alpha|m) = \sum_{k=-\infty}^{\infty} i^k e^{-ik\alpha}J_k(\omega\bar{L})\mathbf{P}_n(\omega,\alpha|k+m) \times [r(-1)^{m+k}\mathbf{F}(2m+2k)+t], \tag{9}$$

where $\mathbf{F}(m)=\frac{1}{2}\int_{-\pi/2}^{\pi/2}e^{im\gamma}F(\gamma)d\gamma$ and

$$J_k(z) = \frac{1}{2\pi i^k}\int_{-\pi}^{\pi} e^{iz\cos\theta}e^{-ik\theta}d\theta \tag{10}$$

is the kth-order Bessel function. Since we are only interested in the Taylor coefficients $Q_{1,n}(\alpha|m)$ and $Q_{2,n}(\alpha|m)$, we insert Eq. (7) into Eq. (9). Using the Taylor expansion of the relevant Bessel functions $J_k(z)$ ($|k|\leq 2$) and $J_k(0)=\delta_{0,k}$ [43] and collecting all terms with the same power in ω results in the following recursion relations for the $Q_{i,n}(\alpha|m)$:

$$Q_{0,n+1}(\alpha|m) = [t+r(-1)^m\mathbf{F}(2m)]Q_{0,n}(\alpha|m),$$

$$\begin{aligned}Q_{1,n+1}(\alpha|m) &= [t+r(-1)^m\mathbf{F}(2m)]Q_{1,n}(\alpha|m) \\ &+ \frac{e^{-i\alpha}}{2}[t+r(-1)^{m+1}\mathbf{F}(2m+2)]Q_{0,n}(\alpha|m+1) \\ &+ \frac{e^{i\alpha}}{2}[t+r(-1)^{m-1}\mathbf{F}(2m-2)]Q_{0,n}(\alpha|m-1),\end{aligned}$$

$$\begin{aligned}Q_{2,n+1}(\alpha|m) &= [t+r(-1)^m\mathbf{F}(2m)]Q_{2,n}(\alpha|m) \\ &+ e^{-i\alpha}[t+r(-1)^{m+1}\mathbf{F}(2m+2)]Q_{1,n}(\alpha|m+1) \\ &+ e^{i\alpha}[t+r(-1)^{m-1}\mathbf{F}(2m-2)]Q_{1,n}(\alpha|m-1) \\ &+ \frac{1}{2}[t+r(-1)^m\mathbf{F}(2m)]Q_{0,n}(\alpha|m) \\ &+ \frac{e^{-2i\alpha}}{4}[t+r(-1)^{m+2}\mathbf{F}(2m+4)]Q_{0,n}(\alpha|m+2) \\ &+ \frac{e^{2i\alpha}}{4}[t+r(-1)^{m-2}\mathbf{F}(2m-4)]Q_{0,n}(\alpha|m-2).\end{aligned} \tag{11}$$

We solve this set of coupled linear difference equations using the method of the z transform [34,44]. The z transform $Q(z)$ of a function Q_n of a discrete variable $n=0,1,2,\ldots.$ is defined by $Q(z)=\Sigma_{n=0}^{\infty}Q_nz^n$. One then derives the z transform of Q_{n+1} simply as $Q(z)/z-Q_{n=0}/z$. Note the similarities of this rule with the Laplace transform of the time derivative of a continuous function [43]. The z transform of Eqs. (11) leads to a set of algebraic equations which immediately gives

$$Q_0(z,\alpha|m)=\frac{Q_{0,n=0}(\alpha|m)}{1-z[t+r(-1)^m\mathbf{F}(2m)]},$$

$$Q_1(z,\alpha|m)=\frac{Q_{1,n=0}(\alpha|m)}{1-z[t+r(-1)^m\mathbf{F}(2m)]}+\frac{z}{2\{1-z[t+r(-1)^m\mathbf{F}(2m)]\}}\times\left\{\frac{e^{-i\alpha}[t+r(-1)^{m+1}\mathbf{F}(2m+2)]Q_{0,n=0}(\alpha|m+1)}{1-z[t+r(-1)^{m+1}\mathbf{F}(2m+2)]}+\frac{e^{i\alpha}[t+r(-1)^{m-1}\mathbf{F}(2m-2)]Q_{0,n=0}(\alpha|m-1)}{1-z[t+r(-1)^{m-1}\mathbf{F}(2m-2)]}\right\},$$

$$Q_2(z,\alpha|m)=\frac{Q_{2,n=0}(\alpha|m)}{1-z[t+r(-1)^m\mathbf{F}(2m)]}+\frac{zQ_{0,n=0}(\alpha|m)}{2\{1-z[t+r(-1)^m\mathbf{F}(2m)]\}^2}+\frac{z}{1-z[t+r(-1)^m\mathbf{F}(2m)]}\times\Bigg\{e^{-i\alpha}[t+r(-1)^{m+1}\mathbf{F}(2m+2)]Q_1(z,\alpha|m+1)+e^{i\alpha}[t+r(-1)^{m-1}\mathbf{F}(2m-2)]Q_1(z,\alpha|m-1)+\frac{e^{-2i\alpha}[t+r(-1)^m\mathbf{F}(2m+4)]Q_{0,n=0}(\alpha|m+2)}{4\{1-z[t+r(-1)^{m+2}\mathbf{F}(2m+4)]\}}+\frac{e^{2i\alpha}[t+r(-1)^m\mathbf{F}(2m-4)]Q_{0,n=0}(\alpha|m-2)}{4\{1-z[t+r(-1)^{m-2}\mathbf{F}(2m-4)]\}}\Bigg\}, \tag{12}$$

where $\mathbf{F}(m)=(1-m^2)^{-1}\cos(m\pi/2)$, especially $\mathbf{F}(0)=1$. The above expressions contain the sum of several terms whose inverse z transforms are readily accessible:

$$1\leftrightarrow\frac{1}{1-z},$$

$$n\leftrightarrow\frac{z}{(1-z)^2},$$

$$a^n\leftrightarrow\frac{1}{1-az},$$

$$na^n\leftrightarrow\frac{az}{(1-az)^2}. \tag{13}$$

Here a is an arbitrary real number whose absolute magnitude is less than 1.

For an arbitrary initial distribution $P_0(x,y|\theta)$, the relevant function $Q_{1,n}(\alpha|0)$ contains terms which are either constant or behave as a^n with $|a|<1$. They are associated with the randomization of the initial distribution of the random walkers, but are not essential for large n. According to Eq. (8),

$$\langle x\rangle_n=\langle y\rangle_n=0. \tag{14}$$

The behavior of the mean-square displacements is associated with $Q_{2,n}(\alpha|0)$; see Eq. (8). We checked that for large n or in the long-time limit it is purely diffusive, i.e.,

$$\langle x^2\rangle_n=2D_x\tau,$$

$$\langle y^2\rangle_n=2D_y\tau, \tag{15}$$

where we introduced the time $\tau=n\bar{L}/v$ which passes when the random walker makes n steps at a speed v. We extract the diffusion constants from $Q_{2,n}(0|0)$ and $Q_{2,n}(\frac{\pi}{2}|0)$:

$$D_x=D_y=\frac{1}{4}\bar{L}v\left(\frac{3}{2r}-1\right). \tag{16}$$

As already mentioned, we write the master equation (4) to describe photon diffusion in the grains and in the host medium. Equation (16) immediately leads to

$$D_{in}=\frac{1}{4}\bar{L}_{in}\frac{c}{n_{in}}\left(\frac{3}{2r}-1\right),$$

$$D_{out}=\frac{1}{4}\bar{L}_{out}\frac{c}{n_{out}}\left(\frac{3}{2r}-1\right). \tag{17}$$

The task is now expressing $\bar{L}_{in}$, $\bar{L}_{out}$, f_{in}, and f_{out} in terms of the model parameters (R, ϕ, and r), and using the two-state model of Lennard-Jones to derive D_m. First, we note that $\bar{L}_{in}=<2R\cos\gamma'>$, where γ' is the incidence angle of photons moving in the disk and here $\langle\cdots\rangle$ denotes averaging with respect to the probability distribution $F'(\gamma')=\cos\gamma'$; see the Appendix A and Fig. 1(b). Second, $\phi=\bar{L}_{in}/(\bar{L}_{in}+\bar{L}_{out})$. Hence we find

$$\bar{L}_{in}=\frac{\pi R}{2},$$

$$\bar{L}_{out}=\frac{\pi R}{2}\frac{1-\phi}{\phi}. \tag{18}$$

The evaluation of f_{in} is more exacting. Each step length $\bar{L}_{in}$ inside a disk takes a time $\bar{\tau}_{in}=\bar{L}_{in}n_{in}/c$. The probability of m internal steps before leaving the disk is tr^m. Hence the average time that a photon spends in the disk is $\Sigma_{m=0}m\bar{\tau}_{in}tr^m=\bar{\tau}_{in}r/t$. The photon spends a time $\bar{\tau}_{out}=\bar{L}_{out}n_{out}/c$ before reaching a disk. Hence

$$f_{in} = \frac{\bar{\tau}_{in} r/t}{\bar{\tau}_{out} + \bar{\tau}_{in} r/t} = \frac{n_{in}\frac{r}{1-r}}{n_{in}\frac{r}{1-r} + n_{out}\frac{1-\phi}{\phi}},$$

$$f_{out} = \frac{\bar{\tau}_{out}}{\bar{\tau}_{out} + \bar{\tau}_{in} r/t} = \frac{n_{out}\frac{1-\phi}{\phi}}{n_{in}\frac{r}{1-r} + n_{out}\frac{1-\phi}{\phi}}. \quad (19)$$

Now we utilize Eq. (3) to derive the diffusion constant of photons in the granular medium as

$$D_m = \frac{\pi R c}{8} \frac{\left(\frac{3}{2r} - 1\right)\left(\frac{r}{1-r} + \left(\frac{1-\phi}{\phi}\right)^2\right)}{n_{in}\frac{r}{1-r} + n_{out}\frac{1-\phi}{\phi}}. \quad (20)$$

In two-dimensional space, the transport mean free path follows from

$$l^* = 2D_m/c_m, \quad (21)$$

where c_m is the transport velocity of light in the medium. To a first approximation,

$$c_m = \phi\frac{c}{n_{in}} + (1-\phi)\frac{c}{n_{out}}. \quad (22)$$

Note that the velocity of light in the disks (the host medium) covering a fraction $\phi(1-\phi)$ of the plane is $c/n_{in}(c/n_{out})$. From Eqs. (20)–(22), we find the transport mean free path l^* mentioned already in Eq. (2) in the Introduction.

B. Numerical simulations

We presented an *analytic* theory to calculate the diffusion constant of photons. Now we carefully examine this analytic result by performing numerical simulations.

In order to generate random configurations of monodisperse disks with a desired packing fraction, we compress a dilute system of rigid disks into a smaller space. Simulation methods based on a confining box generate a packing whose properties in the vicinity of walls differ from those in the bulk. Hence we utilize the compaction method of Ref. [45], which combines the contact dynamics algorithm [46,47] with the concept of the Andersen dynamics [48]. This combined simulation method involves a variable area of the simulation box with periodic boundary conditions in all directions. Due to the exclusion of sidewalls, the algorithm generates homogenous packings.

We let photons perform a random walk in our packing of disks by applying the rules introduced in Sec. II. For improving the speed of our ray-tracing program, we adopt the cell index method commonly used in the molecular dynamics simulations [49]. The square simulation box is divided into a regular lattice of $J \times J$ cells. We maintain a list of disks in each of these cells. A photon moving in the cell $j(1 \leqslant j \leqslant J^2)$ probably hits the disks in the cell j or its neighbor cells. Thus it is not necessary to check the collision between the photon and *all* disks of the medium.

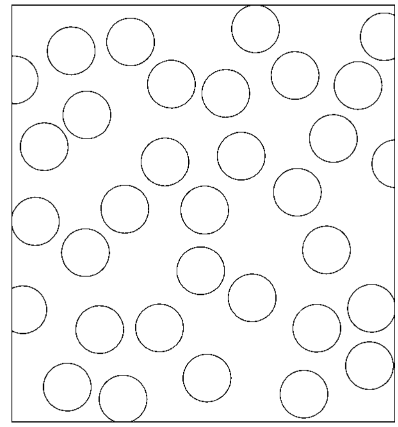

FIG. 2. Part of a packing of 10^4 disks, covering a fraction $\phi = 0.35$ of the plane.

Our computer program shrinks an initial dilute sample of 10^4 nonoverlapping disks randomly distributed in a two-dimensional simulation box. In the course of shrinking the packing, the program saves snapshots of the grain positions if the packing fraction $\phi \in [0.15, 0.25, \ldots, 0.75]$; see Fig. 2. For each medium, the program takes 10^4 photons at an initial position and launches them in a direction specified by angle θ_0 relative to the x axis. Then it generates the trajectory of each photon following a standard Monte Carlo procedure and evaluates the statistics of the photon cloud at times $\tau \in [7000, 7100, \ldots, 9900]$ (in units of R/c). As detailed in Ref. [26], we determine the diffusion constant D_m from the temporal evolution of the average mean-square displacement of the photons: $\langle x^2 + y^2 \rangle = 4D_m\tau$.

For angles $\theta_0 \in [20°, 40°, \ldots, 320°]$, the simulation is repeated for each intensity reflectance $r \in [0.1, 0.2, \ldots, 0.9]$. As a reasonable result, no dependence on the starting point and the starting direction is observed. In Fig. 3 we plot the diffusion constant D_m (in units of the disk radius R times the velocity of light c) as a function of the intensity reflectance r for glass disks ($n_{in}=1.5$) immersed in air ($n_{out}=1.0$). For this medium, Fig. 4 shows the diffusion constant D_m (in units of the disk radius R times the velocity of light c) as a function of the packing fraction ϕ for the intensity reflectances $r = 0.1$ and 0.4. The error bars reflect the the standard deviations when we average over all diffusion constants for different starting positions and angles. We also performed simulations for the other examples ($n_{in}=1.34$, $n_{out}=1.0$), ($n_{in}=1.0$, $n_{out}=1.34$), ($n_{in}=1.0$, $n_{out}=1.5$), etc., but the results are not shown here. We observed the overall agreement between the numerical results and our theoretical value for D_m. Quite remarkably, Eq. (20) involves no free parameters, but reasonably agrees with the numerical results in a wide range of ϕ, r, n_{in}, and n_{out}.

IV. DISCUSSION, CONCLUSIONS, AND OUTLOOK

Diffusing-wave spectroscopy provides invaluable information about the static and dynamic properties of granular

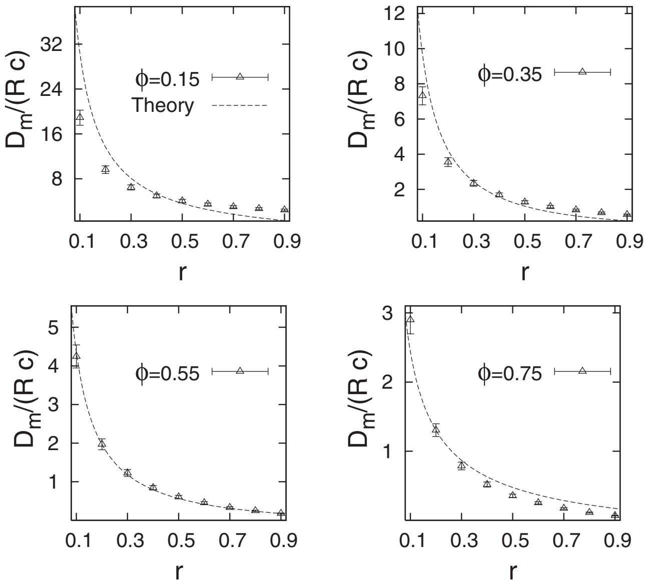

FIG. 3. The diffusion constant D_m (in units of the disk radius R times the velocity of light c) as a function of the intensity reflectance r for various packing fractions. Here $n_{in}=1.5$ and $n_{out}=1.0$ are assumed. Monte Carlo simulation results and $D_m(r)$ are denoted, respectively, by points and the line.

media [6–9] and foams [10–16]. The transport mean free path l^* in terms of the microscopic structure, however, remains to be fully elucidated. In this paper, we consider a *simple* model for photon diffusion in a two-dimensional packing of disks. Our *analytical* result for l^* provides insights into light transport and promotes more realistic models.

We have studied the photon's persistent random walk in a two-dimensional packing of monodisperse disks. We employed ray optics to follow a light beam or photon as it is reflected by the disks. We used a constant-intensity reflectance r. Moreover, we assumed that on hitting a disk, the incident and transmitted rays have the same direction. To achieve a better understanding of photon diffusion in granular media and wet foams, we are extending our studies by considering Fresnel's formulas for the intensity reflectance, Snell's law of the refraction, and the three-dimensional packing of polydisperse spheres. We are also improving our estimate of l^* by taking into account the distribution of photons' step length and the transport velocity of photons [50]. We discuss these points in the following.

Many *two*-dimensional systems offer a rich and unexpected behavior. For an experimental observation of photon diffusion in a two-dimensional packing, a set of parallel fibers can be used. Photons injected in a plane perpendicular to the axis of fibres perform a planar diffusion. One can also drill parallel cylinders in a host medium and fill all the cylinders with a liquid: the l^* dependence on the refractive index n_{in} can be measured. Note that a ray maintains its polarization state on hitting a disk and Fresnel's intensity reflectance depends on the polarization state: Quite interesting, the transport mean free paths for the transverse electric and transverse magnetic polarizations are different. Inspired by the rich optics of a two-dimensional packing and following a step-by-step approach to a real system, our attention is naturally directed to estimate l^* of a *three*-dimensional granular medium or wet foam. We speculate that one can multiply Eq. (2) with a factor of about 1 to estimate the transport mean free path for a three-dimensional packing. Apparently, only a detailed study will approve or disapprove our speculation which is based on the following observation: To understand the role of liquid films for light transport in dry foams, we studied two- and three-dimensional Voronoi foams [27,31]. The interesting result is that the transport mean free paths for these dry foams are determined by the same factor $(1-r)/r$ for a constant-intensity reflectance r in spite of the difference in dimension: $l^*_{2\mathrm{D\ Voronoi}}(r)\approx 1.10R(1-r)/r$ and $l^*_{3\mathrm{D\ Voronoi}}(r)\approx 1.26R(1-r)/r$, where R denotes the average cell radius [51].

Our "mean-field" theory presented in Sec. III A relies on the *average* step lengths $\bar{L}_{in}$ and $\bar{L}_{out}$. We note that $L_{in}=2R\cos\gamma'$ and the probability distribution of the incidence angle γ' is $F'(\gamma')=\cos\gamma'$; see the Appendix and Fig. 1(b). It follows that the probability distribution of L_{in} is $G(L_{in})=L_{in}/(2R\sqrt{4R^2-L_{in}^2})$, $\bar{L}_{in}=\pi R/2$, and $\overline{L_{in}^2}=\int L_{in}^2 G(L_{in})dL_{in}=8R^2/3$. Figure 5 delineates the probability distribution $G(L_{out})$ as a function of L_{out}/R for various packing fractions ϕ. After reaching its pronounced maximum, the distribution $G(L_{out})$ decays exponentially. The pioneering work of Heiderich *et al.* [52] suggests that the broadness of $G(L_{in})$ and $G(L_{out})$ affects the value of l^* by a factor about 1.

In two-dimensional space, l^* and D_m are related as $D_m=l^* c_m/2$, where c_m is the transport velocity of light. In a medium composed of dielectric spheres comparable to the light wavelength λ (Mie scatterers), the transport velocity differs by an order of magnitude from the phase velocity

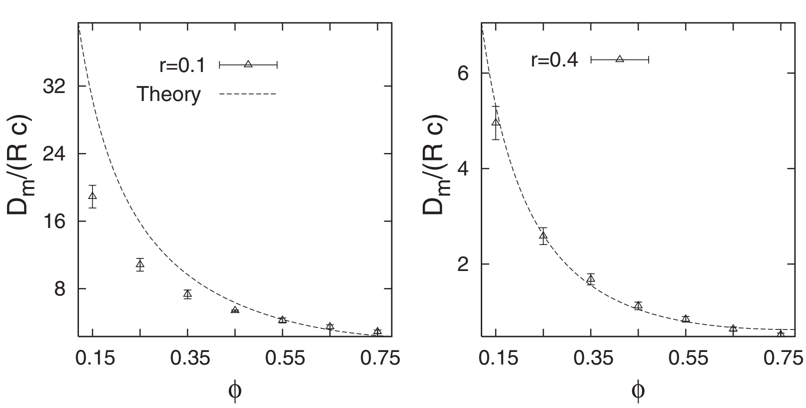

FIG. 4. The diffusion constant D_m (in units of the disk radius R times the velocity of light c) as a function of the packing fraction ϕ for $r=0.1$ and 0.4. Here $n_{in}=1.5$ and $n_{out}=1.0$ are assumed. Monte Carlo simulation results and $D_m(\phi)$ are denoted, respectively, by points and the line.

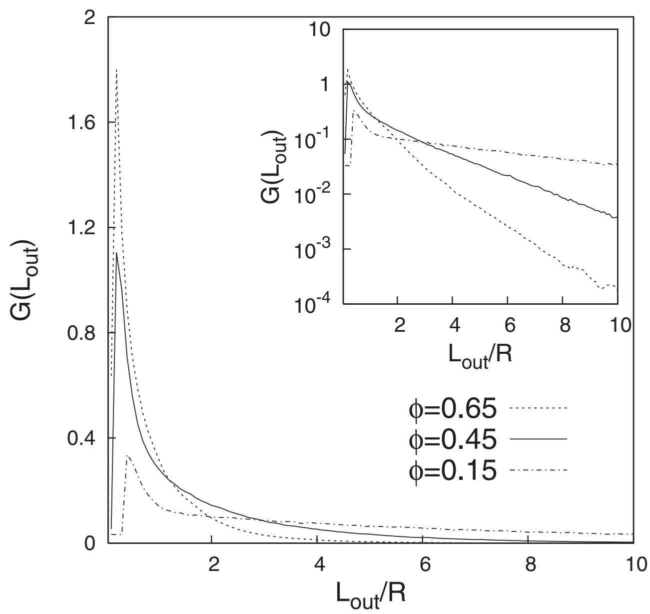

FIG. 5. The probability distribution $G(L_{out})$ as a function of L_{out}/R for various packing fractions ϕ. Inset: the same plot in the logarithmic scale. Note that $G(L_{out})$ decays exponentially.

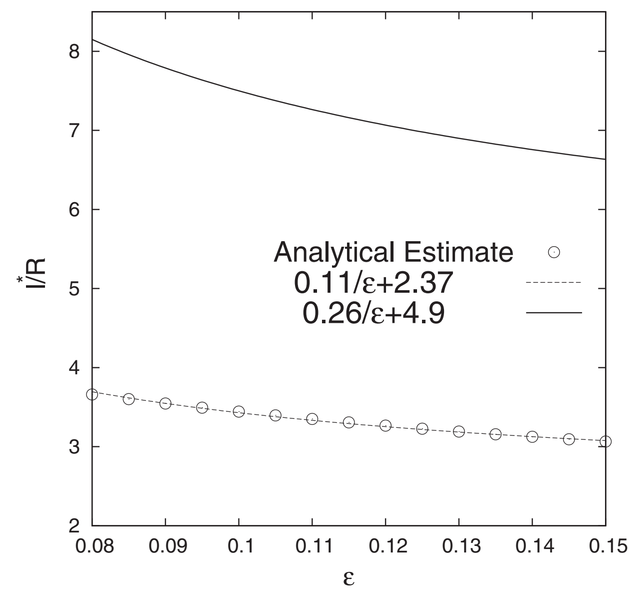

FIG. 6. The transport mean free path l^* of our model wet foam, as a function of the liquid volume fraction ε. l^* is measured in units of the bubble radius R. $n_{in}=1.0$ and $n_{out}=1.34$ are assumed. Our *analytical* result agrees well with the relation $l^*/R \approx 0.11/\varepsilon+2.37$. Numerical simulations of Ref. [24] are well described by $l^*/R \approx 0.26/\varepsilon+4.90$.

[53]. When spheres are much larger than the light wavelength, the difference between the two velocities becomes unimportant. However, our first approximation to c_m [Eq. (22)] can be improved by considering the real (rather than infinite) size parameter of disks and the pair-correlation function of disks.

We assumed that on hitting a disk, the incident and transmitted rays have the same direction. Note that in the case $n_{out}>n_{in}$, a photon moving in the host medium and hitting the disks experiences an average scattering angle $\int_0^{\pi/2}(\pi-2\gamma)F(\gamma)d\gamma=2$ (in radians) due to the reflections. Taking into account the inequality of the incidence and refraction angles and the total internal reflection of rays with incidence angle greater than $\gamma_c=\arcsin(n_{in}/n_{out})$, the average scattering angle due to the transmissions is $\int_0^{\gamma_c}[\arcsin(n_{out}/n_{in}\sin\gamma)-\gamma]F(\gamma)d\gamma=0.132$ (in radians), where $n_{out}=1.34$ and $n_{in}=1$ are assumed. Reflections are more efficient than transmissions in randomizing the direction of photons. Thus the assumption that the incident and transmitted rays have the same direction leads to a plausible estimate of l^*.

Fresnel's intensity reflectance depends on the incidence angle and the polarization state of the light. However, an average (constant) intensity reflectance r may describe the photon diffusion. To estimate r, we first consider the case $n_{out}>n_{in}$ and define the critical angle $\gamma_c=\arcsin(n_{in}/n_{out})$. The probability distributions of the incidence angles γ and γ' are $F(\gamma)=\cos\gamma$ and $F'(\gamma')=\cos\gamma'$. We define $r_{out\to in}=\int_0^{\pi/2}r_{out\to in}(\gamma)F(\gamma)d\gamma$. Here $out\to in$ indicates a ray incident from the host medium onto the disk. The incident electric field can be decomposed into a component parallel (p) to the plane of incidence and a component perpendicular (s) to this plane. To address the photon diffusion in a *three*-dimensional random packing, as reflectance $r_{out\to in}(\gamma)$ we have taken an average over the p and s polarizations: $r_{out\to in}(\gamma)=0.5[r_{out\to in}(\gamma,p)+r_{out\to in}(\gamma,s)]$. Note that for $\gamma>\gamma_c$, Fresnel's intensity reflectances are 1. Similarly, we define $r_{in\to out}=\int_0^{\pi/2}0.5[r_{in\to out}(\gamma',p)+r_{in\to out}(\gamma',s)]F'(\gamma')d\gamma'$ and then use $r=0.5(r_{out\to in}+r_{in\to out})$ in Eq. (2) to estimate l^*.

In the case $n_{out}<n_{in}$, the appropriate critical angle is $\gamma_c=\arcsin(n_{out}/n_{in})$. Here $0<\gamma<\pi/2$ and $0<\gamma'<\gamma_c$. The fact that $\gamma'<\gamma_c$ ensures that a photon moving in the disk is able to enter the host medium, since Fresnel's intensity reflectance is 1 for $\gamma'>\gamma_c$. An extension of the Appendix and numerical simulations indicate that the probability distributions of the incidence angles γ and γ' are $F(\gamma)=\cos\gamma$ and $F'(\gamma')=\cos\gamma'/\sin\gamma_c$, respectively. We define $r_{out\to in}=\int_0^{\pi/2}0.5[r_{out\to in}(\gamma,p)+r_{out\to in}(\gamma,s)]F(\gamma)d\gamma$ and $r_{in\to out}=\int_0^{\gamma_c}0.5[r_{in\to out}(\gamma',p)+r_{in\to out}(\gamma',s)]F'(\gamma')d\gamma'$ to estimate $r=0.5(r_{out\to in}+r_{in\to out})$. In the limit $n_{in}=n_{out}$, we find $r=0$. In this limit the photon transport is ballistic and Eq. (2) correctly predicts $l^*\to\infty$.

For glass disks ($n_{in}=1.5$) immersed in air ($n_{out}=1.0$), we estimate $r\approx 0.12$. Thus for a packing fraction $\phi=0.64$, we find $l^*\approx 7R$. For glass disks immersed in water ($n_{out}=1.34$), we find $r\approx 0.09$ and $l^*\approx 9R$. Our transport mean free paths are smaller than the experimental values [6,8] by a factor of about 2. Now we consider a simple model for wet foams. For air bubbles ($n_{in}=1$) immersed in water ($n_{out}=1.34$), we estimate $r\approx 0.20$. Figure 6 delineates l^* (in units of R) as a function of the liquid volume fraction $\varepsilon=1-\phi$. From Fig. 6 we find that in the range $0.08<\varepsilon<0.15$, our analytical result agrees well with the relation

$$l^* \approx R\left(\frac{0.11}{\varepsilon}+2.37\right). \tag{23}$$

Using the hybrid lattice gas model for two-dimensional foams and Fresnel's intensity reflectance, Sun and Hutzler performed a numerical simulation of photon transport [24]. For $0.02 < \varepsilon < 0.16$, their numerical results can be fitted to $l^* \approx R(0.26/\varepsilon + 4.90)$. Figure 6 compares our analytic prediction with the numerical result of Ref. [24]. Again our analytic estimate is smaller than the numerical simulations by a factor of about 2.

Quite remarkably, our *analytic* estimate of the transport mean free path l^* quoted in Eq. (2), sheds some light on the empirical law of Vera *et al.* and the numerical simulation of Sun and Hutzler: l^*/R is a *linear* function of $1/\varepsilon$; see Eqs. (1) and (23). For a better understanding of the empirical law (1), we aim at a more realistic model which not only considers Fresnel's intensity reflectance with its significant dependence on the incidence angle, but also the broad distribution of the photon step lengths. Also an extension to the three-dimensional packing of polydisperse spheres is envisaged.

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APPENDIX: THE PROBABILITY DISTRIBUTIONS $F(\gamma)$ AND $F'(\gamma')$

To find the probability distribution of the random variable γ, we assume that the impact parameter s in Fig. 1(c) has a uniform distribution in the interval $[0,R]$. In other words, we assume that the number of incident rays with an impact parameter less than s is *proportional* to s. The cumulative distribution function $F_c(\gamma) \equiv \int_0^\gamma F(\psi)d\psi$ is then $F_c(\gamma) = \text{Prob}(s < R\sin\gamma) = \sin\gamma$. It follows that

$$F(\gamma) = \frac{dF_c(\gamma)}{d\gamma} = \cos\gamma. \tag{A1}$$

Now we consider the path of photons inside the disk; see Fig. 1(d). Each ray can be characterized by its distance s' from the center of the disk. We assume that the random variable s' has a uniform distribution in the interval $[0,R]$. Since $s' = R\sin\gamma'$, the cumulative distribution function is $F'_c(\gamma') = \sin\gamma'$ and

$$F'(\gamma') = \frac{dF'_c(\gamma')}{d\gamma'} = \cos\gamma'. \tag{A2}$$

Further numerical simulations of our model confirm these analytical results for $F(\gamma)$ and $F'(\gamma')$.

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