

Theory of diffusive light scattering from disordered materials

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Abstract. It is suggested that the measurement of the angular distribution of light transmitted through a thick slab of diffusively scattering material may provide information about the nature of the local scattering events. Monte Carlo simulations indicate that the angular distribution depends on ℓ/ℓ^* , where ℓ and ℓ^* are the true and transport mean free paths, respectively, and this dependence is confirmed analytically using a modified diffusion model. Preliminary experimental results for liquid and solid foams are explicable in terms of strong forward scattering in the individual scattering events.

Subject terms: foams; diffusive light scattering.

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Many common substances, such as milk, foam, and white paint, owe their white appearance to diffusive light scattering. In such a case, light is multiply scattered within the material and not significantly absorbed. Durian et al.¹ have shown that the measurement of the transmitted light through a sample of slab geometry can be used to monitor the statistics of the local structure that is responsible for the scattering. In particular, the scaling behavior of the coarsening of a foam can be identified by the experiment, on the assumption that the structure remains self-similar.

The procedure of Durian et al. makes only limited use of the available data. In particular, the angular dependence of the transmitted light (Fig. 1) is easily measured. We have made such measurements for both liquid and solid foam slabs, and have found that the angular dependence is well expressed by

$$T(\theta) = a \cos\theta + b \cos^2\theta, \quad (1)$$

where a and b are constants. This prompts the question, what is the significance of the ratio b/a , and is it a useful probe of structure? Because of its importance in many fields of physics, diffusive light scattering has been addressed by many authors, for example in Ref. 2. Nevertheless, there seems to be no appropriate analysis of the angular distribution shown in Fig. 1, and its dependence on the details of the diffusion mechanism. In this paper, we relate b/a to the character of the local scattering events and argue that it should be incorporated in the phenomenology associated with this experimental method.

In common with many previous discussions, we adopt the diffusion equation as an accurate description of light prop-

agation in the bulk of the sample (for sufficiently thick samples). This model does indeed predict a distribution of the form of Eq. (1), but the ratio of the parameters b/a is determined by the boundary conditions employed, and these cannot be rigorous because the diffusion approximation fails in the surface region. We offer, however, an approximate scheme to relate the behavior of b/a for transmitted light to the nature of the individual scattering processes within the diffusive material.

We assume that the material in question has a uniform distribution of scattering centers (particles, bubbles, etc.), each having the same scattering properties, and randomly oriented. For each scatterer, the cross section $\sigma(\hat{\mathbf{k}}, \hat{\mathbf{k}}')$ (averaged over the orientation of the scatterer and polarization of the light) for scattering from the propagation direction $\hat{\mathbf{k}}'$ to direction $\hat{\mathbf{k}}$, is assumed to depend only on $\hat{\mathbf{k}}' \cdot \hat{\mathbf{k}} = \cos\theta_s$. Of particular significance in transport theory is the mean of the cosine of the scattering angle for each scattering event:

$$\langle \cos\theta_s \rangle = \frac{\int_{-1}^{+1} d(\cos\theta_s) \cos\theta_s \sigma(\cos\theta_s)}{\int_{-1}^{+1} d(\cos\theta_s) \sigma(\cos\theta_s)}. \quad (2)$$

The diffusion approximation can be used even when the scattering is not isotropic provided the mean free path ℓ in the usual expression $D = c\ell/3$ for the diffusion constant (where c is the effective velocity of light in the diffusive medium) is replaced by the transport mean free path ℓ^* . In the case of negligible absorption, this is defined by

$$\ell^* = \frac{\ell}{1 - \langle \cos\theta_s \rangle}. \quad (3)$$

Large ℓ^*/ℓ corresponds to the case of strong forward scattering. In the cases of interest, both ℓ and ℓ^* can be assumed

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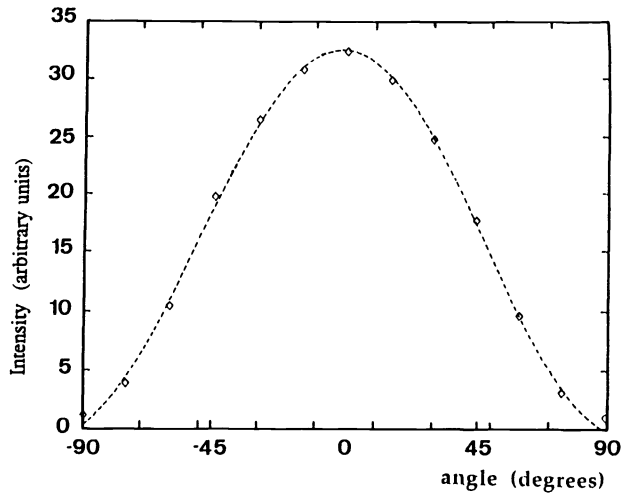


Fig. 1 Angular distribution of light transmitted through a solid foam slab of thickness 1.5 cm, fitted by a function of the form given in Eq. (1) with $b/a=2.7$.

to be much less than the sample dimensions. They are also much greater than the wavelength of light, so we are nowhere concerned with diffraction or interference effects.

We performed model Monte Carlo simulations using an ensemble of rays that propagate from a source within a finite slab. They undergo scattering with a fixed scattering length ℓ and a cross section defined by the simple rule

$$\cos\theta_s = \frac{\mu + \cos\gamma}{(1 + 2\mu \cos\gamma + \mu^2)^{1/2}}, \quad (4)$$

where γ defines a random direction and the parameter $\mu = 0$ for isotropic scattering, whereas $\mu > 0$ biases the scattering toward forward directions. Integration of Eq. (4) over all random directions gives

$$\begin{aligned} \langle \cos\theta_s \rangle &= \frac{2\mu}{3} \quad 0 \leq \mu \leq 1 \\ &= 1 - \frac{1}{3\mu^2} \quad \mu > 1. \end{aligned} \quad (5)$$

The obvious rule is adopted for the escape of a ray from the slab without refraction or reflection at the surface, that is, a ray that crosses the surface plane $z=L$ ($> \ell, \ell^*$) is taken to have been emitted. Data are gathered for the exit angles of such rays.

By the choice of the parameter μ , we vary the strength of forward/backward scattering, and hence ℓ^*/ℓ . We have found that for a wide range of values, the emitted angular intensity profile was fitted well by Eq. (1), as shown in Fig. 2. In Fig. 3, we show the variation of the shape parameter b/a with ℓ^*/ℓ . It increases strongly with increasing forward scattering until saturating at a value of approximately 2.2. The same data are plotted as a function of the inverse variable ℓ/ℓ^* in Fig. 4, exhibiting a straight-line variation for $\ell/\ell^* \leq 1$.

A diffusion model *per se* cannot predict effects that depend on the characteristics of the local scattering events. The relationship between the diffusion constant and the transport mean free path, $D = c\ell^*/3$, is derived when the diffusion

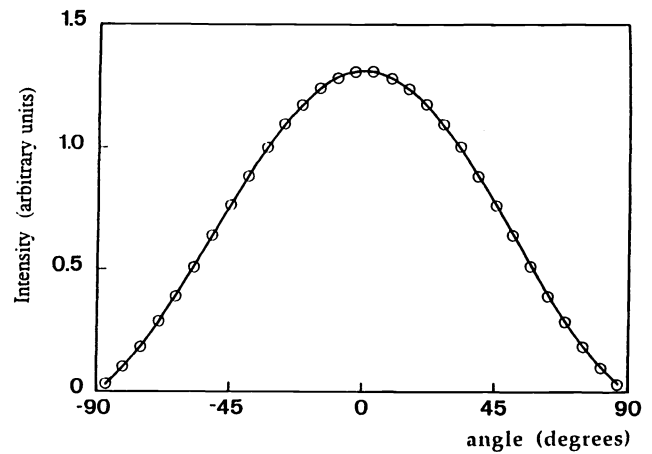


Fig. 2 Results of the Monte Carlo simulation for the angular distribution of particles transmitted through a slab, in the case $\ell^*/\ell=2$, fitted by a function of the form given in Eq. (1) with $b/a=1.65$.

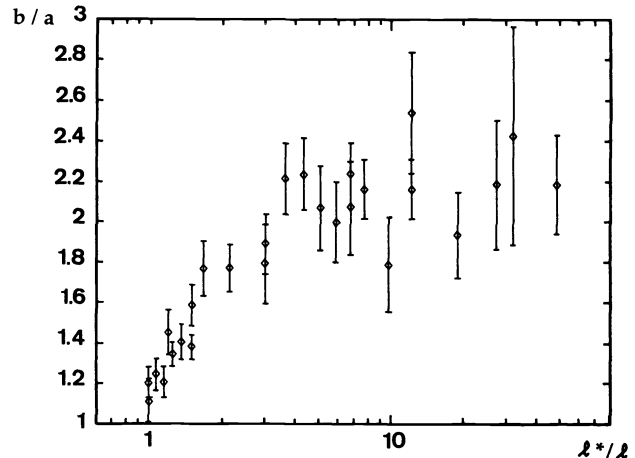


Fig. 3 Results of the Monte Carlo simulation for the shape parameter b/a plotted against ℓ^*/ℓ .

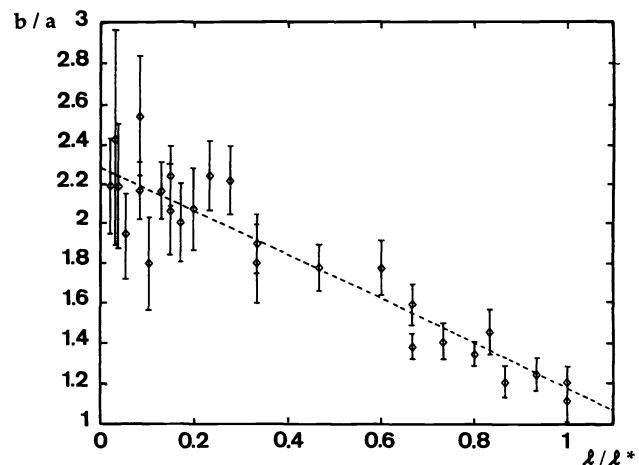


Fig. 4 Results of the Monte Carlo simulation for the shape parameter b/a plotted against ℓ/ℓ^* , showing the best linear fit to the data.

model is obtained as an approximation to transport theory.³ In this way, it can be shown that for light multiply scattered in a slab with plane surfaces normal to the z axis, a diffusion equation for $I_0(z)$ results when the light intensity is approximated by

$$I(z, \hat{\mathbf{k}}) = I_0(z) + (\hat{\mathbf{k}} \cdot \hat{\mathbf{z}}) I_1(z) . \quad (6)$$

Here $I(z, \hat{\mathbf{k}})$ is the rate at which electromagnetic energy propagating in the $\hat{\mathbf{k}}$ direction crosses unit area perpendicular to $\hat{\mathbf{k}}$ at a depth z into the slab, and $(4\pi/3)I_1(z)$ is the net flux of electromagnetic energy in the z direction. Equation (6) is an expansion of the intensity in terms of the Legendre polynomials $P_\ell(\cos\theta)$, where $\cos\theta = \hat{\mathbf{k}} \cdot \hat{\mathbf{z}}$, with all but the first two terms neglected. In this approximation, we find

$$I_1(z) = -\ell^* \frac{dI_0}{dz} , \quad (7)$$

which is Fick's law. The angular distribution of light transmitted through the surface $z=L$ of the slab is then given by $I(L, \hat{\mathbf{k}}) \cos\theta$. (We assume that the width L is sufficiently large that there is no direct transmission through the slab, so that we are only concerned with the diffuse light intensity, and ℓ/L , $\ell^*/L \ll 1$.) Thus the diffusion approximation predicts the angular distribution of transmitted light to be of the form shown in Eq. (1), with

$$a = I_0(L) = \frac{1}{2} \int_{-1}^{+1} d(\cos\theta) I(L, \hat{\mathbf{k}}) , \quad (8)$$

$$b = I_1(L) = \frac{3}{2} \int_{-1}^{+1} d(\cos\theta) P_1(\cos\theta) I(L, \hat{\mathbf{k}}) .$$

The experimental results for solid foam (Fig. 1) are well fitted by the form given in Eq. (1), containing no powers of $\cos\theta$ higher than the second. Similar results are obtained with liquid foam, although the fit is not so straightforward because of reflection and refraction at the walls of the container. These results indicate that the diffusion model is adequate to describe the propagation of light through both solid and liquid foams. However, the magnitude of b/a predicted by the usual boundary conditions is independent of ℓ and ℓ^* . This is in contradiction with the results of the Monte Carlo calculations, shown in Fig. 3, and the discrepancy can be attributed to the crudity of the boundary conditions, as we see later.

In the simple diffusion model, we require that there should be no net diffuse flux into the slab at a boundary. So the diffuse flux integrated over all inwardly directed angles at a boundary is set equal to zero. At $z=L$, this condition becomes

$$\int_{-1}^0 d(\cos\theta) I(L, \hat{\mathbf{k}}) \cos\theta = -\frac{1}{2} I_0(L) + \frac{1}{3} I_1(L) = 0 , \quad (9)$$

giving $b/a = 3/2$. Given the approximate nature of the boundary condition, and the fact that the form of $I(z, \hat{\mathbf{k}})$ determined by the diffusion model is not correct within a few mean free paths of the boundary, this value is not expected to be accurate. The analytic solution to the corresponding transport problem in the case of isotropic scattering events in a semi-infinite slab (the Milne problem) gives $b/a \approx 1/0.71 \approx 1.4$.

The stringent requirement, that there should be no diffuse flux entering the medium at any angle through the boundary,

cannot be satisfied by an intensity with the simple angular distribution of Eq. (6). In contrast, the Monte Carlo simulation does impose this more stringent condition, because any ray that crosses the plane $z=L$ cannot return. It is therefore of interest to see whether any modification of the boundary condition imposed in the simple diffusion model can lead to a dependence of b/a on ℓ^*/ℓ in accordance with the results of the simulation.

In the simulation, where the length ℓ is fixed, a ray crossing the boundary plane $z=L$ at an angle θ to the z axis must have been last scattered in the strip between $z=L-\ell \cos\theta$ and $z=L$, and the position of the last scattering event defines the boundary beyond which the ray cannot be scattered back into the material. Correspondingly in the diffusion model, we suppose that the average depth below the surface at which the last scattering event occurs determines the boundary across which there is no inward diffusion for light emitted from the slab at angle θ to the z axis. For each angle θ , we impose a boundary condition of the form given in Eq. (9) at the average position of the last scattering event, which we take to be $z=L' = L - \alpha \ell \cos\theta$. For fixed ℓ and uniformly distributed scatterers, we should expect $\alpha = 1/2$ (at least in the case of predominantly forward scattering). This gives

$$I_0(L') = \frac{2}{3} I_1(L') . \quad (10)$$

The intensity at the surface $z=L$ [Eq. (6)] is then expanded about $z=L'$:

$$I_0(L) + I_1(L) \cos\theta = \left[I_0(L') + (L-L') \frac{dI_0}{dz} \Big|_{L'} + \dots \right] + \cos\theta \left[I_1(L') + (L-L') \frac{dI_1}{dz} \Big|_{L'} + \dots \right] . \quad (11)$$

In the absence of absorption the diffusion equation gives $d^2 I_0/dz^2 = 0$ at points far from the source, and use of this expression, together with Eqs. (7) and (10), in Eq. (11) reduces it to

$$I_0(L) + I_1(L) \cos\theta = I_0(L') + \cos\theta \left(1 - \alpha \frac{\ell}{\ell^*} \right) I_1(L') = \frac{2}{3} I_1(L) \left[1 + \frac{3}{2} \left(1 - \alpha \frac{\ell}{\ell^*} \right) \cos\theta \right] .$$

The resulting angular distribution of transmitted light, $I(L, \hat{\mathbf{k}}) \cos\theta$, is of the form given in Eq. (1) with

$$\frac{b}{a} = c \left(1 - \alpha \frac{\ell}{\ell^*} \right) , \quad (12)$$

where $c = 3/2$. If we require Eq. (12) to reproduce the Milne result in the case of isotropic scattering, $\ell^* = \ell$, then the value $3/2$ should be replaced by

$$c \approx \frac{1.4}{1 - \alpha} = 2.8, \quad \text{if } \alpha = 0.5 . \quad (13)$$

The best linear fit to the simulation data, shown in Fig. 4,

gives $\alpha = 0.496 \pm 0.02$, in good agreement with the expected value $1/2$, and $c = 2.268 \pm 0.05$.

In the case of a foam with a small liquid or solid fraction, as used in our experiments, the scattering can be assumed to result from the Plateau borders (the thickened lines at the junctions of films), which act as prisms with a characteristic concave cross section.⁴ Pittet⁵ performed a Monte Carlo calculation for such a prismatic scattering element, with the refractive index of water, and found ℓ^*/ℓ to be 3.23. The data of Fig. 4 give $b/a \approx 2.0$ for such a case, and preliminary experimental measurements give $b/a \approx 2.7 \pm 0.5$ for solid foam. It would appear that forward scattering will dominate in many cases of practical interest, but systems with more isotropic scattering can be made, and will offer a detailed test of the validity of Eq. (12).

In a recent paper, Gonatas et al.⁶ recognized the importance of the angular distribution of diffusely scattered light, and Freund et al.⁷ measured it in a quasi-two-dimensional system. These authors, however, have not related the angular distribution to the characteristics of the local scattering events as we do in this paper. In conclusion, we believe this work opens up a new dimension in the use of diffusive light scattering for structural characterizations.

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James Lunney: Biography and photograph appear with the paper "In situ monitoring of metal multilayer growth by optical reflectometry" in this issue.



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