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Importance of Boundary Reflections in the Theory of Diffusive Light Scattering

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Abstract

This PDF file contains the letter "Letter: Importance of boundary reflections in the theory of diffusive light scattering [see 33(12)3849-3852(Dec1994)]" for OE Vol. 34 Issue 11

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At the time of publication, author Douglas J. Durian was affiliated with University of California, Los Angeles. Currently, he is a faculty member at the Physics Department at the University of Pennsylvania.

COMMUNICATIONS

Comment

Importance of boundary reflections in the theory of diffusive light scattering

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In a recent paper, McMurry, Weaire, Lunney, and Hutzler[1] claimed that the angular dependence of exiting photons diffusively transmitted through a disordered multiple-light-scattering material is controlled by scattering anisotropy in terms of the ratio ℓ^*/ℓ_s of the transport to scattering mean free paths. Motivated by discussion with Weaire, I considered the same problem but reached a different conclusion.[2] Namely, the angular dependence is approximately independent of scattering anisotropy and depends strongly on the reflectivity of the sample boundary. Within the confines of a diffusion approximation, transport is best described by the photon concentration field $U(\mathbf{r})$ satisfying a diffusion equation with $D=(1/3)c\ell^*$ and boundary conditions such that $U(\mathbf{r})$ extrapolates to zero at distance $z_e\times\ell^*$ outside the sample. The value of the extrapolation length ratio z_e is chosen so that the fictitious flux of photons entering the sample equals the boundary reflectivity times the flux leaving. This gives $z_e=(2/3)(1+R_2)/(1-R_1)$ where $R_n=(n+1)\int_0^1\mu^n R(\mu)d\mu$ and $R(\mu)$ is the total reflection probability for a photon striking the sample boundary at angle $\cos^{-1}\mu$ with respect to the normal. Given such a concentration field, the angular dependence of the exiting photons can be found by straightforward kinetics. Ignoring refraction, I calculated that the probability $P(\mu)d\mu$ for a transmitted photon to exit between $\cos^{-1}\mu$ and $\cos^{-1}(\mu+d\mu)$ from the normal is given by

$$P(\mu)/\mu=\frac{z_e+\mu}{z_e/2+1/3}\ . \quad (1)$$

It thus contains a mixture of cosine and cosine-squared dependence that depends on the boundary reflectivity through the value of z_e.

In Ref. 2 I tested Eq. (1) by comparison with random walk computer simulations incorporating both scattering anisotropy and boundary reflectivity. Walkers were launched from one edge of a slab with thickness $L=15\ell^*$ and allowed to wander according to the values of ℓ^*/ℓ_s and $R(\mu)$ until they exited at either edge. Figure 1 shows new simulation data for isotropic scattering and several constant boundary reflectivities $R(\mu)=R$. Evidently, $P(\mu)/\mu$ varies dramatically with R but is nearly linear in μ and compares quite well with the prediction of Eq. (1).

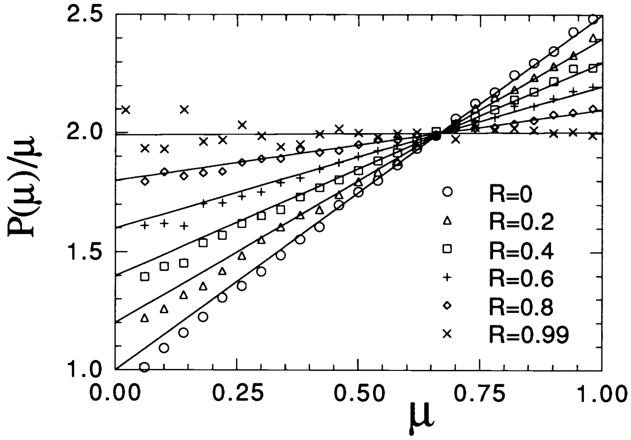

Fig. 1 Simulation results for the angular distribution of exiting photons transmitted through slabs of optical thickness $L/\ell^*=15$, with boundary reflectivities as labeled, for the case of isotropic scattering. Each data set is based on 10^6 transmission events, giving statistical error bars that are typically smaller than the symbol size and vary as $1/\sqrt{\mu}$. Data compare well with the solid lines, which represent the predictions of Eq. (1) with no adjustable parameters.

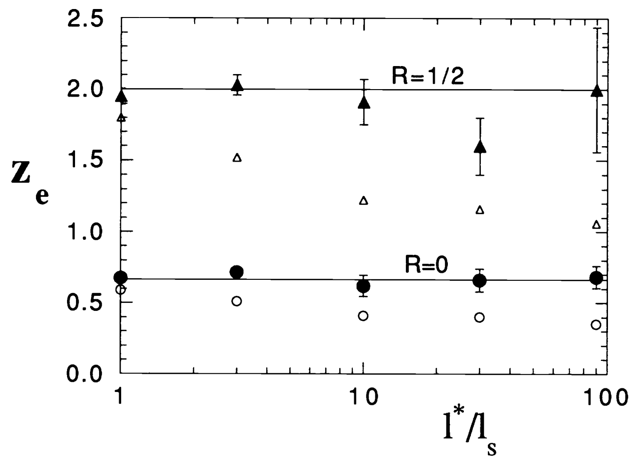

Fig. 2 Values of z_e obtained by fitting Eq. (1) to Ref. 2 simulation data for $P(\mu)$. Solid symbols are for data within 45 deg of the surface normal, where excellent fits are obtained; the resulting z_e are in good agreement with predictions, as shown, independent of scattering anisotropy. Open symbols are for the full data set, where poor fits are obtained.

When anisotropy is included, the angular dependence is unaffected except for a subtle dip at glancing angles (see Fig. 5 of Ref. 2). As demonstrated in Fig. 2, the mixture of cosine and cosine-squared dependence near the forward direction is still controlled by boundary reflectivity. Results for z_e based on Eq. (1), multiplied by a new normalization factor, fit to simulation data for walkers exiting within 45 deg of the normal are shown by solid symbols versus ℓ^*/ℓ_s. To within statistical uncertainty, these values of z_e are constant and equal to the prediction $(2/3)(1+R)/(1-R)$. If the data are instead fit to Eq. (1) over the entire range of μ, including the dip near glancing angles where $P(\mu)/\mu$ deviates from linearity, then the resulting values of z_e decrease systematically with ℓ^*/ℓ_s, as shown by the open symbols, but have little significance since the functional form is incorrect. This could explain the increase in cosine-squared dependence with increasing ℓ^*/ℓ_s obtained by McMurry et al. from fits to their own simulation data. Perhaps the theoretical ideas they advance could be used to quantitatively explain the slight discrepancy at small μ between simulation results and Eq. (1) in terms of the scattering anisotropy. However, it should be cautioned that such deviations can be small compared to refraction effects and may depend on more details of the scattering form factor than just the value of ℓ^*/ℓ_s.

In conclusion, the angular dependence of diffusely transmitted light is set primarily by boundary effects independent of scattering anisotropy. This is also born out by experimental work on suspensions of polystyrene spheres of variable size, where refraction and polarization are also important.[3] Scattering anisotropy has at most a subtle influence on behavior at glancing angles; there, detected photons originate in a region very close to the boundary where diffusion approximations are least accurate. Contrary to the conclusion of Ref. 1, the functional form of $P(\mu)$ thus tells little about the structure of the scattering material itself. Rather, its importance is in revealing the nature of the sample boundary and in showing what value of z_e should be used in diffusion theory predictions for the transmission probability[2] and the diffusing-wave spectroscopy autocorrelation function.[4] With proper choice of z_e, accuracies on the order of 1% can be obtained without recourse to numerical solution of the exact transport equations.

References

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Response

Response to "Importance of boundary reflections in the theory of diffusive light scattering"

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In a recent paper[1] we addressed the problem of the angular dependence of light transmitted through a foam, which is predicted by the diffusion model in the case of slab geometry to be of the form

$$T(\theta)=a\ \cos\theta+b\ \cos^2\theta\ , \tag{1}$$

where θ is the angle the transmitted light makes with the normal to the exit face, and $b/a=3/2$ if the usual boundary condition, that there is no net *inward* flux of diffuse light at the exit face, is applied. Measurements on real foam samples indicated higher values of b/a, and random walk simulations showed that this ratio increases with the degree of anisotropy of the local scattering (measured by the ratio ℓ/ℓ^* of the mean free path to the transport mean free path).

Durian[2] has called these results into question on the following grounds:

1. The angular distribution of the transmitted light is strongly affected by reflection at the boundary, when a glass container is used.
2. His random walk simulations showed no dependence on anisotropy.

We fully agree with Durian on point 1. However, this problem did not arise in our measurements on solid foam, which needs no container. Our random walk program, with no reflection at the boundary, is therefore an appropriate simulation for these measurements. These simulations showed a variation of b/a that was produced only by the anisotropy of the local scattering.

Our experimental results for liquid foam, in a glass container, indicated that b/a was larger than 3/2. Our simulations showed such an increase in the case of predominantly forward local scattering. However, incorporating the effects of reflection through introducing an average reflectance R in the diffusion model[3] decreases, rather than increases, the ratio b/a, giving $[3(1-R)]/[2(1+R)]$ instead of 3/2.

With regard to point 2 we have two comments to make in reply. First, Durian uses an exponential step-length distribution in his simulations,[3] in contrast to the fixed step-length used in Ref. 1. Our simulations using the exponential step-length distribution show a reduction in the range over which b/a varies compared to the case of fixed step length. This is shown in Fig. 1. However, an increase in b/a as ℓ/ℓ^* decreases is still apparent. (The scattering function used in Ref. 3, and in the results displayed in Fig. 1, is equally anisotropic for all values of ℓ/ℓ^*, since it is essentially a delta function, selecting a particular value of the scattering angle. In particular, it does not approach isotropic scattering as $\ell/\ell^*\rightarrow 1$. However, the data we obtained with it show similar trends to that which we obtained using the scattering function of Ref. 1 or the Henyey-Greenstein function.)

Second, the variation of b/a with ℓ/ℓ^* is somewhat obscured if $[T(\theta)]/\cos\theta$ is plotted as a function of $\cos\theta$ as in Ref. 3. If the simulation data used to produce Fig. 1 are replotted in this way the result is similar to the case $R=0$ in Fig. 5 of Ref. 3. We have used these simulation data to calculate b/a in two different ways for each of five different values of ℓ/ℓ^*: