Surface reflections and boundary conditions for diffusive photon transport

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(Received 3 February 1992)

Boundary conditions for the photon-diffusion equation with internal surface reflections are obtained for one, two, and three dimensions, both from transport theory and from approximate solutions of the corresponding Milne equations.

PACS number(s): 42.25.Fx, 42.68.Ay

I. INTRODUCTION

Internal surface reflections have recently been shown, both experimentally and theoretically [1–4], to be of considerable importance in the diffusive transport of light through highly random media. Large effects come about when multiply scattered light that attempts to leave the medium at angles exceeding the critical angle is prevented from doing so by total internal reflection. Optical photons reinjected into the medium execute a second, independent random walk before once again attempting to exit. Although the fraction of the light that is reflected into the medium can vary greatly from one system to another, and depends strongly upon surface properties, an angle-averaged reflectivity $R \sim 50\%$ per pass is not atypical. Thus the average time a photon spends in the medium, the average path length it traverses, and the average number of scattering events it undergoes before exiting are all significantly increased due to surface reflections.

Three approaches have recently been put forward for incorporating surface reflections into the theory of diffusive photon transport. Lagendijk, Vreeker, and DeVries modified the diffusion-equation Green's function to include these reflections, and re-solved for an number of optical properties [1]. Freund and Berkovits showed how an already known result that had been obtained by neglecting surface reflections could be resummed over all passes to yield a final result that included these reflections [3]. Zhu, Pine, and Weitz showed that the surface reflections could be included by introducing a simple modification of the boundary conditions to the diffusion equation [4].

Generally speaking, the diffusion-equation boundary conditions involve specifying how the diffusive photon density ρ goes to zero near a boundary. Although a "natural" condition would be to have ρ vanish at the physical boundaries of the medium, this is not required by the physics of the problem and is known not to be the case. An alternative possibility would be to specify some distance from the boundary at which ρ vanishes. But the diffusion equation itself contains no suitable length scale that could be used for this purpose, so one must go outside the theory. Turning to a microscopic theory of photon transport, one typically solves for some property, compares this with the corresponding diffusion-equation solution, and based upon this comparison one extracts a boundary condition by matching the two solutions in some way. However, since transport theory and diffusion theory are not fully consistent, choosing different properties for the comparison will generally result in somewhat different boundary conditions.

The final results of Zhu, Pine, and Weitz [4] have an especially appealing simplicity. The physical basis for these results, however, appears to be unclear. Zhu, Pine, and Weitz obtain a transport theory expression for the partial photon fluxes *inside* the random medium that they write in terms of scatterer properties and the photon density and its gradient. They then extend this expression into the vacuum *outside* the medium, thereby populating the vacuum with a fictitious set of scatterers that have the same scattering properties (mean free path) as the real scatterers inside the medium. They also assume that the vacuum contains a fictitious diffusive photon density ρ_v, which is a linear extrapolation of the density ρ inside the medium. A boundary condition for the real photon density ρ is then obtained by requiring the total flux of fictitious photons that diffuse out of the vacuum back into the medium to be zero, so that the populated vacuum has no net effect on the photon density inside the medium. But the back-diffusing flux from the populated vacuum can only be made to vanish if the vacuum photon density ρ_v contains both positive and *negative* components, so that the back-diffusing flux of fictitious positive photons is canceled by the back-diffusing flux of fictitious negative photons. Since ρ_v is taken to be a linear extrapolation of ρ, the point at which ρ_v must cross from positive to negative values in order to meet the above requirement is then used as the boundary condition for ρ itself, i.e., that point outside the medium at which ρ extrapolates to zero.

In view of the importance of the boundary conditions, a derivation whose physics is transparent would clearly be desirable. Here, I derive a set of boundary conditions for one, two [5], and three dimensions (1D, 2D, and 3D) by comparing the (real) photon current *leaving* the medium as obtained from transport theory with the photon current that is obtained from diffusion theory. I also compare the photon density obtained from diffusion theory with the photon density obtained from a Milne equation [6] that has been modified to include surface reflections, and I obtain a second set of boundary conditions for 1D, 2D, and 3D. All these boundary conditions have the same general form

$$\rho_B=\Delta_d(R)\lambda_S \left|\frac{\partial\rho}{\partial z}\right|_B , \tag{1}$$

where the z axis is taken along the normal to the boundary, λ_S is the scattering mean free path, and the subscript B implies evaluating the indicated quantities at the boundary. The sought-after constant $\Delta_d(R)$ depends upon dimensionality d and the average surface reflectivity R. Equation (1), which is sometimes called the "radiation boundary condition" for reasons unrelated to our problem [7], is also of the same general form as the 3D boundary condition obtained by Zhu, Pine, and Weitz [4].

II. DIFFUSION THEORY

In obtaining intrinsic boundary conditions for the diffusion equation, we need only consider the simple case in which external sources are absent from the boundary region and photon absorption may be neglected. The photon density $\rho(\mathbf{r},t)$ in the medium is assumed to obey the continuity equation $\partial\rho/\partial t=-\nabla\cdot\mathbf{J}$, where $\mathbf{J}(\mathbf{r},t)$ is the photon current density. Diffusion theory enters when the current density is taken to be diffusive,

$$\mathbf{J}=-D\nabla\rho , \tag{2a}$$

where in d dimensions the diffusion constant is

$$D=\frac{\lambda_S^2}{d\tau_S} , \tag{2b}$$

and τ_S, which is the mean time between scattering events, includes both the transit time and a possible time delay $\Delta\tau=\partial\varphi/\partial\omega$ [8] due to phase shifts φ arising from scatterer resonances at the incident frequency ω [9]. Inserting $\mathbf{J}$ into the continuity equation and assuming the steady state yields the diffusion equation,

$$D\nabla^2\rho=0 . \tag{2c}$$

Taking the boundary to be the plane $z=0$, and assuming for simplicity that $\rho=\rho_0(Z)$, with $Z=z/\lambda_S$,

$$\rho_0=C(\Delta+Z) , \tag{3}$$

where C depends upon the strength of the (distant) source and Δ is to be determined from the boundary conditions. With these assumptions, the photon current density at the boundary $\mathbf{J}_B$ is

$$\mathbf{J}_B=-J_{\text{diff}}\hat{\mathbf{z}} , \tag{4a}$$

$$J_{\text{diff}}=D\frac{\partial\rho}{\partial z}\bigg|_B . \tag{4b}$$

If the sample is a slab of thickness L, and photons are injected with uniform intensity I_0 in the plane $Z=Z_0$, then the photon density ρ_{slab} which satisfies the boundary condition Eq. (1) is

$$\rho_{\text{slab}}=\begin{cases} \dfrac{I_0\lambda_S}{D}\left[\dfrac{L'+\Delta-Z_0}{L'+2\Delta}\right](\Delta+Z) \quad (0\le Z\le Z_0) & \text{(5a)}\\[2ex] \dfrac{I_0\lambda_S}{D}\left[\dfrac{\Delta+Z_0}{L'+2\Delta}\right](L'+\Delta-Z) \quad (Z_0\le Z\le L') , & \text{(5b)}\end{cases}$$

where $L'=L/\lambda_S$.

III. TRANSPORT THEORY

The number of photons dn_S/dt scattered per unit time by a volume element dv is

$$\frac{dn_S}{dt}=\frac{1}{\tau_S}\rho\, dv , \tag{6}$$

so that each volume element acts as a secondary source that emits a total photon flux $f_T=dn_S/dt$ isotropically into 4π. An initial flux f_0 emitted into some particular solid angle is attenuated by scattering. After some distance r, the flux f that still maintains the original direction is $f=f_0\exp(-r/\lambda_S)$. This form is assumed to hold on all length scales, including $r<\lambda_S$, so that implicit in this, the usual form of transport theory, is a continuum white-noise model for the random medium rather than the usually claimed model of discrete pointlike scatteries.

The photon flux $S(\theta)$ escaping from the medium through the surface into a cone of solid angle $d\Omega$ that makes an angle θ with the normal to the boundary may be measured using a collimator constructed from a bundle of long tubes, each of which accepts $d\Omega$. At $\theta=0$, the total cross-sectional area of the colimator equals the sample surface area A. As θ is increased, however, the number of tubes that see the sample decreases as $\cos\theta$, but this loss is exactly compensated for by the $1/\cos\theta$ increase in sample area seen by each of the remaining tubes. Since the transmission coefficient of the surface is $1-R$, in 3D for example, $S(\theta)$ is

$$S(\theta)=(1-R)\frac{A}{\tau_S}\left[\frac{d\Omega}{4\pi}\right]\int_0^\infty dz\,\rho(z)\exp\left[-\frac{z}{\lambda_S\cos\theta}\right] , \tag{7}$$

where in view of the exponential cutoff, the sample depth along the z axis has been extended to infinity. Equation (7) may now be used to develop a boundary condition for ρ for arbitrary surface reflectivity R.

Diffusion theory itself is incapable of providing an expression for $S(\theta)$, since all the theory "knows" about the migration of photons is the diffusive current relationship Eq. (4). Writing the corresponding transport current density $\mathbf{J}_T=-J_{\text{transp}}\hat{\mathbf{z}}$, where in 3D, for example,

$$J_{\text{transp}}=\frac{1}{A}\int_0^{2\pi}d\varphi\int_0^{\pi/2}d\theta\sin\theta\cos\theta S(\theta) , \tag{8}$$

and equating $J_{\text{transp}}=J_{\text{diff}}$ yields a relationship between an integral of ρ and its normal derivative at the boundary that could, in principle, serve as a boundary condition. Of course, such a boundary condition is too difficult to work with in practice, and so we expand the photon density near the boundary in a two-term Taylor series

$$\rho(z)=\rho_B+z\frac{\partial\rho}{\partial z}\bigg|_B , \tag{9}$$

which corresponds to Eq. (3). We then obtain Eq. (1) with

$$\Delta_1=\frac{1+R}{1-R} , \tag{10a}$$

$$\Delta_2=\frac{6\pi-8+8R}{3\pi(1-R)} , \tag{10b}$$

$$\Delta_3=\frac{5+3R}{4(1-R)} . \tag{10c}$$

IV. MILNE THEORY

In the steady state, the rate at which a volume element Δv emits photons equals the rate at which it captures photons radiated by neighboring regions. This leads to an integral equation for the photon density known as the Milne equation [6]. Using Eq. (6), and setting for the moment $R=0$, we may write for 3D, for example,

$$\rho(\mathbf{r})\Delta v=\int_{\text{vol}} d^3r'\rho(\mathbf{r}')\left[\frac{\Delta\Omega(\mathbf{r}'-\mathbf{r})}{4\pi}\right]\exp\left[-\frac{|\mathbf{r}'-\mathbf{r}|}{\lambda_S}\right] , \tag{11a}$$

where the solid angle $\Delta\Omega$ subtended at $\mathbf{r}$ by Δv as seen from the point $\mathbf{r}'$ is $\Delta\Omega=\Delta a/|\mathbf{r}'-\mathbf{r}|^2$, and $\Delta a=\Delta v/\lambda_S$ is an element of area. Assuming as before $\rho=\rho(Z)$ and defining an integration variable $u=1/\cos\theta$, where θ is the polar angle measured from the inward-directed boundary normal, and again extending the sample depth to infinity, we obtain the 3D Milne equation in standard form,

$$\rho(Z)=\tfrac{1}{2}\int_0^\infty dZ'\rho(Z')E_1(|Z'-Z|) , \tag{11b}$$

where

$$E_n(x)=\int_1^\infty \frac{du}{u^n}\exp(-xu) \tag{12}$$

are the generalized exponential integrals [10].

When the surface reflectivity R is nonzero, then in addition to the direct line-of-sight path connecting $\mathbf{r}'$ to $\mathbf{r}$, there may be additional ballistic paths that connect these points via surface reflections. The most convenient model for calculation is a perfectly flat, specularly reflecting surface, for which there is only one additional path connecting $\mathbf{r}'$ to $\mathbf{r}$ that needs be included. As the surface reflectivity R is anyway taken to be an angle-independent average, there is little point in considering surface models of greater complexity, and with a specular surface we obtain the modified Milne equations

$$\rho(Z)=\int_0^\infty dZ'\rho(Z')K_d(Z',Z) , \tag{13}$$

where the kernels K_d depend upon the dimensionality d, and are

$$K_1=\tfrac{1}{2}\{\exp[-|Z'-Z|]+R\exp[-(Z'+Z)]\} , \tag{14a}$$

$$K_2=\frac{1}{\pi}[F_0(|Z'-Z|)+RF_0(Z'+Z)] , \tag{14b}$$

$$K_3=\tfrac{1}{2}[E_1(|Z'-Z|)+RE_1(Z'+Z)] , \tag{14c}$$

with

$$F_n(x)=\int_1^\infty\frac{du}{u^n(u^2-1)^{1/2}}\exp(-xu) . \tag{15}$$

Approximate solutions to Eq. (13) are easily developed by iteration. Since our ultimate purpose is to obtain forms for Δ by matching the diffusion and Milne equation solutions, we chose as our zeroth-order iterate ρ_0 the diffusion solution Eq. (3). Inserting this into the right-hand side of Eq. (13) yields a first iteration

$$\rho_1(Z)=C[\Delta+Z+\epsilon_d(Z;R)] , \tag{16}$$

where

$$\epsilon_1=\tfrac{1}{2}[1+R-\Delta(1-R)]\exp(-Z) , \tag{17a}$$

$$\epsilon_2=\frac{1}{\pi}[(1+R)F_2(Z)-\Delta(1-R)F_1(Z)] , \tag{17b}$$

$$\epsilon_3=\tfrac{1}{2}[(1+R)E_3(Z)-\Delta(1-R)E_2(Z)] . \tag{17c}$$

In 1D, Δ can be chosen such that $\rho_1=\rho_0$ for all Z, in which case ρ_0 is an exact solution to the problem. This is not possible in 2D and in 3D, but for any order n of iteration we can achieve equality on average by choosing Δ such that

$$\int_0^\infty dZ[\rho_n(Z)-\rho_{n-1}(Z)]=0 . \tag{18}$$

Using the exact result for 1D and performing the calculation to second order for 2D and 3D yields

$$\Delta_1(R)=\frac{1+R}{1-R} , \tag{19a}$$

$$\Delta_2(R)=\frac{\pi(1+R)}{4(1-R)}\Phi_2(R) , \tag{19b}$$

$$\Delta_3(R)=\frac{2(1+R)}{3(1-R)}\Phi_3(R) , \tag{19c}$$

where

$$\Phi_2(R)=\frac{1-(2/\pi^2)(1-R)}{1-(1-\pi/4)(1-R)} , \tag{20a}$$

$$\Phi_3(R)=\frac{1-\frac{3}{16}(1-R)}{1-\frac{2}{3}(1-\ln 2)(1-R)} . \tag{20b}$$

Comparing Eqs. (10) and (19) we observe that in 1D transport theory, diffusion theory, and Milne theory are all consistent. In 2D and 3D, however, inconsistencies arise, and the transport values of Δ are always larger than those obtained from Milne theory.

In addition to the value of Δ, the solutions for ρ are also of interest. The second-order solutions are rather complicated and will not be presented, but the first-order solutions are already a good approximation. This may be seen by comparing our 3D result with the exact 3D solution for $R=0$ (the exact 2D solution does not appear to be available). Using the high-accuracy approximation to the exact solution given by Morse and Feshbach [6], we find good agreement between Eq. (17c) and the exact solution for all values of Z down to and including the boundary itself, with the maximum error never exceeding 1.6%. Since our first-order results are exact in 1D, and are a good approximation in 3D, we surmise that they will also be good in 2D. Thus in light of the approximate nature of the calculations that assume scalar waves, a simplified model for the surface, etc., the increased level of complexity involved in going to higher order clearly negates any resulting (largely illusory) increase in accuracy.

Worth noting is that the diffusion theory solutions ρ_0 in 2D and 3D also closely match the Milne-equal solutions to within a small fraction of a mean free path from the boundary. Indeed, for $Z=0.1$ the error is already less than 5%, for $Z=0.2$ the error has dropped to less than 2%, and for large Z the error decays to zero as the functions $E_n(Z)$ and $F_n(Z)$ decrease rapidly with increasing Z.

V. DISCUSSION

Since transport theory and Milne theory yield significantly different values for $\Delta_d(R)$, the question naturally arises as to which provides the better approximation? Milne theory appears preferable since it involves the photon density directly, while transport theory involves the photon currents. There is also another reason for using Eqs. (19) rather than Eqs. (10). One method for experimentally determining the diffusion constant D and hence the mean free path is to measure the diffuse transmittance. In performing such experiments one normally measures the transmitted *intensity,* collecting all or some known fraction of the scattered light. But in comparing the data with theory in order to extract D, one invariably uses an expression for the diffusive *current,* such as may be obtained from Eqs. (4) and (5). Since there is a fundamental difference between intensities and currents, this approach must inevitably lead to an erroneous value for D unless compensated for by the second error. The requisite compensating error is to calculate $\Delta_d(R)$ by equating the diffusive current density J_{diff} to the transport intensity I_{transp}. In 3D, for example, I_{transp} is obtained from $S(\theta)$ as

$$I_{\text{transp}}=\frac{1}{A}\int_0^{2\pi}d\varphi\int_0^{\pi/2}d\theta\sin\theta S(\theta)\ . \tag{21}$$

This procedure recovers Eqs. (19) with the minor correction terms $\Phi_d(R)$ set equal to unity.

It is also of interest to compare our present results with those obtained previously. By summing over all intermediate passes, Freund and Berkovits [3] obtained for the diffuse transmittance

$$T=\frac{T_0}{1-R+2RT_0}\ , \tag{22}$$

where T_0 is the transmittance in the absence of surface reflections. Denoting by Δ_0 the value of Δ for $R=0$, assuming for simplicity $Z_0=1$, and using Eq. (5), Eq. (22) yields

$$T=\frac{1+\Delta_0}{(1-R)L'+2(R+\Delta_0)}\ . \tag{23}$$

Equations (5) and (19) with $\Phi_d(R)$ set equal to unity, on the other hand, yield

$$T=\frac{1+\Delta_0-R(1-\Delta_0)}{(1-R)L'+2(R+\Delta_0)-2R(1-\Delta_0)}\ . \tag{24}$$

As R approaches unity, both Eqs. (23) and (24) pass correctly to the physically required limit $T=\frac{1}{2}$ [3], but only in 1D where $\Delta_0=1$ are they the same for arbitrary R.

The present treatment, in line with previous approaches [1,4], has assumed isotropic (i.e., pointlike) scatterers. For anisotropic scattering, it is well known that diffusion theory is still valid if λ_s is replaced by the transport mean free path $\lambda_T=\lambda_S/\langle 1-\cos\theta\rangle$, where θ is the scattering angle [6]. However, in transport theory [Eq. (7)], or in Milne theory [Eq. (11a)], one may not simply replace λ_S by λ_T, since even for anisotropic scatterers the attenuation of the photon flux is determined by $\exp(-r/\lambda_S)$ and *not* by $\exp(-r/\lambda_T)$. Although one often makes the ansatz that the boundary conditions obtained by considering isotropic scattering hold also in the anisotropic case, this has not been proven, and indeed recent experimental results [9] suggest that there may be significant differences. Worth noting in this respect is that the treatment of surface reflections by Freund and Berkovits [3] is independent of such considerations, and may also be applied to data obtained from computer simulations or from experiment. On the other hand, in keeping with the approach of Zhu, Pine, and Weitz [4], the present approach, though strongly theory dependent, has the great advantage that an expression that neglects surface reflections can be easily modified to include these reflections simply by replacing Δ_0 with $\Delta(R)$.

ACKNOWLEDGMENTS

I am pleased to acknowledge useful discussion with E. Kogan, and the support of the U.S.-Israel Binational Science Foundation (Jerusalem).

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