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Transmission and Surface Intensity Profiles in Random Media.

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Abstract. – We have performed a series of measurements of transmission and surface intensity profiles *vs.* thickness in which the coherent depth of penetration and the surface reflectivities are varied independently. The excellent agreement of diffusion theory which includes interfacial coupling in a natural way allows us to determine the domain of validity of the diffusion model and to obtain the first accurate values for the coherent penetration depth and the optical transport mean free path.

The photon diffusion model provides a framework for describing steady-state electromagnetic propagation in random media [1, 2]. The model has been used to infer the transport mean free path l from a variety of measurements including coherent backscattering [3, 4] and scale dependence of the total transmission [5, 6]. The very confidence that the diffusion theory provides a straightforward description of steady-state transport in random media, however, has led to its consistent misapplication. Transport parameters are generally found by fitting diffusion theory to the results of a single experiment using simplifying assumptions regarding the coupling at the sample interface. But the question remains as to whether these parameters would adequately describe a broader range of independent experiments with the same system without introducing additional parameters. Our goal is first to provide a description of transport in a finite system utilizing the diffusion formalism with a *minimum* number of parameters and then to test the consistency of the model by performing a series of *independent* experiments in which these parameters are overdetermined. The issues raised here are relevant to propagation in any random medium in which questions of the coupling between media exist, as they do, for example, in connection with the contact resistance or impedance mismatch in electronic or acoustic systems, respectively. The need to resolve these questions becomes all the more compelling with the realization that, in the presence of resonances with the microstructure of the sample, the transport mean free path l cannot be determined from measurements of the diffusion coefficient D obtained using time or frequency domain techniques because the ratio of D and l is not constant but itself has a resonant structure [6, 7].

The experiments reported in this letter allow us to parameterize interfacial optical interactions within the framework of the photon diffusion model and to determine the regime of validity of the model. The importance of properly accounting for the randomization of the wave as it enters the sample [8] and for the reflection of the wave reaching the surface from the interior of the sample [9-12] has been emphasized in recent work. In the model presented below, steady-state diffusive transport is described using three interfacial coupling parameters. These parameters are the coherent penetration depth z_p from the surface at which the incident wave is effectively randomized, and the extrapolation lengths z_b beyond the input and output boundaries at which the intensity inside the sample extrapolates to zero. Optical scattering lengths in sintered alumina are determined from a subset of the measurements reported here and are consistent with a set of new experiments which critically test the physical meaning of these scattering lengths. Each of these lengths is varied independently by changing either the index mismatch at the sample surface or the incident angle of the beam. The interfacial scattering parameters in an alumina wedge are found from measurements of the scale dependence of optical transmission in air and in index matching fluid. The surface extrapolation lengths can be expressed in terms of l and the internal reflection coefficients R at the sample's interfaces. These can both be obtained from a comparison with measurements in a sample immersed in index-matching fluid, in which case $R = 0$. Using parameters found from measurements of $T(L)$ we find diffusion theory is in excellent agreement with measurements of the dependence of transmission upon incident angle $T(\theta)$. Such measurements provide a remarkable simple method for determining propagation parameters. A test of the validity of diffusion theory is whether the microscopic intensity distribution on the sample surface can be predicted using these scattering lengths. These results again confirm the adequacy of diffusion theory and make possible a detailed study of the transition from ballistic to diffusive transport. The intensity distribution at a surface [2, 13, 14] is the essential particle aspect of propagation corresponding to interference phenomena in the scattered wave which are observed in coherent backscattering and angular intensity correlation functions [15].

Measurements of $T(L)$ were made on a 0.0182 rad wedge fabricated by polishing a slab of 99.7% purity polycrystalline alumina with 0.97 solid fraction provided by Valley Design Corporation. Electron-micrographs of the material show random grains of average size 2 μm. A 3 mW He-Ne laser beam is focused to a 5 μm spot on the sample surface. Near normal incidence the specular reflection coefficient is measured to be 7.2%. This suggests the effective refractive index for the sample is 1.70 which is close to the index of 1.76 of crystalline alumina. We use an integrating sphere to measure the energy of the incident, transmitted and reflected light. The ratio of the transmitted light to the part of the incident beam which is not specularly reflected from the input surface gives the transmission coefficient. The intensity profile at the sample surfaces *vs.* transverse coordinate $I(\rho)$ is imaged near the normal to the surface with an $f/1.4$, 5.5 cm focal length Nikkon lens. The image is magnified 30 times and is recorded by scanning a photomultiplier tube with an affixed 20 μm aperture in the image plane. The results reported for $I(\rho)$ are incoherent averages of speckle patterns obtained by scanning the sample at fixed thickness as the data is collected.

We consider a simple version of diffusion theory in which boundary reflectivity is taken into account. Inside a slab positioned perpendicular to the z axis between $0 < z < L$ the intensity obeys the diffusion equation

$$\frac{\partial I(\boldsymbol{r}, t)}{\partial t} - D\nabla^2 I(\boldsymbol{r}, t) + \frac{1}{\tau_a} I(\boldsymbol{r}, t) = Q(\boldsymbol{r}, t), \tag{1}$$

where τ_a is the absorption time, $D = (1/3)vl$ is the diffusion coefficient, v is the transport velocity and $Q(\boldsymbol{r}, t)$ is a source function. We consider stationary transport due to a focused beam. We replace the incoming focused coherent flux by a source of isotropic radiation at a point $z = z_p$, $\rho = 0$, with a strength equal to the incident flux. The source function can then be written as $Q(\boldsymbol{r}) = qv\delta(\rho)\delta(z - z_p)/\rho$, where q is the intensity of the point source. Now eq. (1) takes the form

$$\nabla^2 I(\rho, z) - \alpha^2 I(\rho, z) = -\frac{3q}{l}\frac{1}{\rho}\delta(\rho)\delta(z - z_p), \tag{2}$$

where $\alpha = (1/D\tau_a)^{1/2}$ is the absorption coefficient.

Because the source is assumed to lie inside the sample, the only flux coming from a boundary towards the inside of the slab is the reflected part of the internal flux directed toward a boundary. The boundary conditions for eq. (2) can, therefore, be written as [11, 12]

$$\left[\frac{I(\rho, z)}{z_0^{(\mathrm{in})}} - \frac{\mathrm{d}I(\rho, z)}{\mathrm{d}z}\right]_{z=0^+} = 0; \quad \left[\frac{I(\rho, z)}{z_0^{(\mathrm{out})}} + \frac{\mathrm{d}I(\rho, z)}{\mathrm{d}z}\right]_{z=L^-} = 0, \tag{3}$$

where $z_0^{(\mathrm{in,\,out})} = 2l(1 + R^{(\mathrm{in,\,out})})/3(1 - R^{(\mathrm{in,\,out})})$ and $R^{(\mathrm{in,\,out})}$ are reflection coefficients of the input/output surfaces of the slab. Since the index mismatch is the same at the input and output surfaces, we will assume for the sake of simplicity $R^{(\mathrm{in})} = R^{(\mathrm{out})}$ and $z_0^{(\mathrm{in})} = z_0^{(\mathrm{out})}$.

Solving eq. (2) with these boundary conditions gives the intensity distributions on the output surface of the slab,

$$I(\rho, L) = \frac{3qz_0}{l}\int_0^\infty \mathrm{d}\lambda\,\lambda \frac{J_0(\lambda\rho)\{\sinh[k(\lambda)z_p] + z_0 k(\lambda)\cosh[k(\lambda)z_p]\}}{[1 + z_0^2 k^2(\lambda)]\sinh[k(\lambda)L] + 2z_0 k(\lambda)\cosh[k(\lambda)L]}. \tag{4}$$

Here $J_0(x)$ is the Bessel function of zero order and $k^2(\lambda) = \lambda^2 + \alpha^2$.

Integrating the transmitted flux corresponding to the intensity distribution given by eq. (4) gives

$$T(L) = \frac{1}{\alpha z_b}\frac{\sinh[\alpha(z_p + z_b)]\sinh(\alpha z_b)}{\sinh[\alpha(L + 2z_b)]}, \tag{5}$$

where $z_b = \ln[(1 + \alpha z_0)/(1 - \alpha z_0)]/2\alpha$. For the case of weak absorption, $\alpha L \ll 1$ and $\alpha z_b \ll 1$, realized in our experiments, we have $z_b = z_0$. Equation (5) then reduces to

$$T(L) = (z_b + z_p)/(L + 2z_b). \tag{6}$$

We note that in the case of a plane wave incident on a slab of disordered medium z_b corresponds to the length beyond the boundary at which the intensity extrapolates to zero. In the absence of absorption, diffusion theory gives $z_b = 2l/3$, whereas transport theory gives the Milne result, $z_b = 0.7104l$ [1]. This correction is incorporated below.

Measurements of total transmission and reflection *vs.* thickness for the sample in air are shown in fig. 1. Within the experimental error of 1% their sum is unity indicating the low level of absorption in the sample. The inverse of $T(L)$ shown in fig. 1 is plotted in fig. 2. For

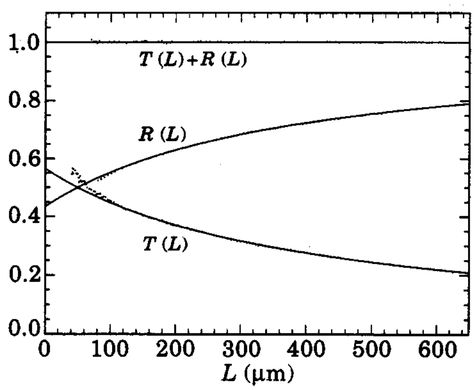

Fig. 1

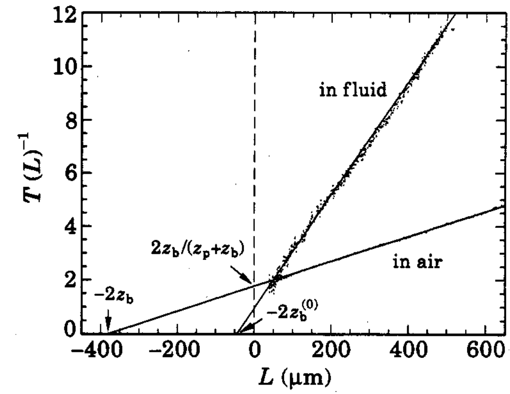

Fig. 2

Fig. 1. – Total transmission and reflection and their sum *vs.* thickness. The solid lines are fits of eq. (6) to the data (····). The reflection data is fitted by $1 - T(L)$.

Fig. 2. – The inverse of transmission in the alumina slab in air and in index-matching fluid. —— theory, ···· data.

$L > 100\,\mu\text{m}$, $T^{-1}(L)$ is a straight line following the prediction of diffusion theory in eq. (6). Fitting eq. (6) to the linear portion of the curve gives $z_p = 24.8\,\mu\text{m}$ and $z_b = 190.9\,\mu\text{m}$ with standard deviation σ, respectively, of $0.1\,\mu\text{m}$ and $0.3\,\mu\text{m}$. From measurements of the variation in intensity readings with the angle at which the laser beam enters the integrating sphere we estimate that relative transmission is uncertain to 0.5%. In addition uncertainties of $1\,\mu\text{m}$ in the thickness of the thinnest part of the sample result in uncertainties of $1\,\mu\text{m}$ in z_p and of $1.5\,\mu\text{m}$ in z_b.

The mean free path can only be determined from the measurement of z_b once the reflectivity is known. We therefore measured the relative transmission with the sample immersed in the index-matching fluid. This eliminated internal reflection giving $R = 0$. These measurements are shown in fig. 2. From the x-intercept to the linear fit of $T^{-1}(L)$ to the data, we find $z_b = (22.3 \pm 1.5)\,\mu\text{m}$. Since $z_b = 0.7104l$ in this case, we find $l = (31.4 \pm 1.5)\,\mu\text{m}$. Using this value for l and the value of z_b in air in the expression for z_b gives $R = 0.81$ at the sample/air interface for diffusing waves. Using the value of z_b to calculate the diffusive angular distribution at the surface gives an angle-averaged Fresnel reflection coefficient of 0.76 which is consistent with the measured value of R.

Scattering parameters may be obtained independently of measurements of the angular dependence of total transmission. Within the framework of the present simple diffusion model, we expect that the longitudinal penetration depth varies with angle of incidence θ as $z_p \cos\theta_r$, where θ_r is the angle of refraction in the random medium, whereas the surface extrapolation length remains the same. We then have

$$T(\theta) = (z_p \cos\theta_r + z_p^{(\text{in})})/(L + z_b^{(\text{in})} + z_b^{(\text{out})})\,. \tag{7}$$

A measurement of $T(\theta)$ for the sample in which the input surface is index matched and the output is in air is shown in fig. 3. In this case $\theta = \theta_r$. The solid line is $T(\theta)$ defined by eq. (7) using values of z_p and $z_b^{(\text{in})} = z_b$ found from measurements of $T(L)$. The close agreement of experimental and theoretical curves is confirmation of the uniqueness of the parameters which appear in the present theory.

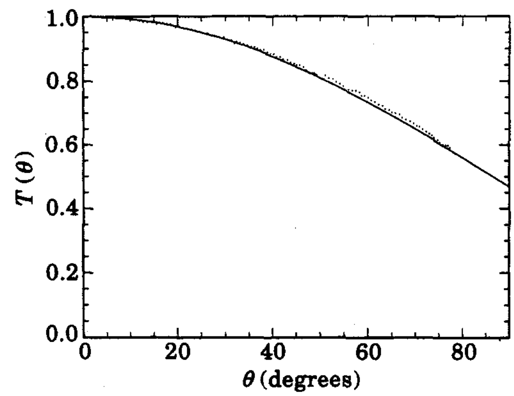

Fig. 3

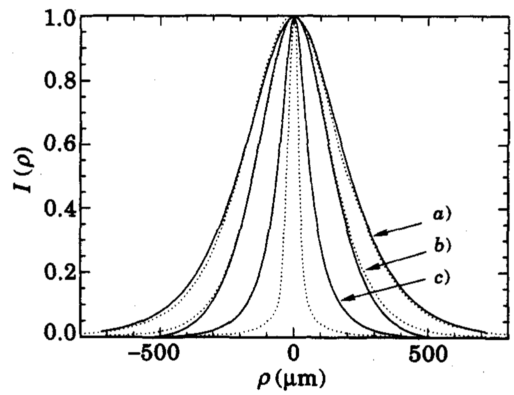

Fig. 4

Fig. 3. – Normalized transmission *vs.* incident angle. The solid line is calculated from eq. (7). —— theory, ···· data.

Fig. 4. – Intensity profiles on the output surface. *a*) $L = 280\,\mu\text{m}$ in air, *b*) $L = 280\,\mu\text{m}$ in fluid, and *c*) $L = 89\,\mu\text{m}$ in air. The solid lines are calculated from eq. (4) with parameters obtained from the fit shown in fig. 2; ···· data.

An independent test of the adequacy of diffusion theory and the accuracy of values of the scattering parameters is whether the parameters found in measurements of integrated transmission can be used to predict the intensity distributions at the output surface. The normalized intensity distribution for different thicknesses in air and in index-matching fluid is shown in fig. 4. Intensity measurements are shown along a line going through the centre of the distribution which is taken as the origin in the figure. For the sample in air we find good agreement with diffusion theory whenever $L \geqslant 150\,\mu\text{m}$. In this limit we find that the measured angular intensity correlation function with rotation of the sample is the square amplitude of the Fourier transform of $I(\rho)$ illustrating that wave aspects of propagation are also determined by photon diffusion. At smaller length scales, diffusion fails to describe $I(\rho; L)$ even though measurements of $T(L)$ are in accordance with diffusion theory.

In conclusion, we have demonstrated that diffusion theory can *quantitatively* describe a broad array of independent optical measurements of integrated and local intensity using three interfacial parameters. These results allow us to determine the range of validity of diffusion theory and accurately determine l and z_p without making any sample-specific assumptions.

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