Optical bandpass characteristics of reflectance from random media

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Experiments show that the optical reflectance of a random medium can have a nonmonotonic dependence on wavelength even when its absorption spectrum is flat and its scattering coefficient decreases monotonically with wavelength. Measured at a particular radial distance from the illumination point, the reflectance is almost independent of wavelength over a wide band. The peak wavelength of the reflectance increases as the optical distance to the detector increases. We explain that this behavior is a consequence of the interplay between direct backscattered and diffuse photon fluxes, whose magnitudes at the surface of the medium have opposite dependencies on the mean scattering length. [S1063-651X(98)00805-8]

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During the past decade the study of optical propagation in random media has uncovered a number of intriguing physical phenomena related to the multiple scattering of photons through paths of different lengths. Several of these phenomena, which include weak localization [1], polarization preservation [2], and the optical memory effect [3,4], demonstrate that the wave nature of light can reveal itself even after the light has scattered multiple times. At the boundary between ballistic and diffusive propagation of photons, in the so-called quasiballistic region, lies one of the most fruitful but least well understood areas in the investigation of the optical physics of random media. The transition between ballistic and diffusive propagation is abrupt and depends critically on the mean scattering length ℓ_s in relation to the distance between the points of illumination and detection [5]. In recent studies, researchers discovered that the sensitivity of the reflectance of a random medium to small variations in ℓ_s falls to zero at a particular distance away from the point of illumination [6,7]. The insensitivity has been attributed to a balance between the ballistic and diffuse fluxes of photons that reach the surface from a secondary source set up below the surface of the medium by the incident beam [6].

This paper describes a related phenomenon that arises from the wavelength dependence of the distribution of path lengths of photons reflected from a random medium. We present experimental results that illustrate the peculiar characteristics of reflectance spectra measured at short optical distances from a white-light source. The shapes of the spectra are found to be extraordinarily sensitive to the mean scattering length in the medium. To explain the results, we give a theoretical argument based on a modified photon-diffusion model.

The scattering samples used in the experiments consisted of a suspension of either 0.2- or 1.0-μm-diam polystyrene microspheres in water containing a black carbon-based ink. According to numerical calculations based on Mie theory, the scattering cross sections of the spheres decrease monotonically according to a power law, $\sigma_s^{0.2\,\mu\mathrm{m}} \sim \lambda^{-2.75}$ and $\sigma_s^{1.0\,\mu\mathrm{m}} \sim \lambda^{-1.06}$ for $400 \leqslant \lambda \leqslant 800$ nm. Diluted in water, the ink has an absorption coefficient that is nearly independent of wavelength, but its transmittance decreases slightly with wavelength because the microscopic carbon particles in the ink behave as weak Rayleigh scatterers. In these experiments, the extra scattering contributed by the ink was negligible compared to that of the polystyrene particles. To measure the reflectance $R(\lambda)$ of a sample, we placed two 600-μm-diam optical fibers in light contact with the surface of the liquid. The source fiber was fixed in position and the detection fiber was attached to a translation stage for precise adjustment of the center-to-center distance r_0 between the fibers. The light source was a tungsten lamp filtered by a step-scanning monochromator with a passband of about 10 nm. The intensity of the light that scattered back to the surface into the detection fiber was measured by a photomultiplier tube which was connected to a lock-in amplifier synchronized with a light chopper at the input of the monochromator. To convert the intensities measured at different wavelengths to normalized reflectance spectra, the intensities were divided by the reflectances of a calibrated diffuse-reflectance standard.

A set of reflectance spectra measured from suspensions of 0.2-μm-diam spheres and ink is shown in Fig. 1 for different fiber separations r_0. The particle and ink concentrations in these suspensions were adjusted to obtain an absorption coefficient $\mu_a = 0.04\ \mathrm{mm}^{-1}$ and a mean transport-corrected scattering length of $\ell_s^* = 2$ mm [Figs. 1(a) and 1(b)] or $\ell_s^* = 1$ mm [Figs. 1(c) and 1(d)] at $\lambda = 800$ nm. A remarkable characteristic of this set of spectra is the complete reversal of the slopes of the spectra measured at the largest and smallest fiber separations. For $r_0 = 1$ mm, the steep *decrease* in reflectance with wavelength throughout most of the band contrasts with the steep *increase* with wavelength in reflectance for $r_0 = 4$ mm. Although it takes place over a narrow range of fiber separations, the transition is smooth between the two extremes. In the intermediate range, $r_0 = 2-4$ mm, the reflectance increases at shorter wavelengths, becomes almost flat, and then decreases again at long wavelengths. This non-

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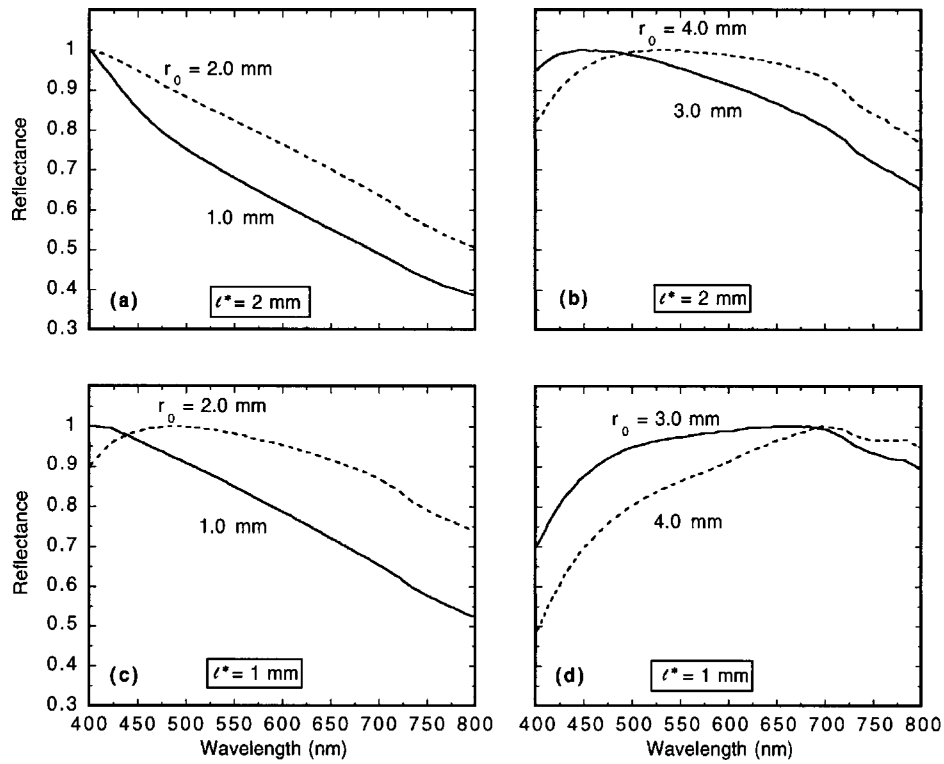

FIG. 1. Reflectance spectra measured from suspensions of 0.2-μm-diam microspheres at different source detector separations r_0. The values of ℓ_s^* calculated at $\lambda = 800$ nm are shown at the bottom of each plot. The spectra were normalized to their peak values.

monotonic behavior appears to be at odds with the optical attenuation that one would expect in a medium containing particles whose cross sections decline monotonically with wavelength. Comparing Figs. 2(a) and 2(b) with Figs. 2(b) and 2(c), we see that halving the particle concentration to increase ℓ_s^* from 1 to 2 mm caused the peaks of the reflectance spectra measured at a given r_0 to shift to longer wavelengths. We found that the shapes of the spectra measured for $\ell_s^* = 1$ mm at a given probe separation resemble those of the spectra measured for $\ell_s^* = 2$ mm at about twice the probe separation, which suggests that ℓ_s^* scaled the effective optical distance between the points of illumination and detection without altering the underlying scattering mechanism.

To examine the relationship between $R(\lambda)$ and the wavelength dependence of the scattering cross sections of the spheres, we repeated the experiments using suspensions of 1.0-μm spheres with their concentrations adjusted to give the

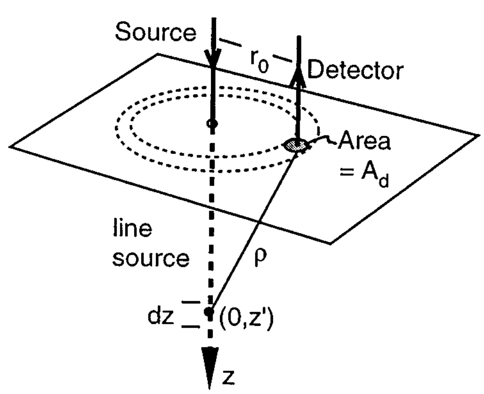

FIG. 2. Geometry of the modified diffusion model.

same magnitudes of ℓ_s^*. The same general changes in the shapes of the reflectance spectra at different probe separations were found, with the exception that the slopes of the spectra were flat over a wider band of wavelengths. The weaker wavelength dependence of the cross sections of the 1.0-μm spheres broadened the effective passband. For $r_0 = 2$ mm, the separation distance that gave the flattest spectrum, the measured reflectance varied less than 5% over the 400–800-nm band.

Our experimental findings can be explained by considering a simplified model of a semi-infinite medium illuminated by a narrow collimated beam (Fig. 2). We assume that the medium is composed of homogeneous random distribution of particles that scatter photons isotropically and that the absorption takes place in the spaces between the particles. The photons that leave the medium through a small area A_d are collected by a detector located a radial distance r_0 from the incident beam. We first consider the limit in which the scattered photons can reach the detector only via a diffusion-like process. In the diffusion approximation, the equation describing the steady-state density of photons in the interior of the medium is [8]

$$-D\nabla^2\phi(r,z) + \mu_a\phi(r,z) = S(r,z), \tag{1}$$

where D is the diffusion coefficient, which is defined in terms of the transport mean free path as $D = \ell_s^*/3$, and μ_a is the absorption coefficient which equals the reciprocal of the average length that photons travel in the medium before absorption. $S(r,z)$ is a function that represents the source of the diffused photons. Although the diffusion equation is not strictly valid at the boundary of the medium, a previous

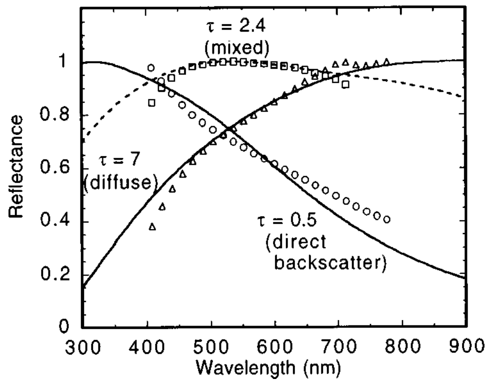

FIG. 3. Comparison of the measured and predicted shapes of the reflectance spectra in three scattering regions characterized by the optical distance $\tau = r_0/\ell_s$.

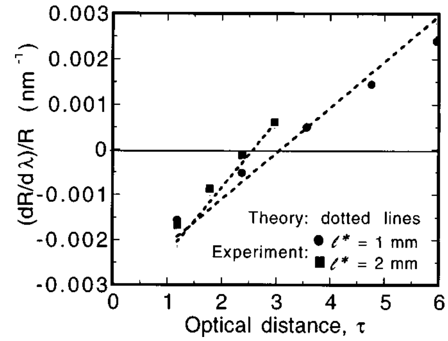

FIG. 4. Spectral slopes obtained by averaging the normalized derivatives of the center portion of the measured and predicted reflectance spectra ($500 \leqslant \lambda \leqslant 700$ nm) in Fig. 1. The optical distance is the unitless quantity $\tau = r_0/\ell_s$.

study [9] has shown that Eq. (1) gives a good estimate of the flux of photons directed out of a bounded medium if $S(r,z)$ is treated as an embedded point source located at a fixed distance below the surface and an equivalent negative source (photon sink) located the same distance above the surface. Here we go a step closer to reality and assume that the incident beam creates an exponentially distributed line source as it penetrates the medium and write the intensity of the light scattered into the detector as

$$I_d = A_d I_0 \int_0^\infty S_0(0,z') G(r_0,0;0,z') dz', \tag{2}$$

where I_0 is the incident intensity, $S_0(0,z') = \mu_s \exp[-(\mu_s + \mu_a)z']\delta(r-r_0)\delta(z-z')$ is the embedded source function, $\mu_s = 1/\ell_s$ is the single-scatter coefficient, and $G(r_0,0;0,z')$ is the transmission function that specifies the attenuation that occurs between a point on the source line and the detector. For the isotropically scattering medium assumed here, the source function has a simple form because ℓ_s and ℓ_s^* are identical, but for a medium composed of particles that scatter light preferentially in the forward direction, backward and forward scattering would need to be distinguished. Under the assumed conditions, the purely diffusive component of $G(r_0,0;0,z')$ is given by the solution of Eq. (1) for the point source S_0 and its image. However, direct backscatter from the source to the detector must also be taken into account for small values of r_0. To account for both propagation mechanisms, we approximate $G(r_0,0;0,z')$ as a sum of ballistic and diffusive propagators,

$$\begin{aligned} G(r_0,0;0,z') &= G_b + \{1-\exp[-(\mu_s+\mu_a)\rho]\} G_d \\ &= \frac{\exp[-(\mu_s+\mu_a)\rho]}{4\pi\rho^2} \\ &\quad + \{1-\exp[-(\mu_s+\mu_a)\rho]\} \frac{z'}{2\pi\rho^2} \\ &\quad \times [(\alpha+1/\rho)\exp(-\alpha\rho)], \end{aligned} \tag{3}$$

where $\rho = \sqrt{r_0^2 + z'^2}$ is the distance from the embedded point source to the detector (Fig. 2) and $\alpha = \sqrt{\mu_a/D}$ is the effective attenuation coefficient for the diffusive component of the photon flux. Combined with Eq. (3), Eq. (2) describes the essential characteristics of the wavelength dependence of the reflectance, $R(\lambda) = I_d(\lambda)/I_0$. In the limit $\exp(-\mu_s\rho) \ll 1$ in which the contribution of the directly backscattered flux is negligible, $R(\lambda) \sim \exp[-\alpha(\lambda) r_0]$. Since $\alpha(\lambda) = \sqrt{3\mu_a/\ell_s(\lambda)}$ for a medium that scatters isotropically, it follows that $R(\lambda)$ is a monotonically increasing function of the mean scattering length. In the opposite limit, $\exp(-\mu_s\rho) \to 1$, the directly backscattered light dominates and $R(\lambda) \sim A_d/\rho\ell_s$. Therefore, in contrast to its behavior in the diffusive region, $R(\lambda)$ in this limit has a monotonically decreasing dependence on the mean scattering length. These opposing tendencies lie at the heart of a general explanation of the nonmonotonic wavelength dependence of reflectance from a random medium: When the increase in the scattering cross sections of the particles with wavelength is sufficiently steep, light propagation at short wavelengths is predominately diffusive and $R(\lambda)$ increases with wavelength as ℓ_s increases. However, as ℓ_s increases further, $R(\lambda)$ eventually flattens and then begins to decrease as direct backscatter becomes more likely. With the direct backscatter dominant, $R(\lambda)$ continues to decrease with wavelength as the embedded source moves deeper into the medium away from the detector. This behavior can be observed only when r_0 is adjusted to approximately balance the contributions of the ballistic and diffuse components of the total flux that reaches the detector from the embedded source.

The shapes of the reflectance spectra predicted by the modified diffusion model are illustrated in Fig. 3 for the direct backscatter, mixed, and diffusive scattering regions. To permit comparison of theory and experiment, data from Fig. 1 are replotted in this figure and the spectral slopes $(dR/d\lambda)/R$, calculated from the spectra in Fig. 3 as averages over the central portion of the band, are plotted in Fig. 4 versus the unitless optical distance $\tau = r_0/\ell_s$. The measurements correspond well with the predictions of the model. Both show a reversal in the spectral slope in the quasiballistic region, $2 < \tau < 3$, with a bias toward longer optical distances with increasing particle concentration. We attribute the discrepancies between the shapes of the predicted and measured spectra to the simplifying assumptions that underlie the derivation of Eq. (2). In particular, the modified diffusion theory does not account properly for multiple forward scattering, nor does it account for the finite width and numerical aperture of the source and detector fibers. The influence of these variables is greatest for small values of r_0. Nevertheless, the modified diffusion model appears to explain the main features of the observed spectra and their dependence on ℓ_s^* and r_0.

In summary, we have studied and explained the characteristics of the reflectance of a semi-infinite random medium measured close to the point of illumination. We found that the wavelength at which the reflectance peaks at a given source-detector separation r_0 shifts in a predictable manner toward longer wavelengths as the mean scattering length in the medium decreases. A particular r_0 exists at which the spectral slope is flattest within a given band of wavelengths. We anticipate that similar effects can also be observed for off-axis transmission through random media and for acoustical and other types of waves. These findings have practical implications for spectroscopy of optically thick materials and optical communications through turbid medium.

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