When Does the Diffusion Approximation Fail to Describe Photon Transport in Random Media?

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The transport of photons through a slab of random medium is shown to deviate from the diffusion approximation when z/l_t is small, where z is the thickness of the slab and l_t is the transport mean free path. When $z/l_t=10$ and $z=10$ mm, the average time of arrival is about 0.9 times that predicted by diffusion theory. Photons are found to arrive earlier than that predicted by the diffusion theory as z/l_t becomes smaller or the anisotropic scattering increases.

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Diffusion theory is widely used to describe the transport of particles and waves in random media. The analytical solutions for the diffusion equation are generally available and are found to be valid[1,2] when $z/l_t \gg 1$, where z is the thickness of the random medium and l_t is the transport mean free path. For many physical media the particle or wave transport often occurs in the region where z/l_t is small. This is particularly true for the ultrasmall electronic devices of nanometer size, where electrons traverse only a few transport mean free paths. Recently, the diffusion approximation has been extensively used in many of the pioneering light-scattering studies[2-8] and in applications of light scattering to the study of physical and biological media.[9-11] It is important to know at what value of z/l_t the diffusion approximation begins to fail. In this Letter, the transport of photons through a slab of random medium is *directly* studied using ultrafast laser pulses and time-resolved detection. Here, we provide the first direct insight on when and how the diffusion approximation deviates from the actual transport of photons through a slab of random medium. Our results resolve the current controversy[9-11] on whether or not photon transport is diffusive after passing through a slab of random medium of only a few transport mean free paths thick. Weitz *et al.* using a diffusing-wave spectroscopy[10] technique showed that photons are diffusive after propagating through a slab of few transport mean free paths thick. However, the results of Freund, Kaveh, and Rosenbluh[11] using a similar technique gave a contrary conclusion in which they claimed the possibility of ballistic transport. Our time-resolved studies show that the prediction of the diffusion approximation deviates monotonically from the measured scattered pulse as the value of z/l_t decreases below 10. The scattered photons are found to arrive much earlier than predicted by the diffusion approximation. Ballistic photon transmission has been shown to exist in a small-diameter random-bead medium.[12]

A direct method to probe the transport of photons in a random medium is to measure the distribution of photon arrivals in the time domain. When a plane front ultrafast pulse is incident normally onto a slab of random medium, the temporal distribution of photon arrivals at a point on the opposite side of the slab is predicted by the diffusion theory as

$$I_z(t)=\frac{D}{\pi z^2}\sum_{m=1}^{\infty} m\left(\frac{\pi z}{d}\right)^2 \sin\left(\frac{m\pi z}{d}\right) \times\exp\left[-Dt\left(\frac{m\pi}{d}\right)^2\right], \qquad (1)$$

where $D=vl_t/3$ is the diffusion coefficient, $d=z+2z_0$, $z_0=0.71l_t$, v is the speed of photons, and z and l_t have been defined earlier.

Our experimental arrangement is shown schematically in Fig. 1 which closely imitates the geometry governed by Eq. (1). Ultrafast laser pulses of 100 fs were generated at a repetition rate of 82 MHz by a colliding pulse mode-locked dye-laser system. The laser power was 10

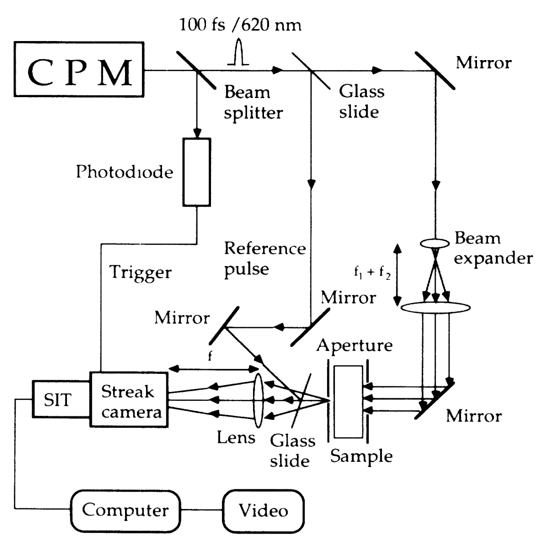

FIG. 1. Schematic diagram of the experimental setup.

mW at a wavelength centered at 620 nm. The laser beam of 4 mm diam was split into two beams: a reference beam to mark the zero time of the signal beam which was expanded to 35 mm diam. The central portion of the expanded laser beam was selected by a diaphragm and passed through the random medium consisting of latex beads suspended in water contained in a cylindrical glass cell of diameter 50 mm and of thickness 10 mm. Photons scattered out of the cell will be lost, i.e., both sides of the cell resemble an absorbing boundary condition. A black pinhole of 2 mm diam was placed at the center on the opposite side of the cell. The temporal distribution of the photons scattered out from this pin hole in the forward direction at an angle of 10 mrad was measured by a synchroscan streak camera. The diameter of the incident beam was set to 20 mm; any further increase in the beam diameter will not change the temporal profile of the scattered photons. The measured scattered pulse profile can be described by Eq. (1), if the diffusion approximation is valid.

Experimental studies were carried out for different bead diameters, each at a series of different concentrations. For highly concentrated random media, where $z/l_t \gg 1$, the diffusion equation holds, which was not surprising. In this case, the transport mean free path was obtained by fitting the scattered pulse profile by Eq. (1). Figure 2(a) illustrates an example where the diffusion theory fits the experimental data. The photon transport mean free path l_t was found to be 0.309 mm which is close to the value of 0.25 mm computed from Mie theory using $l_t^m = 1/n\sigma_m$, where n is the number density and σ_m is the momentum-transfer scattering cross

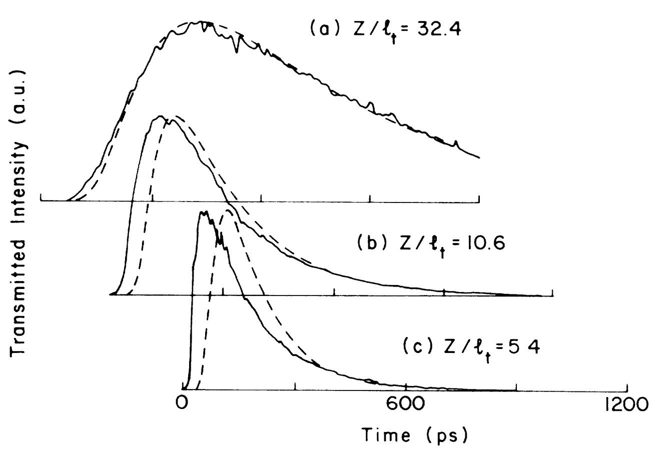

FIG. 2. Transmitted pulse profiles of 10-mm-thick random media consisting of latex beads of diameter 0.296 μm. The bead concentrations n, l_t, and z/l_t are, respectively, (a) 6.65 $\times 10^8$ mm^{-3}, 0.309 mm, and 32.4; (b) 2.18×10^8 mm^{-3}, 0.941 mm, and 10.6; and (c) 1.10×10^8 mm^{-3}, 1.853 mm, and 5.4. l_t is computed from Eq. (2). Solid curves are the experimental results and dashed curves are computed from the diffusion theory where their peak intensity is normalized.

section.

The following procedure is used to determine when and how the diffusion theory fails to describe photon transport when z/l_t is decreased. The transport mean free path l_t for a smaller particle concentration (n) medium is extrapolated from a larger particle concentration (n') with known transport mean free path l_t' by

$$l_t = l_t' n'/n\,. \tag{2}$$

The scattered pulse profiles for various values of l_t are computed from the diffusion theory [Eq. (1)]. These computed pulse profiles are then compared with the corresponding experimental results at the same number density.

The relationship of Eq. (2) is valid if bead arrangements in the media are not correlated in either short or long range. The short-range (nearest-neighbor) particle-particle correlation[13-15] can strongly reduce the scattering of light when there is more than one bead in the volume of λ^3, where λ is the wavelength of the light in the medium. The effect of short-range correlation can be neglected in our media because there is less than one bead in unit volume λ^3. There is negligible long-range particle correlation[13] in the media used because of (1) the distance between beads are large (low-bead concentration), (2) large-bead size, and (3) the presence of ions in the water. These conditions reduce the effective Coulomb interaction and thus the long-range correlation among the beads is negligible. The medium are homogenized in an ultrasonic bath so that there is a negligible amount of bead aggregation. The relationship of Eq. (2) has been verified by us and others[14,16] in the dilution studies of highly concentrated media (but with less than one particle per λ^3).

Figures 2 and 3 display a series of transmitted pulse profiles for media with decreasing bead concentrations for bead diameters of 0.296 and 3.134 μm, respectively. Figures 2(a) and 3(a) show that the experimental (solid) curves can be fitted by the diffusion theory (dashed curves) where z/l_t is large as indicated in the figures. Figures 2(b) and 3(b), where $z/l_t \approx 10$, clearly show deviations. The measured photons arrive earlier than that predicted by the diffusion approximation. The peaks of the theoretical (dashed) computed curves are normalized to the peak of the experimental data, whereas the dash-dotted curves are normalized to the total intensity of the experimental results. None of the theoretical plots from the diffusion approximation agrees with the experimental results at early time when $z/l_t < 10$.

The temporal distribution of photons for anisotropic scattering media and small z/l_t can differ significantly from the prediction of the diffusion approximation. To illustrate this point, the solid curve in Fig. 3(d), where $z/l_t = 2.8$, shows that photons arrive substantially earlier than predicted. These plots show that more photons arrive at early times. At later times ≈ 300 ps, the theoret-

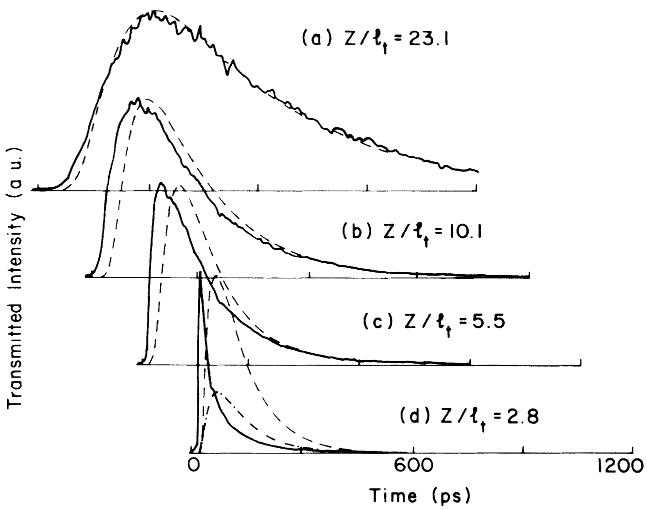

FIG. 3. Transmitted pulse profiles of 10-mm-thick random media consisting of latex beads of diameter 3.134 μm. The bead concentrations n, l_t, and z/l_t are, respectively, (a) 9.62 $\times 10^5$ mm^{-3}, 0.433 mm, and 23.1; (b) 4.20×10^5 mm^{-3}, 0.989 mm, and 10.1; (c) 2.28×10^5 mm^{-3}, 1.823 mm, and 5.5; and (d) 1.16×10^5 mm^{-3}, 3.578 mm, and 2.8. Solid curves trace the experimental results. Dashed and dash-dotted curves are computed from the diffusion theory where the peak and total intensity are normalized to the experimental curves, respectively.

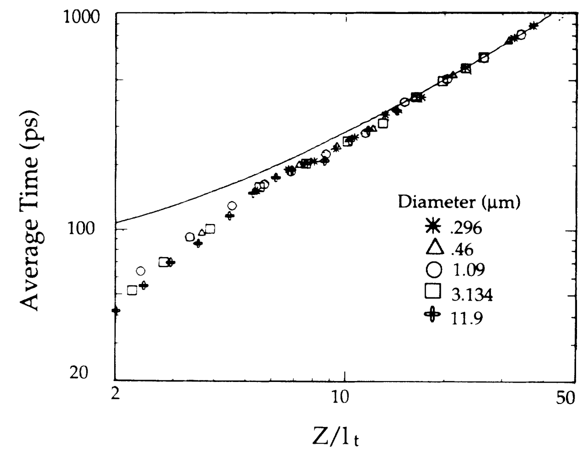

FIG. 4. Plots of average time of arrival $\bar{t}$ vs z/l_t. The solid curve is the $\bar{t}$ predicted by diffusion theory, and the dotted curve is $0.9\bar{t}$. Experimental results (corresponding bead diameter) are plotted by stars (0.296 μm), triangles (0.46 μm), circles (1.09 μm), squares (3.134 μm), and plusses (11.9 μm).

ical (dash-dotted curve) and experimental curves merge.

A quantitative indication where the diffusion approximation breaks down is the average time of photon arrival. The average arrival time is computed by

$$\bar{t}=\int_0^\infty dt'\,t'I(t')\Big/\int_0^\infty dt'\,I(t')\,, \tag{3}$$

where $I(t')$ is the temporal distribution of the scattered pulse measured experimentally. The theoretical prediction of the average arrival time can be obtained exactly from Eq. (1) as

$$\bar{t}=\frac{1}{2}\frac{z(z+4z_0)}{vl_t}\,. \tag{4}$$

The average time of arrival $\bar{t}$ as a function of z/l_t predicted by Eq. (4) is plotted by the solid curve in Fig. 4. The average time of arrival of the experimentally measured photons are computed from Eq. (3). The results from random media of beads with diameters $d=0.296$, 0.46, 1.09, 3.134, and 11.9 μm are represented by plots of stars, triangles, circles, squares, and plusses, respectively, in Fig. 4. The dotted curve is obtained by multiplying Eq. (4) by 0.9 to indicate where the experimental data deviate by more than 10%. The data displayed in Fig. 4 clearly demonstrate that the average time of arrival deviates by 10% when $z/l_t\approx 10$. The deviation increases as z/l_t is reduced further below 10 and many more photons arrive earlier than predicted by diffusion theory. Beads of larger diameter will increase the amount of light scattered in the forward direction. This anisotropic scattering will result in reducing the average arrival time of the photons, which is apparent in Fig. 4 when z/l_t is less than 5. For a given value of z/l_t (<5), the largest-bead-diameter (11.9-μm) medium (plusses) shows the smallest average arrival time, while the smallest-bead-diameter (1.09-μm) medium (circles) shows the largest average arrival time, and the intermediate-bead-diameter (3.134-μm) medium (squares) shows that its average arrival time falls between that of the 11.9- and 1.09-μm beads. Anisotropic scattering as well as coherent interference among the multiple-scattered waves may account for the earlier arrival of the photons.

In conclusion, the temporal distribution of photon arrival after propagating through a slab of an anisotropic scattering random medium is shown to deviate from the diffusion approximation when z/l_t is small. When $z/l_t=10$ and $a=10$ mm, the average time of arrival of the photon is about 0.9 times that predicted by diffusion theory. The photons are found to arrive increasingly earlier as z/l_t is further decreased or anisotropy of the scattering increases.

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