Dynamic Multiple Scattering: Ballistic Photons and the Breakdown of the Photon-Diffusion Approximation

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We find experimentally that in the forward direction the length scale of quasiballistic photons dominates the dynamic intensity-intensity correlation function of light multiply scattered from randomly diffusing spheres. This implies a breakdown of the photon-diffusion approximation, which is characterized by a single length, the transport mean free path. We suggest why this breakdown occurs, introduce phenomenologically a dynamic correlation length as an additional length scale, and with this extension reconcile the theory with experiment.

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We have measured the frequency spectrum of light strongly multiply scattered from a system of 0.46-μm spheres undergoing random diffusive motion, and find that in the forward direction the spectrum is a simple Lorentzian with a half-width that scales linearly with sample thickness at the rate of 56 kHz/mm. Although these results may appear perfectly innocuous, they represent, in fact, a major surprise which is at complete variance with the almost universally used photon-diffusion approximation[1-16] for light propagation in highly random media, an area currently of intense interest.[1-20] The photon-diffusion approximation, which yields correct results for static correlations,[13] fails in every respect for our dynamic system, since it predicts the wrong shape for the frequency spectrum, the wrong value for the half-width, and the wrong dependence on sample thickness.

In the photon-diffusion approximation, which is characterized by a single length scale, the transport mean free path l, multiply scattered photon trajectories are assumed to be purely diffusive in nature. Photons traversing the sample trace out long tortuous paths, with the mean trajectory length scaling as the *square* of the sample thickness s. For a static scattering system this leads to the experimentally verified[11] prediction[13] that in the forward direction the half-width of the wavelength-dependent intensity correlation function, $C(\Delta\lambda)$, scales as $1/s^2$. In contrast, our experimental results for a dynamic system show that in the forward direction the half-width of the time-dependent intensity correlation function, $C(\Delta t)$, varies as $1/s$, and that $C(\Delta t)$ is dominated by a short, quasiballistic length which scales *linearly* with sample thickness. This departure from s^2 scaling heralds the essential failure of the photon-diffusion approximation when applied to *dynamic* systems.

In Fig. 1 we display in detail the predictions of the photon-diffusion approximation for $C(\Delta t)$. The curves calculated for (curve a) a point source in an infinite medium and for (curve b) a finite slab are obtained by an extension of our previous theory[10] (carried out in real space), which yields results in complete agreement with the most recent diagrammatic treatment of Stephen.[21] Within the framework of the photon-diffusion approximation, this latter is the most rigorous and complete theory currently available. Also shown for comparison are (curve d) our previous results for backscattering[10] (both theoretical and experimental), and (curve c) our present experiments for forward scattering. Since the dephasing time of a single photon trajectory, or loop, varies inversely with loop length,[10,22] $C(\Delta t)$ for a point source in an infinite medium decays extremely rapidly as a result of the presence of many very long loops. The boundary conditions for a slab of finite thickness cut off these long loops and lead to a much slower rate of decay for $C(\Delta t)$. Since only short loops are important for backscattering, on the scale of Fig. 1, $C(\Delta t)$ for backscattering decays extremely slowly. Our present experi-

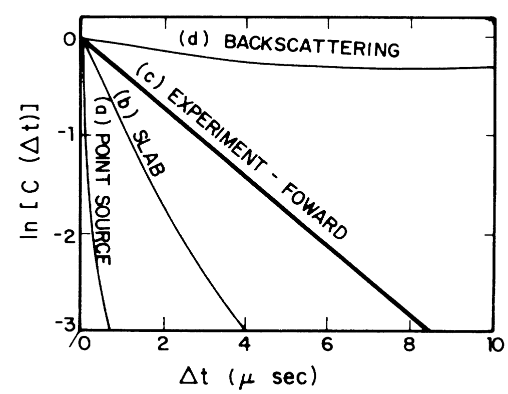

FIG. 1. Natural logarithm of the dynamic intensity-intensity correlation function $C(\Delta t)$ vs time Δt for $s = 1$ mm. Curve a: Forward scattering calculated for a point source in an infinite medium, with s the source-receiver separation. Curve b: Forward scattering calculated for a finite slab of thickness s. Curve c: Our experimental results for a 1-mm-thick cell. Curve d: Backscattering, for which both experiment and theory agree.

mental results (curve c) show that for forward scattering $C(\Delta t)$ decays much more slowly than is predicted even for the slab, leading to the conclusion that *there exists an additional mechanism for damping out the contributions of most of the remaining long loops.* Moreover, the fact that our experiments indicate that $C(\Delta t)$ is well described by a simple exponential implies the existence of a single characteristic length which dominates the time dependence, a length which our data show scales linearly with sample thickness s. As mentioned, none of these new experimental findings are in accord with the photon-diffusion approximation, indicating clearly the need for theories that go beyond this framework.

Our samples were a 10% (by volume) sonicated aqueous suspension of 0.46-μm-diam polystyrene spheres.[23] Such spheres have recently been used in a wide variety of multiple-scattering experiments.[1,4–7,9,10,22] The frequency spectrum of Ar-ion laser light scattered in the forward direction was measured between 100 Hz and 100 MHz with the homodyne technique.[24] The data are shown in Fig. 2, where the solid lines are best-fit Lorentzians. In Fig. 2(b) we plot the inverse of the intensity as a function of the square of the frequency shift. Such a plot is the classic test of a Lorentzian line shape, and as may be seen, all thicknesses display this line shape to high accuracy. In Fig. 3 we plot the measured half-width of the frequency spectrum as a function of sample thickness s. Within error, the data lie on a straight line of slope 56 kHz/mm. Since the frequency spectrum in a homodyne measurement is the Fourier transform of the dynamic intensity-intensity correlation function $C(\Delta t)$, the observed Lorentzian line shapes indicate that over the range studied, $0<\Delta t<3\tau$, $C(\Delta t)$ is well approximated by a simple exponential,

$$C(\Delta t)=\exp(-\Delta t/\tau), \tag{1a}$$

with a multiple-scattering correlation time τ that varies inversely with sample thickness, which we write as

$$\tau=A\tau_1(l/s). \tag{1b}$$

Here A is a constant, and τ_1 is the single-scattering correlation time averaged over all directions of scattering. Using the average of values previously determined experimentally, $l=19$ μm (Ref. 6) and $\tau_1=1300$ μsec,[10,22] we find $A=0.11$.

We have also determined experimentally that neither absorption nor the finite longitudinal coherence length of our laser causes the observed cutoff of long loops. The transmission of the sample was found to decrease as $\alpha l/s$, with α approximately equal to unity, exactly as is expected[3,11] for normal energy diffusion without absorption. From these transmission measurements, and from measurements of the laser linewidth, lower limits were found for the intrinsic absorption length of the sample and the laser coherence length which were both almost 2 orders of magnitude longer than the characteristic length which dominates $C(\Delta t)$. We emphasize that normal diffusive energy transport is a consequence of the fact that the velocity of light is very much greater than the particle velocities, so that as far as energy transport is concerned, one does not expect significant differences between a dynamic and a static system. Accordingly, the transmission teaches nothing about the time evolution of $C(\Delta t)$, other than that it is unaffected by absorption. Having

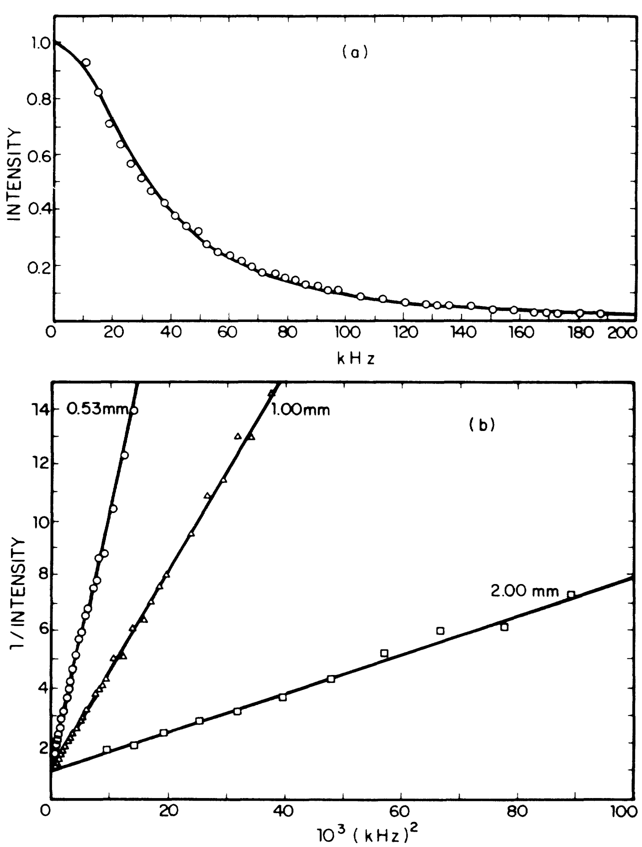

FIG. 2. (a) Measured forward-scattered frequency spectrum for a 0.53-mm-thick cell. The solid line is a best-fit Lorentzian. (b) Inverse of intensity vs square of frequency shift for forward-scattered light from 0.53-, 1.00-, and 2.00-mm-thick cells.

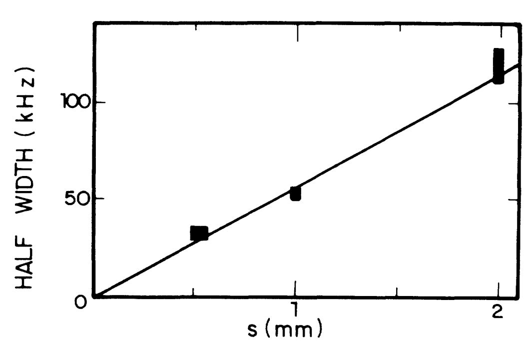

FIG. 3. Half-width of frequency spectrum vs cell thickness s.

eliminated these (and many other) possible sources of experimental error, we turn now to an examination of the assumptions which underlie the theory.

Do the scatterers in our dense system really undergo simple diffusive, i.e., Brownian, motion, as has been assumed in the theory of Fig. 1? Since our measurements correspond to times which are much shorter than τ_1, on these short time scales diffusion might not yet be operative, and so the scatterer dynamics could be described by a Maxwell-Boltzmann velocity distribution. For this distribution the photon-diffusion approximation predicts for a slab[21] $C(\Delta t) = \exp[-\Delta t/\tau']$, which has both the right functional form and the right s dependence on the decay time $\tau' = (\lambda l/4\pi s)(M/k_B T)^{1/2}$, where M is the particle mass, λ the optical wavelength, and T the absolute temperature. The scale of τ', however, is all wrong, since for a 1-mm cell $\tau' = 0.06$ μsec, some 45 times shorter than the measured value. Accordingly, if plotted in Fig. 1, $C(\Delta t)$ would fall even faster than for the point source, so that this possibility is eliminated by the data.

A second source of concern is strong particle-particle interactions. Hard-sphere repulsions (i.e., crowding) do slow somewhat the rate of scatterer diffusion, but, as found both experimentally and theoretically by Pusey and co-workers,[25] at the 0.1 concentration of our samples, and on the fast time scale of our experiments, this effect is small, and may be accounted for simply by use of an effective diffusion constant somewhat less than the free-particle value. This conclusion is consistent with previous experimental determinations,[10,22] which yield an average value of $\tau_1 = 1300$ μsec that corresponds to a diffusion constant 25% smaller than for free particles.

A final possible source of concern is long-range Coulomb interactions due to adsorbed charges which might not be completely screened by the solvent. This problem has also been considered by Pusey,[25] who concludes again that the initial decay of the particle number-density correlation function is determined largely by free-particle Brownian motion. We summarize the situation by stating that only Brownian motion produces the rapidly varying fluctuations which are relevant to our measurements, and that on the time scales probed by our experiments, the particles undergo normal diffusive motion. This conclusion is further supported directly by our previous results on backscattering,[10] in which good agreement was found between experiment and theoretical calculations based upon normal (i.e., diffusive) particle dynamics.

Returning to the photon-diffusion approximation, we note that the forward-scattering curves a and b of Fig. 1 can be obtained by our extending the real-space arguments developed previously[10,22] to treat backscattering. For the forward direction this yields

$$C(\Delta t) = \left[\int_s^\infty dL\, W(L,s) \exp\left(\frac{-L\Delta t}{2l\tau_1} \right) \right]^2, \qquad (2)$$

where the $W(L,s)$ are normalized random-walk probabilities for advancing a distance s via a diffusive trajectory of length L. With the use of the appropriate $W(L,s)$,[26] Eq. (2) reproduces both the point-source and finite-thickness–slab results of Stephen,[21] which were obtained with rigorous diagrammatic methods. Accordingly, this equation provides a useful alternative approach to the photon-diffusion approximation.

As mentioned, our experimental data clearly show the existence of some mechanism whereby the contributions of long trajectories to $C(\Delta t)$ are cut off. We believe that this is due to positive correlations between the time evolution of the optical phases of different trajectories, an effect which grows with trajectory length. Since the time evolution of the intensity fluctuations depends upon the rate at which different trajectories beat against each other, i.e., upon phase *differences* between trajectories, a positive correlation in the time evolution of these phases acts to reduce the effective length of long trajectories. Thus, in terms of their contribution to $C(\Delta t)$, long, correlated trajectories behave as if they were effectively shorter. This implies that in Eq. (2) the weights $W(L,s)$, which were obtained from the photon-diffusion approximation with the assumption of independent trajectories, are too large for large L and too small for short L. Accordingly, we correct this deficiency phenomenologically by multiplying the $W(L,s)$ of the photon-diffusion approximation by an exponential correlation function of the form $\exp(-L/\Lambda)$, and then renormalize these weights. With this modification, Eq. (2) now yields our experimental results, Eq. (1a), for the region $0 < \Delta t < 3\tau$, which covers the range of our measurements. The correlation time τ is given by

$$\tau = (2\tau_1 l/\sqrt{3}s)(l/\Lambda)^{1/2}. \qquad (3)$$

Both the point source and the finite slab yield this same result, since it is the exponential cutoff which is now dominant, rather than the boundaries. For long times $\Delta t \gg \tau$, however, for which only short trajectories are important, we recover the usual results of the photon-diffusion approximation, since here the exponential cutoff is no longer important. We emphasize that Eq. (3) completely fits our data for all sample thicknesses with a single value of $\Lambda/l = 4/3A^2$, which yields for the dynamic correlation length $\Lambda = 2.0$ mm. From Eq. (3) and our data, we also obtain for the characteristic length which dominates $C(\Delta t)$ the quasiballistic value $s/A = 9s$, which clearly illustrates the importance of the length scale of the ballistic trajectory s.

In summary, we believe that the dramatic departure of our experimental results from the predictions of the photon-diffusion approximation represent a major theoretical challenge. We sharpen the issue by stating that this theory predicts a functional dependence of the form $C = C(s^2\Delta t)$, while our experiments require $C = C(s\Delta t)$. The corollary of this is that the theory predicts that the relevant dynamic length scale of the important photon trajectories is the diffusive length s^2/l, while our experimental results show that in actual fact it is the ballistic length s. Our experiments thus clearly demonstrate the need for a theory which goes beyond the framework of the photon-diffusion approximation. We suggest that the problem with this approximation as applied to dynamic systems is the neglect of positive correlations between the time evolution of the optical phases of different trajectories, a problem which appears to worsen as the trajectory lengths increase, since the number of trajectories grows exponentially with length. We introduce an exponential correlation function as a phenomenological means of incorporating the necessary correlations into the theory, and find that this successfully reconciles theory with experiment. We expect that these ideas can be given a rigorous foundation using suitable diagrammatic techniques, and will lead to a theory which goes beyond the framework of the photon-diffusion approximation. Finally, we note that the effects described here may be expected to occur also for other types of waves in dynamic multiply scattering media, and that there may be practical implications for propagation of radar, laser beams, or acoustic waves, through seawater, fogs, clouds, dust, and aerosols.

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[1]Y. Kuga and A. Ishimaru, J. Opt. Soc. Am. A **1**, 831 (1984); L. Tsang and A. Ishimaru, J. Opt. Soc. Am. A **2**, 2187 (1985).

[2]S. John, Phys. Rev. Lett. **53**, 2169 (1984).

[3]P. W. Anderson, Philos. Mag. B **52**, 505 (1985).

[4]M. P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985); M. P. van Albada, M. P. van der Mark, and A. Lagendijk, Phys. Rev. Lett. **58**, 361 (1987).

[5]P. E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).

[6]E. Akkermans, P. E. Wolf, and R. Maynard, Phys. Rev. Lett. **56**, 1471 (1986).

[7]S. Etemad, R. Thompson, and M. J. Andrejco, Phys. Rev. Lett. **57**, 575 (1986).

[8]M. Kaveh, M. Rosenbluh, I. Edrei, and I. Freund, Phys. Rev. Lett. **57**, 2049 (1986).

[9]G. H. Watson, Jr., P. A. Fleury, and S. L. McCall, Phys. Rev. Lett. **58**, 945 (1987).

[10]M. Kaveh, M. Rosenbluh, and I. Freund, Nature (London) **326**, 778 (1987); M. Rosenbluh, M. Hoshen, I. Freund, and M. Kaveh, Phys. Rev. Lett. **58**, 2754 (1987).

[11]A. Genack, Phys. Rev. Lett. **58**, 2043 (1987).

[12]A. A. Golubentsev, Zh. Eksp. Teor. Fiz. **86**, 47 (1984) [Sov. Phys. JETP **59**, 26 (1984)].

[13]M. J. Stephen, Phys. Rev. Lett. **56**, 1809 (1986); M. J. Stephen and G. Cwilich, Phys. Rev. B **34**, 7564 (1986); M. J. Stephen and G. Cwilich, to be published.

[14]B. Shapiro, Phys. Rev. Lett. **56**, 1809 (1986).

[15]P. Sheng and Z. Zhang, Phys. Rev. Lett. **57**, 1879 (1987).

[16]C. A. Condat and T. R. Kirkpatrick, Phys. Rev. Lett. **58**, 226 (1987).

[17]K. Arya, Z. B. Su, and J. L. Birman, Phys. Rev. Lett. **57**, 2725 (1986).

[18]M. Kohmoto, B. Sutherland, and K. Iguchi, Phys. Rev. Lett. **58**, 2436 (1987).

[19]S. John, Phys. Rev. Lett. **58**, 2486 (1987).

[20]For some relevant earlier work see P. C. Colby, L. M. Narducci, V. Bluemel, and J. Baer, Phys. Rev. A **12**, 1530 (1975); F. C. van Rijswijk and U. L. Smith, Physica (Amsterdam) **83A**, 121 (1976); C. M. Sorensen, R. C. Mockler, and W. J. O'Sullivan, Phys. Rev. A **14**, 1520 (1976); A. Boe and O. Lohne, Phys. Rev. A **17**, 2023 (1978); F. Gruner and W. Lehman, J. Phys. A **13**, 2155 (1980).

[21]M. J. Stephen, Phys. Rev. B **37**, 1 (1988).

[22]G. Maret and P. Wolf, Z. Phys. B **65**, 409 (1987).

[23]Sigma Chemical Company, Post Office Box 14508, St. Louis, MO 63178.

[24]N. A. Clark, J. H. Lunacek, and G. Benedek, Am. J. Phys. **38**, 575 (1970); *Dynamic Light Scattering: Applications of Photon Correlation Spectroscopy,* edited by R. Pecora (Plenum, New York, 1985).

[25]P. N. Pusey, J. Phys. A **8**, 1433 (1975); J. C. Brown, P. N. Pusey, J. W. Goodwin, and R. H. Ottewill, J. Phys. A **8**, 664 (1975); P. N. Pusey and R. J. A. Tough, J. Phys. A **15**, 1291 (1980).

[26]For a point source $W(L,s) \propto L^{-3/2}\exp(-3s^2/4L)$; for a finite-thickness slab see I. Edrei and M. Kaveh, Phys. Rev. B **35**, 6462 (1987).