Time Evolution of Universal Optical Fluctuations

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We show that in a dynamic multiply scattering system, the intensity-intensity correlation function of universal optical fluctuations does not decay exponentially, but rather drops rapidly to half its initial value and then tails off very slowly to zero. We illustrate this dramatic behavior, and the approach to the ensemble average, with high-speed video images. We also present quantitative results for the correlation function, and provide a theory which is in excellent agreement with the data.

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Multiple scattering of light has recently been the focus of intense interest.[1-14] Among the dramatic effects observed have been coherent backscattering of light from diffusely scattering systems,[1-6] the existence, and the disappearance under ensemble averaging,[4,5] of optical fluctuations which are the analog of universal conductance fluctuations,[15,16] and long-term diffusive trapping of photons[7] corresponding to an amazing optical Q exceeding 10^8. Still sought is unambiguous evidence of the Anderson localization of light,[17,18] predicted to occur when the optical mean free path approaches the wavelength λ. The application of antilocalization[19] to coherent backscattering of light has also been considered very recently.[20] All these studies have involved either time-dependent or ensemble-averaged properties. Here, we discuss the dramatic effects of strong multiple scattering on the dynamic, time-dependent properties of light scattered from disordered systems which evolve randomly in time. In particular, we consider the time evolution of universal optical fluctuations[4,5] and the rate at which these approach their ensemble average. We find as a very general result that quasielastic multiple scattering markedly accelerates the time evolution of these universal fluctuations and thus the rate of approach to the ensemble average. For the particular case of strong multiple, as opposed to single, backscattering of light from diffusing particles, we find a universal, approximately 25-fold initial acceleration in the time evolution of the optical fluctuations. We also find that the intensity-intensity correlation function is totally nonexponential, and that after its initial very rapid decay to one-half, it falls exceedingly slowly, ultimately approaching zero at the single-scattered rate. All these effects we observe directly using a high-speed video technique[21,22] to record images of the universal optical fluctuations and their time evolution. We also present quantitative results obtained by digitizing these images. Finally, we extend the theory of the time dependence of the intensity-intensity correlation function to the case of multiple scattering, and relate the rather unexpected results observed here for backscattering to the different rates at which multiply scattered photon trajectories of different lengths lose phase as a result of random motion of the scatterers.

Our samples were a 10% (vol/vol) sonicated suspension in distilled water of 0.46-μm-diameter polystyrene spheres. Such spheres have become something of a *de facto* standard in recent studies of multiple scattering,[1-4,6,7] and the optical properties of the particular diameter chosen here have been especially well characterized as a result of the analysis of coherent backscattering by Akkermans, Wolf, and Maynard.[11] The time evolution of the universal optical fluctuations produced by this system is shown in Figs. 1-3. Figures 1-2 graphically illustrate the unexpected way in which the optical fluctuations change with time, while Fig. 3 provides a quan-

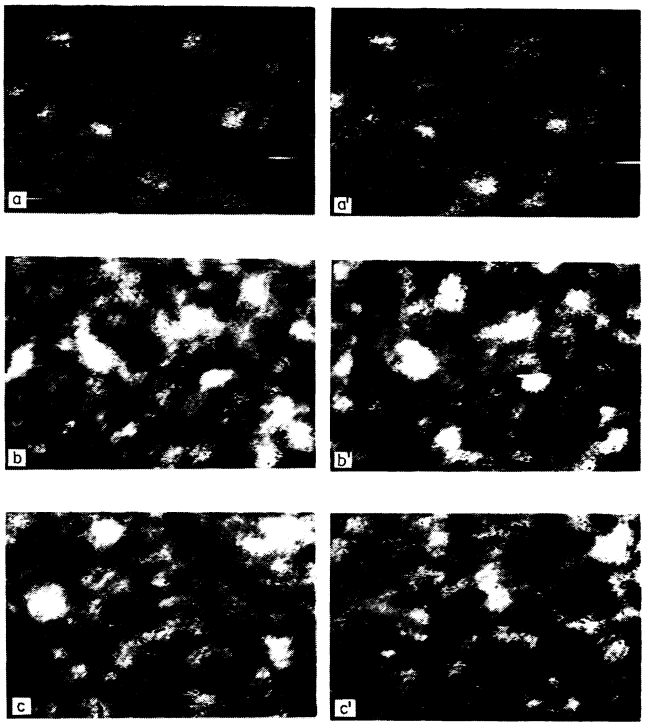

FIG. 1. Time evolution of universal optical fluctuations. Displayed are snapshots separated in time by (a),(a') 15 μsec; (b),(b') 40 μsec; and (c),(c') 100 μsec.

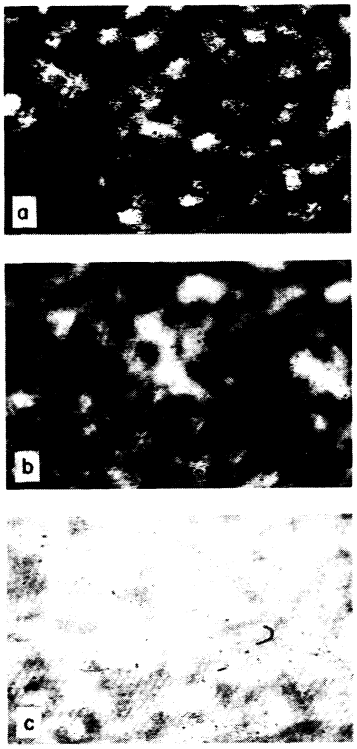

FIG. 2. Approach to the ensemble average: Time evolution of the contrast of the optical fluctuations for a single optical pulse of width (a) 40 μsec, (b) 400 μsec, and (c) 4000 μsec.

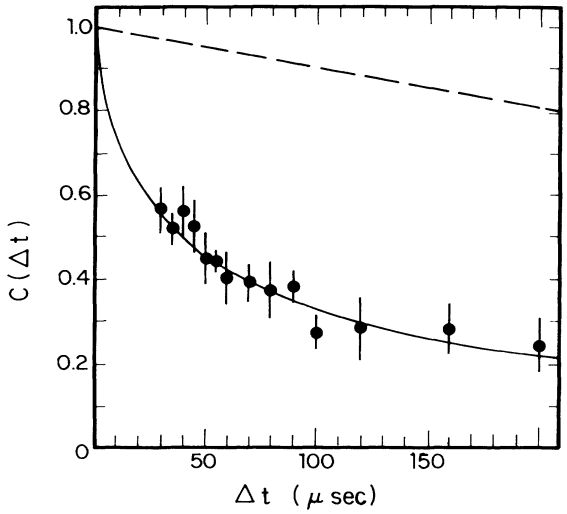

FIG. 3. Multiple-scattering intensity-intensity correlation function $C(\Delta t)$. The experimental points were obtained by digitizing images similar to those in Fig. 1. The solid curve is the theoretical expression given in Eq. (5), while the dashed curve shows the exponential decay appropriate to single scattering.

titative comparison between our theory and experiment. These data were obtained with an apparatus which included a 2-W Ar-ion laser operated in the green at 514.5 nm, a sensitive (0.7 lux) video camera, and a high-contrast acousto-optic modulator used to produce 10–20-μsec-wide optical pulse pairs of variable separation Δt. Because of the very small pulse energies and low system losses, heating effects were entirely negligible.[22] The scattering pattern from each member of an optical pulse pair was stored individually in a separate frame on video cassette. This was accomplished[21,22] by the synchronization of the optical pulse pairs with the camera readout in such a way that the first optical pulse arrived immediately before readout of a given video field and hence was stored in that field, while the second optical pulse arrived immediately after readout and was thus stored in subsequent fields. In this way optical scattering patterns separated in time by only 10 μsec could be stored in sequential video frames separated by 33.3 msec.

Displayed in Fig. 1 are snapshots of the time evolution of the universal optical fluctuations which illustrate the dramatic effects of multiple scattering. In Figs. 1(a) and 1(a') are shown two snapshots separated in time by an amount $\Delta t = 15$ μsec. This time difference is less than $\frac{1}{60}$ the average single-scattering correlation time $\langle \tau_1 \rangle$. Although these two images are obviously very similar and highly correlated, noticeable differences have already begun to appear. In Figs. 1(b) and 1(b'), Δt is increased to 40 μsec, $\frac{1}{25}$ the single-scattering time. Here a significant time evolution of the scattering pattern with a concomitant very substantial loss of correlation is immediately apparent, while in Figs. 1(c) and (c') for which $\Delta t = 100$ μsec, most correlation seems to have disappeared. These images of the time evolution of the universal optical fluctuations thus graphically illustrate the very rapid initial loss of correlation induced by multiple scattering.

Since the unaided eye cannot easily distinguish between no correlation and a comparatively small degree of correlation, we display in Fig. 2 images of the universal optical fluctuations obtained with a single laser pulse of variable width ΔT. These images illustrate the way in which the system approaches its ensemble average. In Fig. 2(a), a short laser pulse with $\Delta T = 40$ μsec is used, and the high degree of contrast of the scattering pattern is readily apparent. As the pulse width ΔT is increased, and ensemble averaging proceeds, one expects that the pattern will smear out and that the contrast will be lost. This loss of contrast, however, sets in surprisingly slowly. In Fig. 2(b), $\Delta T = 400$ μsec, and although this is 10 times longer than the 40 μsec required to lose half of the initial correlation, a very significant degree of contrast is still visible. Not until ΔT reaches 4000 μsec, a 100 times the half-width of the correlation function, does the contrast finally approach zero. Thus Figs. 1 and 2 dramatically illustrate the very rapid initial loss of correlation, and its subsequent very slow rate of decay, which we

have found to be the unique signature of multiple scattering.

After digitization, images such as those in Figs. 1 and 2 yield a wealth of quantitative information on all aspects of the time evolution of the universal optical fluctuations. In Fig. 3, we plot our measurements of the intensity-intensity correlation function $C(\Delta t)$ defined by

$$C(\Delta t)=N[\langle\langle I(t)I(t+\Delta t)\rangle\rangle-\langle\langle I(t)\rangle\rangle^2], \tag{1}$$

where the normalization constant N is chosen such that $C(0)=1$, and where the $\langle\langle\rangle\rangle$ represent an average over sets of patterns similar to those in Fig. 1, and therefore imply both a time and restricted momentum-space average of the intensity fluctuations $I(t)$. As may be seen, the quantitative form of $C(\Delta t)$ fully conforms to the qualitative results illustrated so graphically by Figs. 1 and 2; $C(\Delta t)$ initially falls to 0.5 very rapidly, faster than exponential, with 40 μsec, and then begins to tail off very slowly, slower even than $1/\Delta t$, towards zero. For comparison, we also show in Fig. 3 the known exponential decay[23] of $C(\Delta t)$ for the case of single scattering (dashed line), and our theoretical results for the case of multiple scattering (solid line).

Starting from Eq. (3) of Kaveh *et al.*,[5] we now extend the calculation of $C(\Delta t)$ to the case of multiple scattering. With $\langle\rangle$ representing an ensemble average over the system of diffusing scatterers, and A the complex amplitude of the scattered wave, we have

$$C(\Delta t)=\left|\sum_{n=0}^{\infty} W_n\langle A_n^*(t)A_n(t+\Delta t)\rangle\right|^2, \tag{2}$$

where the sum is over all photon trajectories or loops, and the relative weight of a loop consisting of n steps is given by the normalized probability W_n. The $n=0$ term corresponds to single scattering, and larger values of n correspond to successively higher-order scattering processes. In our optically thick, dense, strongly multiply scattering sample, all orders are of importance. Although the accumulated optical phase[5] ϕ_n of a loop of length L_n is simply $\phi_n=(2\pi/\lambda)L_n$, for our present purposes it is convenient to incorporate the loop end points and recast this in the form

$$\phi_n=\sum_{j=1}^{n+1}\Delta\mathbf{k}_j\cdot\mathbf{r}_j, \tag{3}$$

where the sum is over the $n+1$ scatterers which comprise the loop, the $\mathbf{r}_j$ are the positions of the scatterers, and the $\Delta\mathbf{k}_j$ are the changes in photon wave vector due to scattering at the corresponding sites. In the specific case of randomly diffusing particles considered here, we assume that the motions of individual scatterers are uncorrelated, and may be described by standard diffusion theory. With D the diffusion constant of the scatterers, we obtain

$$\langle A_n^*(t)A_n(t+\Delta t)\rangle = \exp\left[i\omega\Delta t-D\Delta t\sum_{j=1}^{n+1}|\Delta\mathbf{k}_j|^2\right]. \tag{4}$$

For the particular case of double scattering ($n=1$), Eq. (4) is, in effect, verified by the results of a very fine experiment performed some time ago by van Rijswijk and Smith.[24] For the case of single scattering ($n=0$) and our experimental geometry, the appropriate value of $\Delta\mathbf{k}$ is that for backscattering, and we separate out this term explicitly. For higher-order processes, we replace the sum in Eq. (4) by $(n+1)\langle|\Delta k|^2\rangle$, where the average is over all directions of scattering. This neglects a small degree of correlation in the directions of the scattering vectors whose sum is constrained by our backscattering geometry. This approximation clearly improves as n increases, but it is also numerically correct even for the case $n=1$. Writing $\langle\tau_1\rangle=\lambda^2/16\pi^2D$ for the average value of the single-scattering correlation time, we finally obtain

$$C(\Delta t)=-\left[W_0\exp(-\Delta t/\langle\tau_1\rangle)+\sum_{n=1}^{\infty}W_n\exp[-(n+1)\Delta t/2\langle\tau_1\rangle]\right]^2. \tag{5}$$

Since this result for $C(\Delta T)$ is independent of the optical mean free path, it is a universal function of $\Delta t/\langle\tau_1\rangle$.

For single scattering we take W_0 to be 0.12 as given by Milne theory,[6,25] while for higher-order processes we employ the same set of relative weights used so successfully in describing the coherent backscattering.[9,11,12] With these weighting factors we then fit Eq. (5) to our experimental data using $\langle\tau_1\rangle$ as the single adjustable parameter. The solid curve in Fig. 3 is the calculated result with $\langle\tau_1\rangle=1.4$ msec. It may be seen that the overall fit is excellent, and that the calculated $C(\Delta t)$ indeed initially falls very rapidly as implied by the data contained in Fig. 1, and then tails off extremely slowly as implied by the results contained in Fig. 2. Equations (4) and (5) show that this initial rapid falloff is due to the long loops which dephase with characteristic time $\langle\tau_1\rangle/n$, while the slow asymptotic approach to zero is due to the much slower rate of dephasing of the short loops.

When absorption is important, it may be incorporated in Eq. (5) by the multiplication of the W_n by $\exp[-(n+1)\alpha l]$, where α is the absorption coefficient and l the transport optical mean free path. This has the effect of cutting off the long loops, thereby both rounding and markedly broadening $C(\Delta t)$. We verified this behavior directly by adding to our samples a strongly absorbing dye with $\alpha\sim 1/l$, and observing an approximate fivefold increase in the half-width of $C(\Delta t)$.

We note that our measured value for $\langle \tau_1 \rangle$ implies a diffusion constant D which is 30% smaller than that calculated for free particles. This is consistent with expectation for these dense systems in which the average center-to-center spacing between particles is only two diameters, and suggests the utility of our results in studying the dynamics of dense, highly scattering systems which are of special interest in many areas of pure and applied physics.

In summary, we have found that quasielastic multiple scattering drastically shortens the phase correlation time and thus markedly accelerates the rate of approach of a dynamic system to its ensemble average. For backscattering, we have found a universal 25-fold initial acceleration factor. We have also found that the time decay of the backscattered intensity-intensity correlation function is totally nonexponential, falling initially extremely rapidly for short times, and then tailing off exceedingly slowly for long times. We have illustrated this unique behavior with high-speed images of the time evolution of the universal optical fluctuations, and we have presented a quantitative theory, applicable to any wave phenomenon, which is in excellent agreement with our experimental results.

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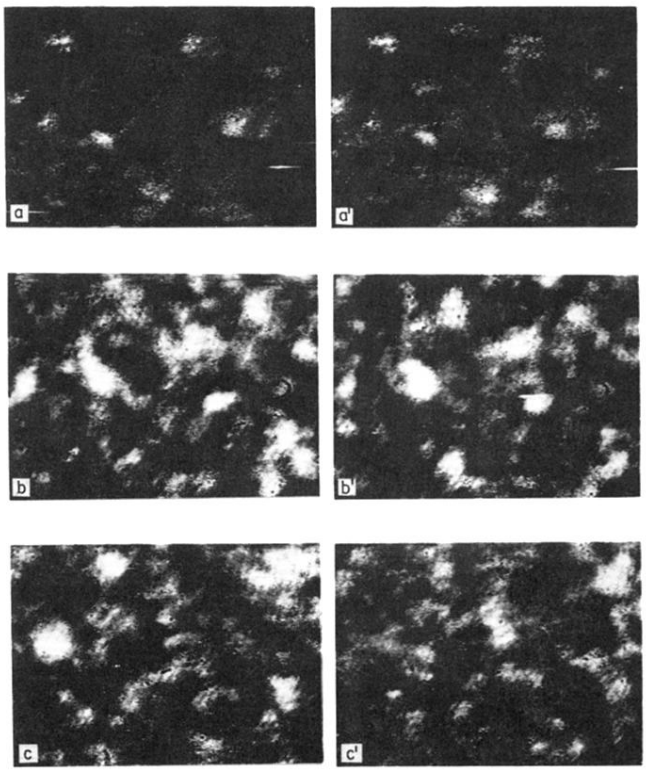

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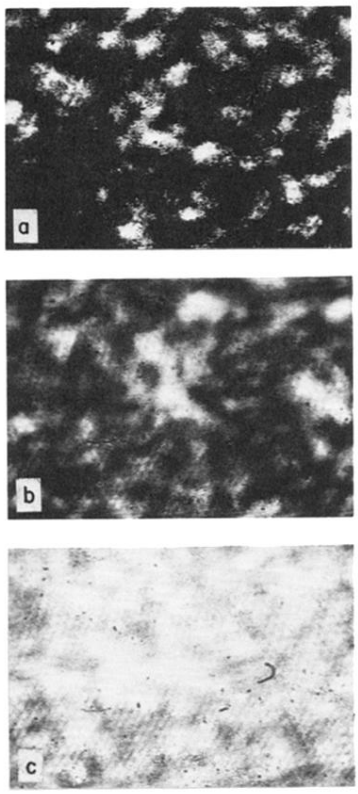

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