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Geometric scaling of the optical memory effect in coherent-wave propagation through random media

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Geometric scaling of the optical memory effect is studied for transmission and reflection. Two-dimensional momentum matching, and two-dimensional scaling of the correlation function, is obtained for smooth-surfaced samples from theory and from experiment. Very rough-surfaced samples exhibit three-dimensional scaling. Internal surface reflections are found to play an important role in determining the width of the correlation function, and are shown substantially to account for previous discrepancies between theory and experiment.

Recently, a striking memory effect in the propagation of coherent optical waves through highly-random media was observed experimentally,[1] verifying earlier theoretical predictions of Feng, Kane, Lee, and Stone.[2] One important manifestation of this memory effect is the ability of the emitted speckle pattern to track the (invisible) laser beam through a random, highly multiply scattering medium. This tracking, however, is strongly dependent upon geometry, an effect not explicitly considered previously.[1-3] Theory predicts,[1-3] and the experiments verified,[1] that the angular range over which such tracking occurs is limited by the sample thickness t for transmission, and by the photon-transport mean free path l for reflection. When the change $\mathbf{k}$ in momentum transfer due to a change in direction of the laser beam is such that kt for transmission, or kl for reflection, is of order unity, memory loss sets in, and the emitted speckle pattern loses track of the laser-beam direction. A quantitative measure of the extent of the memory effect is thus the degree of cross correlation C between initial and final speckle patterns as the laser direction is varied. The angular halfwidth of this correlation function is also strongly dependent upon geometry, and shows an important, general scaling property which will be our major concern. In this Rapid Communication, we explicitly introduce the requisite geometric effects into the theory, and present the first experimental and theoretical results on the geometric scaling of the memory effect in both transmission and reflection. We also show that internal surface reflections play an important role in determining the width of the correlation function C, and that these reflections account for much of the previous discrepancy between theory[2] and experiment.[1]

Our experiments are performed on free-space speckle patterns, which are approximated here as being in the far field, since wave-front curvature is unimportant. The (assumed scalar) optical field $E(\mathbf{r})$ at the output face is related to the field $A(\mathbf{r}')$ at the input face by

$$E(\mathbf{r})=\int T(\mathbf{r},\mathbf{r}')A(\mathbf{r}')d^3r' , \quad (1)$$

where the complex transfer function T describes propagation of light through the random medium. Neglecting correlations between optical fields at different points in the sample, and denoting initial and final configurations by subscripts 1 and 2, the far-field intensity-intensity correlation function is $C=|\langle E_1(\mathbf{K}_1)E_2^*(\mathbf{K}_2)\rangle|^2$, where $E(\mathbf{K})$ is the Fourier transform of $E(\mathbf{r})$, the $\mathbf{K}$ are outgoing wave vectors, and the angular brackets imply an ensemble average. With $A(\mathbf{r})=\exp[i\mathbf{k}\cdot\mathbf{r}]$, we then have

$$\langle E_1(\mathbf{K}_1)E_2^*(\mathbf{K}_2)\rangle=\delta(\mathbf{K}_\parallel-\mathbf{k}_\parallel)\int\int\int \exp[i(\mathbf{K}_\parallel\cdot\mathbf{R}+K_\perp z'-k_\perp z)P(R,z,z';t)d^2R\,dz\,dz' , \quad (2)$$

where $\mathbf{K}=\mathbf{K}_2-\mathbf{K}_1$, $\mathbf{k}=\mathbf{k}_2-\mathbf{k}_1$, $\parallel$ and $\perp$ refer to directions parallel and perpendicular to the sample surface, $\mathbf{R}$ is a vector parallel to this surface, and z and z' are inward-directed distances measured from the input and output faces, respectively. In obtaining Eq. (2) we have set

$$\langle T(\mathbf{r},\mathbf{r}')T^*(\mathbf{r}'',\mathbf{r}''')\rangle=\delta(\mathbf{r}-\mathbf{r}'')\delta(\mathbf{r}'-\mathbf{r}''')\langle|T(\mathbf{r},\mathbf{r}')|^2\rangle ,$$

and

$$\langle|T(\mathbf{r},\mathbf{r}')|^2\rangle=P(R,z,z';t) ,$$

where $R=|\mathbf{r}'_\parallel-\mathbf{r}_\parallel|$. P, which differs for transmission and reflection, describes the probability that a photon injected at depth z from the input surface will exit at depth z' from the output surface, while having moved a transverse distance R. For reflection, it may be noted that the Fourier transform appearing in Eq. (2) is essentially the same as that which describes the coherent backscattering peak.[4]

Since the incident beam width $W\gg z$, the Fourier transform of the product of incident-field distributions $A_1A_2^*$ is well approximated as being two dimensional, leading to the δ function appearing in Eq. (2), which requires that only the components of momentum parallel to the sample surface be conserved. Experimentally, we use a geometry in which $\mathbf{k}$, $\mathbf{K}$, and the normal to the sample surface all lie in a plane, so that the angular rotation Θ of the emitted speckle pattern is related to the angle of rotation of the laser, Δ_L, and of the sample, Δ_S, by

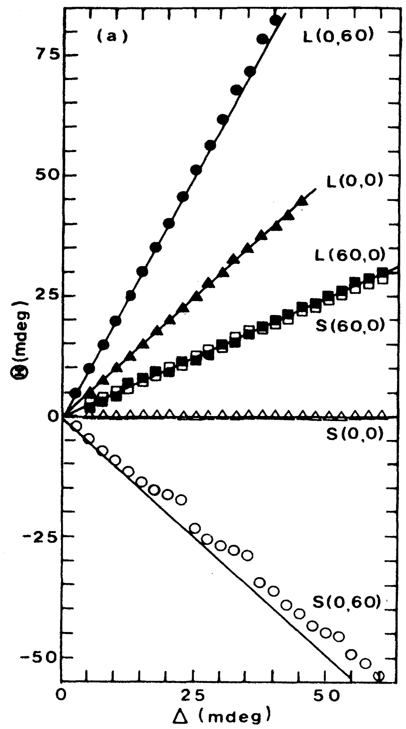
$$\Theta = -(\cos\theta_i/\cos\theta_o)\Delta_L\ , \tag{3a}$$

$$\Theta = (1+\cos\theta_i/\cos\theta_o)\Delta_S\ , \tag{3b}$$

where θ_i is the angle of incidence of the incoming laser beam, and θ_o is the angle of emission of the outgoing speckle pattern. In Eqs. (3) both these angles are measured from the normal to the input face, thereby avoiding the need for separate formulas for reflection and transmission. Experimentally, we verify Eqs. (3) by identifying Θ with the angular displacement of the peak of the correlation function C as either Δ_L or Δ_S is varied. Our experimental methods are described in Ref. 1, which may also be consulted for graphic illustrations of how the speckle pattern tracks the rotation of the laser beam, and how this is mirrored in the angular displacement of the peak of the correlation function C.

The tracking data in transmission for various geometries are shown in Fig. 1(a) for a 0.27-mm-thick sample containing 0.5% by weight micrometer-sized TiO_2 particles dispersed in a polystyrene matrix. We note that the diffusely transmitted light was almost completely depolarized, while its angular dependence approximated the predictions of Milne theory,[5] so that optical transport in this sample is highly diffusive. The curves displayed in Fig. 1(a) are labeled by $L(\theta_i,\theta_o)$ for rotation of the laser beam, and by $S(\theta_i,\theta_o)$ for rotation of the sample, both rotations being about an axis perpendicular to the scattering plane and passing through the middle of the sample at $t/2$. Note that for certain geometries, say $\theta_i=0°$, $\theta_o=60°$, very different results are obtained if the laser rotates, $L(0,60)$, in which case the speckle pattern moves in the same direction as the laser, but at *twice* the rate, or if the sample rotates, $S(0,60)$, in which case the speckle motion is *retrograde*. Overall, the data for both transmission and reflection verify the two-dimensional momentum matching conditions predicted by Eqs. (3), which are shown in Fig. 1(a) by the straight lines. Although perhaps not immediately obvious from Eq. (2), the correlation function for a given geometry is predicted to be an identical function of Δ_L and of Δ_S. This is verified in Fig. 1(b) for the two extreme cases $S(0,60)$ and $L(0,60)$ of Fig. 1(a), and was found to hold true in all geometries, both for transmission and reflection.

As was the case previously,[1] the half width of the correlation function C, shown in Fig. 1(b), is substantially narrower than the theoretical width.[2,3] We have identified the cause of this discrepancy as being mostly due to internal reflections from the sample surfaces. For a typical sample refractive index of 1.5, half of the internally scattered light will exceed the critical angle for total internal reflection. As a result of such reflections, the photon moves a transverse distance R which is much greater than predicted by diffusion theory, leading to a significant narrowing of the correlation function. These internal reflections may be minimized by index matching the sample surfaces to thick optical flats, while masking out the large-angle reflections from the free surfaces of the flats. In Fig. 2, we display the results for transmission through our TiO_2-polystyrene sample. The substantially improved



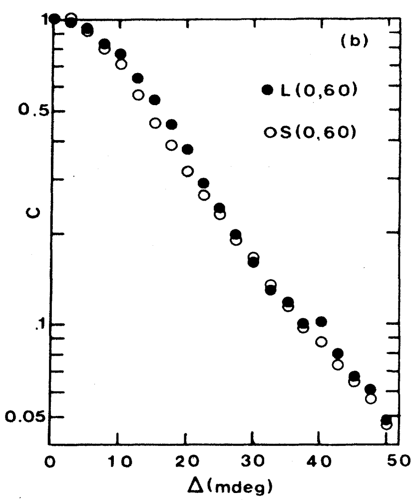


FIG. 1. (a) Angular displacement Θ of the peak of the correlation function C in transmission vs the angular rotation Δ of either the laser ($\Delta=\Delta_L$) or the sample ($\Delta=\Delta_S$). The curves are labeled $L(\theta_i,\theta_o)$ for rotation of the laser, and $S(\theta_i,\theta_o)$ for rotation of the sample, where θ_i is the angle of incidence of the laser, and θ_o is the angle of emission of the speckle pattern. Both the angles are given in degrees. For convenience and clarity, in the labels to all graphs θ_o is measured from the normal to the *output* face and θ_i from the normal to the *input* face. The straight lines are the theoretical predictions of Eqs. (3). Negative Θ implies that the speckle motion is *retrograde*. (b) Memory effect correlation function C vs angular rotation Δ of either the laser ($\Delta=\Delta_L$) or the sample ($\Delta=\Delta_S$). The data correspond to the two extreme cases $L(0,60)$ and $S(0,60)$ shown in (a). In spite of their very different tracking behavior, the correlation functions for these two extreme cases are the same within experimental error, in full accord with theory.

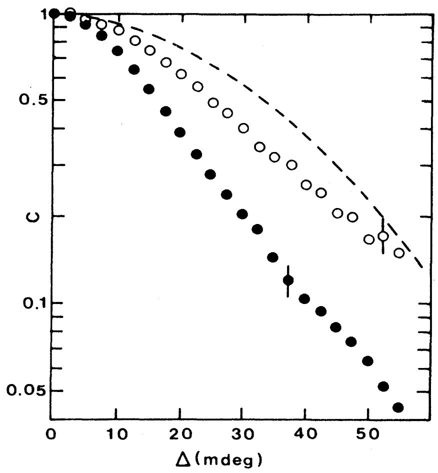


FIG. 2. Reduction of internal surface reflections in transmission for $L(0,0)$ geometry. The solid circles are the correlation function C for a native TiO_2-polystyrene sample, and the open circles are for the same sample in which surface reflections have been reduced by index matching. The dashed line is the theoretical prediction of Ref. 2.

agreement between theory and experiment is self-evident. We also find a similar degree of improvement for reflection.

We turn now to a consideration of the geometric scaling behavior of the correlation function C. For transmission, the transverse displacement R is of order the sample thickness t, and is thus much greater than z, and z', which are both of the order of the transport mean free path l. Accordingly, the Fourier transform in Eq. (2) is dominated by the $\mathbf{K}_{\parallel}\cdot\mathbf{R}$ term, and is thus nearly two dimensional.

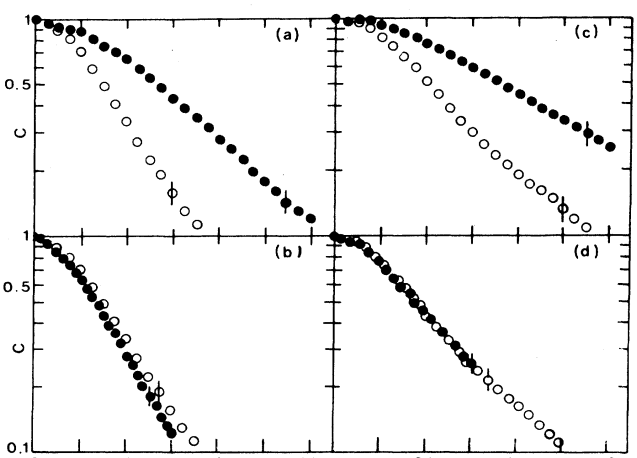


FIG. 3. Scaling of the correlation function in transmission (a),(b), and in reflection (c),(d), for a TiO_2-polystyrene sample. The incident laser direction θ_i is shown by either open circles —normal to the input face in transmission, or 25° from the normal in reflection; or by solid circles—at an angle of incidence equal to 60°. The output angle $\theta_o=0°$. (a) and (c) are plotted unscaled, $\Delta=\Delta_L$, while (b) and (d) are plotted using two-dimensional scaling, $\Delta=\cos(60°)\Delta_L$.

This implies that when plotted against Δ_L, for example, C will appear to be strongly geometrically dependent, whereas it will exhibit scaling when plotted against $\cos(\theta_i)\Delta_L$. This prediction is verified in Figs. 3(a) and 3(b) for transmission through our TiO_2-polystyrene sample.

For reflection, it is not at all obvious what degree or

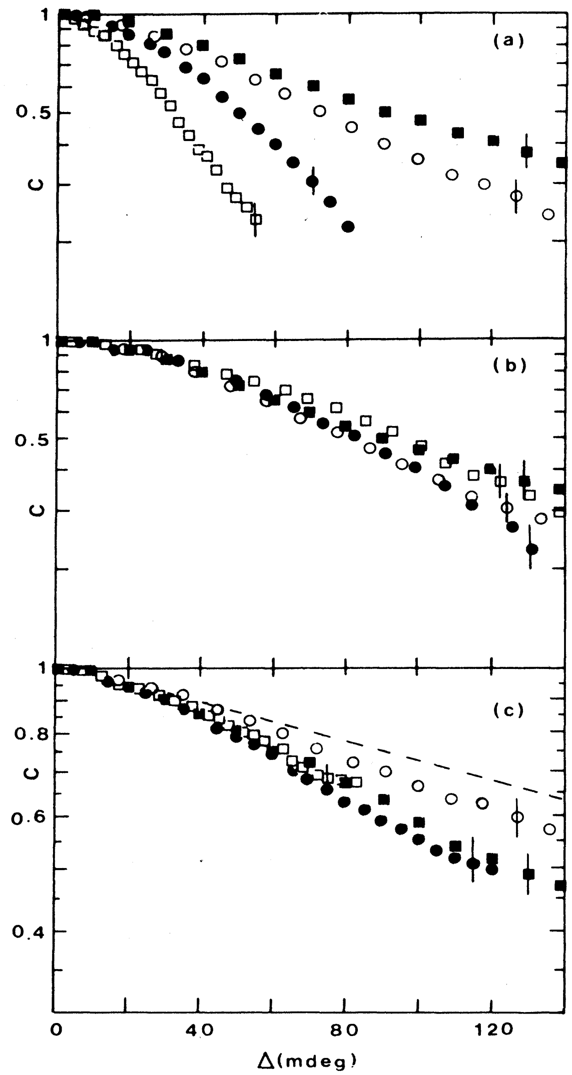


FIG. 4. Scaling of the correlation function in reflection for $BaSO_4$ coatings. (a) and (b) are for a rough-surfaced sample, while (c) is for a smooth-surfaced sample prepared as described in the text. Data are shown for four different angles of incidence: ■, 5°; ○, 25°; ●, 60°; and □, 75°. The angle of emission $\theta_o=0°$. The rough-surface data in (a) are plotted using two-dimensional scaling, $\Delta=\cos(\theta_i)\Delta_L$, which fails completely. In (b) these same data are replotted using three-dimensional scaling, Eq. (5), with $\langle z^2\rangle/\langle R^2\rangle=(0.75)^2$, which is reasonably successful. In (c) the smooth-surface data are scaled using two-dimensional scaling, which in marked contrast to (a) is successful for this sample. Note also the expanded scale for the correlation function C. The dashed line is the square of the coherent backscattered peak as measured in Ref. 9.

what kind of scaling is to be expected, since R, z, and z' are all of order l, and the Fourier transform in Eq. (2) appears three dimensional. We have extended prior treatments[1–3] of the memory effect by explicitly including the normal components of **k** and **K**, and have obtained C for reflection from a sample of arbitrary thickness t using continuous photon injection. Our general expression is too long to be reproduced here, and will be published separately.[6] In the limit $t \rightarrow \infty$ it yields for the normalized correlation function,

$$C=\delta(\mathbf{k}_\parallel-\mathbf{K}_\parallel)\,|\,(2k_\parallel z_0\lambda_i\lambda_o)^{-1}\{(\lambda_i+k_\parallel+ik_\perp)^{-1}(\lambda_o-k_\parallel-iK_\perp)^{-1} -2k_\parallel[(\lambda_o-iK_\perp)^2-k_\parallel^2]^{-1}[\lambda_i+\lambda_o+i(k_\perp-K_\perp)]^{-1} -(\lambda_i+k_\parallel+ik_\perp)^{-1}(\lambda_o+k_\parallel-iK_\perp)^{-1}\exp(-2k_\parallel z_0)\}\,|^2\,, \tag{4}$$

where $\lambda_{i,o}=1/l\cos(\theta_{i,o})$, and $z_0=0.71l$ from the boundary conditions to the photon diffusion equation.[5] Both our general expression, and the limiting form in Eq. (4), exhibit the following quite surprising result: excellent (although not exact) $\cos(\theta_i)\Delta_L$ scaling is obtained for *any* combination of input and output angles within the limits $\theta_{i,o}\le 75°$.[7] Experimental verification of this striking prediction is given for our TiO_2-polystyrene sample in Figs. 3(c) and 3(d). We note that this scaling implies that also for reflection the Fourier transform in Eq. (2) is very nearly two dimensional. We may describe this quasi-two-dimensional behavior as resulting in part from the fact that for small angles $k_\perp$ and $K_\perp$ are, anyway, not important, while for large angles both z and z' go to zero with $\cos\theta$.

When the sample surface is very rough, this two-dimensional scaling can be broken, and a crossover forced to three-dimensional scaling. Since it is now the arbitrary surface roughness which determines z, and also influences R, we could expect that z and R will be more or less uncorrelated random variables, and that a heuristic scaling function of the form

$$\Delta_{\text{scaled}}=(\cos^2\theta_i+(\langle z^2\rangle/\langle R^2\rangle)\sin^2\theta_i)^{1/2}\Delta_L \tag{5}$$

would provide a reasonable description of the data. In Fig. 4 we present our results for diffuse reflection from a suitable rough-surfaced scatterer consisting of $BaSO_4$ microparticles.[8] In Fig. 4(a) we first plot our data using two-dimensional, $\cos(\theta_i)\Delta_L$ scaling, which fails completely. In Fig. 4(b), we replot these same data scaled according to Eq. (5) with $\langle z^2\rangle/\langle R^2\rangle=(0.75)^2$, which yields a quite reasonable degree of scaling. We note that even for this rough-surfaced sample, Eqs. (3) were well satisfied experimentally over the full range of angles studied, in accordance with expectation.

How does the correlation function for a smooth-surfaced $BaSO_4$ sample scale? We prepared such a sample by forming the coating of microparticles onto the surface of a thick optical flat, and studied the diffuse reflection from the inner surface of the coating which was in optical contact with the glass, while masking out the large-angle reflections from the remaining free glass surfaces. The data, which are shown in Fig. 4(c), are scaled using $\Delta=\cos(\theta_i)\Delta_L$ (i.e., $\langle z^2\rangle/\langle R^2\rangle=0$), which produced the best scaling results. This once again indicates that for smooth-surfaced samples the scaling of the correlation function is essentially two dimensional. Also shown in this figure as the dashed curve is the square of our previous measurement[9] of the coherent backscattering peak from similar $BaSO_4$ coatings. As may be seen, our present data for the memory effect correlation function in reflection, and our previous data for the coherent backscattering peak,[9] are in near agreement with each other, in full accord with theory.

We conclude by noting that our methods may be extended to the study of important correlation effects predicted for propagation of coherent optical waves through highly random media,[2,10–12] as well as to the study of other interesting memory effects which also occur in such media.

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[7] As the θ increase, the scaled curves lie slightly one above the other. For our samples, for which $t < 15l$, this effect is very small, so that for *either* θ_i or $\theta_o = 75°$, the deviation from perfect scaling is less than 5%, but increases to 20% when *both* θ_i *and* $\theta_0 = 75°$. These deviations increase with t, reaching 20% for $\theta = 60°$ and $t \rightarrow \infty$.

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