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Wavelength dependence of static intensity correlation functions

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We develop a real-space theory for the wavelength dependence of the intensity correlation function $C(\Delta\lambda)$ in the framework of the diffusion approximation. We calculate $C(\Delta\lambda)$ for various practical geometries and boundary conditions and find that they play a crucial role in determining the functional form.

The propagation of optical waves in random media has recently aroused much interest.¹⁻⁷ The multiple scattering of the wave causes new weak localization effects, which result in a narrow coherent backscattered peak, that were observed³⁻⁷ for disordered media. For random solids, in addition to the coherent backscattered peak one gets^{6,7} intensity fluctuations. These fluctuations are closely related to the universal conductance fluctuations.^{8,9} The time dependence of the backscattered intensity fluctuations were studied experimentally^{10,11} and related theoretically^{10,11} to the dynamic intensity-intensity correlation function $C(\Delta t)$ by the concept of light trajectories caused by multiple scattering.

These effects were also studied ¹² by diagrammatic techniques for various geometries and recently shown ¹³ to coincide with the real-space method. ^{10,11} The sensitivity of the transmitted speckle to the wavelength of the wave was demonstrated experimentally. ¹⁴ The static autocorrelation function $C(\Delta\lambda)$ for a point source was studied diagrammatically by Shapiro, ¹⁵ and Stephen and Cwilich ¹⁶ have shown that Shapiro's result is correct only for two points which are apart less than the transport mean free path *l*. For large distances long-range correlations exist. ^{16,17}

In this Rapid Communication we develop a real-space theory for calculating $C(\Delta \lambda)$ for various geometries. We show how $C(\Delta \lambda)$ depends on the phase acquired by the real-space photon trajectory and its sensitivity to the source wavelength change $\Delta \lambda$. Larger photon trajectories are more sensitive to $\Delta \lambda$, and therefore are more strongly affected by the surrounding boundaries which act as a cutoff for long photon trajectories. We derive analytical expressions for $C(\Delta \lambda)$ for various practical geometries which may be studied in future experiments.

The motion of an optical wave undergoing multiple elastic scattering performs a random walk in a random media where each step is l, the transport mean free path. This picture has led to a very successful model¹⁸ which accounts^{19,20} for the coherent backscattered peak and which was found to be in agreement with rigorous diagrammatic approaches.^{16,21,22} The concept of photon trajectories was also used successfully to calculate $C(\Delta t)$, the dynamic correlation functions.^{10,11,13,23} Here we use the randomwalk theory^{13,24} for each geometry for the probability W_N for performing N random steps. The phase ϕ_N acquired by the wave-traveled N steps depends explicitly on λ and is given by¹⁰

$$\phi_N = (2\pi/\lambda) L_N \,, \tag{1}$$

where L_N is the length of the trajectory of N steps. On the average, $\langle L_N \rangle = Nl$. The intensity-intensity correlation function at a given point in the diffusion approximation is factorized to yield

$$C(\Delta\lambda) = \left| \sum_{N,N'} P_N P_{N'} \langle e^{i[\phi_N(\lambda) - \phi_{N'}(\lambda + \Delta\lambda)]} \rangle \right|^2, \qquad (2)$$

where $\langle \rangle$ denotes an ensemble average and P_N is the amplitude probability⁷ ($P_N^2 = W_N$). The sensitivity to $\Delta \lambda$ enters through the optical phases in (2). The random-walk amplitudes P_N are not sensitive to small changes of $\Delta \lambda$. The change in phase of a photon trajectory of N steps is given by

$$\phi_N(\lambda) - \phi_N(\lambda + \Delta \lambda) = (2\pi/\lambda)(\Delta \lambda/\lambda)L_N$$
(3a)

and is proportional to the actual trajectory length. Thus, longer trajectories are more sensitive to the wavelength changes. The correlation function $C(\Delta\lambda)$ will depend strongly on the particular geometry through P_N in (2). Smaller values for W_N for large values of N will correspond to a *broader* fall-off of $C(\Delta\lambda)$.

We perform the ensemble average in (2) by using the equality

$$\langle \exp[i(2\pi/\lambda)(\Delta\lambda/\lambda)L_N] \rangle = \exp[i(2\pi/\lambda)(\Delta\lambda/\lambda)Nl]$$
(3b)

and neglecting higher moments of L_N which leads to another factor $\exp[-\frac{1}{2}(2\pi/\lambda)^2\langle(\delta L_N)^2\rangle]$ in (3b), where δL_N is a fluctuation in the length of a trajectory of N steps. We find that this approximation is almost always justified.²⁵ Using (3a) and (3b) in (2), we get a normalized $C(\Delta \lambda)$ [namely, we divide all our $C(\Delta \lambda)$ by C(0)]:

$$C(\Delta\lambda) = \left|\sum_{N} W_{N} \exp[i(2\pi/\lambda)(\Delta\lambda/\lambda)lN]\right|^{2}, \qquad (4)$$

where W_N is the probability of performing N random steps. Equation (4) is our key result and shows how $C(\Delta \lambda)$ depends explicitly on $\Delta \lambda$.

We now derive $C(\Delta\lambda)$ for the following geometries: (i) a point source in an infinite medium (no boundaries); (ii) an injected point source outside a "half-infinite" space geometry (one boundary) with point detection inside the medium; (iii) an infinitely wide light source injected from

<u>38</u> 950

951

outside a "half-infinite" space geometry (one boundary) and detection on the boundary; (iv) the same as (iii) but from a slab width S (two boundaries); and (v) transmitted light through a slab.

These geometries were recently used by Edrei and Kaveh¹³ to calculate the dynamic correlation function $C(\Delta t)$. Some of these geometries were also used for measuring ^{10,11,23} $C(\Delta t)$. The explicit experimental form of $C(\Delta \lambda)$ for these geometries was not yet determined. Nevertheless, the half-width of $C(\Delta \lambda)$ for transmission through a slab was measured¹⁴ and our results are in agreement with these data. The above geometries differ by their different forms for W_N which result from solving the diffusion equation for the particular boundary conditions. For a point source in an infinite random medium where the light is collected at a point R from the source, we use for W_N the standard well-known random-walk solution with discrete time¹⁰ t = Nl/c, where c is the velocity of light in the medium:

$$W_N = C_1 N^{-3/2} \exp\left[-\frac{3}{4} (R/l)^2 / N\right], \qquad (5)$$

where $C_1 = (4\pi l^2/3)^{-3/2}$.

Inserting (5) in (4) yields, for $C(\Delta\lambda)$,

$$C(\Delta\lambda) = \exp[-(R/l)\sqrt{12\pi l\Delta\lambda/\lambda^2}]$$
(6)

in precise agreement with the result first obtained by Shapiro¹⁵ by diagrammatic methods. Thus, the real-

space method which leads to (4) is capable of accounting for the correlations between different trajectories to the same accuracy of the Shapiro approach which is believed ^{16,17} to yield correct results for correlations at a given point from the source. For correlations at *different* points one needs the Stephen-Cwilich (Ref. 16) type diagrams.¹⁷ The stretched exponential in (6) comes about because there is no characteristic length scale in the problem. The averaged diffusive trajectory of the optical wave between the source and a point at distance R is $\langle L \rangle = \sum W_N Nl$ and *diverges*. This leads to the extra sensitivity of $C(\Delta \lambda)$ to small $\Delta \lambda$ which corresponds to large photon trajectories. Indeed, the derivative $dC(\Delta \lambda)/d(\Delta \lambda)$ $\rightarrow \infty$ as $\Delta \lambda \rightarrow 0$.

We now calculate $C(\Delta\lambda)$ for the geometry of an injected point source on the surface of a "half-infinite" medium and collected at a point with distance $|R| = (z^2 + \rho^2)^{1/2}$ from it, where $\rho = (x^2 + y^2)^{1/2}$. In this case, W_N can be obtained by using the image method^{7,10,18,23,24} in which W_N is given by subtracting from Eq. (5) an image term, and is given by

$$W_{N} = C_{1} N^{-3/2} \{ \exp[-\frac{3}{4} (\bar{R} - \bar{d})^{2} / l^{2} N] - \exp[-\frac{3}{4} (\bar{R} + \bar{d})^{2} / l^{2} N] \}, \quad (7)$$

where $d = d\hat{z}$ (\hat{z} is the direction perpendicular to the boundary) and d = 1.7l. Inserting W_N as given by (7) in (4), we obtain

$$C(\Delta\lambda) = [AB/(B-A)]^{2} \{A^{-2} \exp[-(\rho/l)XA] + B^{-2} \exp[-(\rho/l)XB] -2(AB)^{-1} \exp[-(\rho/l)X(A+B)/2] \cos(\rho/l)X(A-B)/2\},$$
(8)

where $A = [1 + (z + d)^2/\rho^2]^{1/2}$, $B = [1 + (z - d)^2/\rho^2]^{1/2}$, and $X = (12\pi/\Delta\lambda/\lambda^2)^{1/2}$. The effect of the boundary is to reduce the probability for large trajectories. Here the asymptotic form of W_N is $N^{-5/2}$ instead of [as follows from (3)] $N^{-3/2}$. The reduction in W_N for large loops causes a *broadening* of $C(\Delta\lambda)$. In Fig. 1, we plot $C(\Delta\lambda)$ as given by (8) [curve (b)] for z = d and $\rho = 20/$, and compare the result with a point source in an infinite medium (no boundaries) for R = 20/ [curve (a)]. We see that the effect of the boundary is to cut the large loops and therefore to broaden $C(\Delta\lambda)$.



FIG. 1. $C(\Delta \lambda)$ as a function of $\Delta \lambda l/\lambda^2$ for (a) point source in infinite medium for R/l=20; (b) point source with one boundary for $\rho/l=20$; (c) transmitted intensity from a slab with S/l=20; and (d) backscattered intensity from a slab with S/l=20.

952

For the case where $\rho \gg z$ and for $(\rho/l)x \ll 1$, expanding Eq. (8) and reexponenting it yields a simplified expression for $C(\Delta \lambda)$:

$$C(\Delta\lambda \to 0) \simeq \exp[-1/6(\rho/l)^3(12\pi\Delta\lambda l/\lambda^2)^{3/2}].$$
(9)

This expression is a good approximation for $C(\Delta \lambda)$ for the backscattered intensity-intensity correlation function (where the light is collected at z = d).

We now turn to a plane source. In this case, ρ disappears from W_N and therefore $C(\Delta \lambda)$. For a "half-infinite" space (one boundary) W_N is now obtained by integrating Eq. (7) over ρ , which yields

$$W_N = (4\pi l^2/3)^{-1/2} N^{-1/2} \left\{ \exp\left[-\frac{3}{4}(z-d)^2/l^2N\right] - \exp\left[-\frac{3}{4}(z+d)^2/l^2N\right] \right\}.$$
(10)

For the correlation function $C(\Delta \lambda)$ for a given point z inside the medium, we find

$$C(\Delta\lambda) = \frac{1}{2} (l/dX)^{2} \exp[-(z-d)X/l] + \exp[-(z+d)X/l] - 2 \exp(-zX/l) \cos(dX/l) \}, \quad (11)$$

and again $X = (12\pi l \Delta \lambda / \lambda^2)^{1/2}$.

The most practical case is to measure $C(\Delta\lambda)$ for the *backscattered* intensity. Here we make the usual assumption that the intensity at z = d is actually the measured intensity. Inserting z = d in (11) and expanding for $\Delta\lambda \rightarrow 0$, we expect to obtain the same result as (6) for a point source but with R replaced by d. The reason for this lies in the fact that for z = d the asymptotic form of W_N is $N^{-3/2}$ exactly as for a point source. Thus, our result for $C(\Delta\lambda)$ for backscattered light from a plane source injected from outside an infinite "half-space" random medium is

$$C(\Delta\lambda) = \exp[-(d/l)\sqrt{12\pi l\Delta\lambda/\lambda^2}].$$
(12)

This result coincides with (11) for X < 1, up to 2%.

We have also calculated $C(\Delta \lambda)$ for the backscattering from a slab of width S. In this case W_N is obtained by solving the diffusion equation for the geometry of a slab as done in Ref. 24 and is given by

$$W_N = S^{-1} \sum_k \sin(k\pi d/S) \sin(k\pi z/S) \\ \times \exp[-(k\pi l/S)^2 N/3].$$
(13)

For backscattering correlations we have to take z = d, and for transmission z = S - d. The correlation function $C(\Delta \lambda)$ for backscattering is given below by Eq. (14). However, for large values of S/l, we find that the result coincides with Eq. (12) (see Fig. 2). The effect of small values of S/l is to cut large trajectories. In particular, trajectories with large number of steps for which $N > (S/l)^2$ will be cut off, causing a rounding of $C(\Delta \lambda)$ for $\Delta \lambda \rightarrow 0$. This is demonstrated in Fig. 2 for S/l = 5, 10, 20, and $S/l = \infty$. We see a systematic broadening of $C(\Delta \lambda)$ as S/l becomes smaller, and how it rounds off for $\Delta \lambda \rightarrow 0$. The full expression for the correlation function $C(\Delta \lambda)$ for backscattering is given by

$$C(\Delta\lambda) = X_1^{-2} (1 - d/S)^{-2} [F(X_1)/F(X_2)] \times [F(X_1)G(X_2) + G(X_1)F(X_2) -F(X_1 + X_2) + F(X_2 - X)],$$
(14)

where $X_1 = Xd/l$, $X_2 = Xs/l$, $F(y) = \cosh y - \cos y$, and $G(y) = \cosh y + \cos y$. In contrast to (12), $C(\Delta \lambda)$ due to finite values of S/l as given by (14) is very flat for $\Delta \lambda \rightarrow 0$.

We now provide our final result for $C(\Delta\lambda)$ for the transmitted light, a case studied experimentally by Genack.¹⁴ Inserting W_N as given by (10) with z = S - d in (4) yields

$$C(\Delta \lambda) = (S/dX_1)^2 \times \frac{[1 - \cos(X_1)\cosh(X_1)]^2 + \sin^2 X_1 \sinh^2 X_1}{\cosh X_2 - \cos X_2}.$$
(15)



FIG. 2. $C(\Delta\lambda)$ for backscattered intensity from a plane wave from a slab for (a) $S/l = \infty$, (b) S/l = 20, (c) S/l = 10, and (d) S/l = 5.

In Fig. 1 [curve (c)], we plot $C(\Delta\lambda)$ for S/l=20. We see that it decays much faster than $C(\Delta\lambda)$, which corresponds to backscattering from a slab with S/l=20 [curve (d)]. This follows from the fact that for backscattering the other boundary has little effect on W_N . Thus, Fig. 1 demonstrates clearly the role of the boundaries on $C(\Delta\lambda)$ for the different geometries. From (15) we find that the halfwidth for $C(\Delta\lambda)$ is proportional to $(l/S)^2$ in agreement with experiment.¹⁴ For $X_2 \gg 1$ and $X_1 \ll 1$, Eq. (15) leads to $C(\Delta\lambda) \simeq \exp[-(S/l)X]$, which coincides with the point-source result as given by (6) but with R replaced by S.

In summary, we have calculated the wavelength dependence of the intensity-intensity correlation functions $C(\Delta\lambda)$ in the diffusion approximation [which leads to Eq.

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(4) for various geometries]. Our main results are demonstrated by Figs. 1 and 2, where we see clearly the effect of the boundaries on $C(\Delta\lambda)$. The sharpest decay of $C(\Delta\lambda)$ is for a point source in an infinite medium. When a boundary is introduced, $C(\Delta\lambda)$ decays more slowly. The slowest decay of $C(\Delta\lambda)$ corresponds to backscattering from a plane source where only short light trajectories are effective.

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