Long-Range Intensity Correlation in Random Media

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Measurements of the spatial and spectral intensity correlation of microwave radiation allow us to identify the leading terms in a perturbation expansion of the correlation function. This function is shown to depend upon two average properties of damped eigenstates of the random medium. These are the average level width δv and the number of eigenstates within the level width δ , whose inverse is the expansion parameter for the correlation function.

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The random phasing of partial waves in the presence of disorder leads to large fluctuations in the intensity of classical waves and in the current density of quantummechanical waves. Fluctuations occur as the point of observation is moved¹ or at a fixed point as the frequency of the incident field² or energy of injected carriers is varied. We report here on the first direct measurements of long-range intensity correlation. These measurements reveal that the apparently random intensity variations in different coherence volumes of the sample are not statistically independent but rather mask a remarkably high degree of correlation throughout the sample. Because of this correlation, fluctuations in transport through quantum and classical mesoscopic systems do not self-average as the size of the sample increases. Thus, these results provide the microscopic origin of universal conductance fluctuations³⁻⁵ observed in magnetoresistance experiments³ and of large fluctuations in optical transmission.⁶⁻¹⁰

In this Letter we report measurements of the intensity cross-correlation function of microwave radiation as a function of detector separation and frequency difference. These observations allow us to identify the contributions to the correlation function associated with distinct orders of perturbation theory. Spatial correlation is shown to be a consequence of spectral correlation at a point. These results can be described in terms of two statistical properties of damped eigenstates for a wave which is both elastically and inelastically scattered in a finite sample. These properties are the ensemble averages of the width of eigenstates δv , ¹¹⁻¹³ and of the number of eigenstates within the level width δ , ^{12,14} which is $\delta = (dN/dv)\delta v$, where dN/dv is the density of states.

In the weak-scattering limit, in the absence of inelastic scattering, the intensity correlation function has been calculated as a perturbation expansion in a small parameter which is the inverse of the dimensionless conductance g. ⁶⁻⁸ For classical waves, g corresponds to the total transmission summed over all input transverse momentum channels of a wave at a given frequency with unit incident flux density.¹⁵ Feng, Kane, Lee, and Stone⁸ calculated the three leading terms in the expression for the cumulant intensity correlation function as a function of

angle, within the transmitted speckle pattern, $C = C_1 + C_2 + C_3 + \cdots$. The leading term C_1 has dominated previous measurements of spectral,^{11,13} temporal,¹⁶ and angular¹⁷ intensity correlation. It is obtained in the field factorization approximation for $C.^{6,11,17,18}$ In terms of the intensities I_1 and I_2 at two points in the sample, which are normalized to their ensemble-average values, this approximation gives

$$C = \langle (I_1 - 1)(I_2 - 1) \rangle - 1 \approx |\langle E_1 E_2 \rangle|^2 = C_1 = |\tilde{G}^E|^2,$$

where E_1 and E_2 are the associated normalized complex scalar fields, the brackets denote the ensemble average, and $G^E = \operatorname{Re}(\tilde{G}^E)$ is the field correlation function. The field in the medium due to an incident monochromatic wave can be expressed as a linear superposition of eigenstates of the wave in the random medium which are driven at the excitation frequency $v, E = \sum c_i u_i (r) e^{i2\pi v t}$. The amplitudes c_i of modes with natural frequencies v_i are only appreciable when the frequency offset is less than the level width, $|v-v_i| < \delta v$. The value of $G^E(\Delta v)$, therefore, only changes significantly for frequency shifts $\Delta v > \delta v$. $G^E(\Delta v)$ is the Fourier transform of the time-of-flight distribution of particles that have not been inelastically scattered, and its half-width can be identified with δv .¹¹

The influence of long-range correlation upon intensity statistics was observed recently in microwave experiments.¹³ The dependence of C_2 upon separation between the points R and frequency shift Δv is reported here. C_2 is responsible for enhanced fluctuations in the total transmission for samples irradiated with a single-input mode, ⁶⁻¹⁰ which were observed recently in optical experiments.¹⁰ Long-range correlation and enhanced transmission fluctuations are a consequence of spectral correlation, which limits the number of statistically independent parameters which influence the transmitted intensity to the number of eigenstates in a linewidth, δ . We expect, therefore, that the fractional fluctuation from the average intensity $(I_1 - 1)$ at point 1 in the output plane of the sample is correlated with the fluctuations $(I_2 - 1)$ at another point in the same plane to a degree $\sim 1/\delta$. To lowest order in δ^{-1} , the cross correlation of these intensity fluctuations is, therefore, given by

$$\langle (I_1-1)(I_2-1)\rangle = \langle (I_1-1)[\delta^{-1}(I_1-1)]\rangle$$
$$= \delta^{-1} \operatorname{var}(I).$$

For $\delta \gg 1$, $\operatorname{var}(I) = 1$ and $\langle (I_1 - 1)(I_2 - 1) \rangle = \delta^{-1}$. Thus, δ^{-1} is a natural expansion parameter for the correlation function. Indeed δ^{-1} is the probability that a coherence volume, through which a typical path reaching point 1 passes, is also crossed by a typical path reaching point 2.¹²

The degree of intensity correlation between point 1 on a transverse plane in the interior of the sample and point 2 on the output face depends upon the separation between the planes, R, since only particles following paths which emerge from the sample at plane 2 contribute to intensity correlation. Therefore, for a sample with transverse dimensions less than its length L for planes which are sufficiently separated such that $C_2(R) \le \delta^{-1}$,

$$C_2(R) = \delta^{-1} f(R/L),$$
 (1)

where f(R/L) is the fraction of photon paths passing through plane 1 which emerge through the output surface. f(R/L) is independent of absorption and is shown below to decrease linearly with R/L. The predicted linear falloff of $C_2(R)$ is confirmed experimentally below. This behavior holds only for the waveguide geometry since it relies on the assumption that the pathlength distribution for waves reaching a point on a given transverse plane is the same for all the points on that plane. Our results, therefore, differ from the algebraic falloff of $C_2(R)$ in powers of 1/R in a number of different geometries predicted by Stephen and Cwilich⁶ and by Pnini and Shapiro.⁹ Calculations by Feng et al.⁸ indicate that within the limits of the diffusion approximation C_3 is constant independent of scattering angle in the far field. This suggests that C_3 is also independent of separation between points. Evidence for such a term is reported here.

In the absence of absorption, in the limit $\delta \gg 1$, the Einstein relation gives $\delta = g.^{14,19}$ When absorption is introduced, the degree of correlation is expected to decrease, whereas g^{-1} increases. On the other hand, the exponential suppression of long paths by absorption narrows the width of the time-of-flight distribution δt and leads to a reduction in δ^{-1} , since $\delta^{-1} \sim \delta v^{-1} \sim \delta t$. This supports the argument that δ^{-1} rather than g^{-1} is a universal correlation parameter. We note further that δ is related to the Thouless number^{12,14,19} and describes the proximity to the localization threshold which occurs at $\delta \approx 1$.

For separations between points R such that the change in the half-width of $C(\Delta v)$ is small, we find that the dependence upon Δv and R of each order of perturbation theory can be factorized. Thus, in the presence of absorption, for $R \ll L$ we can write

$$C(qL,R) = \sum_{i=1}^{N} A_i \delta^{1-i} F_i(qL) H_i(R) , \qquad (2)$$

where q is the square root with negative imaginary part of $a^2 + i 2\pi\Delta v/D$, $a = (D\tau_a)^{-1/2}$ is the absorption coefficient for $L > a^{-1}$, D is the diffusion coefficient, and τ_a is the photon absorption time. The A's are functions of δ and are of order unity with $A_1 = 1$ for $\delta \gg 1$. The F's and H's are normalized for $\Delta v = 0$ and R = 0, respectively; $F_i(\alpha L) = H_i(0) = 1$.

The results reported here are for a sample of $\frac{1}{2}$ -in. polystyrene spheres with volume filling fraction f = 0.58. The sample is contained in two 7.3-cm-diameter copper tubes and has a length L = 150 cm as shown schematically in Fig. 1. The spheres fill the volume uniformly but are loosely enough packed that all the spheres move when the cylinders are rotated so that new sample configurations are readily produced. Intensity spectra at various points in the sample are obtained simultaneously using Schottky-diode detectors. The frequency of the microwave oscillator is swept from 20.6 to 21.7 GHz in 1000 steps by a computer-generated voltage and the data are stored in the computer. The radiation is launched from a horn placed 20 cm in front of the sample. Diodes are mounted on the face of the plunger directly behind the sample and inside a $\frac{1}{4}$ -in. quartz tube in a narrow section between the two copper tubes. The separation between the detector inside the sample and those on the face of the plunger can be varied without changing L by translating both the plastic tube at the input and the plunger at the output of the sample. After each spectrum is taken, the copper cylinders are rotated about their axes for a few seconds to produce a new sample configuration.

In the limit $\delta \gg 1$, $C \approx C_1 = |\tilde{G}^E|^2$ for R = 0, and the measured correlation function with frequency shift of optical and microwave radiation is in agreement with the result of the field factorization approximation, ^{11,13}

$$F_1(qL) = \frac{|\sinh(qa)/\sinh(qL)|^2}{[\sinh(\alpha a)/\sinh(\alpha L)]^2}$$

where $a = \gamma l$, *l* is the transport mean free path, and γ is a constant, of order unity, such that the transmission coefficient is $T(L) = \gamma l/L$ for $l \ll L \ll L_a \equiv \alpha^{-1}$. Since $H_1(R)$ decays as a result of the randomization of the phase of the field, its correlation length is just the field



FIG. 1. Schematic diagram of the experimental setup.



FIG. 2. Cross-correlation functions of intensities between a point in the output face and points inside the sample, at various distances R from the output face. The solid line is a fit to the points, as explained in the text.

correlation length $\sim 1/k$, where k is the wave number. Calculations by Shapiro for a wave in an infinite medium give $H_1(R) = [\sin(kR)/kR]^2 e^{-R/l}$.¹⁸ In contrast, higher-order corrections decay much more slowly with R and dominate the cross-correlation function $C(\Delta v; R)$ for $R \gg 1/k$ and are particularly large for small values of δ .

Measurements of the cross-correlation function using the experimental arrangement described above are shown in Fig. 2. Since $R \ge 4$ cm = 25/k, the contribution of C_1 is expected to be negligible. The results for R = 4 cm, averaged over 1500 spectra (triangles), R = 20cm averaged over 2800 spectra (circles), as well as for the average of 11000 spectra taken at R = 4, 6, 8, 10, and 20 cm (points) are shown. The frequency dependence of $C_2(\Delta v; R)$ is seen to be independent of R for 4 $cm \le R \le 20$ cm. This supports the conjecture that the spectral and spatial dependence of C_2 can be factorized as in Eq. (2). An analytic expression for $F_2(qL)$ has not been calculated theoretically. To obtain an analytic expression for $F_2(qL)$ to be used in a comparison of Eq. (2) to the measured intensity correlation function, we express F_2 as the sum $F_2(qL) = B_1F_1(qL) + B_2\overline{F}_2(qL)$, where B_1 and B_2 are constants and

$$\overline{F}_2(qL) = \frac{\left| (a/q) \left[\coth(qL) - qL/\sinh^2(qL) \right] \right|}{\coth(aL) - aL/\sinh^2(aL)}$$

is a normalized function that falls asymptotically as $\Delta v^{-1/2}$. The form of $F_2(qL)$ is suggested by the calculation by Feng *et al.*⁸ of the far-field intensity correlation function with angle. Apart from a small base-line shift, the points in Fig. 2 are fitted using the form given above for $F_2(qL)$ with $B_1=0.46$ and $B_2=0.54$. This fit is shown as the solid line in Fig. 2.

The autocorrelation function for a point at the output face of the sample with L = 150 cm averaged over 30000



FIG. 3. (a) Intensity autocorrelation function for a point at the output face of the 150-cm-long sample. (b) Intensity cross-correlation function for points on the output plane separated by 0.6 cm.

spectra is given by the circles of Fig. 3(a). The solid line is a fit of Eq. (2) to the data using the forms for $F_1(qL)$ and $F_2(qL)$ given above and $L_a = 25$ cm as determined from measurements of the exponential falloff of T(L). The fit gives $A_1 = 0.90$, $D = (3.0 \pm 0.2) \times 10^{10}$ cm²/s, and $A_2\delta^{-1} = 0.13$. Using this value of D, we find the halfwidth of $G^E(\Delta v)$ is $\delta v = 3.1 \pm 0.2$ MHz. The density of states per unit frequency in a sample of volume V is $dN/dv = (2k^2/\pi v)V$, where v is the wave velocity in the sample. For this sample dN/dv = 6.3 MHz⁻¹ and $\delta = (dN/dv)\delta v = 20 \pm 2$. The scattering is locally weak since $l = 3D/v = 4.0 \pm 0.2$ cm and kl = 23.¹³

 $C(\Delta v; R)$ for two ponts on the output face of the sample which are separated by R = 0.6 cm is shown by the circles in Fig. 3(b). A fit of Eq. (2) to the data using the values of A_1 and A_2 found above gives $H_1 = 0.008$ and $H_2 = 0.43$. The rapid falloff of H_1 is consistent with the calculation by Shapiro.¹⁸ On the other hand, the much



FIG. 4. Spatial dependence of the cross-correlation function.

slower falloff of $H_2(R)$ is a direct manifestation of the existence of long-range correlation.^{6,9}

The measured values of $C(R;\Delta v=0)$ between a point on the output surface and points on the axis of the cylinder for distances R > 2 cm into the sample are plotted as the points in Fig. 4. The predicted form of $C_2(R)$ is given by Eq. (1) as a function of x = R/L. Since all paths in the sample emerge either through the input or the output face, we have the condition

$$f(x) + f(1 - x) = 1.$$
(3)

f can be expanded as a polynomial in its argument, $f(x) = \sum_{i=0}^{\infty} a_i x^i$, with a linear term which is nonvanishing for diffusive transport. The sum of the left-hand side of Eq. (3) is independent of x only if all coefficients a_i with i > 1 vanish. Hence $f(x) = a_0 + a_1 x$. Since $f(1) = \gamma l/L$, and from Eq. (3), $f(0) = 1 - \gamma l/L$, we find

$$f(x) = (1 - \gamma l/L) - (1 - 2\gamma l/L)x.$$
(4)

 $C_2(R)$ is given by Eqs. (1) and (4). The value of γ can be determined from the slope of Fig. 4. We find $\gamma = 1.93$. The value of δ can be readily evaluated from this line since f(0.5) = 0.5 and $C_2(R = L/2) = 0.5\delta^{-1}$. This gives $\delta = 21 \pm 2$, in agreement with the value obtained from the measurement of C_1 in Fig. 3(a). From Eqs. (1) and (4), $C_2(R)$ is expected to extrapolate to zero at a distance $\approx \gamma l \approx 8$ cm beyond the sample, i.e., at L = 158 cm. The line in Fig. 4, however, extrapolates to zero at 168 cm. This discrepancy could be explained if one assumes that the data are the sum of $C_2(R)$ and a constant term equal to 0.0026. This constant term is of order δ^{-2} which is the expected value of C_3 . This conjecture is supported by the observation that the intensity cross-correlation function with frequency shift between points in the output plane of samples composed of a mixture of aluminum and Teflon spheres with $\delta \sim 10$ contains a constant contribution of magnitude δ^{-2} .

In conclusion, we have observed the leading terms in the intensity correlation function. The results support the conjecture that long-range correlation is a consequence of spectral correlation and that intensity correlation may be expressed in terms of statistical characteristics of the states of the random system.

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FIG. 1. Schematic diagram of the experimental setup.