Anomalous Photon Diffusion at the Threshold of the Anderson Localization Transition

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(Received 16 July 1990)

Microwave transmission through a random mixture of aluminum and Teflon spheres at a metal filling fraction of 0.30 is found to fall initially as $1/L^2$ in agreement with predictions of the scaling theory of localization. The renormalized diffusion coefficient is determined from measurements of transmission and of the spectral intensity correlation function. This function is found to be an expansion in δ^{-1}, where δ is the product of the width and density of states in the medium. From these results we find the Ioffe-Regel criterion for photons to be $kl_c = 1.6 \pm 0.4$.

PACS numbers: 42.20.−y, 72.15.Rn, 78.20.Dj

Understanding the localization transition has remained a challenge since Anderson first showed that a metal-insulator transition could be driven by disorder in the independent-electron approximation.[1-4] The electronic localization transition is complicated by the important role of electron correlations associated with the Mott transition and by a variety of inelastic processes which influence the temperature dependence of the conductance.[5,6] Anderson localization is the result of the constructive interference of backscattered waves which reduces transport from a point.[7,8] Since localization is a general wave phenomenon, rather than a specifically quantum-mechanical effect, it may also occur for classical waves such as sound and electromagnetic (EM) radiation in the presence of sufficiently strong scattering.[9-12] Because photons do not mutually interact, they should exhibit a pure Anderson transition.

In this Letter, we report the scale dependence of the effective diffusion coefficient $D(L)$,[3,10,12-14] as well as large intensity fluctuations and correlation in the critical regime of the Anderson localization transition for microwave radiation. The samples are composed of randomly positioned aluminum and Teflon spheres. Since their configuration can be changed, the ensemble average of local properties at a given length can be measured. The sample dimensions may be scaled while maintaining homogeneous structural disorder. From the results, we determine the minimum value of the transport mean free path for extended waves, l_c. We find kl_c is of order unity, where $k = 2\pi/\lambda$ is the wave number, as predicted by Ioffe and Regel.[15] More generally, we establish a universal criterion for localization in terms of the dimensionless ratio δ of the width $\delta\nu$ to the spacing $(d\nu/dN)$ of resonance modes of a finite random medium in the presence of inelastic processes.[2,16]

Weak localization[8] and the approach to localization have been observed in a number of recent optical and microwave experiments. (1) Coherent backscattering has been observed in the weak-scattering regime, $kl \gg 1$, as enhanced optical reflection in a narrow cone about the incident beam direction of angular width $\sim 1/kl$.[17-20] (2) The cumulant intensity correlation function with spatial separation and frequency shift has been found to be an expansion in the parameter δ^{-1}.[21-23] In the present study, in which $kl \sim 1$, we observe a large contribution to the spectral correlation function of order δ^{-2}, which is independent of frequency shift. The buildup of correlation is associated with the approach to localization which occurs when $\delta = \delta_c \sim 1$.[2,16] (3) The distribution function of the intensity for a single polarization component in the weak-scattering regime was found to change from negative exponential to stretched exponential as the length of a sample of constant cross section increases and δ decreases.[21] In the present study of strong scattering, we find that the intensity distribution is a stretched exponential even for $L \sim \lambda$. The enhancement of large fluctuations is a precursor to localization since the transmitted intensity for localized waves is exponentially small except when the incident wave is resonant with a localized state, in which case the transmission coefficient can be of order unity.[24,25] (4) Small values of the optical diffusion coefficient were observed in studies of pulse transmission in a random sample of titania spheres.[26] But in that case, the sphere diameter d was greater than λ and the wave may have been successively trapped in individual spheres. In the present study of metallic spheres with $d < \lambda/2$ this possibility is avoided.

In the critical regime, transmission is reduced by wave interference and can no longer be described in terms of an intensive diffusion coefficient $D = vl/3$, where v is the velocity of the wave in the medium. The breakdown of the diffusion approximation is parametrized by a scale-dependent diffusion coefficient such that the transmission coefficient $T(L)$ is given by the classical diffusion result with $D(L)$ substituted for D. For diffusive transport we find

$$T(L) = \sinh(\alpha a)/\sinh(\alpha L), \tag{1}$$

where $\alpha = (D\tau_a)^{-1}$ is the absorption coefficient, τ_a is the absorption time, $a = \gamma l = 3\gamma D/v$, and γ includes the internal reflection at the sample interface and is defined such that $T(L) = \gamma l/L$ for $L < L_a$.[21] In the strong-scattering regime, John[10] and Anderson[12] argued that in a one-parameter scaling theory, transmission is given by the classical result with the substitutions $\alpha \rightarrow [D(L)\tau_a]^{-1/2}$ and $a \rightarrow 3\gamma D(L)/v$. The effective diffusion coefficient for extended waves in the critical regime is[3,13]

$$D(L) = \frac{vl}{3}\frac{l}{\xi}, \tag{2}$$

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where ξ is given by

$$\frac{1}{\xi}=\frac{1}{\xi_0}+\frac{1}{L_a}+\frac{1}{L}, \tag{3}$$

and ξ_0 is the coherence length in an infinite sample in the absence of absorption,

$$\xi_0=l^2/(l-l_c). \tag{4}$$

Equation (3) does not correctly include the effects of sample length when L is approximately equal to both ξ_0 and l, since it then leads to the unphysical result $\xi<l$.

We have studied propagation of K-band microwave radiation in a sample which is a mixture of $\frac{3}{16}$-in. aluminum and Teflon spheres at a total volume filling fraction of 0.60 contained in a 7.3-cm-diam copper tube. The metallic volume filling fraction f is adjustable. New random configurations can be obtained by rotating the cylinder about its axis. The experimental setup has been described elsewhere.[21] Strong scattering is expected because the waves are confined to a tortuous network of narrow channels[12] and because the wave is near the dipole resonance of the aluminum spheres.[27] Because the wave can scatter freely in three dimensions on length scales greater than the sphere diameter d, we expect that for $L>l,d$ the scaling of transmission can be described by a universal theory in which details of the scattering mechanism do not appear.

$T(L)$ is obtained from the average intensity detected by a Schottky diode at the output of the sample as the sample is tumbled. The average of the intensity at a point of 6×10^4 sample configurations for each value of L, at 19 GHz, is shown in Fig. 1. The vertical scale is in arbitrary units because the absolute transmission was not determined. Previous optical[14] and microwave[21] measurements of $T(L)$ were consistent with the photon diffusion approximation [Eq. (1)] which gives $T(L)\sim1/L$ for $l<L<L_a$. This behavior is also observed in measurements made in a sample of aluminum spheres with $f=0.20$, as shown by the open circles in Fig. 1(a). In the critical regime, however, in which $(1/\xi_0+1/L_a)^{-1}\gg l$ we expect that initially $T(L)$ will fall as $D(L)/L\sim1/L^2$. Such behavior is observed for the sample with $f=0.30$ as shown by the solid circles in Fig. 1(a). The smaller value of L_a for $f=0.30$ ($L_a=2.1$ cm) as compared to that for $f=0.20$ ($L_a=5.6$ cm), seen in Fig. 1(b), is a consequence of the smaller renormalized diffusion coefficient and the smaller absorption time at $f=0.30$, where $\tau_a=3.0\times10^{-9}$ s as compared to $\tau_a=8.8\times10^{-9}$ s for $f=0.20$. This unexpected change in τ_a may represent a redistribution of EM energy in the medium due to coherent backscattering.

The values of D for $L>L_a$ are determined from an analysis of the cumulant intensity correlation function with frequency shift $C(\Delta\nu)$.[14,21,22] The results for $f=0.30$ are shown in Fig. 2 and discussed below. This gives $D=1.45\times10^9$ cm^2/s at $L=12$ cm. Using the value of D for $L>L_a$ and the form of Eq. (1), in which the diffusion coefficient is allowed to be scale dependent, the measurements in Fig. 1 are expressed in terms of $D(L)$ with the result shown in Fig. 3. A fit of Eqs. (2)–(4) to the measured values of $D(L)$ for $f=0.30$ using l and l_c as fitting parameters gives $l=0.45\pm0.05$ cm and $l_c=0.4\pm0.1$ cm and is shown by the curve in Fig. 3. This gives $\xi_0=4.0$ cm and $\xi=1.2$ cm at $L=12$ cm. These results indicate a reduction of the diffusion coefficient from the classical prediction by a factor of $\xi/l=2.7$. A comparison of the scaling theory to measurements at $L=1$ cm at which $L\sim\lambda$ and $L\sim2d$ may appear to stretch the theory. However, the requirement of the scaling theory that δ not vary in the critical regime on length scales $L<\xi$ and that δ_c is a fixed point can only be achieved if $D(L)$ approximately follows Eqs. (2)–(4) once $L>l$.

An analysis of the results in Fig. 3 for $f=0.20$ using Eqs. (2)–(4) and the value $l_c=0.38$ cm obtained by

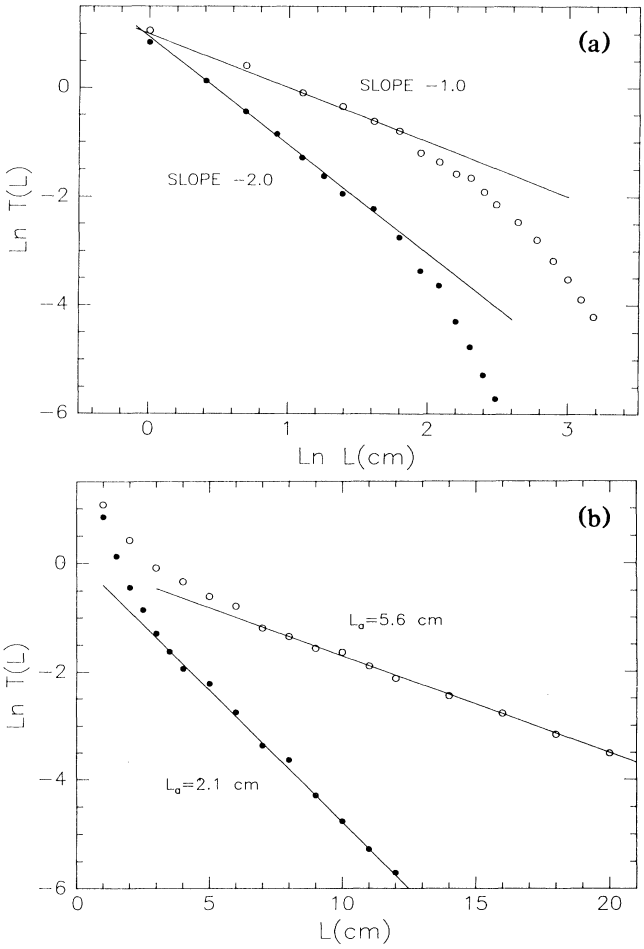

FIG. 1. (a) Log-log plot of transmission vs thickness at $\nu=19$ GHz for a random sample of aluminum spheres. For $L<L_a$, $T(L)\sim L^{-1}$ in a sample with a filling fraction $f=0.20$ (open circles), while $T(L)\sim L^{-2}$ for a sample with $f=0.30$ (solid circles). (b) Semilogarithmic plot of $T(L)$ for the two samples in (a).

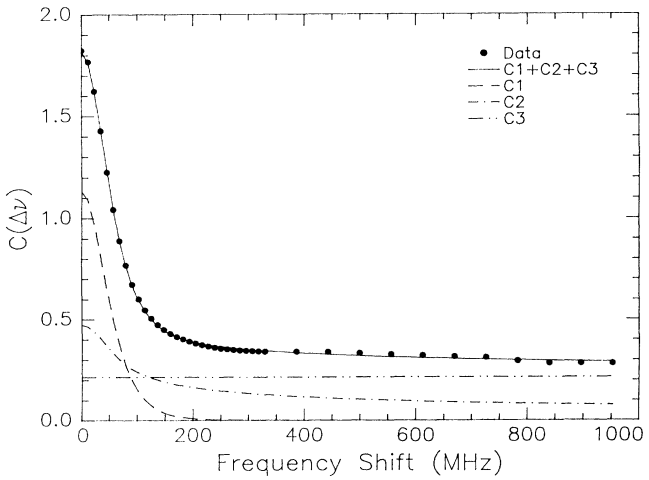

FIG. 2. The points represent the average of the cumulant autocorrelation function for 4000 sample configurations for $L=12$ cm at $f=0.30$. The decomposition of $C(\Delta\nu)$ into its components C_1, C_2, and C_3 is explained in the text.

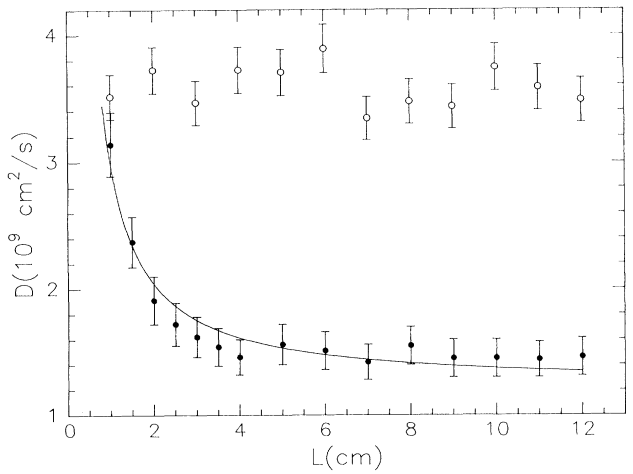

FIG. 3. Scale dependence of the diffusion coefficient $D(L)$ for the two samples in Fig. 1. The solid line is a fit by Eqs. (2)–(4) to the data.

modifying the result for $f=0.30$ to reflect the change in f, as discussed below, gives $l=0.7\pm0.2$ cm, $\xi_0=1.5$ cm, and $\xi=1.1$ cm at $L=12$ cm. The renormalization factor for D in the $f=0.20$ sample is $\xi/l=1.5$. Most of the reduction in D occurs for $L<2$ cm. The reduction for $L\geq 2$ cm is within the experimental uncertainty. We note that Eq. (3) does not apply for $L=1$ cm because $L\sim\xi_0\sim l$ in this case.

We note that alternative explanations of these results such as Faraday-cage effects as well as purely topological considerations would lead to a monotonic decrease of D with increasing values of f. We find, however, that though the value of D decreases to 0.60×10^9 cm^2/s at $f=0.35$, it increases for higher values of f. D is 0.90×10^9 cm^2/s at $f=0.40$ and 1.3×10^9 cm^2/s at $f=0.45$. The dependence of D upon f is consistent with the interpretation that the sample at $f=0.30$ is near a mobility edge separating extended waves from a narrow window of localized states in which the renormalization of the diffusion coefficient is limited by L_a.

There has been considerable theoretical and experimental effort to ascertain the value of kl_c in electronic systems.[28] The analogous quantity for photons can be obtained for the $f=0.30$ sample by evaluating $k=2\pi\nu/v$. From Eq. (2) we have $v=3\xi D/l^2=2.6\times10^{10}$ cm/s. This is in good agreement with the geometric-optics estimate for v given by

$$v=c[(0.40/0.70)+(0.30/0.70)n]^{-1}=2.5\times10^{10}\ \text{cm/s},$$

where c is the speed of light in air, $n=1.45$ is the index of refraction of Teflon, and 0.40 and 0.30 are the volume fractions of air and Teflon, respectively. This confirms that the sample is truly disordered and that the velocity is not reduced by backscattering in a nearly periodic structure as might occur near a photon band gap. Using the value of v gives $k=4.7$ cm^{-1} and $kl_c=1.9\pm0.5$ in this sample.

The critical value of kl depends upon the density of states and strictly applies to infinite systems in which inelastic scattering is absent. A universal condition for localization, which holds for any wave for any combination of internal structure, reflectivity at the boundary, and size can be obtained, however, by considering the scaling behavior of $\delta(L)=(dN/d\nu)\delta\nu(L)$.[2,3,16]

δ has the same scaling behavior as $\delta\nu$ except for the factor of $dN/d\nu$ which scales as the volume V. $\delta\nu$ may be defined as the half-width of the field-field correlation function,[21,22,29] $G^E(\Delta\nu)=\langle E(\nu)E(\nu+\Delta\nu)\rangle$. In the absence of absorption, the half-width of $G^E(\Delta\nu)$ is $\delta\nu=0.96D/L^2$. In the critical regime, we expect that $\delta\nu=0.96D(L)/L^2$ in the absence of absorption.[16]

The value of $\delta\nu(L)$ and hence of δ at the mobility edge can be inferred from the value of kl_c, which gives the localization condition in a sample with the same internal structure as the one we have studied, but in which the volume V diverges and absorption is absent. For such a sample, $\xi=L$ and Eq. (2) gives $D_c(L)=vl_c^2/3L$. The density of states per unit volume is $dn/d\nu=2k^2/\pi v$. At the mobility edge δ is given by $\delta_c=(dn/d\nu)V[0.96D_c(L)/L^2]$, where V is the volume which contains typical paths traversing the sample. For the present case in which photons are excluded from the metallic regions of the sample we have $V=L^3(1-f)$. This gives the universal criterion for localization $\delta_c=0.20(1-f)(kl_c)^2$. Using the experimentally determined value of kl_c for our sample, we obtain $\delta_c=0.5\pm0.2$. The Ioffe-Regel criterion for photons is, therefore, $kl_c=[\delta_c/0.20(1-f)]^{1/2}\approx1.6/(1-f)^{1/2}$.

The value of $D(L)$ at $L=12$ cm for $f=0.30$ is obtained from the measurement of the intensity correlation function, $C(\Delta\nu)=\langle\delta I(\nu)\delta I(\nu+\Delta\nu)\rangle$, where $\delta I(\nu)$ is the fractional fluctuation of intensity from the ensemble-

average value. The points shown in Fig. 2 are the average of the correlation function of intensity spectra normalized by the average of all spectra taken, from 18.5 to 19.5 GHz for 4000 sample configurations. The correlation function is presented in the figure as the sum of the three leading terms of an expansion in the correlation parameter δ^{-1}, $C=C_1+C_2+C_3$.[22,30] $C_1(\Delta\nu)$ is obtained by factorizing the fields, $C_1(\Delta\nu)=|\langle\tilde{E}(\nu)\tilde{E}(\nu+\Delta\nu)\rangle|^2$, where $\tilde{E}$ is the complex field.[21,29,31,32] It does not include the influence of spatial intensity correlation. Because ξ is limited by L_a, the frequency dependence of C_1 is still given by the photon diffusion model for $L>L_a$, but with the renormalized diffusion coefficient.[29] The functional form of $C_2(\Delta\nu)$ was found from measurements of the cross-correlation function for two detectors on the output face of the sample separated by 1.9 cm.[22] In agreement with recent predictions,[30] the contribution of C_3 to both the autocorrelation and cross-correlation functions is found to be independent of frequency. When $L>L_a$, the effective diffusion coefficient is found by fitting $C(\Delta\nu)$ by the sum of the three terms using the value of α determined from measurement of $T(L)$ shown in Fig. 1 and using D and the amplitudes of the three terms as fitting parameters. The fit gives the contributions of the three terms as shown in Fig. 2 and $D=1.45\times10^9$ cm^2/s. This value of D is smaller than the classical value $D=vl/3=3.8\times10^9$ cm^2/s by a factor of 2.6. This is consistent with the degree of renormalization found above for the scale dependence of transmission. Using the measured values of D and α in Eq. (5) gives $\delta\nu=22.5$ MHz. The density of states is given by $dN/d\nu=(2k^2/\pi v)(1-f)V$. For $L=12$ cm, $dN/d\nu=2\times10^{-7}$ s, this gives $\delta=4.5$.

We note that the variance of the intensity is given by $\mathrm{var}(I)=C(\Delta\nu=0)=1.8$. This is in contrast to results in the diffusive regime, in which we find $\mathrm{var}(I)=1.0$.[21] The larger variance here is associated with large intensity fluctuations which are manifest in the distribution of intensities $P(I)$. We find that $P(I)\sim\exp(-I^{0.5})$ as compared to negative exponential statistics, $P(I)=\exp(-I)$, found in the weak-scattering regime.[21]

In conclusion, we have observed suppressed scale-dependent transmission and enhanced correlation and fluctuations of microwave radiation in the critical regime of the Anderson localization transition. The proximity to the mobility edge is determined from measurements of the scale-dependent diffusion coefficient and of spectral correlation. Our results confirm that the scaling theory of localization holds in the case in which giant intensity fluctuations occur and in which the functional form of the intensity distribution changes as the sample size increases. The results allow us to determine the Ioffe-Regel criterion for photons.

We thank W. Polkosnik for assistance with the data analysis and E. Kuhner for technical assistance. The work was supported by the NSF under Grant No. DMR-9001335 and by two Professional Staff Congress–City University of New York awards.

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