Observation of Photon Localization in a Three-Dimensional Disordered System

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The diffusion and absorption coefficients of microwave radiation in random mixtures of metallic and dielectric spheres are determined from measurements of the intensity correlation function with frequency shift and of the scale dependence of transmission. We find a narrow window of localization in frequency and metallic concentration. The transition is rounded by the presence of absorption.

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The particle diffusion picture for wave propagation within a random medium breaks down[1] when the probability of a scattered wave returning to a coherence volume in the sample approaches unity. Constructive interference of time-reversed backward-scattered waves then suppresses transport and gives rise to localization.[2,3] In three dimensions, localization due to disorder can only occur when the wave is scattered within a coherence length $l < 1/k$, where l is the transport mean free path and $k = 2\pi/\lambda$ is the wave number. This gives the Ioffe-Regel criterion for electron localization, $kl < 1$.[4] Since localization is a wave interference phenomenon, it may occur in principle for classical waves[5-11] as well as for quantum-mechanical electron waves, where it is widely observed.[12] Recent measurements of the scale dependence of the effective diffusion coefficient of microwave radiation through random metallic samples suggest that photon localization can be achieved when $kl < 1.6/(1-f)^{1/2}$, where f is the fraction of the total sample volume from which the wave is excluded by metallic surfaces.[13] In this Letter, we report the first observations of photon localization in a random three-dimensional sample. Localization of microwave radiation is found in a narrow range of frequency and metallic concentration in random close-packed mixtures of $\frac{3}{16}$-in. aluminum and Teflon spheres. Whereas electron localization occurs as a result of the combined effects of disorder scattering, associated with the Anderson transition, and of electron correlation, associated with the Mott transition, the localization of electromagnetic (EM) radiation is a pure Anderson transition since photons do not mutually interact. Moreover, because intensity measurements can be made in an ensemble of samples, the microscopic statistical character of the transition can be determined.

There has been considerable interest in specifying the structural characteristics which lead to localization since Anderson first showed that electron localization could be the reason for the vanishing conductivity of glasses as $T \rightarrow 0$.[1] Using an independent-electron, tight-binding model on a periodic lattice with diagonal disorder, Anderson found that the entire band is localized when the diagonal disorder is greater than the coupling between sites. Generally, however, electronic localization is found in systems in which the density of states is low, such as in impurity bands of lightly doped semiconductors and in the pseudogap of amorphous semiconductors.[12] Photon localization has proven to be harder to achieve. The tight-binding model is not applicable because photons cannot be bound to a single particle. Photon-photon interactions are not available to facilitate localization. The density of EM states in random samples is only low at long wavelength. But in this case, the wavelength is much greater than the scale of dielectric fluctuations d and scattering is weak. It is described by Rayleigh scattering for isolated particles and by effective-medium theory for a high density of scatterers. The strongest scattering for samples with positive dielectric function occurs for $d \sim 1/k$. But localization can then occur only if the scattering cross section is significantly greater than the geometrical area.

An alternative scattering system in which the wave propagates between metal spheres is considered here.[7-9] Anderson suggested that localization could be achieved in a random tangle of interconnected metallic waveguides.[8] Strong scattering is anticipated when microwave cutoff is approached as π/k becomes comparable to the channel diameters. The continuous pore network accessible to the wave in the present sample may be similar to such structures and the scattering strength may increase as the ratio of the spacing between spheres to the wavelength decreases over some range. However, a turn around in scattering strength may occur as this ratio decreases beyond some point because the increasing coherent coupling between neighboring channels enhances the effective channel diameter. Another approach to this problem has been taken by Condat and Kirkpatrick.[9] Using a pseudosphere approximation, they predicted that maximum scattering occurs as the wavelength approaches the sphere resonance and that localization occurs in random samples of metallic spheres above a critical density sufficiently near resonance. We expect that peak scattering occurs when the requirements given above for the relationship of the wavelength to both the pore width and sphere diameter hold simultaneously. A window of localization may occur since scattering is weakened in the limit of long wavelengths

and high metallic density, by effective-medium corrections, as well as in the short-wavelength limit in which geometric optics may be applicable.

Strictly speaking, waves can only be localized by disorder in an infinite medium in which inelastic processes are absent. Only then will the wave not leak out of the sample, lose phase coherence, or be absorbed. Localization can nonetheless be defined in real systems by the condition that the universal scaling parameter[14,15] δ, which is the product of the level width and the density of states $\delta=\delta\nu dN/d\nu$,[16] is less than a critical value δ_c for some sample length L. δ represents the typical number of modes of the sample within the level width. The level width is the field-field correlation function with frequency shift of the transmitted radiation.[16,17] It is related to the diffusion coefficient D since the field correlation function with frequency shift is the Fourier transform of the time-of-flight distribution.[17] For $\delta\gg\delta_c$ and L smaller than the absorption length, given by $L_a=(D\tau_a)^{1/2}$, where $1/\tau_a$ is the photon absorption rate, $\delta\nu=0.96D/L^2$.[17] However, if $\delta\leq\delta_c$ on some length scale of the medium, wave transport is reduced as a result of coherent backscattering on all length scales.[2,3] Propagation then cannot be described in terms of an intensive diffusion coefficient. However, propagation can be described in terms of a scale-dependent, effective diffusion coefficient $D(L)$. For example, for $L<L_a=[D(L)\tau_a]^{1/2}$, the level width is given by $\delta\nu=0.96D(L)/L^2$. For extended waves, the scaling theory of localization gives[6,8,15]

$$D(L)=(vl/3)l/\xi\,, \tag{1a}$$

$$1/\xi=1/\xi_0+1/L_a+1/L\,. \tag{1b}$$

To leading order in an ϵ expansion in $2+\epsilon$ dimensions, $\xi_0=l^2/(l-l_c)$.[7] In this Letter, localization is determined from a comparison of the measured diffusion coefficient $D(L)$ to the effective diffusion coefficient $D_c(L;\tau_a)$ that the sample would have if it were at the localization threshold with the absorption rate measured for the sample. At the mobility edge $l=l_c$ and ξ_0 diverges. Hence, using Eq. (1), the implicit dependence of D_c upon $1/\tau_a$ is given by

$$D_c(L;\tau_a)=(vl_c^2/3)(1/L+1/L_a)\,. \tag{2}$$

At this point $\delta=\delta_c$ is seen to be constant for $L<L_a$, since $dN/d\nu\sim L^3$ and $\delta_c=[0.96D_c(L)/L^2]dN/d\nu$. In samples in which $\delta<\delta_c$ and $L<L_a$, δ decreases exponentially with L whereas in samples in which $\delta>\delta_c$, δ increases with L.[15] For $L>L_a$, however, $D(L)$ approaches a constant value and δ increases with L. The Anderson transition is therefore rounded by the presence of absorption. The existence of scaling behavior, which is consistent with localization for $L<L_a$, can nonetheless be determined in samples for which $L>L_a$, by the condition $D(L)<D_c(L;\tau_a)$. This condition can only be satisfied if $\delta<\delta_c$ for $L<L_a$. In such samples $l<l_c$.

The samples studied here are contained in a 7.3-cm-diam copper tube at a sphere filling fraction of 0.60. The radiation from a tunable K-band oscillator is launched from a horn placed 20 cm in front of the sample. Two Schottky diode detectors are placed 2.5 cm apart at the output face of the sample. $T(L)$ is obtained from the average value of the intensity measured by the diodes while the sample is tumbled as the cylinder is rotated about its axis. 30000 readings are taken at each thickness at intervals of the intensity correlation time which is of order 10 msec. $C(\Delta\nu)$ is obtained from the average of the intensity correlation function of 4000 spectra which are normalized by the average of these spectra. New configurations are obtained by rotating the tube momentarily after each spectrum is taken.

The value of $D(L)$ is determined from measurements of the cumulant intensity correlation function with frequency shift, $C(\Delta\nu)=\langle\delta I(\nu)\delta I(\nu+\Delta\nu)\rangle$, where the intensity $I(\nu)$ is normalized to the ensemble-average intensity at the frequency ν and $\delta I(\nu)=I(\nu)-1$ is the fractional intensity fluctuation from the average.[18,19] The three leading terms in the expansion of $C(\Delta\nu)$ in terms of the correlation parameter δ^{-1} are observed and distinguished by their different dependence upon frequency shift.[19] In terms of functions $F_i(\Delta\nu)$, which are normalized at $\Delta\nu=0$, the correlation function can be expressed as

$$C(\Delta\nu)=\sum_{i=1}C_i(\Delta\nu)=\sum_{i=1}A_i\delta^{1-i}F_i(\Delta\nu)\,. \tag{3}$$

The leading term is given by factorizing the complex fields $C_1=|\langle E(\nu)E^*(\nu+\Delta\nu)\rangle|^2$.[20,21] For diffusive transport,[17,19,22]

$$F_1(\Delta\nu)=\frac{|\sinh(qa)/\sinh(qL)|^2}{[\sinh(\alpha a)/\sinh(\alpha L)]^2}\,, \tag{4}$$

where q is the root with negative imaginary part of $q^2=\alpha^2+i2\pi\Delta\nu/D$, $a=\gamma l$, $\alpha=L_a^{-1}$, and γ is a number of order unity which includes the effect of internal reflection at the boundaries of the sample. For $L>L_a$, Eq. (4) holds even in the critical regime with the substitutions $a=\gamma l\rightarrow 3\gamma D(L)/v$ and $\alpha\rightarrow\alpha(L)=[D(L)\tau_a]^{-1/2}$. $C_3(\Delta\nu)$ is independent of $\Delta\nu$, giving $F_3(\Delta\nu)=1$.[13,23] $F_2(\Delta\nu)$ is found from measurements of the cross correlation function of intensity at two points at a separation $R\approx 10/k$ on the output face of the sample.[19] C_1 in this case does not contribute significantly to the intensity cross correlation function since it depends upon the field correlation function which falls in a coherence length $1/k$.[19,20] We find that the cross correlation function can be expressed as the sum of a constant background and a term which is found to fall asymptotically as $\Delta\nu^{-1/2}$ and which we associate with $F_2(\Delta\nu)$.[19,21,23] $D(L)$ for $L\gg L_a$ is obtained from a fit of Eq. (3) to the measured intensity autocorrelation function, using the value of α obtained from measurements of $T(L)$, and utilizing $D(L)$ and the $A_i\delta^{1-i}$ as fitting parameters.

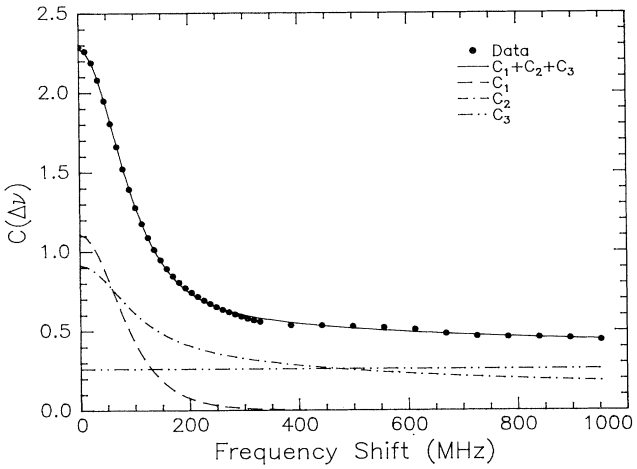

FIG. 1. Cumulant intensity correlation function with frequency shift in the range $\nu=18.5$–19.5 GHz for $f=0.35$. The solid line is a fit to the data of the sum of the first three terms in a perturbation expansion for $C(\Delta\nu)$ [Eq. (3)]. The contribution of each of these terms is also shown.

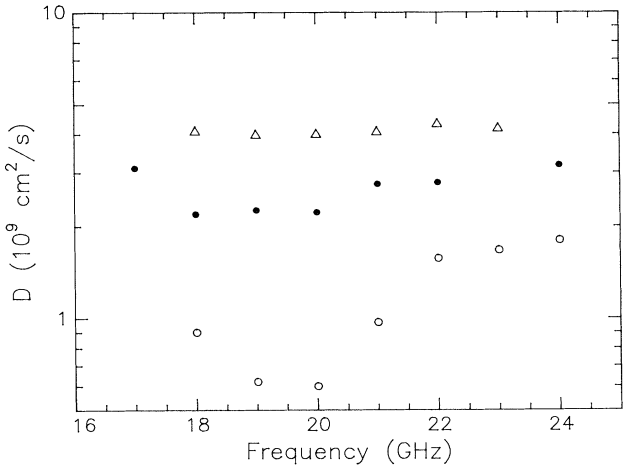

FIG. 2. The frequency dependence of D for $f=0.20$ (△), $f=0.25$ (●), and $f=0.35$ (○). D is determined from measurements of $C(\Delta\nu)$ at $L=24$, 12, and 6 cm for $f=0.20$, 0.25, and 0.35, respectively.

The strongest scattering is observed at a metallic filling fraction $f=0.35$ and $\nu=19$ GHz. At this frequency, the exponential tail of $T(L)$ gives $\alpha=0.98\pm0.1$ cm^{-1}. The corresponding value of D for $L=6$ cm is determined from the measurement of $C(\Delta\nu)$ in the frequency range 18.5–19.5 GHz. The results are shown as the points in Fig. 1. A fit of Eq. (3) to the data, using Eq. (4) for F_1 and the measured value of α, gives the three contributions to $C(\Delta\nu)$ as shown in Fig. 1 and $D=(6.0\pm1.0)\times10^8$ cm^2/s. Using the values of α and D obtained for this sample gives $\tau_a=1.7\times10^{-9}$ s. The relation[10] $1/v=[0.40/c+(0.60-f)n/c]/(1-f)$, where 0.40 is the volume fraction of air, $0.60-f$ is the volume fraction of Teflon, c is the speed of light in air, and $n=1.44$ is the index of refraction of Teflon, gives $v=2.57\times10^{10}$ cm/s. The value of v obtained from this relation for a sample with $f=0.30$ was found to agree with the value obtained from D and l which were determined independently in that sample.[13] Using this value of v gives $k=2\pi\nu/v=4.7$ cm^{-1} and $l_c=1.6/k(1-f)^{1/2}=0.42$ cm. From Eq. (2), $D_c(L;\tau_a)=1.3\times10^9$ cm^2/s. Since $D(L)<D_c(L;\tau_a)$, we conclude that the wave is localized. In the absence of absorption in a sample with equivalent scattering strength $D(L)$ would vanish as $L\to\infty$. In the present case, D approaches an asymptotic value for $L\gg L_a$, because coherent interference is cut off on length scales greater than L_a.

The occurrence of localization is also established here from measurements of the absorption coefficient. These results are in agreement with predictions of John[6,7] and Anderson.[8] Empirically we find that the transmission coefficient for diffusive transport for $L>l$ is given by[13,18]

$$T(L)=\sinh(\alpha a)/\sinh(\alpha L). \quad (5)$$

This result is predicted to hold as well in the presence of strong scattering if we make the same substitutions for a and α which are made in Eq. (4).[6,8] For $L>L_a$, $T(L)$ falls exponentially. For localized waves, $\alpha>\alpha_c=[D_c(L;\tau_a)\tau_a]^{-1/2}$.[6] For this sample at 19 GHz we find $\alpha_c(L;\tau_a)=0.70$ cm^{-1}. The fact that the measured α is greater than $\alpha_a(L;\tau_a)$ confirms that the wave is localized. Because of the small value of D when the wave is localized, L_a is as short as the wavelength itself. Nonetheless, in our sample D is reduced from the classical diffusion value, given by $D=vl/3\sim vl_c/3\approx3.4\times10^9$ cm^2/s, by a factor of ~6.

The variation of D and α with ν and f is shown in

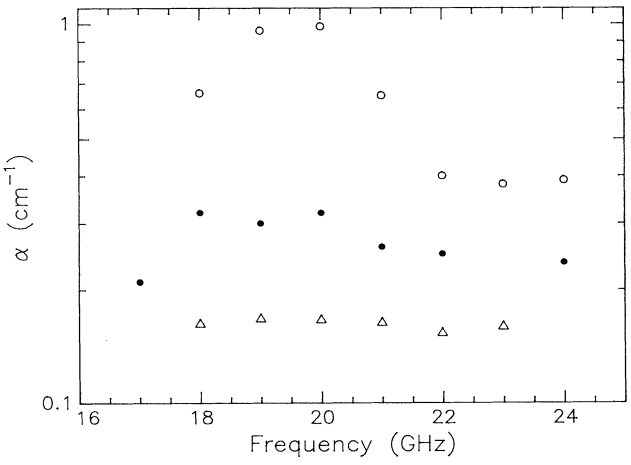

FIG. 3. The frequency dependence of α for $f=0.20$ (△), $f=0.25$ (●), and $f=0.35$ (○). α is the absorption coefficient for the transmission for $L>L_a$.

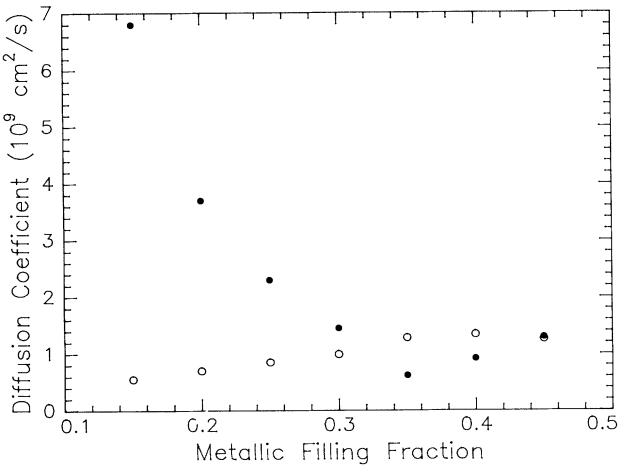

FIG. 4. A comparison of the measured value of D (●) and the critical value D_c calculated using the measured absorption rate $1/\tau_a$ (○).

Figs. 2 and 3, respectively. For $f=0.20$, D and α are independent of frequency. $T(L)$ is given by Eq. (5) for $L>1$ cm using a scale-independent value of D. For $f>0.20$, we observe a dip in D and a peak in α centered at $\nu\sim19.5$ GHz. At $f=0.30$ and $\nu=19$ GHz, $D(L)$ was found to have a form close to that of Eq. (2), indicating that this point is close to the mobility edge with $l\sim l_c\sim 1/k(1-f)^{1/2}\sim 0.4$ cm.[13] For $f=0.35$ at $\nu=19$ and 20 GHz, we find $D(L)<D_c(L;\tau_a)$ and $\alpha(L)>\alpha_c(L;\tau_a)$ at $L=6$ cm, indicating that the wave is localized. In this case D is smaller than near the mobility edge at $f=0.30$ even though L_a is smaller. This is inconsistent with predictions for extended waves [Eq. (1)] and can only occur for localized waves. A rounded localization transition as a function of f at $\nu=19$ GHz is seen in Fig. 4 in a comparison of $D(L)$ and $D_c(L;\tau_a)$. Structure in the frequency dependence of D and α occurs only when D becomes scale dependent and when $l\sim l_c$. This suggests that the structure is not ascribable to an appreciable change in l with frequency but rather to a large fractional change in $l-l_c$ as the localization threshold is approached.

In conclusion, we have observed a narrow window of localization for EM radiation as the frequency and density of metallic spheres is varied. The extent of renormalization of D and the sharpness of the transition are limited by absorption in the medium. The results are consistent with the scaling theory of localization when the influence of absorption is included.

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