Breakdown of Diffusion in Dynamics of Extended Waves in Mesoscopic Media

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We report the observation of nonexponential decay of pulsed microwave transmission through quasi-one-dimensional random dielectric media that signals the breakdown of the diffusion model of transport for temporally coherent extended waves. The decay rate of transmission falls nearly linearly in time due to a nearly gaussian distribution of the coupling strengths of quasi-normal electromagnetic modes to free space at the sample surfaces. The peak and width of this distribution scale as $L^{-2.05}$ and $L^{-1.81}$, respectively.

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The diffusion model is widely applied to electronic, neutron, and thermal conduction as well as to acoustic and electromagnetic propagation in multiply scattering media. The model is used not only when the phase of the wave is scrambled by inelastic scattering, but also when the wave is temporally coherent in mesoscopic samples [1, 2]. Although wave interference leads to large intensity fluctuations within a particular sample, the ensemble average of the flux reaching a point is generally assumed to be the incoherent sum of contributions of randomly phased, sinuating Feynman paths reaching that point. In this model, the average intensity varies smoothly in space and time and is governed by the diffusion equation.

Diffusion has been taken as the counterpoint to wave localization [2, 4, 5]. On one side are sharply defined localized modes with the average level spacing $\Delta \nu$ exceeding the typical level width $\delta \nu$ and on the other are the quasi-normal modes in a random ensemble should be a continuum. As time progresses, long-lived quasimodes would contribute more substantially and the rate of flow out of the sample would slow down continuously. Nonexponential decay has been observed in acoustic scattering in reverberant rooms [6] and solid blocks [7] as well as in microwave scattering in cavities whose underlying ray dynamics is chaotic [8]. Similarly, the decay rate of electronic conductance has been predicted to fall as a result of the increasing weight of long-lived, narrow-linewidth states [4]. The leading correction to the diffusion prediction for the electron survival probability $P_s(t)$ was calculated by Mirlin [10] using the supersymmetry approach [11] to be, $-\ln P_s(t) = (t/\tau_1)(1-t^2/2\pi^2g\tau_1)$, where $g$ is the dimensionless conductance, which can be expressed as $g = \delta \nu/\Delta \nu$.

An ideal way to investigate the applicability of the diffusion model to mesoscopic systems is to consider pulsed electromagnetic transmission. Previous studies [12, 13, 14, 15] have found exponential decay for $t > \tau_1$, as predicted by diffusion theory. However, measurements of optical transmission [14] indicate that the pulse rises earlier than predicted by diffusion theory. Even more puzzling is the finding by Kop et al. [15] of an increase in the inferred value of the diffusion coefficient with increasing $L$.

In this Letter, we report a dramatic breakdown of diffusion in microwave measurements in nominally diffusive random samples for which $\delta \nu > \Delta \nu$. We find that the decay rate of pulsed transmission falls nearly linearly in time, as predicted by Mirlin [10]. These results are interpreted in terms of the distribution of decay rates of quasimodes of the sample, which is found by taking the inverse Laplace transform of the decaying signal. The distribution of the modal decay rates is nearly gaussian, with an average value that scales as $L^{-2.05}$, which is close to the inverse square scaling for the decay rate in the diffusion model. The width falls as $L^{-1.81}$, which is faster than predicted by Ref. [10].

Spectra of the in- and out-of-phase components of the steady-state transmitted microwave field are measured in low-density collections of dielectric spheres using a Hewlett-Packard 8772C vector network analyzer. These spectra are multiplied by a gaussian envelope of width $\Delta f$ centered at $f_c$ and then Fourier-transformed to give

\[ H(f) = 11.8 f_c \]
the response to a gaussian pulse in the time domain. Circular horns are positioned 30 cm in front of and behind the sample. Linearly polarized microwave radiation is launched from one horn and the cross-polarized component of the transmitted field is detected with the other to eliminate the ballistic component of radiation. The sample is composed of alumina spheres of diameter 0.95 cm and refractive index 3.14, contained within a 7.3-cm-diameter copper tube at an alumina filling fraction of 0.068. This low density is produced by embedding the alumina spheres in Styrofoam spheres of diameter 1.9 cm and refractive index 1.04. Measurements for random ensembles are obtained by momentarily rotating the tube about its axis to create new random configurations before each spectrum is taken. In this way, measurements are carried out in ensembles of 10,000 sample realizations at lengths of 61, 90, and 183 cm. In addition, measurements are made in an ensemble of 2,300 realizations of a more strongly absorbing sample of 90 cm length (L = 90 cm) produced by covering 40% of the inside surface of the tube with a thin strip of titanium foil laid from end to end. The measurements are made within the frequency interval 14.7-15.7 GHz for L = 61, 90 and 90* cm and 15.0-15.4 GHz for L = 183 cm. The frequency intervals are chosen to be far from sphere resonances, so that the dynamics of transmission is uniform over the frequency range and the sample is far from the localization threshold. The closeeness to localization, even in the presence of absorption, is indicated by the variance of the steady-state transmitted intensity normalized to its ensemble average value, var(I/⟨I⟩) [17]. In the absence of absorption, var(I/⟨I⟩) ≈ 1 + 4/3g [17,18]. At the localization threshold, var(I/⟨I⟩) ≈ 7/3. The values of var(I/⟨I⟩) in the samples of L = 61, 90*, 90, and 183 cm are 1.18, 1.25, 1.26, and 1.50, respectively.

The envelope of the temporal response to a gaussian pulse peaked at t = 0 is squared to give the transmitted intensity I(t) for a particular sample realization. The average transmitted intensity ⟨I(t)⟩ is found by averaging over the ensemble, and then over the frequency interval by shifting f. The results are shown on a logarithmic scale in Fig. 1a. We find that, when Δf > δν, the tail of ⟨I(t)⟩ does not depend on Δf. We use Δf = 15 MHz for L = 61, 90, and 90* cm and Δf = 7.5 MHz for L = 183 cm, so that Δf > δν in all cases. The noise in the frequency spectra produces a constant background intensity in the time domain. Once this background is subtracted, a dynamic range of more than 6 orders of magnitude is achieved. This makes it possible to study transmission on time scales an order of magnitude longer than the time of peak transmission, though the longest times are still smaller than the inverse level spacing, 1/Δν, which gives the time required for a photon to explore each coherence volume of the sample. The measured decay rate due to leakage out of the sample and absorption, γ = −d ln⟨I(t)⟩/dt, is plotted in Fig. 1b. This rate is not constant as predicted by diffusion theory, but falls nearly linearly with time. The constant increase in the decay rate for the L = 90* sample over that for the L = 90 sample, seen in Fig. 1b, indicates that the absorption rate 1/τa is constant in time. The decrease in γ with time is thus attributable solely to propagation out of the sample, which proceeds at a rate, π²D(L+2z₀)². The absorption rate is found from a fit of the diffusion model to measurements of ⟨I(t)⟩ up to the time at which 95% of the full pulse energy has been transmitted (Fig. 2). The fit is obtained by taking D(t) to be a constant, D*, and by minimizing the parameter χ² = (σ²)⁻¹ ∑(|I(t)|i − I(t₀))², where ⟨I(t₀)⟩i are the values of the measured intensity, σ is the uncertainty in I(t₀)i, averaged over the time of the fit, and I(t₀) are the values of the model intensity calculated at t₀. The parameters D*, τa, and z₀ obtained from the fit are listed in Table I and the corresponding decay rates are shown by thin solid lines in Fig. 1b. For L = 61 cm, χ² at the minimum depends only weakly on τa since the temporal range used in the fit is smaller than the τa. For this reason, we use the value τa = 97 ns obtained in the fit to the data for L = 183 cm for this length.

The diffusion coefficient D* obtained from the fit is found to decrease slightly with increasing L. The equal-
decrease in the slope of the curves do not extrapolate to a constant bare diffusion coefficient, $D$, as shown in Table I, which indicates that these parameters are not sensitive to the absorption rate. Moreover, when $1/\tau_\alpha$ is subtracted from $\gamma$ to give the decay rates without absorption in Fig. 3a, the same time-dependent decay is found for both samples with length of 90 cm.

The leakage rates in Fig. 3a give the “time-dependent diffusion coefficient”, $D(t) = (\gamma - 1/\tau_\alpha)(L + 2z_0)^2/\pi^2$. The values of $D(t)$ found from the measurements may be compared to those from the theory of Ref. [10] for the survival probability, which yields $D(t) = D(1 - t^2/\pi^2g\tau_1 + \ldots)$. This is done by plotting $D(t)$ as a function of the dimensionless time $t' = t/g\gamma_1$ in Fig. 3b. The time $g\gamma_1$ is proportional to $1/\Delta\nu$ and all the data are predicted to fall on a single curve. Although the data for samples with different values of $L$ at a given time appear to coincide within the noise, there is a clearly discernable decrease in the slope of $D(t')$ as $L$ increases. As a result, the curves do not extrapolate to a constant bare diffusion coefficient at $t = 0$. When $D(t)$ is plotted instead versus the dimensionless time $t'' = t/\sqrt{g\gamma_1}$ in Fig. 3c, the slope of $D(t'')$ appears to be the same for all values of $L$, though the curves do not overlap. A strong deviation from exponential decay at $t'' \approx 1$ has also been found in numerical simulations [20].

The changing slope of $D(t)$ with $L$ is associated with the scaling of the width of the distribution $P(\alpha)$ of decay rates of quasi-normal modes of the sample. These are hypothesized to form a complete set [19], even when $\delta\nu > \Delta\nu$. Since the time evolution is given by the superposition of these modes [19], the average transmission can be expressed as $\langle I(t) \rangle \propto \int_0^\infty P(\alpha) \exp(-\alpha t) d\alpha$, provided the mode coupling is, on the average, independent of $\alpha$. To find $P(\alpha)$, we use an approximate Laplace

### Table I: Values of the diffusion coefficient $D^*$, absorption time $\tau_\alpha$, and extrapolation length $z_0$, obtained from fitting Eq. (1) of [14] to the short-time transmitted intensity in Fig. 2.

| $L$ (cm) | $D^*$ (cm$^2$/ns) | $\tau_\alpha$ (ns) | $z_0$ (cm) |
|----------|------------------|------------------|-----------|
| 61       | 39.4 ± 0.3       | 97               | 9.6 ± 0.3 |
| 90       | 37.9 ± 0.3       | 104 ± 7          | 9.8 ± 0.6 |
| 90       | 37.4 ± 0.4       | 46 ± 2           | 8.7 ± 0.8 |
| 183      | 37.0 ± 0.8       | 97 ± 4           | 12.1 ± 2.5|

FIG. 2: Fit of the diffusion model to 95% of the full pulse energy (circles) transmitted in the alumina samples of $L = 61$ (b), 90° (c), 90 (d), and 183 cm (e). The solid curves show the fit. (a) is the incident pulse of $\Delta f = 15$ MHz used to obtain (b), (c), and (d); a pulse of $\Delta f = 7.5$ MHz was used to obtain (e). All the curves are normalized.

FIG. 3: (a) Leakage rates in alumina samples with $L = 61$ (red), 90° (purple), 90 (blue), and 183 cm (green). The black curves are the best fit to the data by a polynomial of power 2. (b) “Time-dependent diffusion coefficients”, $D(t) = (\gamma - 1/\tau_\alpha)(L + 2z_0)^2/\pi^2$, plotted versus $t' = t/g\gamma_1$. (c) $D(t)$ plotted versus $t'' = t/\sqrt{g\gamma_1}$.
and active random media. The statistics of these modes is fundamental to understanding electromagnetic quasi-normal modes. The level width exceeds the spacing between levels, even for the longest-lived modes, lasing would occur in the longest-lived modes, samples may be the source of the sharp spectral peaks in pulsed transmission through disordered media in which the dynamics observed is characteristic of extended waves and is not associated with the approach to localization with increasing $L$. The width of the distribution scales as $\sigma_\alpha \propto (L + 2z_0)^{-1.5}$. This is close to the scaling $\sigma_\alpha \propto \sqrt{L} \propto 1/g^{1/4} \tau_1 \propto L^{-1.75}$, and differs from the scaling predicted by Ref. 11. The wide distribution of the modal decay rates in thin samples may be the source of the sharp spectral peaks observed in amplifying random media 22, 23. In these samples, lasing would occur in the longest-lived modes, which have the lowest critical gain 24.

In conclusion, we have found nonexponential decay of pulsed transmission through disordered media in which the level width exceeds the spacing between levels, even at long times and in thick samples. This departure from diffusion theory is interpreted in terms of the decay rate statistics of electromagnetic quasi-normal modes. The statistics of these modes is fundamental to understanding the static and dynamic behavior of waves in both passive and active random media.

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