Breakdown of Wave Diffusion in 2D due to Loops

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The validity of the diffusion approximation for the intensity of multiply scattered waves is tested with numerical simulations in a strongly scattering 2D medium of finite extent. We show that the diffusion equation underestimates the intensity and attribute this to both the neglect of recurrent scattering paths and interference within diffusion theory. We present a theory to quantify this discrepancy based on counting all possible scattering paths between point scatterers. Interference phenomena, due to loop paths, are incorporated in a way similar to coherent backscattering.

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Interference phenomena are essential in the multiple scattering of classical waves. Their consideration led to the discovery of coherent backscattering [1] and brought about new measurement techniques, such as diffusive wave spectroscopy [2] and coda wave interferometry [3]. Recently, attention has been paid to more complicated interactions of waves and scatterers [4,5], especially recurrent scattering [6,7].

Recurrent scattering takes place when wave energy bounces off a particular scatterer, proceeds to bounce off at least one other scatterer, and subsequently returns to the original scatterer. This process forms a loop in the wave path. The connection has been made by van Tiggelen and Lagendijk between the simplest type of recurrent scattering and induced dipole-dipole coupling in atomic physics [8]. Recurrent scattering has also been shown to decrease the magnitude of coherent backscattering [9], and it may be crucial in the Anderson localization of light [10].

In view of these far-reaching implications, it is remarkable that standard energy transport theory, or radiative transfer, ignores the contribution of recurrent scattering events by accounting for self-avoiding scattering paths only [11]. This omission follows from the two approximations needed to render the complete multiple scattering problem tractable—the independent scattering approximation for the coherent beam, and the Boltzmann approximation for the diffuse intensity. These approximations are so ubiquitous in the literature that they are frequently invoked without justification. The redeeming aspect of these simplifications lies in their ability to conserve energy [5].

For a finite number of scatterers, the self-avoiding assumption must break down for late times. After a time equal to the number of scatterers multiplied by the average time between successive scattering events, τ_s , a wave must have revisited a scattering site on average at least once. Therefore, the predictions of radiative transfer, and the diffusion approximation [12], should, over time, progressively underestimate the amount of energy emerging from a scattering medium of finite extent.

In this Letter, we represent multiple scattering as a summation over all possible paths, including recurrent paths, and quantify the underestimation of the intensity by the diffusion approximation in 2D. We do this by counting the self-avoiding and recurrent paths and comparing the relative numbers of each at different orders of scattering. We test this theory with an exact numerical solution of multiple scattering for 300 isotropic point scatterers based on a numerical implementation of Foldy's method [13–15]. Because of recurrent scattering, the numerically computed intensity is larger than predicted by the diffusion approximation at late times. After independently measuring the scattering mean free path ℓ_s and the group velocity v_g , we show that this observed enhancement agrees well with our theory.

The geometry of the numerical experiment is depicted in Fig. 1, and the employed parameters are listed in Table I. A ring of receivers, shown as triangles, encircles the scattering region. At the center of the scattering region, a point source (S) at r = 0 emits a band limited pulse centered about t = 0. Within the diffusion approximation, the Green's function for the average total intensity, I(r, t), is the solution of the diffusion equation

$$D\nabla^2 I(r,t) - \frac{\partial I(r,t)}{\partial t} = \delta(r)\delta(t), \tag{1}$$

with $D = \frac{1}{2} v_E \ell_{tr}$ the diffusion constant, v_E the energy

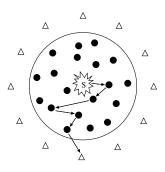


FIG. 1. The geometry for the numerical experiment: receivers are shown as triangles, scatterers as black spheres, and the source as S. The scattering region contains 300 scatterers.

TABLE I. Values of parameters for the numerical experiment and experimentally measured quantities.

Quantity	Value
Number of scatterers	300
Radius of scattering region, R	5 mm
Radius to the arc of receivers	6 mm
Dominant wavelength, λ	2.5 mm
Scattering mean free path, ℓ_s	$1.25 \pm 0.1 \text{ mm}$
Background velocity, c_o	1500 m/s
Group velocity, v_g	792 ± 36 mm/ms

velocity, and ℓ_{tr} the transport mean free path. The boundary conditions require that the solution is finite at the center of the scattering region and that it vanishes at a distance $\pi\ell_{tr}/4$ outside the edge of the scattering region:

$$I\left(r = R + \frac{\pi\ell_{\rm tr}}{4}, t\right) = 0,\tag{2}$$

where *R* stands for the radius of the scattering region [12]. Supplemented with these boundary conditions, the solution of Eq. (1) is given by

$$I(r,t) = \sum_{m=1}^{\infty} \frac{\exp[-(\frac{z_m}{R + \pi \ell_w/4})^2 Dt] J_0(\frac{z_m r}{R + \pi \ell_w/4})}{J_1(z_m)^2}, \quad (3)$$

where the Bessel functions of order zero and one are shown as J_0 and J_1 , respectively. The terms z_m denote the *m*th zero of the Bessel function of order zero.

In the numerical simulations, the total intensity was computed by averaging the squared wave field at 12 receiver locations over 20 different configurations of the scattering medium. For a single realization, the receivers were sufficiently far apart to yield uncorrelated signals. We verified that, in obtaining the estimator of the average, or coherent, wave field, the incoherent energy diminished with additional realizations by the factor $1/\sqrt{N_R}$, with N_R the total number of realizations. To characterize the transport of energy, ℓ_s and ν_g were measured independently from the numerical data. Since the individual scatterers radiated isotropically with a scattering cross section $\sigma = 4/k$, the transport mean free path, $\ell_{\rm tr}$, equaled ℓ_s . Rayleigh scattering $(\sigma \sim k^3)$ was not necessary for the purpose of this study. The physical constraint imposed on the scatterers was that energy be conserved via the optical theorem [15]. To estimate ℓ_s , we looked at the average wave field emerging from four scattering regions of radii 2, 2.5, 3, and 5 mm while keeping the density of scatterers constant [16]. By squaring the average wave field at the receivers and picking its maximum, we obtained the estimate of $\ell_s = \ell_{tr} = 1.25 \pm 0.1$ mm. When compared to the size of the scattering region ($R \sim$ $4\ell_{tr}$), this confirms that we are in the strong scattering regime where wave propagation should be diffusive [17]. The group velocity was estimated by monitoring the arrival time of the maximum of the coherent intensity with distance. Because of the absence of resonant point scatterers in our simulations, the energy velocity, v_E , could be taken as equivalent to v_g . We find $v_g = v_E = 792 \pm 36$ mm/ms, almost half of the background velocity.

By inserting the estimated values for $\ell_{\rm tr}$ and v_E into Eq. (3), the accuracy of the self-avoiding assumption can be evaluated. As seen in Fig. 2, the solution of the diffusion equation approximates the early arriving energy as well as can be expected. However, the growing number of recurrent paths with time eventually causes the diffusion approximation to underpredict the transmitted energy. Dividing the measured total intensity by the diffusion solution yields the measured enhancement of energy due to recurrent scattering, shown in Fig. 3. The recurrent paths cause the enhancement to grow in time. Given the poor fit of the diffusion approximation to the measured total intensity in Fig. 2 for late times, a quantitative explanation of the enhancement, taking into account the self-avoiding assumption, is necessary.

The enhancement can be explained with an argument based on counting the number of different paths through the scattering region. Ishimaru utilized path counting to illustrate the self-avoiding approximations made by Twersky [18]. A combinatoric factor was employed in determining the decrease of the coherent backscattering cone due to two-particle recurrent scattering in 3D [9]. In our theory, all orders of many-particle recurrent scattering are counted. The inclusion of all orders of recurrent paths is consistent with the theory of random walks in 2D [7].

As seen from their "most-crossed" diagram, two distinct scattering paths traverse a loop in different directions. Since the two paths have equal length, they constructively interfere with each other [19]. Taking this into account, it is evident that all single loop paths have their intensity doubled by interference, in a way similar to coherent backscattering. Paths with two loops have their intensity quadrupled. The complete solution of the multiple scattering problem includes all the scattering paths and the interference between them. Only

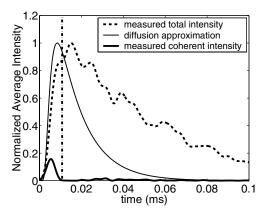


FIG. 2. The measured total intensity and the diffusion approximation. Also shown is the measured coherent intensity and a vertical line marking its termination.

093902-2

self-avoiding scattering paths are accounted for within the diffusion approximation. The enhancement of multiple wave scattering over the diffusion approximation for scattering of order n in a system of N scatterers is thus given by

$$E(N, n) = \frac{\text{\#self-avoiding} + 2(\text{\#1loop}) + 4(\text{\#2loops}) + 8(\text{\#3loops}) \cdot \cdot \cdot + \text{\#allotherpaths}}{\text{\#self-avoiding}},$$
(4)

where #allotherpaths refers to all paths that have at least one folded event [9]. The number of scatterers encountered after a time t is on average equal to t/τ_s . Therefore, the enhancement in Eq. (4) can be calculated at times $t = n\tau_s$. Since we have measured τ_s independently, all that remains is the counting of the different types of paths.

After performing the combinatorics needed to count the number of different types of paths, the predicted enhancement of Eq. (4) can be expressed as a finite series

$$E(N, n) = \frac{N(N-1)^{n-1}(N-n)!}{N!} + \sum_{m=0}^{f(n)} \frac{2^m - 1}{2^m} \frac{(n-m-1)!(N-n)!}{m!(n-3m-1)!(N-n+m)!},$$
(5)

$$f(n) = \frac{n - 1 - [(n - 1) \bmod 3]}{3}.$$
 (6)

In the appendix, we briefly describe how the enumeration leads to these equations. The enhancement factor of Eq. (5) suggests that the time decay of transmitted intensity is nonexponential, in contrast to diffusion, Eq. (3). Recently, Chabanov *et al.* reported nonexponential decay in an experiment with microwaves in which the deviation from exponential decay was seen as an enhanced transmission [20]. The leading-order correction term to diffusion derived by Mirlin [21] from supersymmetry calculations predicts a similar enhancement.

Using the independently measured values for ℓ_s and v_g to obtain τ_s , the theoretical prediction of Eq. (4) is compared in Fig. 3 to the measured enhancement of the intensity from the numerical experiment over the diffusion approximation. At times past the arrival of the coherent wave (marked by a vertical line), agreement is seen over 20 orders of scattering. Note that the theory predicts quite well the rate of increase in enhancement on the logarithmic plot of Fig. 3. Any change of the normalization used in Fig. 2 would only shift the measured and theoretical enhancements up or down; their rates of increase with time would still agree. This shows that, although the diffusion approximation grossly underestimated the measured intensity, the discrepancy can be understood on the basis of a random walk model.

Past 30 orders of scattering (~ 0.05 ms), the theoretical enhancement overestimates the measured enhancement. A number of factors are responsible for this. The deviation of late-time transmission from diffusion has been studied numerically by Weaver in infinite 2D media [22]. Such a deviation in infinite media suggests that a finite number of scatterers in a region on the order of the mean free path contribute in the path enumeration, Eq. (5).

Additionally, radiative transfer may not be accurately described by the diffusion approximation, Eqs. (1) and (2), for our finite geometry. Finally, the theory predicts an enhancement factor of infinity at times past $N\tau_s$, since the number of self-avoiding paths, the denominator of Eq. (4), vanishes at those times. One possibility to improve the theory is to make τ_s a distribution, instead of simply an average quantity [23,24].

An important step is the extension of this theory to 3D. The fact that 2D is the lower critical dimension for localization can be understood by the recurrence of the random walk in 2D [25]. For a finite scattering volume in 3D, it is not yet clear how the effects of recurrent scattering will depend on $k\ell_s$, the measure of scattering strength.

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Appendix.—Equation (5) follows from Eq. (4) by counting the different paths. For N total scatterers and n scatterings, the number of self-avoiding paths is

#self-avoiding =
$$\frac{N!}{(N-n)!}$$
. (7)

The number of one-loop paths can be expressed with a similar combinatoric factor describing how many different combinations of n-1 scatterers can be made from N total scatterers. Additionally, there is a factor F(1, n) that represents how many different ways one loop can be made

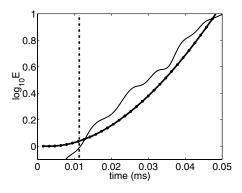


FIG. 3. The logarithm of the measured enhancement and the theoretical prediction. The vertical line marks the termination of coherent energy, after which the diffusion approximation should apply. The theoretical curve has dots along it that indicate time intervals τ_s or, equivalently, different orders of scattering.

093902-3 093902-3

from the n-1 scatterers:

#1loop =
$$\frac{N!}{[N - (n-1)]!}F(1, n)$$
. (8)

To illustrate what F(1, n) is counting, consider a one-loop scattering path that visits the four scatterers labeled a, b, c, and d. For this example, n, the number of scatterings, equals five. Given these scatterers, there are three different ways that one loop can be made: (a, b, c, a, d), (a, b, c, d, a), and (a, b, c, d, b). Therefore, F(1, 5) = 3 for the one-loop paths.

For the calculation of the function F(1, n), suppose there is a scattering path with n scatterings. In order to make a loop, a path must revisit a scatterer after at least two scatterings off different scatterers. Hence, after one scattering, there are n-3 possibilities for a return. After two scatterings, there are n-4 possibilities. Extending this reasoning over n scatterings, it is clear that the total number of possibilities is equal to the sum of the numbers 1 through n-3, or

$$F(1,n) = \frac{(n-3)(n-2)}{2}. (9)$$

Applying similar logic to paths with two loops, the function F(2, n) is equal to

$$F(2,n) = \frac{(n-6)(n-5)(n-4)(n-3)}{8}.$$
 (10)

This expression is important since it enters into the equation describing the number of two-loop paths,

#2loops =
$$\frac{N!}{[N - (n-2)]!}F(2, n)$$
. (11)

A general form for F(m, n) can be written that is consistent with Eqs. (9) and (10),

$$F(m,n) = \frac{1}{2^m} \frac{(n-m-1)!}{(n-3m-1)!m!},$$
(12)

where m represents the number of loops. Note that in the case of no loops (m = 0), the function F(m, n) equals 1. A general equation for the number of m-loop paths can be obtained by inserting F(m, n) into Eqs. (8) and (11) and by making the generalization

$$\#\text{mloops} = \frac{N!}{\lceil N - (n-m) \rceil!} F(m, n). \tag{13}$$

Also appearing in Eq. (4) is the number of paths that are not self-avoiding and do not form loops, #allotherpaths. By definition

#allotherpaths = #allpaths - #self-avoiding - #1loop

$$-$$
 #2loops $-$ #3loops \cdots , (14)

where #allpaths = $N(N-1)^{n-1}$. Inserting this expression for #allpaths and Eq. (14) into Eq. (4) gives the enhancement

$$E(N,n) = \frac{1}{\text{\#self-avoiding}} [N(N-1)^{n-1} + 1\text{loop} + 3(\text{\#2loops}) + 7(\text{\#3loops}) \cdots].$$
(15)

Substituting Eqs. (7) and (13) into Eq. (15) leads to Eq. (5).

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093902-4 093902-4