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The question of classical localization
A theory of white paint?

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Abstract

The expected behaviour of localizing media for classical wave propagation is analysed. Some possible examples in electromagnetic and acoustic phenomena are given.

The phenomenon of localization has its primary application in the problem of electron transport, and has been related to the diffusion of electron or nuclear spins—with some success—and of optical excitation in solutions—with considerably less. The question of whether localization can ever be convincingly observed in classical systems, such as microwave, acoustic, or light propagation in random media, has been discussed in a number of cases, but not very realistically, except for John (1984)†. One serious problem is that many of the discussions have emphasized one-dimensional systems, which do not exhibit truly delocalized extended-wave behaviour, so that a clear physical distinction is not possible.

In all higher dimensionalities there is a clear distinction in principle. Imagine a slab of thickness W of material containing random non-absorptive scatterers embedded between two non-random identical propagating media. If the slab is thick compared with the conventionally defined mean-free-path l, coherent radiation will be exponentially attenuated: if a wave of amplitude $\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t)]$ is incident from the left, the transmitted wave of the *same* $\mathbf{k}$ and ω will have amplitude

$$t \exp i(\mathbf{k}\cdot\mathbf{r}-\omega t), \text{ with } t=\exp[i\delta - W/l].$$

Most of the incident radiation will be scattered diffusely and, since we have postulated that there is no absorption, it must all come out on one side or the other of the slab W. First let us consider the classical theory, which assumes that the radiation diffuses in a random walk of wave velocity c, step length l, and mean free time $\tau=l/c$, and hence satisfies a diffusion equation

$$D\nabla I = -J$$

with

$$D \simeq l^2/3\tau = cl/3.$$

† Many of the ideas in the present paper were also suggested in this excellent paper, but there is at least one major difference: we find that absorption (as measured by the reflection coefficient) *decreases* near the mobility edge, in contradiction to this reference.

Here J is the average current of radiation which satisfies the continuity equation

$$\frac{\partial I}{\partial t} = -\nabla \cdot J,$$

so that

$$D\nabla^2 I = \frac{\partial I}{\partial t}.$$

In the steady state,

$$D\nabla^2 I = 0,$$

so $J = -D\nabla I$ is constant.

A simple way to satisfy outgoing boundary conditions on the right-hand side of the slab is to estimate that all radiation starting to the right within l of the right-hand boundary exits and is not back-scattered, while no radiation enters from the right. This can be mimicked by setting $I = 0$ at $x = W + l$ in an infinite slab. Thus

$$I = |-\nabla I|(W + l - x),$$

$$I(0) = W + l; \qquad I(W) = l,$$

and it is easy to see that

$$J_{\mathrm{inc}}/J_{\mathrm{refl}} = W/W - l, \qquad J_{\mathrm{trans}}/J_{\mathrm{inc}} \simeq l/W.$$

Thus the slab has a reflection coefficient for diffuse radiation, $1 - l/W$, and a resistance $1/l$ per unit thickness; the opacity grows only linearly with thickness, like heat resistance, not exponentially as it would for a random one-dimensional system. This behaviour has been discussed in some classical cases, e.g. light propagation in clouds (Ishimaru 1978) or colloidal suspensions (Kerker 1969).

This contrasts with the behaviour expected as localization takes over. The standard criterion for localization is $kl = 2\pi l/\lambda \simeq 1$; as l/λ approaches unity, the effective opacity per unit thickness will rise faster than $1/l$ and become infinite at some finite value of $l = l_c$ (l being the classically calculated mean free path). A good way of describing the phenomenon is to have recourse to the scaling theory of localization, which assumes that at the 'microscopic' scale of the wavelength λ or of l, the mean free path and transmission coefficient T are varying relatively continuously. However, at longer scales the effect of coherent backscattering is that T falls more rapidly than $1/W$ with thickness W until we reach a length scale L_l, above which the system resumes simple diffusive behaviour. Present estimates suggest that the opacity ρ increases linearly with scale and at scale L is approximately L/l^2. The critical behaviour is

$$L_l \sim l\frac{l}{l - l_c},$$

so that

$$\rho = \frac{1}{l - l_c}, \qquad D = \frac{c}{3}(l - l_c)$$

once the system becomes diffusive. Beyond l_c, there is a localization length L_l which also probably diverges at $l^2/l_c - l$ near the mobility edge l_c. The transmission decreases exponentially with W beyond l_c,

$$T = ((l_c - l)^2/l^2)\exp(-W(l_c - l)/l^2).$$

Thus there is a clear distinction between the transmission of diffuse, incoherent radiation of a 'localized' and an 'extended' system, once the system is sufficiently large. Unfortunately, 'sufficiently large' also means 'sufficiently opaque' and the experimental problem may not be all that simple.

It is worth discussing what happens in the presence of absorption. In the simple diffusive regime, we can define an l_i, (i for inelastic), some multiple of the elastic mean-free-path l, and l_i/l, the average number of scatterings before absorption. If the thickness W is greater than $(ll_i)^{1/2}$, attenuation because of absorption will begin to be important, and the linear diffusive behaviour will not appear. Absorption will also play a role in changing the reflection coefficient; the sample will behave as though its thickness were $W_{\text{eff}} = (ll_i)^{1/2}$, and hence the reflection coefficient will be

$$R \sim 1 - (l/l_i)^{1/2}.$$

The square root is probably the reason why many white paints do not reflect white light perfectly ($R \sim 0{\cdot}5$–$0{\cdot}8$) even though the pigments are almost non-absorptive.

What happens in the presence of incipient localization? In the critical regime, the effective diffusion constant decreases with scale and hence with thickness W. We can define an effective thickness W_i at which the wave is absorbed by the scaled diffusion equation,

$$D_{\text{eff}}\tau_i = W_i^2,$$

with

$$D_{\text{eff}} = \frac{cl^2}{W_i}.$$

Hence $W_i^3 = l^2 l_i$. The reflection coefficient will then be the same as that for this thickness of material, as calculated from the non-absorbing case,

$$R = 1 - \frac{l^2}{W_i^2} = 1 - (l/l_i)^{2/3}$$

Finally, it is a relatively trivial exercise to obtain the localized case.

I have collected most of these rather confusing results in the table. In the absorbing case, I feel that the reflection coefficient is the most important result since, when the sample is thick, there will simply be exponential attenuation in all cases. The most striking results are, for weak absorption, the $1/W^2$ and then exponential transmission; and for finite absorption, the change in reflection coefficient, which, while only a $\frac{1}{2}$–$\frac{2}{3}$ power, may make localizing systems considerably more reflecting.

Now let me discuss, rather briefly, possible instances and how they might be implemented. First let us consider electromagnetic radiation. Here I think the ideal system could be constructed in the form of a microwave or millimetre wave transmission experiment through a system composed essentially of random wave-guides near cut-off and random resonators, such as might be realized by a random packing of metallic balls of the right size. Another possibility is foil sheets with random holes cut in them, or even with random spot patterns applied to them, stacked either regularly or randomly perpendicular to the propagation direction. With modern lithographic techniques, the same trick might even be tried at infrared or optical wavelengths.

Table of expected behaviour

Case		T	R	L_{loc}	L_i
Without absorption					
Classical		l/W	$1-l/W$	–	–
Critical:	$W<L_{loc}$, $l \simeq l_c \simeq \lambda$	l^2/W^2	$1-l^2/W^2$	–	–
Critical:	$W>L_{loc}$, $l>l_c$	$(l/l_c)/W^2$	$1-(l-l_c)/W$	$l^2/l-l_c$	–
Localized:	$W>L_{loc}$	$(l_c-l)^2/W^2$	$1-T$	l^2/l_c-l	–
	$l>l_c$	$\times \exp(-W/L_{loc})$	$1-T$	l^2/l_c-l	–
With absorption					
Classical:	$W>L_i$	$\exp(-W/L_i)$	$1-(l/l_i)^{1/2}$	–	$(ll_i)^{1/2}$
Critical:	$L_i<L_{loc}$, $W>L_i$	$\sim\exp(-W/L_i)$	$1-(l/l_i)^{2/3}$	–	$(l^2l_i)^{1/3}$
Critical:	$L_i>L_{loc}$	–	$1-(l-l_c)/(l^2l_i)^{1/3}$	–	–
Localized:	$L_i>L_{loc}$	–	$1-(l_c-l/L_i)^2$	–	–

A relatively tricky example is high-dielectric-constant balls or particles embedded in vacuum, air, or low-dielectric-constant material such as linseed oil or other transparent polymerizing liquid. (White paint is composed of just such a system.) Here the problem is one which we will encounter very severely in acoustical systems, namely that of an inhomogeneous mixture of two propagating media. If the dielectric particles are in too intimate contact, we are in danger of developing a propagating 'slow wave' which may localize more or less easily than the faster wave in the interstices. We note that where two types of coupled propagation can occur at the same frequency, they both must localize or diffuse together.

A careful matching of obstacle size to wavelength must be made. If the obstacles are small compared to the wavelength, they scatter very weakly—the famous k^4 scattering law implies that for a dense array of scatterers of size a, $kl \sim 1/(ka)^3$. For obstacles which are larger when compared to the wavelength, the cross-section is the geometric size a^2 and for a dense array $kl \sim ka$. Thus optimum scattering requires structures comparable to the wavelength. I cannot esimate what contrast in dielectric constant makes it actually possible to localize electromagnetic waves, but surely the fact that high-dielectric-constant pigments such as TiO_2 or PbO are used, is evidence that a ratio in n of 1·5–2 could be near the critical one. Clearly the correct configuration is high dielectric constant, low-velocity particles and high-velocity interstices to make the interstices below waveguide cut-off.

Acoustic systems are probably harder to localize: one has few materials which simply do not propagate sound the way metals do not propagate electromagnetic waves, and there is a tendency for rigid bodies to be of high rather than low velocity—hence the 'framework' wave is the fast one. Expanded silica gel might be a very interesting material to fill with denser liquids, or even with air. With sufficient contrast in velocity, i.e. a high velocity framework and soft interstitial material, it could be that localization of the 'slow wave' could be studied meaningfully, treating the coupling to the framework as a weak perturbation. Undoubtedly there are localization aspects which have not been adequately taken into account in the standard 'Biot' theory of acoustic wave propagation in random media (Johnson and Plona, to be published).

In conclusion, I feel that localization in classical wave propagation is a phenomenon which should, in carefully prepared systems, be easily observed, and in such systems the basic laws of the localization phenomena could be conveniently studied, for example, the critical exponents and the phenomenon of anomalous fluctuations. In addition, although great complexity may be encountered, the work may impinge on a number of highly interesting and practical systems, such as paints, porous media, etc.

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