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Looking through the ground glass*

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Abstract

We initiate, and to an extent, motivate our discussion of wave propagation through a random medium by asking whether we can view or image an object through a light scattering medium. We answer in the affirmative by arguing that the image-bearing ballistic component of light can be time-resolved with respect to the image-blurring diffusive component that has to traverse relatively much longer distance. This leads us to the question of diffusion of light or its absence (localization) in disordered media. We discuss some essential differences between photon localization *vis-a-vis* electron localization. One of these that makes photon localization much harder to realize experimentally is that the photon energy multiplies the dielectric disorder in the Maxwell equation, as a result of which localization is missed in the limit of both the long wavelength (Rayleigh scattering) and the short wavelength (geometrical optics). The narrow 'window of localization' requires drastic enchancement of effective scattering which is possible by the coincidence of the Mie-resonant scattering and the Bragg-reflection (umklapp) conditions. Photon localization at microwave frequencies (as also the complete photon band gap) has already been achieved by several workers. Localization at visible wavelengths is awaited. We also discuss some fundamental QED effects of photon localization, such as the suppression of spontaneous emission from an excited atom embedded in a random dielectric. We end the discussion with some speculations on photon localization in an active (amplifying) medium and the possibility of a 'mobility-edge' laser.

Key words: Wave propagation, light scattering medium.

1. Introduction

Can we view or image an illuminated object through a turbid medium or, more precisely through an optically inhomogeneous medium that scatters light strongly, but does so without absorbing it? Answers to this question may range from commonsense to deep thought, and involve some good and interesting physics. The correct answer is also not entirely devoid of importance to practical solutions of some real problems. For, the object to be viewed may well be a small, or ultrasmall ($\ll 1$ mm) tumour embedded in a scattering medium, that is soft tissues. It may involve a cataract patient unable to see through his clouded (or occluded) eyelense that

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diffuses light. It could also be an aircraft flying under the cover of clouds, an automobile lost in the fog, or a submarine hidden in murky waters. Examples multiply. At a fundamental level of interest to condensed-matter physicists, however, it has to do with the question of coherent (ballistic) *versus* incoherent (diffusive) light wave propagating through a disordered dielectric and the associated possibility of disorder-induced (Anderson) localization of the light waves in space. Also, its proper study leads naturally to what is now known as the diffuse wave spectroscopy (DWS). This is a powerful technique that uses the very multiple scattering (diffusion) of light to probe the dynamics of the particulate scatterers on length scales ~ wavelength of light in certain model fluids and soft-condensed matter, *e.g.*, micron-sized polystyrene balls (polyballs) or latex beads suspended in water.

In this talk, we will briefly discuss some of these very interesting aspects of scattering[1] of light and end with some speculations on light scattering in a random but active (amplifying) optical medium. However, much of what I say is known and is applicable, more or less unchanged, to almost any wave propagating through an appropriately defined random medium. It is the wave character giving interference and diffraction which is the operative feature here. Thus, instead of light (classical electromagnetic waves) obeying Maxwell's equations, we may have an electron (the quantum-mechanical de Broglie waves) obeying Schrödinger's equation. The optically random medium consisting of randomly distributed polarizable particles is here replaced by the potential due to the randomly distributed impurities in a dirty metal. Light localization then translates to metal-to-insulator transition. One could, of course, equally well discuss the scattering and localization of sound (acoustic waves) in an inhomogeneous elastic medium containing randomly distributed bubbles, say. Similarly for the various water or seismic waves whose localization can lead to dangerous concentration of energy. (The devastation of Mexico city in the recent earthquake may be a case in point). Or, for that matter, even for the gravitational waves scattered by the random background metric—the foam-like metric of John A. Wheeler perhaps.

Studying these general wave phenomena with light, however, has certain advantages. First, light is clean. There is no light–light interaction except in nonlinear media and at high intensities. Photons carry no charge. Second, light from a coherent source (laser) is athermal—it is essentially at absolute zero of temperature even at room temperature ($K_B T \ll$ photon energy). Thus, the inelastic scattering effects and the resulting decoherence is negligible (unlike the case of electrons in solids except at very low temperatures). The relative weakness of electromagnetic interaction due to the smallness of the fine-structure constant does pose a disadvantage though. Further, the Bose character of photons (whose phase coherent state represents the Maxwell electromagnetic field) admits the possibility of coherent amplification which, when combined with disorder-induced confinement (Anderson localization) in an active (lasing) random medium, leads to novel possibilities, not open to electrons (Fermions). There are, of course, some complications in the case of light due to its polarization and the associated anholonomy, extinction by absorption and the nonlinear optical effects. But all these bring in richness of possibilities important for the fast-expanding field of photonics. Finally, these are novel radiative (electrodynamic) effects of fundamental significance such as the suppression of spontaneous emission due to photon localization that open up new vistas of basic research[2].

But let us now begin by addressing the question posed at the very beginning. The answer is yes!, we can. In order to see how, consider a well-collimated beam of coherent light at wavelength λ from a laser incident on the disordered medium, a slab of thickness L, say. The latter may be conveniently, but not necessarily, viewed as a dilute random distribution of scatterers—dielectric spheres of radius a $\ll \lambda \ll 1$, mean spacing between the scatterers. The wave amplitude then splits into two distinct partials : (1) a so-called ballistic (coherent) component that results from the interference of the unscattered primary amplitude with the coherently forward-scattered amplitude, and is transmitted within a small forward cone angle. The scattering medium effectively offers a refractive index $n = k' / k$, with $k'^2 = k^2 + 4\pi\ Nr\ f(k' \leftarrow k')$, where $k(k')$ is the incident (ballistic) wave vector magnitude, $N = l^{-3}$, the number density of scatterers with the forward scattering amplitude $f(k' \leftarrow k')$, and $r \approx l$ for uncorrelated scatterers. The ballistic component traverses the medium with the shortest optical path length $l = nL$, corresponding to a time-delay $T_B = nL/c$. It casts a sharply defined shadow-image of any opaque object embedded in the medium on the screen; (2) the diffuse (incoherent) component that is multiply scattered on the randomly located scatterers, and diffuses with an elastic mean free path $l_e = 1$ corresponding to a diffusion constant $D_e = (l / 3)\ c\ l_e$. It emerges at the far end after traversing an optical path length $l_D = cL^2 / 2D \gg l_B$, in a large cone angle, and produces a general illumination on the screen. This, of course, reduces the fringe visibility (the contrast) and hence blurs the otherwise sharp image cast by the ballistic component. (There is a third component, the 'snake' that meanders around the ballistic component and may be identified with it for the present purpose). The next step is obvious. All we have to do is to use a picosecond pulsed laser and an ultrafast (picosecond) CS_2 Kerr gate (shutter) so timed as to let the early image-bearing ballistic signal pass but shut off the late-arriving image-blurring diffuse signal. Such an imaging by ultrafast time-resolved transillumination has been demonstrated, and immense improvement in resolution and fringe visibility is soon expected with the use of high repetition femtosecond multiple Kerr gate system and 2-dimensional, cooled CCD imaging[3]. Early non-invasive detection of breast tumours, too small (< 1 mm) for X-ray mammography may then be possible.

Theoretically, the difference in the optical path lengths, $l_B \ll l_D$, suggests yet another way of recovering the image. Introduce a certain amount of extinction by absorption (α) through a dye, say. Then, the image-bearing ballistic signal would be attenuated by $e^{-\alpha L} \ll e^{-\alpha c L^2}/2D_e$, the attenuation suffered by the diffuse signal that had a longer way to run. This should enhance the contrast, though at the cost of overall reduction in intensity. In fact, diminished intensity should help enhance the definition of image.

One may ask if we gain by varying the wavelength of light. Well, if we could tune the wavelength close to the mobility edge, assuming that one exists, we would drastically enhance the time-delay of the diffuse component and hence gain in fringe visibility (at the cost of intensity, of course).

Thus, we turn to another aspect of light scattering, namely, that of the possibility of localizing light by disorder. In doing so, we are, of course, motivated by the known phenomenon of Anderson localization of electron (waves) in dirty metals[4]. Thus, in a 3-dimensional disordered conductor there exists a disorder-dependent mobility edge E_c (infinitely sharp in the limit of infinite sample size) separating the conducting (extended) states above E_c (towards midband) from the insulating (localized) states below E_c (towards the band edges). One can define a disorder parameter $\eta = (k_F l_e)^{-1}$ in terms of the Fermiwave vector k_F and the elastic mean free path l_e such that we have the extended or the localized states according as $\eta <$ or $> \eta_{\text{critical}}$, respectively. Here $\eta \approx l$ is the Mott–Ioffe–Regel criterion for the mobility edge which simply means that $2\pi l_e \approx \lambda_F$ (Fermi wavelength).

By analogy, one should expect the same for the case of light waves, but with an important difference that can be readily appreciated by looking at the Maxwell wave equation:

$$- \nabla^2 E(x) + \nabla(\nabla\cdot E(x)) - \frac{\omega^2}{c^2} \epsilon_1(x)E(x) = \frac{\omega^2}{c^2} \epsilon_0(x)E(x) \qquad (1)$$

where $\epsilon_1(x)$ and $\epsilon_0(= 1)$ are, respectively, the spatially random and non-random parts of the local dielectric constant $\epsilon(x) = \epsilon_0 + \epsilon_1(x)$. Except for the second-term LHS and the vector nature of E (which is not crucial to our discussion), this is just the Schrödinger equation with essentially positive eigenvalue $\omega^2 / c^2\epsilon_0$ (hence no truly bound state). But there is an all important difference, namely, that the eigenvalue (ω^2) multiplies the randomness $(\epsilon_1(x))$ which eventually determines the disorder parameter η. Thus, in the low frequency ω or long wavelength $\lambda \gg a$ limit, the disorder too becomes small and gives a scattering mean free path $l_e \propto \omega^{-4} \propto k^{-4} \propto \lambda^4$. This is the well-known l / λ^4 Rayleigh Scattering for $\lambda \gg a$, the disorder correlation length or the scatterer size, that makes the sky blue and the twilight red. Here, it means that the disorder parameter $(k_F l_e)^{-1} \ll l$, and hence misses the MIR criterion for localization. This is not so for electrons where l_e is not a sensitive function of energy and hence $1 / k_F l_e$ must exceed unity as k_F becomes small giving localization at low enough energy, *i.e.*, close to band edges. In the opposite limit of short wavelength $\lambda \ll a$, we are in the limit of geometrical or ray optics, that gives just classical particulate diffusion. Thus, in either limit photons escape Anderson localization, as indicated schematically in Fig. 1. There is at best a narrow window of localization and that demands strong scattering. Let us see how this can be met.

Wave propagation in a random dielectric modelled by the local dielectric constant $\epsilon_1(x)$ in eqn (1) fluctuating continuously in space(x) is characterized by the wavelength λ, the length l_c over which the random fluctuation of $\epsilon_1(x)$ are correlated, and the strength of randomness Δ (the root mean-squared value of $\epsilon_1(x)$, say). One can at once define the dimensionless Kubo number κ (Kappa) = $\frac{1}{2}\Delta k l_c$ = depth of random modulation / spatial speed of random modulation. One usually works in the limit (for mathematical convenience) $l_c \to 0$, but keeping $\Delta^2 l_c$ finite at the actual value, corresponding to the gaussian white noise limit. This leads to the disorder parameter

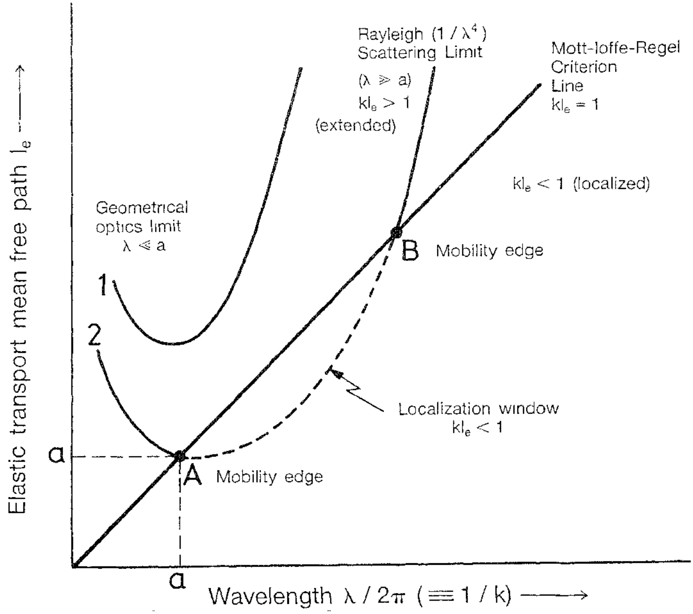

FIG 1. Phase diagram for Anderson localization of photons in the $l_e - \lambda$ plane. Curve 1 is for weakly disordered medium and misses MIR localization condition $kl_e < 1$. Curve 2 is for strong disorder with localization window AB Scattering is weak in both limits $\lambda \gg 2\pi a$ and $\lambda \ll 2\pi a$, for a = correlation length of disorder.

$\eta = l / kl_e$. The problem can be treated by the invariant imbedding method[5] exactly for one dimension (corresponding to a disordered optical fibre, say) and approximately for higher dimensions, or perturbatively by treating the coherent multiple scattering (*see* John[1]). However, the available dielectric constants, *e.g.*, $\epsilon_{air} = 1$, $\epsilon_{water} = 1{\cdot}69$, $\epsilon_{TiO_2} = 7{\cdot}5$, $\epsilon_{Polystyrene} = 2{\cdot}55$, $\epsilon_{polyethylene} = 2{\cdot}25$, $\epsilon_{teflon} = 2{\cdot}1$, etc., are not able to give the condition $l / kl_e > 1$ (Mott–Ioffe–Regel or MIR criterion) for localization. Here, one is really missing certain critical effects, the 'glory' of the scatterers that must be invoked.

Consider a random collection of spherical scatterers of radius a_i, dielectric constant ϵ_i and number density N_i dispersed through a background dielectric constant ϵ_0.

The elastic scattering cross-section σ_e of each scatterer for $\lambda \gg a$ is given by the familiar Rayleigh expression

$$\sigma_e = \left(\frac{8\pi}{3}\right) a_i^2 \left| \frac{\epsilon_i - \epsilon_0}{\epsilon + 2\epsilon_0} \right|^2 (ka)^4 \propto \frac{1}{\lambda^4} \qquad (2)$$

giving the inverse transport mean free path, $\frac{1}{l_e} = N_i\ \sigma_e$. Thus, we miss the MIR localization criterion in the limit of long wavelength. In the limit of short wavelength $\lambda \ll a_i$, the cross-section tends to something of the order of the geometrical cross-section. In between is the oscillatory region of Mie resonances where the radiation is internally multiply reflected by the dielectric sphere of radius $\sim \lambda$. (Not for metallic spheres, of course). Mie resonance can enhance the cross-section by an order of magnitude. One can then obtain further enhancement of scattering by introducing a synergetic effect of the Bragg reflection and the Mie resonance. That is, arrange the scattering by introducing a synergetic effect of the Bragg reflection and the Mie resonance. That is, arrange the scatterers in space periodically so as to produce first an optical band gap : $\mathrm{k}\cdot\mathrm{G} = \frac{1}{2}\mathrm{G}$ with G the reciprocal vector of the lattice. Now, for the photon frequency ω slightly above the band edge ω_c, one can show that the effective guide wavelength $\lambda_{\text{eff}}\ \alpha\ (\omega - \omega_c)^{-1/2}$, the envelope wavelength (Recall that in a rectangular waveguide, the effective guide wavevector close to the cut-off is $k_g = k_0 \sqrt{(1 - \omega^2/\omega_c^2)}$. Thus, the MIR condition $2\pi l_e/\lambda_{\text{eff}} < l$ for localization is readily satisfied, for even a relatively small randomness of the scatterers a pseudogap opens up. This is quite similar to the phenomenon of high resistivity of rare-earth compounds, *e.g.*, $CePd_3$ due even to a small disorder—the $4f$ resonance plays the role of the Mie resonance there. The synergetic effect of strong Mie resonance condition and the Bragg reflection condition is key to photon localization. In point of fact the enhancement of backscattering by Bragg reflection is made use of in RAS (radio acoustic sounding) to measure suspended matter (water droplets) in air that backscatter microwaves. One makes an acoustic beam ride the microwave beam and the acoustic wavelength is adjusted so as to satisfy the Bragg condition, creating a diffraction grating of periodically modulated air density (and hence refractive index). This enhances the backscattering by the random scatterers despite low contrast. The fact that the acoustic grating is moving is not important because of the much greater speed of light. Photon localization[6], as also photon bandgap[7], at microwave frequencies has already been experimentally demonstrated.

At this point a fundamental difference between the photon and the electron localization must be pointed out. Photon localization is always at positive energy analogous to the +ve energy-bound states constructed by Von Neumann and Wigner, and is directly related to coherent backscattering—the fact that the partial amplitudes multiply scattered by the scatterers in the opposite (time-reversed) sequences add in phase and hence give refocussing of the wave in a direction counter to the incident direction to within an angle $= \lambda/2\pi l_e$. This has indeed been observed for light[8]. For electrons this yields the well-known weak localization correction to conductivity on the metallic side[4]. For electrons, however, one can also have a mobility edge lying below the random potential maximum (assume a bounded telegraph-potential disorder, say), where tunneling through potential barriers begins to dominate transport. One could possibly obtain localization even if the coherent backscattering is eliminated by a strong magnetic field. This question needs careful analysis. In any case the question as to how coherent backscattering interpolates between the weak and the strong localization remains open. Experiments with light, free from photon–photon interaction complication, could throw much light on this electronic problem.

With photons one can also measure the distribution of path lengths traversed by the backscattered photons at microwave frequencies by measuring the distribution of rotations of the plane of polarization due to Faraday rotation. One needs disordered magnetic medium for this. The point is that the Faraday rotation is accumulative as the photon is scattered multiply back and forth parallel to the magnetic field.

Localization of photons by disorder has some fundamental consequences for quantum electrodynamics (QED)[2]. Let us consider just one of these, namely, the inhibition of spontaneous emission from an excited (metastable) atomic state and hence the natural line-width narrowing. To see this, consider the atom to be embedded in a lossless dielectric host which is disordered enough to have mobility edges (Fig. 1). Let the transition frequency ω of the photon to be emitted spontaneously fall in the localization window AB. It is clear then that the emitted photon cannot 'really' propagate away (beyond a localization length) and hence must be re-absorbed—it becomes a virtual photon. In fact, we have here a two-level system (a clock): Atom in the exicted state + no photon $\longleftrightarrow$ atom in the ground state + one photon localized in the dielectric medium. One has then a photon–atom-bound state. One has effectively enhanced the lifetime of the metastable atomic state (or effectively switch off the zero-point vacuum fluctuations that are usually viewed as causing the atomic decay). This, of course, should have electrodynamic effects, *e.g.*, anomalous Lamb-shift (for a discussion of this see ref. 2).

There is an exciting possibility following from the above that merits speculating on. Consider a three-level atom embedded in the strongly disordered dielectric medium, with the transition frequencies $\omega_p > \omega_L > \omega_I$ connecting, respectively, the highest atomic level to the ground level, intermediate level to the ground level and the highest level to the intermediate level. (The subscripts P, L, and I have the obvious connotation of pump, lasing and the idler frequencies). Now, let ω_p lie above the upper mobility edge (A in Fig. 1), ω_I below the lower mobility edge (B in Fig. 1) and ω_L in the localization window AB. It is clear then that one can pump the system and populate the meta-stable intermediate level and the bound photons (ω_L) confined within Anderson localization length. The two-level atom–photons system will now be : Atom in the excited state + N photons in the localized state $\longleftrightarrow$ atom in the ground state + $(N + 1)$ photons in the localized state with $N \gg 1$. The frequency of this 'clock', of course, increases towards the pump frequency as the number of photons condensed into localized state is increased by pumping—a nonlinear effect. It is important to remember that the pump field is incoherent and acts stochastically on the atomic electron in the ground state causing excitation. But, the oscillations are coherent though. Now, we can have a finite concentration of such atom-localized photon centres in an intentionally disordered section of an optical fibre doped with the 3-level atomic centres, say, with mean spacing ≈ localization length so that the photons can tunnel to the neighbouring atomic centres by excitation/deexcitation of the neighbouring atom. The coherent dynamics of such a coupled, randomly kicked (pumped) system is not fully understood. But on general grounds, one can expect it to self-organize itself into a dissipative structure—a laser in which the mode frequency gets narrowly selected despite parametric disorder so as to maximize emission. A study of this phenomenon modelled by coupled pseudospin $-\frac{1}{2}$ + local bosons complexes in a static pseudomagnetic field and a stochastic pump field causing pseudospin flips is under investigation. Here, the boson can tunnel by flipping the neighbouring spins. We are talking here of a possible mobility-edge laser! Some similarity with the lasing droplets where the optical feedback is provided by the whispering gallery modes (the Mie resonances) instead of mirrors is clear[9].

Recently, we have studied, using invariant imbedding, the probability distribution of backscattering intensity of coherent radiation in scalar approximation when the medium is attenuating/amplifying coherently as simulated by introducing appropriately signed imaginary part to the local dielectric constant. An important point to note here is that a coherent beam (from a laser) is essentially an eigenstate of the photon annihilation operator. There is no loss of phase coherence due to absorption which is stochastic (acts at the level of fields) rather than deterministic (which would act at the level of intensity).

In conclusion, let me say that scattering of light in a disordered optical medium is a paradigm of wave phenomena. It has already offered and continues to offer novel insights. Much of the study so far has centred on *passive random* media. One could perhaps now look for surprises in the *active random* media.

Trying looking through the ground glass is not a bad business after all!

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