Life before mean free path

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A coherently amplifying medium containing dense random weak scatterers can exhibit mirror-less lasing beyond a threshold of optical pumping even when the active medium has linear dimensions much smaller than the estimated transport mean free path. The threshold pump power decreases with decreasing mean free path. This mirror-less lasing can be understood in terms of the sub-mean free path scatterings which are normally statistically rare, but are now made effective by the coherent amplification that more than offsets their otherwise low probability of occurrence.

WAVE propagation through a spatially random scattering medium holds many surprises of condensed matter physics, of which the best known example is Anderson localization[1] – weak as well as strong. The wave in question may be a complex scalar (e.g. the quantum mechanical probability amplitude as in the case of electrons moving in a disordered solid), or a real scalar (e.g. the sound wave propagating in an elastically disordered medium), or a real vector (e.g. the light wave propagating through a disordered dielectric, which is of interest here). It may, however, even be a real tensor (e.g. the gravitational wave scattering on a random background metric, as perhaps in the early universe!). A closely related phenomenon here is that of the large statistical fluctuations of the wave transmission/reflection coefficient – over an ensemble of macroscopically controllably identical but microscopically uncontrollably distinct realizations of the spatial (quenched) randomness, making the system non-self-averaging. The strong as well as the weak localization, and the sample-to-sample statistical fluctuations (or *the reproducible noise*) are all diverse manifestations of the common phenomenon of intermittency which is favoured by randomness. Here intermittency refers to the occurrence of rare events that are nevertheless intense enough to affect the statistics drastically. For the wave motion in a random medium it is due generally to the coherent multiple scattering, and more specifically to the Coherent Back Scattering (CBS)[2], i.e. the partial wave-amplitudes counter-propagating along the same return path acquire identical phase-shifts (see Figure 1). The amplitude doubling resulting from these time-reversed partial waves returned in phase refocuses the scattered wave in a direction opposite to that of incidence. This sharp feature persists no matter how strong the disorder is so long as the medium is time-reversal symmetric, e.g. no external magnetic field. (Caustics and Glory are the common examples of intermittency known from optics.) The CBS refocussing, in fact, defines a cone of finite opening angle which is typically a few milli-radians for light, and increases with disorder.

Now, the disordered medium referred to above has been so far assumed to be passive, which is the only kind of spatial randomness relevant to the case of electrons (fermions). But, photons are bosons admitting the possibility of coherent amplification by stimulated emission beyond a threshold of optical pumping. This has led to a novel development, namely that of light propagation in a Random Amplifying Medium (RAM)[3–11]. Such a random amplifying medium is readily realized as a colloidal suspension of dielectric microspheres in the solution of a laser-active dye, optically pumped by an appropriate pulsed laser. Thus, for example[4], the random scatterers could be micron-sized spheres of rutile (TiO_2) dispersed in methanol giving a high refractive index contrast. The dye solution could be rhodamine 640 perchlorate in methanol with an emission peak at ~ 617 nm. The pump could be a frequency-doubled pulsed Nd:YAG laser operating at 1.064 μm. The main point to note here is that the coherent amplification

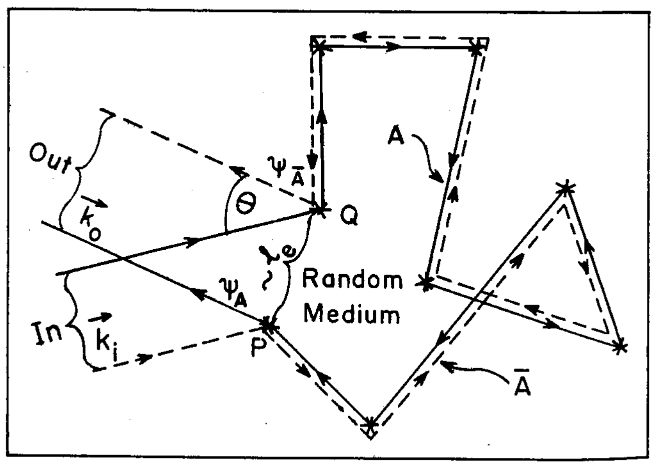

Figure 1. Coherent back-scattering. The path A (solid line) and its time reversed path $\bar{A}$ (dashed line) visit the same scatterers but in the reversed sequence, giving identical phase shifts, for $\theta = 0$.

maintains the condition for coherent back-scattering, and, indeed, it enhances the various intermittency effects noted above, e.g. the higher the amplification the narrower the CBS cone angle; also greater is the tendency to localization inasmuch as the longer return paths now contribute more effectively to CBS.

Mirror-less lasing in such a RAM has been reported by several experimental groups[4,7,11]. Fundamentally, it may involve one of the two distinct mechanisms – the Anderson localization that provides a virtual high-Q cavity giving a resonant positive feedback[3]; or it may result from diffusion, a non-resonant distributed feedback from the enhanced path lengths traversed by the diffusing photon[12]. Such a randomly folded optics in a RAM subtends a high gain. We are concerned here with this latter case. However, it raises an interesting question, namely, what if the medium has linear dimensions much smaller than the transport mean free path l^*? Such a sub-mean free path medium can hardly be expected to subtend diffusively the prolonged path lengths, much less localization; and hence no mirror-less lasing is to be expected. But, there is now experimental evidence of and theoretical support for mirror-less lasing in this case too[11]. This can be physically understood in terms of the statistically rare scattering events occurring before the mean free path, which become effective because of the high-gain RAM. A RAM with Dense Random Weak Scattering (DRWS), indeed, provides an interesting example of what is known in a game of chance as the St. Petersburg paradox, where orders of magnitude lower probability odds have orders of magnitude higher gains, upping thereby the *ante*.

Mirror-less lasing in sub-mean free path RAM: Experimental

Recent experimental results on lasing in a RAM in the limit of sub-mean free path sample size suggest that a novel mechanism is at work. The random amplifying medium studied by us[11] was an aqueous suspension of polystyrene microspheres containing the rhodamine 590 dye. The RAM parameters were:

Dye concentration: 5×10^{-5} M – 5×10^{-2} M (mole/litre); polystyrene microspheres: 0.12 μm diameter; polystyrene microsphere concentration $n_s = 10$ cm^{-3} – 10^{13} cm^{-3}; pump (p) frequency-doubled Nd:YAG; pump wavelength $\lambda_p = 532$ nm; pump pulses duration: 10 ns; pump repetition rate: 10 pps; energy deposited per pulse (E): 50 μJ – 13 mJ; active medium size ≈ 1 mm; estimated transport mean free path l^* (μm) = [(4.35×10^{15})/n_s)] (4.35×10^{14} μm – 4.35×10^2 μm); gain narrowing was observed for $n_s > 10^9$ cm^{-3}.

Figure 2 shows a typical plot of gain narrowing enhancement by random scattering. The surprise, however, is that the effect persists even for the sample size $L \ll l^*$, the photon transport mean free path when the normal diffusion can hardly be expected to dominate.

Thus, for $n_s = 1.24 \times 10^9$ cm^{-3}, $l^* = 350$ cm, and for $n_s = 1.24 \times 10^{11}$ cm^{-3}, $l^* = 3.5$ cm while $L \sim 0.1$ cm. And for proper diffusion, we need to have the sample size $\geq 6l^*$. Thus there seems to be unexpected life *before* the mean free path !

Mirror-less lasing in sub-mean free path RAM: Theoretical

Consider a random amplifying medium characterized by a scattering length l_s, and a gain length l_g in the absence of scattering. (We assume the scatterers to be isotropic so that $l_s = l^*$, the transport mean free path given by $n_s l_s \sigma_s = 1$, where n_s is the number density of the scatterer of scattering cross-section σ_s.) For diffusive motion in a RAM with uniformly excited medium, optical energy-density ρ can be described by the diffusive–reactive equation:

$$\frac{\partial \rho}{\partial t} = D\nabla^2 \rho + \frac{1}{\tau_g}\rho, \qquad (1)$$

where the diffusion constant $D = (1/3)cl_s$ and the gain time $\tau^{-1}{}_g = c/l_g$, with c the speed of light in the averaged

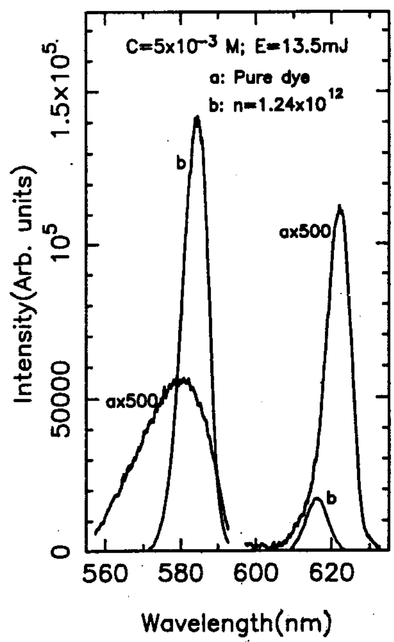

Figure 2. Emission spectrum from a RAM as function of the scatterer density at a fixed pump power. Gain narrowing for higher scatterer density is clearly seen even with the sample size less than transport mean free path (ref. 11).

refractive index medium. Physically this diffusive description is valid over length scales $L \gg l_s$. (Formally, eq. (1) is valid on all length scales in the limit $l_s \to 0$, $c \to \infty$, and $l_g \to \infty$, keeping D and τ_g constant.) This diffusion–reaction equation describes the threshold condition for lasing in much the same way as the corresponding equation for neutrons describes the criticality of chain reaction in a nuclear reactor. The gain length l_g decreases with increasing optical pumping.

Equation (1) is, of course, not valid under the sub-mean free path condition $l_s \gg L \equiv$ the linear dimension of the active medium, as is the case in the experiment discussed above for a RAM with dense random weak scatterers. For the DRWS, we have $(\frac{4\pi}{3} \cdot l_s^3)n_s \gg 1$ (dense), $1/kl_s \equiv$ disorder parameter $\ll 1$ (weakly scattering) with $\lambda = 2\pi/k =$ the wavelength of light and $L \ll l_s$ (sub-mean free path). We will now work out the contribution of the rare sub-mean free path scatterings to lasing in a RAM. First, let us note that for an arc length s traversed by a diffusing photon in a passive DRWS medium, the mean number s of scatterings is $s/l^* \equiv \mu$. Now, for a Poissonian distribution of the discrete scattering events in a continuum, the probability for m events in the arc length s will be $P_s(m) = e^{-\mu}\mu^m/m!$ Thus there is a non-zero, albeit small, probability of undergoing scattering even for $\mu \ll 1$ (i.e. $s \ll l^*$). Normally these rare events are ignorable in any reckoning. For a RAM, however, this statistical rarity may be offset by the medium gain. As a rough estimate of this effect, consider the probability of undergoing at least one scattering back into the finite RAM of size L. It is ~$(1 - \exp(-L/l^*))$, in one-dimension. The associated gain factor is $\exp(L/l_g)$. Now, for high enough gain (pumping), i.e. for small enough l_g, one can have the product $(1 - \exp(-L/l^*)) \exp(L/l_g) > 1$. For sub-mean free path-sized medium, $L \ll l^*$, we get the threshold condition $(L/l^*)\exp(L/l_g) \geq 1$, which is indeed realizable. We expect this result to hold qualitatively even in higher dimensions.

An essentially exact analytic treatment of the lasing threshold is possible for a model RAM in one-dimension as outlined here. This treatment covers the localization as well as the diffusion limit noted above. The Maxwell wave equation for a time-harmonic electric field vector = Re$E(x)\exp(i\omega t)$ propagating in a RAM is:

$$-\nabla^2\mathbf{E} + \nabla(\nabla\cdot\mathbf{E}) - \left(\frac{\omega^2}{c^2}\right)\varepsilon_r(x)\mathbf{E} = \varepsilon_0\left(\frac{\omega^2}{c_0^2}\right)\mathbf{E}, \qquad (2)$$

with ω = circular frequency, and c_0 = the speed of light in vacuum. Disorder in the medium is introduced here phenomenologically through a dielectric constant $\varepsilon(x) = \varepsilon_0 + \varepsilon_r(x)$, where $\varepsilon_r(x)$ is the spatially random part of it modelling quenched disorder and fluctuating about ε_0, the mean dielectric constant. It is now possible to simulate coherent amplification (gain) by adding a negative imaginary part to the dielectric constant. Thus we consider $\varepsilon_0 \to \varepsilon_0' - i\varepsilon_0''$, and take ε_0'' (> 0) to be constant. With this, $\varepsilon(x) = \varepsilon_0' - i\varepsilon_0'' + \varepsilon_r(x)$, where $-i\varepsilon_0''$ amplifies the wave while $\varepsilon_r(x)$ scatters it, both without causing decoherence. We have assumed here local isotropy of the dielectric tensor.

It is readily seen that, but for the second depolarization term on the left hand side of eq. (2), we have a Helmholtz equation well-known in the electronic context. Further simplification results by noting that $(\nabla\cdot\mathbf{E}) = -(\nabla\ln\varepsilon(x))\cdot\mathbf{E}(x) + (1/\varepsilon(x))\nabla\cdot\mathbf{D}(x)$, where $\mathbf{D}(x)$ = displacement field. Assuming no free charges, we have $\nabla\cdot\mathbf{D}(x) = 0$, and hence $\nabla\cdot\mathbf{E} = -(\nabla\ln\varepsilon(x))\cdot\mathbf{E}(x)$. Now, for a transverse electromagnetic mode propagating along an optical fibre (for a 1-dimensional propagation) with the dielectric constant varying randomly along the propagation direction, this quantity vanishes identically, and the wave equation (eq. (2)) reduces to the Helmholtz equation:

$$\frac{\partial^2\mathbf{E}}{\partial x^2} + k^2(1 + \eta_r(x) + i\eta_a)\mathbf{E} = 0, \qquad (3)$$

with $k^2 = \frac{\omega^2}{c^2}\cdot\varepsilon_0'$, $\eta_r(x) = \frac{\varepsilon_r(x)}{\varepsilon_0'}$ and $\eta_a = -\frac{\varepsilon_0''}{\varepsilon_0'}$.

Despite the formal similarity, eq. (3) differs from its electronic counterpart in an important respect even in the absence of the amplification factor ($i\eta_a$). The scattering term $k^2\eta_r(x) \equiv (\varepsilon_r\omega^2/c_0^2)$ involves the eigenvalue ω^2 multiplicatively. Indeed, this term is responsible for the famous $1/\lambda^{(1+d)}$ Rayleigh scattering (in d dimensions in the first Born approximation). Here it becomes small and ineffective in the limit of low frequency/long wavelength. This is not so for the electronic case. Thus, localization of light is suppressed in the low frequency limit, while for the electron the low energy tail is easily localized. In the high frequency limit, of course, the problem is no different from the electronic case and we have the geometrical–optical limit that again makes localization difficult. Indeed, localization for photons is most demanding and requires a combination of strong single-particle resonance scattering (high dielectric contrast for the scatterers) and a pseudo-gap providing a Bragg reflection–resonance condition $\mathbf{k}\cdot\mathbf{G} = ½\mathbf{G}\cdot\mathbf{G}$. This helps satisfy the Mott–Ioffe–Bragg condition for localization. One-dimensionality is, however, an exception and arbitrarily small disorder can cause exponential localization.

The Helmholtz equation will now be studied for super-radiant reflection of an incident wave of unit amplitude. This should reveal the synergetic enhancement of amplification due to the localization effect of the dielectric disorder, as also due to the lengthening of the path due to diffusion. The imbedding equation for the amplitude reflection coefficient $R(L)$ is now[13]:

$$\frac{dR(L)}{dL} = 2ikR(L) + \frac{ik}{2}(1+\eta_r(L)+i\eta_a)(1+R(L))^2, \quad (4)$$

with $R(L) = (r(L))^{1/2}e^{i\theta(L)}$ and $R(L=0) = 0$. Here $r(L)$ is the intensity reflection coefficient.

We take the disorder to be a gaussian white noise

$$\langle \eta_r(L) \rangle = 0,$$

$$\langle \eta_r(L)\eta_r(L') \rangle = \eta_0{}^2\delta(L-L'). \quad (5)$$

In the random phase approximation, where we assume the joint probability density $p(r, \theta, L)$ to factorize and θ to be uniformly distributed over 2π, the Fokker–Planck equation for the marginal distribution $p(r, L)$ is obtained as:

$$\frac{\partial p(r,l)}{\partial l} = r(1-r)^2\frac{\partial^2 p(r,l)}{\partial r^2} + [1+(-6-D_s)r+5r^2]$$

$$\times\frac{\partial p(r,l)}{\partial r} + [(-2+D_s)+4r]p(r,l). \quad (6)$$

Here $l = L/l_s$, the dimensionless sample length, $l_s = 2\eta_0{}^2k^2 =$ the localization length (ξ), and the gain parameter $D_s = 4\eta_a/\eta_0{}^2k = l_s/l_a$, with the amplification length $l_a = 1/2\eta_a k \equiv$ gain length l_g. (In 3-D we can relate the transport mean free path l_g, the gain length l_g and the speed of light c in the medium to the parameters $\eta_0{}^2$, ε_0'' and ε_0' characterizing scattering, amplification, and the mean dielectric constant of the medium as:

$$l_s = \left(\frac{4\pi}{\eta_0^2k^4}\right),\ l_g = \frac{1}{|\varepsilon_0'|k},\ c_s = c\sqrt{\varepsilon_0'},$$

(Dimensionality is important).

Equation (6) has been solved[3] analytically in the asymptotic limit $l \to \infty$. We are, however, interested here in the sub-mean free path limit $l \ll 1$. Numerical solution in this limit confirms super-reflection with $\langle r(l) \rangle \gg 1$ for sufficiently large gain. Such a treatment is directly applicable to a single mode polarization maintaining optical fibre doped with the rare-earth laser-active ion Er^{3+}, pumped optically, and having some refractive index randomness along its length. Recently, a generalization of the above treatment of RAM to the case of N-channels (modes) has been achieved by Beenakker *et al.*[8]. Their treatment is based on the DMPK equation[14] and contains a number of new results.

Conclusions

Lasing observed in a sub-mean free path-sized random amplifying medium can be understood in terms of the statistically rare scatterings over length scales $< l^*$, which become effective due to the high gain in the optically pumped medium beyond a threshold. It will be interesting to analyse what determines the emission wavelength and its line-width as also the effect of non-linearity, e.g. saturation, which is particularly important beyond the threshold for the onset of lasing.

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