Mirrorless Lasing in Random Amplifying Media

S. Anantha Ramakrishna

Raman Research Institute, C.V. Raman Avenue

Bangalore 560 080

In this article the phenomenon of lasing from Random Amplifying Media (RAM) has been reviewed, which evoked much interest in recent years. After briefly describing the transport of light in passive disordered media, the two different pictures of lasing from such random media are described: the light diffusion with gain and the coherent amplification with coherent feedback due to Anderson localization of light.

Can a random system have an ordered output? The mention of a laser evokes the picture of a highly (spatially, temporally and spectrally) coherent source of light. In other words, it is an highly ordered system. True enough, care is generally taken to ensure that the laser gain medium and cavity are homogeneous and non-scattering, for otherwise, the pump and emitted beams would be scattered out of the cavity and the coherence would be lost. Even the presence of a small inhomogeniety in the cavity can cause mode-hopping, intensity fluctuations etc. and can be a major source of noise in an otherwise highly ordered output. Can this seemingly antithetical nature of disorder and lasing be reconciled and can lasing be actually aided by the deliberate introduction of scatterers into a cavity? This question has evoked much scientific debate for the greater part of the past decade. In this article, I shall endeavour to convey some of the excitement that the phenomenon of random lasers have generated.

It all began with the experimental discovery [1] of the original proposition by Letokhov[2] in 1968, that placing random scatterers in a gain medium could enhance the frequency stability of the laser emission. In these experiments, it was found that the introduction of disorder (by suspending Titania microspheres) into a homogeneous Rhodamine laser dye solution caused a drastic spectral narrowing of the emission from the dye above a well-defined threshold of pump light intensity. Sure, this is a well-known phenomenon which occurs even in homogeneous amplifying media due to Amplified Spontaneous Emission (ASE). But the remarkable aspect in these experiments was that, the threshold of the pump laser intensity at which the flourescence spectrum collapsed dramatically was almost two orders of magnitude smaller in the case of the microsphere-laser dye suspension, compared to the case of the ASE in the neat laser dye solution. Later experiments showed that the emitted pulses from the Random Amplifying Medium (RAM) exhibited temporal shortening (much like in high gain lasers) [3], and a bichromatic emission at some intermediate pumping levels [3]. Lasing in these media occur even at very small concentration of the scatterers, i.e. even when the mean free path of light in the medium is larger the sample size [4]. In the recent experiment of Cao *et al.*[5], where the lasing from very strongly scattering semiconductor powder was studied, a connection between lasing and Anderson localization of light has been claimed.

Let us first discuss a typical experiment. It seems deceptively simple at first sight. Take a beautiful laser crystal and grind it into a fine powder (size ~ 1m m). Now excite this powder by pumping it in the absorption band with pulses from a powerful laser and carry out spectral and temporal measurements of the emission from the powder in different directions. Another option would be to suspend scattering microparticles such as titania or polystyrene microspheres in a laser dye solution, or to embed the scatterers into a laser dye-doped polymeric matrix.

Now let us see what happens to light in highly scattering random media (for the time being assumed to have no gain). On a gross macroscopic level, it is treacherously simple. The incoming beam is attenuated due to scattering and a diffuse glow emanates from the medium. On the microscopic level, however, the situation is extremely complicated. The light could be scattered several thousands of times before leaving the sample. It is clear that there are two scattering regimes. One, the single scattering regime, when the scatterers are weakly scattering and the density of the scatterers is also small, so that the probability of light that is scattered once to be scattered again by other scatterers is extremely small. In this case, the mean free path of the light is much larger than the sample size and the scattering acts only as small perturbation on the incident light. The other case is that of strong scatterers present in a high density so that the light is multiply re-scattered many times. In this multiple scattering regime, the scattering is a very strong perturbation on the incident light field and the field amplitude at any given scatterer is strongly modified by the presence other scatterers. It is in the multiple scattering domain that most of the random laser experiments have been performed in and consequently, in which we will be interested here.

The transport of light in multiply scattering media can be described at three levels of varying sophistication. At the first, gross level of description as an incoherent energy transport (shown to be valid for $kl^* >> 1$ where k is the wave vector and l^* is the mean free path of light in the medium), light can be thought of as particles bouncing of randomly placed scatterers with the coefficient of restitution being unity (the bouncing photon picture). Sure enough, this random walk problem is accurately described by a diffusion equation for the light energy density at large length scales ($L>>l^*$). More sophisticated theories of persistent random walks where the photons retain some directional memory have been developed [6] and accurately describe both the ballistic transport at short length scales (L ~ l^*) and the diffusive transport at large length scales ($L >> l^*$). At the second level of description in terms of a wave transport (necessary at $kl^* \sim 1$), interferences of the scattered light become very important. Coherent backscattering of light where an enhanced scattering (doubling of the intensity) occurs within a cone of directions around the backscattered direction is perhaps the most well known phenomenon of such an (constructive) interference effect in disordered media. All important in strongly scattering media are recurrent scattering events, where the light continuously traverses a (closed) loop like path involving the same set of scatterers. As will be seen later, these paths can form random cavities and cause feedback much like a laser ring cavity. In fact, for very strongly scattering media, the effects of such interference and recurrent scattering can result in total spatial localization of light (Anderson localization)[7]. This was predicted to occur when the mean free path of light became very small (kl^* ? 1 - known as the Ioffe-Regel criterion) and has been experimentally found to occur [8]. In this situation, light simply stops flowing over long length scales, i.e. the energy is spatially trapped over a length scale known as the localization length. In other words, the diffusion coefficient goes to zero and ``light stands still". The third level of description at a quantum optical level, involves the photon statistics and field quantization in random media. Beenakker *et al.*[9] have worked extensively on the photon statistics problem in one-dimensional random media, but much remains to be done in this area.

Now the question is, what happens when such highly scattering media become amplifying as well? First of all, let us consider what happens in a conventional laser. The amplifying (homogeneous) medium is placed in a one-dimensional optical cavity, which traps the light emitted spontaneously by the excited medium, forcing it to repeatedly pass through the amplifying medium while undergoing amplification due to stimulated emission in each traverse. Laser action occurs when the amplification in each round trip exceeds the loss at the cavity output couplers and the optical elements in the cavity. Then a coherent mode builds up in the cavity and results in a unidirectional, monochromatic, coherent beam. The frequency of the laser light is at an eigen-frequency of the cavity, near the maximum of the gain spectrum of the amplifying medium. Thus, the laser action occurs due to a coherent feedback of light from the mirrors.

Next, let us look at a multiply scattering amplifying medium with weak, dense scatterers, where the diffusion picture holds. The light emitted spontaneously by the medium will have to undergo a long random walk inside the medium (which could be orders of magnitude longer than in a homogeneous medium) before exiting the medium. During every part of this random walk, the light is continuously amplified due to stimulated emission. Thus, gain can become larger than loss and one can have lasing action caused by diffusive feedback. The output loss is proportional to the surface are of the medium through which the light exits while the gain is proportional to the overall volume of the medium. As the size of the RAM is increased beyond a point, the gain becomes larger than the loss and a strong flash of light will occur in all directions. This is exactly analogous to what happens in a fission nuclear bomb or reactor, where neutrons are scattered and amplified by nuclear fission - the system becomes super-critical beyond a critical size of the system. In such random laser, one notes that there will be a large gain narrowing of the emitted spectrum, as photons at the emission maximum would be much more amplified in their long passage through the medium. Thus, it is a ``feed-forward mechanism" that causes lasing action compared to the feedback mechanism in a conventional laser. It is this large extension of the path length in the RAM due to multiple scattering that is responsible for the lowering of the threshold pump intensity. Note that in comparision to the unidirectional output of a conventional laser, the random laser will emit in all directions. In Fig.1, experimental spectra obtained from a RAM (consisting of polystyrene microspheres suspended in a Rhodamine 6G solution) obtained below (curve-a) and above (curve-b) the threshold of lasing are shown. Above the threshold, the FWHM of the spectrum is about 6nm. Also, due to the very lage intensities which occur during the lasing, the population inversion in the RAM is quickly depleted, leading to a sharp temporal peak or pulse shortening of the emission. If the exciting pump beam is still present, then the population inversion will again build up and deplete successively, resulting in a train of spikes in the emission.

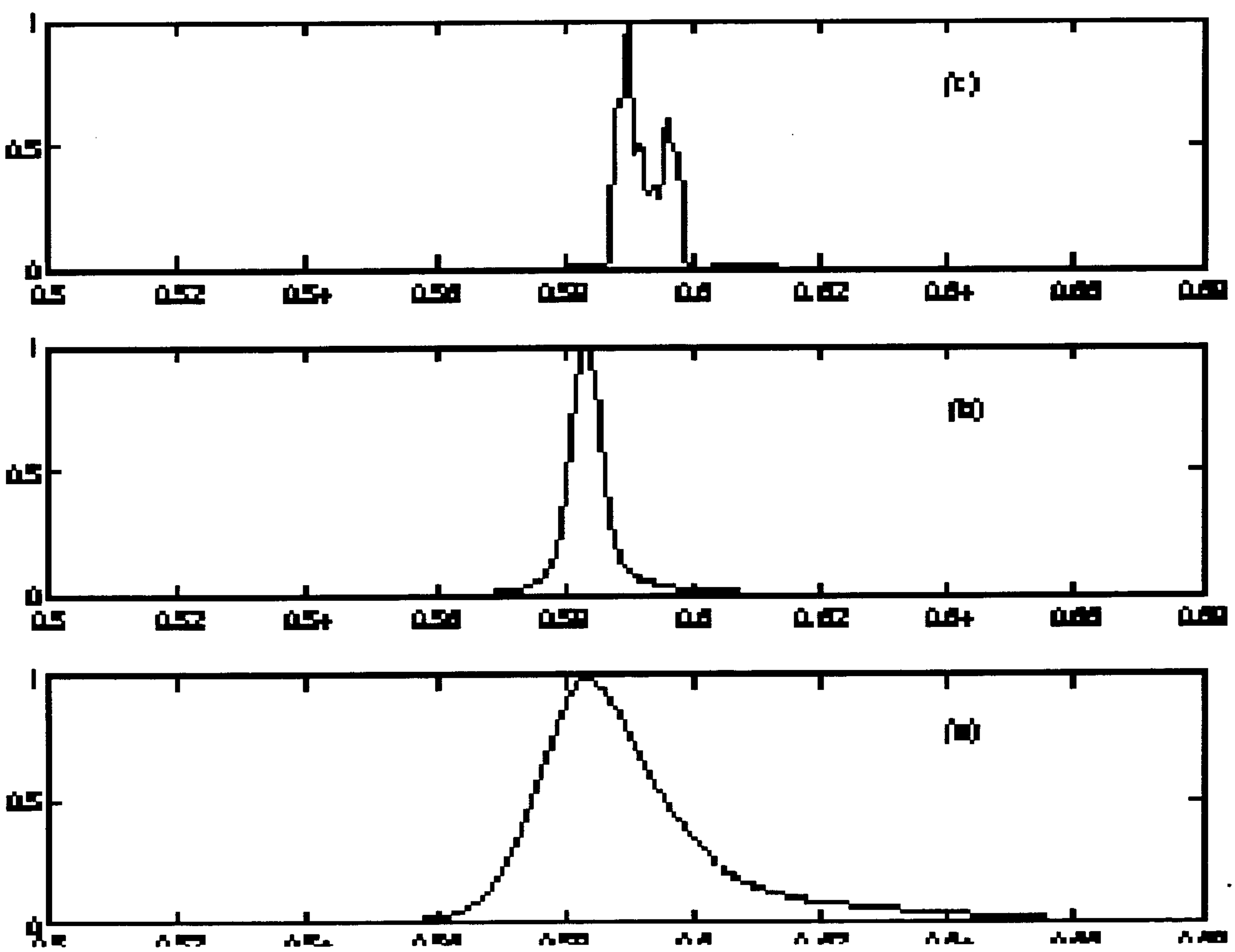

Fig. 1 : The normalized spectral emission from a RAM consisting of polystyrene microspheres suspended in a Rhodamine 6G solution (a) sub-threshold (b) above-threshold (c) bichromatic emission at intermediate pump levels

Coming to the other details of the emission, it was found that at some intermediate pump powers, a bichromatic emission results (See Fig.1, curve-c). This has been explained as a result of the displaced absorption and emission spectra of the RAM, based on Monte Carlo simulations [10]. Another extremely interesting feature observed in these media is that there is a sharp reduction in the threshold of pumping even when the transport mean free path of light is much larger than the sample size [4]. This would naively imply that there are no multiple passes through the medium as the photons escape ballistically. This effect could, however, be explained[4] by considering the effect of the probabilistically rare sub-mean-free-path large-angle scattering events, which now are rendered important by the virtue of high gain in the medium. Thus, a manifestation of the paradox wherein rare event but with a very large multiplier value can become more important than more likely events, but with a small multiplier value.

Finally, we look at very strongly scattering ($kl^* \sim 1$) and amplifying media where the diffusion picture breaks down and localization effects become important. Also, in the above picture of light diffusion with gain, the amplification could very well have been an incoherent process (such as in the generation of neutrons in a nuclear fission reaction where the outgoing neutrons have no phase correlations with the incoming neutrons). Stimulated emission is, however, a coherent process where the emitted photons have the same phase and direction as the incoming photon. In fact, this is a bosonic property without a fermionic analogue. This property of coherent amplification of light coupled with the coherent feedback offered by Anderson localization can result in a new kind of coherent laser output. This enhancement of amplification due to a synergy between coherent amplification and wave confinement by localization was theoretically predicted by Pradhan and Kumar[11] in 1994. In this case, the recurrent multiple scattering events forming closed loops provide feedback much like in the ring cavity lasers. Of course, due to the random nature of the medium, there would exist a distribution of many such random cavities with different Q-factors. As the pump laser intensity is increased, the laser threshold condition (gain =loss) would be satisfied first in the cavities with the highest Q-factors. Laser oscillation would occur at frequencies determined by the cavity resonances. Thus, the laser emission would consist of sharp discrete lines. At higher pump intensities, oscillations would begin even in lossier cavities and more lines would be added to the emission spectrum. One notes that the random cavities formed by recurrent multiple scattering would be totally different in different samples Thus, the lasing lines would vary from sample to sample, a typical effect found in disordered systems. Also, the random cavities would have outputs in different directions and the emitted spectrum would be different in different directions and we have a multi-mode laser in all directions. In the recent experiments of Cao *et al.* [5] with very strongly scattering semiconductor powders (ZnO, GaN), all the above effect were observed to occur. The lasing lines had extremely small spectral linewidths (~ 0.2nm). Thus, a random laser with coherent feedback was claimed. However, one should note that, in these experiments the semiconductor powders were deposited on glass substrates as a thin film of powder(~ 10 m m). Hence, keeping in mind the large dielectric constant of the semiconductor material, it is possible that total internal reflection at the interfaces could have effectively acted as mode-guiding mechanism, thereby effectively reducing the dimensionality of the random system to two. Thus, though there are doubts that the fine spectrum observed in these experiments could be due to a reduced dimensionality of the system when localization effects are easier to achieve, there is no doubt that the coherent feedback mechanism due to Anderson localization has been demonstrated.

In conclusion, I have reviewed the phenomenon of lasing from a random amplifying medium. These media have already found application in photonic textile fibres [12] to produce photonic codes for a variety of civilian and military identification schemes. In the context of photonic bandgap media, disorder can induce pseudo band gaps even in media without a complete photonic bandgap. This, coupled with amplification provides a new frontier for quantum optics. Novel and fascinating avenues for fundamental research and new technical applications seem to be guaranteed.

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