Chapter 8

Optical diode

8.1 Introduction

The extensive theoretical[1, 2, 3] and experimental[4, 5, 6, 7] work carried out on random lasers has made obvious the potential applications of these materials. They may be used as intense, narrowband sources of light, or as sensors, or in a variety of other civilian and military applications. We consider one such application of the random laser in this chapter.

In this chapter, we use the model of the random laser developed in the previous chapter to engineer the emission spectra of the laser. The commonly observed spectral characteristics of liquid amplifying media have been used to design and experimentally realise an optical device that prevents the propagation of a band of wavelengths in one direction and permits it in the opposite, thus acting as an optical diode. Addition of random scattering centres is seen to narrow the width of the forbidden band. A model has been proposed to explain the observations and is verified by Monte-Carlo simulations. In particular, we exploit the critical dependence of the spectral profiles of such media on the concentration of the amplifying medium. By simple manipulation of the concentration profile of the medium, we show that the emissions from the random lasers can be easily tailored to suit our purpose.

8.2 Experimental setup

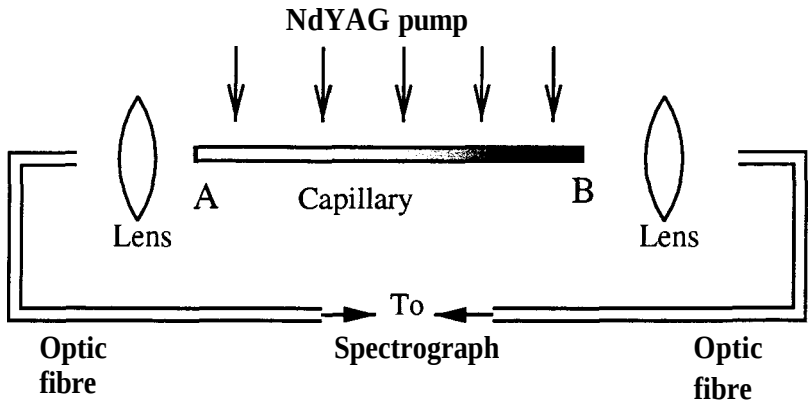

Figure **8.1:** *Schematic of the experimental setup. The capillary is pumped longitudinally and the emissions from the two ends are analysed.*

A diode is an essential element in most electronic circuits, permitting a current in one direction and blocking it in the other. It is known that most optical elements, if transmitting in one direction, transmit in the reverse too. Here we employ the characteristic of the gain medium to an advantage, to break the forward and reverse symmetry and to achieve directional propagation of light.

The device consists of a glass capillary, about **10** cm long and **100** μm in diameter, filled with ethanol. A drop of high concentration **(0.1** M) Rhodamine **B** solution in ethanol was introduced at one end of the capillary. The large concentration gradient sets up diffusion of dye molecules into the region of the ethanol and soon an exponential concentration gradient of the dye is obtained along the length of the capillary. To speed up the process of formation of the gradient, gentle taps from a piezo device at one of the ends of the capillary may be applied. The capillary is held horizontal (Fig 8.1), and pumped uniformly along the length by a line focussed, frequency doubled pulsed NdYAG laser light at 532 nm and the fluorescent emission emanating from the two ends A and B (A with low dye concentration) of the capillary is viewed on a spectrograph. To add randomness to the system, the capillary was first filled with a colloidal suspension of polystyrene microspheres (n = 1.59, diameter 0.21μ, concentration $10^{12}/cc$) suspended in ethanol. The dye was added and an

exponential concentration gradient obtained, now accompanied by a random variation of the refractive index. The degree of randomness could be varied with the number density of microspheres.

8.3 Experimental observations

At low pump powers, the spectra at ends A and B were similar, while at higher powers, the spectrum from the end A peaked sharply at around 588 nm, with a width of 10 nm while that from end B peaked at around 608 nm, with about the same width (Fig. 8.2).

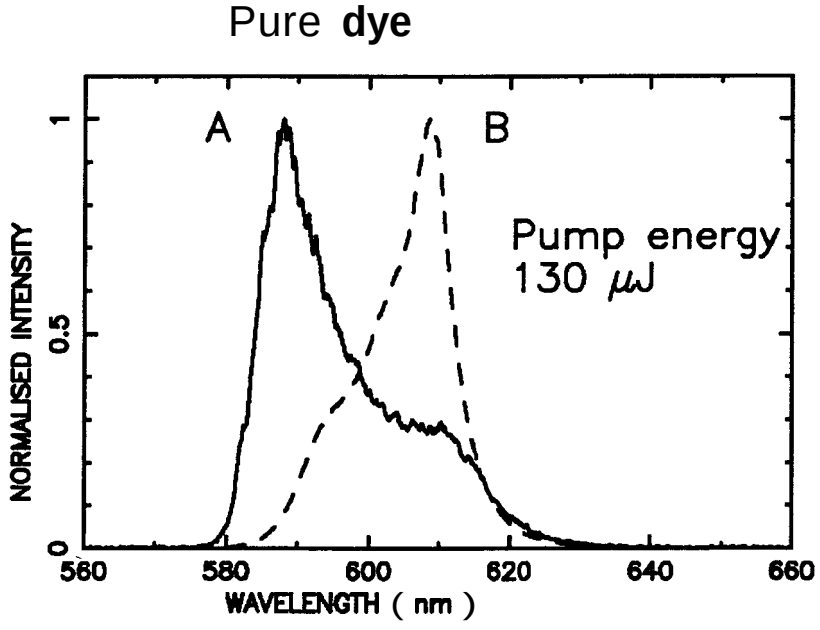

Figure 8.2: *Experimentally observed emission spectra at the two ends of the diode, with homogeneous amplifying medium. End A is the low concentration end.*

The separation between the two peaks was close to 20 nm and the overlap quite small. While the dye emits both along +X and -X directions (assuming the capillary length along X axis) and over a range from the yellow to the red at any point along the length of the capillary, yellow is emergent from end A alone, and red from end B alone. Thus, a narrow band of the yellow wavelengths, of a width of about 10nm centred at around 585 nm could come out of end A but was not allowed to propagate through end B. That is, effective

transmission for some wavelengths takes place along the +X direction, and for others in the - X direction. Thus the device acts as a diode. At quite high pump powers, the two emissions once again look very similar, and there is free propagation of all wavelengths in either direction. We also studied the performance of the diode when the gain medium had refractive index inhomogeneity due to the addition of the the microspheres of various number densities, and found that the pass bands of the two ends could be tuned. We present results for the scatterer concentration of 10^{12}/cc in figure 8.3, where the spectra at ends **A** and B are seen to be centered at 586 nm and 588 nm respectively, and the width of the forbidden band is just over **2** nm. The anisotropy of emission persisted over a smaller range of pump powers in this case.

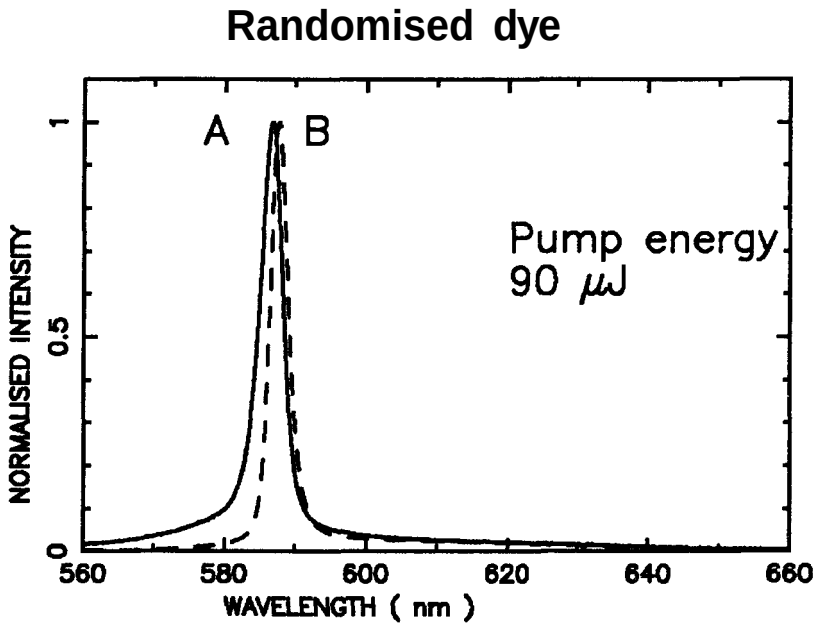

Figure 8.3: *Experimentally observed emission spectra from the diode, with scatterers in the amplifying medium.*

8.4 Simulation results

A Monte Carlo code that simulated the system was written, based on our earlier model[8] that explains the spectral characteristics of random amplifying media in terms of spon-

taneous and stimulated emission and self-absorption. The code essentially creates 10^6 spontaneously emitted photons at random in random directions along the length of the capillary, with the emission statistics being biased by the local concentration of the dye. Each photon is tracked as it traverses the capillary and finally emerges. In the absence of scattering, the photon has a unidirectional path and an amplification given by

$$\alpha(\lambda, x) = exp[-L\{N_0(x)\sigma_{abs}(\lambda) - N_1(x)\sigma_{em}(\lambda)\}] \tag{8.1}$$

where x is the distance from the end B, N_1 and N_0 are the excited state and ground state occupation numbers, respectively, and L is the total pathlength of the photon in the medium. $\sigma_{abs}(\lambda)$ and $\sigma_{se}(\lambda)$ are the absorption and stimulated emission cross-sections at the wavelength A.

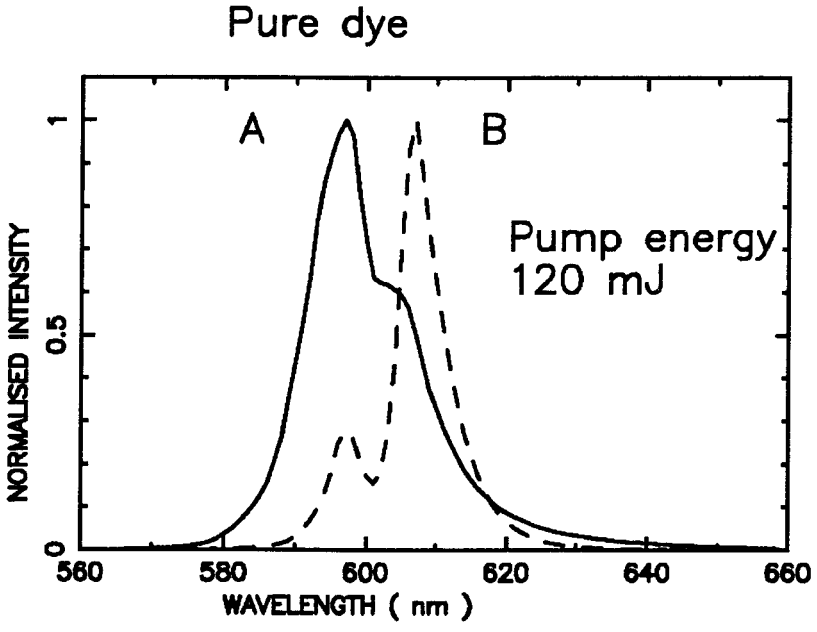

Figure 8.4: *Simulated spectra for a 1-d exponentially graded gain medium. End A is the low concentration end.*

In the presence of scatterers, the photon executes a random walk in one-dimension with an exponential step length distribution with the mean equal to the scattering mean free path. The concentration gradient of the dye was simulated by discretising the X axis into

a suitable number of bins, and populating them according to an exponential distribution. Uniform pumping was implemented by raising the same number of molecules in each bin into the upper state. Consequently, the population inversion assumes a gradation along the length. The code reproduced our experimental findings qualitatively. Figure 8.4 shows the simulated spectra of the device with the pure dye; the extent of the forbidden band is about 10 nm centered at 585 nm. Figure 8.5 shows the simulated spectra in the presence of scatterers and one can clearly see that the emission is narrowed. The experimentally observed overlap is, however, not quite reproduced. We suspect this is because the capillary, due to its finite diameter, is not the one-dimensional system considered in our simulations.

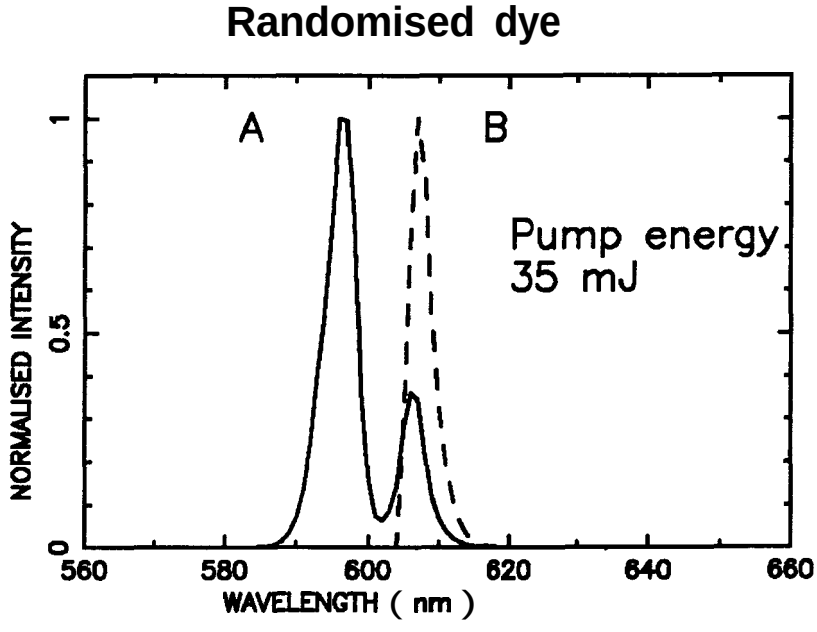

Figure 8.5: *Simulated spectra for a 1-d exponentially graded gain medium with random scatterers.*

The following behaviour of the diode can be immediately deduced, from the model used in the Monte Carlo simulations. It is well known that there is a considerable overlap in the absorption and emission spectra of a typical fluorescent dye, and this overlap decides the spectral characteristics of the homogeneous or even the randomised dye[8]. The change in emission of the dye with concentration is also attributed to this overlap, due to

which, the wavelengths that fall within the overlap are self-absorbed by the ground state dye molecules. Pumping of the molecules at high powers enhances stimulated emission, and the competition between stimulated emission and self-absorption decides the spectral characteristics of the amplifying medium.

8.5 Functionality of the optical diode

In the present case, when the dye is pumped, the spontaneously emitted photons travel either in the +X or in the -X direction, and experience an anisotropy between the two directions due to the concentration gradient of the dye. Photons travelling towards the high concentration end always encounter a stronger absorption due to the presence of more molecules in the ground state, whereas the photons travelling in the opposite direction rapidly cause stimulated emission as the population inversion is larger in that direction. Consequently, the two effects of self-absorption and stimulated emission, which usually compete with each other, now act on different photons depending on their direction. As a result, the emission in one direction, which suffers from self-absorption, is red-shifted, because the yellow wavelengths are absorbed. In contrast, the same yellow wavelengths are amplified in the other direction owing to stimulated emission, which favours those wavelengths at which the emission cross-section is larger. As a result, we notice that the band of yellow frequencies is allowed to propagate only from the high concentration end to the low concentration end, and not in the reverse. The reduced width of the forbidden band on addition of scatterers can be directly attributed to enhanced stimulated emission due to increase in the photon path lengths because of multiple scattering. This stimulated emission forces the excited molecules to emit more in the yellow wavelengths, thus eroding the forbidden band. It should be emphasised that it is the gradation in the amplifying medium profile that is the crux of the matter. Every photon, regardless of where it is born, always sees an anisotropy between the two directions. This would not be possible if, for example, one had two media positioned side-by-side, one with a higher concentration than the other, but both uniform.

Indeed, inside a liquid amplifying medium, there occurs continuous diffusion of molecules and the consequent spatial gain profile changes, limiting the above behaviour upto a certain short time interval only. This can be overcome either by maintaining a concentration gradient by some external means, or by freezing it, as, for example, polymerising the gain medium. The latter provides a means of making solid state, exponentially graded gain media, which will be useful in actual practical applications.

8.6 Conclusions

In conclusion, we have utilised the competing effects of self-absorption and stimulated emission in a fluorescent dye to propose a device that prevents a certain band of frequencies from propagating in one direction, and allows it to do so in the other, thus acting as an optical diode. This has been experimentally realised, and shown to act as an optical diode. The addition of scatterers affects the threshold of stimulated emission, and consequently alters the bandwidth characteristics of the diode. The device may be a precursor to an all-optical switch, one that is controlled by external optical field. Diodes suited for various wavelengths can be constructed using different laser dyes. Using the above mentioned tunable parameters, the diode may be perhaps used as a multiplexer-demultiplexer in all-optical circuits. Indeed, an improvised version of the device can be expected to find sweeping applications in optical networking, circuitry and engineering.

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Chapter 9

Evanescent wave pumped random lasers

9.1 Introduction

In chapter 7, we have presented our studies on the emissions of randomised liquid amplifying media and modelled the spectral features based on a numerical Monte-Carlo analysis in all the three regimes of scattering, *viz.* $L < l^*$, $L \sim l^*$ and $L > l^*$. We could explain the experimentally observed phenomena through our analysis which consisted of a three-dimensional random walk of photons within an excited medium. Most importantly, we could explain the reduction of threshold of lasing when disorder was introduced into the homogeneous amplifying medium. The model enabled us to engineer the emissions of the random amplifier by creating a graded-gain profile inside the medium, as described in the previous chapter. In the present chapter, we study the behaviour of the liquid dye in a quasi-two dimensional system, that is realised using the evanescent modes formed over a dielectric substrate.

The attempt to study such a system is motivated by two causes. We have seen that scattering within a gain medium lowers the pump energy required for the gain medium to lase, a phenomenon known as threshold reduction. Upon addition of disorder, the threshold of lasing from the homogeneous media has been observed to be reduced by orders of magnitude[1, 2, 3, 4]. Weak pump energies, therefore, suffice to generate narrowband

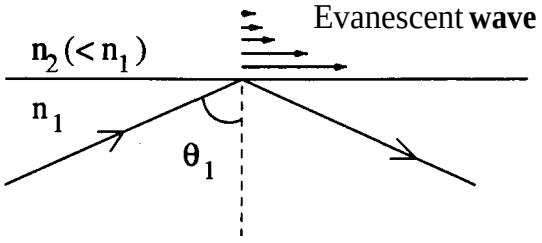

Figure 9.1: *The formation of an evanescent wave at an interface between two dielectrics. The ray is incident onto the interface at an angle greater than the critical angle from the medium with the higher refractive index. Evanescent wave forms inside the rarer medium, propagates along the interface, and has an amplitude that decays exponentially within a wavelength in the direction normal to the interface.*

emissions. If the pumping is coupled to the amplifying medium through evanescent waves, very weak excitation energies are provided to the medium. We study the emissions of the system under such evanescent field excitation, where very little energies are absorbed by the liquid amplifying medium, in our case, a high efficiency laser dye. We investigate whether, and to what efficiency, lasing is possible under such weak excitations. We investigate the effect of addition of disorder to such a system, and look for reduction of threshold in case of random lasing. Further, it is known that all photon states are localised inside a two-dimensional disordered medium[5]. Any little gain, therefore, inside the medium should enhance the emissions vastly. Such a system has been studied in three-dimensions by using strong scatterers[6], and large enhancements in intensity, accompanied by large temporal coherences, have been reported. We investigate whether such enhancements, that can be attributed to localised photon states, are indeed observed inside our system, where, as we shall now show, the pumped region is two-dimensional.

When light is incident on an interface fiom the higher refractive index medium at an angle larger than the critical angle (refer to the figure 9.1), it suffers total internal reflection and an evanescent field is generated inside the rarer medium. This field is non-propagating, and shows an exponential decay in the direction normal to the boundary surface in the rarer medium. The characteristic decay length, called the penetration depth[7], is given

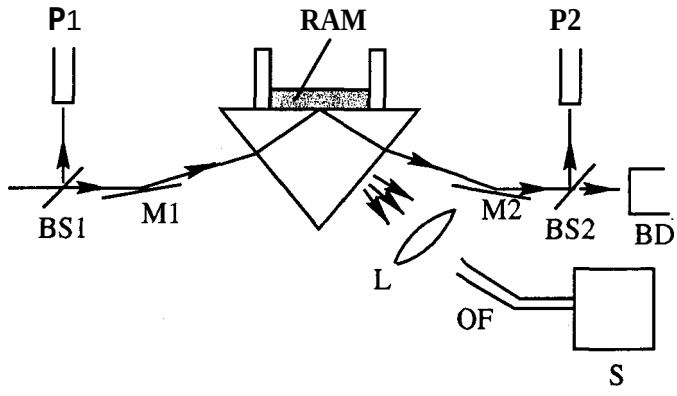

Figure 9.2: *Experimental setup for studying the emission spectrum from a random amplifying medium pumped by an evanescent field. BS1, BS2 : Beamsplitters, M1, M2 : Mirrors, P1,P2 : Probes of an energy ratiometer, L : Lens, OF : Optic Fibre, BD : Beam Dump, S : Spectrometer, RAM : Random Amplifying Medium.*

by $l = \frac{1}{\gamma}$ where

$$\gamma = \frac{\omega}{c}(n_1^2 sin^2\theta_1 - n_2^2)^{\frac{1}{2}} \tag{9.1}$$

Here, n_1 and n_2 are the refractive indices at frequency *w* inside the denser and rarer medium respectively, θ_1 is the angle of incidence and *c* is the speed of light in vacuum. This penetration depth is smaller than the wavelength of the light in the medium, and hence the light can be assumed to occupy a two-dimensional region over the dielectric surface. We utilise this characteristic to realise a two-dimensional random amplifying medium, and study the spectral features of the emissions.

9.2 Evanescent pumping

Figure 9.2 shows the schematic of the experimental setup. A gaussian beam of width 4mm (frequency doubled NdYAG, $\lambda = 532.8nm$) is first expanded using a beam expander to a width of 8mm, and the central circular region of diameter of 3 mm is extracted through an iris as the working beam. This ensures a reasonably flat intensity profile of the beam. The beam splitter BS1 redirects a part of this input beam onto one of the probes of an energy ratiometer. The residual beam is incident onto the mirror M1. The beam reflected

from M1 is incident on one of the isosceles faces of an isosceles right-angled prism, at an angle such that, the refracted beam that enters the prism hits the hypotenuse face at an angle of incidence greater than the critical angle. The beam is totally internally reflected and exits the prism, upon which the mirror M2 deflects it onto another beamsplitter BS2, which reflects a part of the beam into the second probe of the energy ratiometer. The residual beam is dumped in a beam dump. The energy ratiometer registers the amount of energy absorbed by the medium.

The amplifying medium used is the laser dye Rhodamine 6G dissolved in ethanol, which is kept within a cylindrical cavity of diameter 8mm on the hypotenuse face of the prism. The dye remains directly in contact with the glass surface of the prism. The pump beam is incident on this surface from inside the prism at an angle of 66" that is well above the critical angle, θ_c. For the glass-ethanol surface, the critical angle is 63.47" ($n_{glass} = 1.52$ and $n_{eth} = 1.36$). Upon total reflection inside the prism, an evanescent field is generated inside the rarer medium, i.e., ethanol, the decay length of which is 0.2224μ for the present conditions of reflection. The rhodamine dye is pumped by the green evanescence and emits yellow-orange fluorescence that is collected outside one of the isosceles faces of the prism, and focussed by the lens L onto the collection aperture of an optic fibre that guides it into a spectrometer. Disorder is introduced in the dye by suspending polystyrene microparticles (diameter 0.21μ, $n = 1.59$) and the degree of disorder is varied by changing the scatterer concentration. It may be of importance to note that the collection of the fluorescence by the lens is done perpendicular to the isosceles face of the prism, in other words, at 45° to the plane of excitation. Because of the rigidity of the sample holder and other mechanical factors, this angle of detection was not altered.

Due to the reflections and refractions that occur before the beam undergoes total internal reflection, the shape of the illuminated region, and hence the evanescent region, becomes elliptical, with the semiminor axis equal to the radius of the original beam and the semimajor axis approximately twice as much. Thus for radius of 3mm, the pumped region of the dye is approximately an ellipse of area π x $\frac{3}{2}$ x $\frac{6}{2}$ mm^2. The scatterer

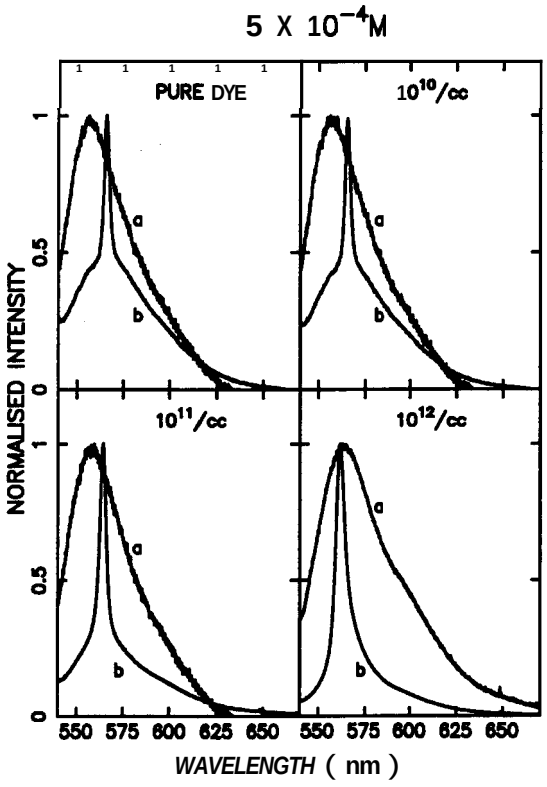

Figure **9.3:** *Emission spectra of the dye at a concentration of* $5 \times 10^{-4} M$*, at various scatterer concentrations, under evanescent pumping. Curve* (a) *is the spectrum at pump energy* $2\mu J$ *and* (b) *at* $9\mu J$*. A marked change in the spectrum at* $9\mu J$ *is seen in the case of the scatterer concentrations* $10^{11}/cc$ *and* $10^{12}/cc$*.*

concentration ranges from $10^{10}/cc$ to $10^{12}/cc$. At larger concentrations, the suspension coagulates. Thus, the transport mean free path is varied from $6.2cm$ to 624μ. Evidently, we have investigated both the diffusion $(L > l^*)$ and the subdiffusion $(L < l^*)$ regime of scattering, since our system length is of the order of ***6mm.***

9.3 Lasing under evanescent pump

We studied spectral emissions from Rhodamine ***6G*** in ethanol at three different concentrations, 5×10^{-4}M, 10^{-3}M and 10^{-2}M.We saw clear evidences of lasing, threshold reduction and linewidth narrowing from the evanescent-wave pumped system at each of the above concentrations.

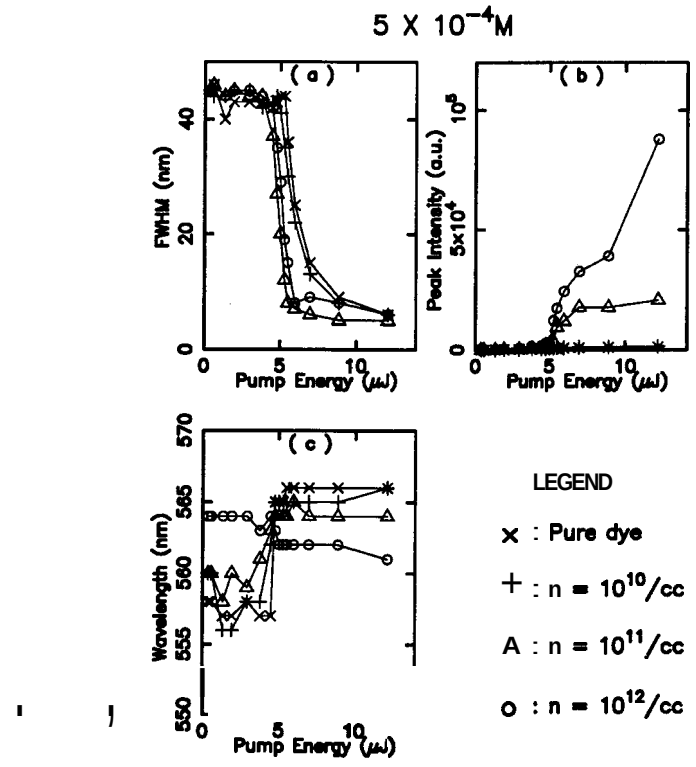

Figure 9.4: *Details of the emission profiles of the dye at a concentration of* $5 \times 10^{-4} M$, *under evanescent pumping. The subplots show, as a function of the pump energy, (a) the reduction of FWHM, (b) the peak intensity, and (c) the wavelength of peak emission. The legend is explained in the figure. The reduction in FWHM and the enhancement in emission intensity upon addition of scatterers is obvious from the figure.*

The figure 9.3 shows the emission spectra of Rh *6G* in ethanol at a concentration of 5×10^{-4}M for various concentration of scatterers. Each subplot shows two curves, curve *a* is the spectrum at the pump energy $2\mu J$ and *b* is at $9\mu J$. In all four cases, linewidth narrowing at increased pump energy is quite evident, but, more noticeably, the effect of scatterers is also clearly seen. The emission spectra show that the spectra from the pure dye and the dye with scatterer concentration $10^{10}/cc$ narrowed only partially, while that from the stronger scattering samples narrowed down completely. The peak wavelength of emission is seen to shift to shorter wavelength for the scatterer concentration of $10^{12}/cc$.

The figure 9.4(a) shows the variation of the FWHM with pump energy for different degrees of scattering. The threshold of lasing, defined as the pump energy at which the

peak intensity starts diverging and the FWHM of the emission spectrum starts falling, is seen to reduce with the addition of disorder to the homogeneous dye. Figure 9.4(b) shows the enhancement in the peak intensity of emission, which is maximum at a scatterer concentration of $10^{12}/cc$. Fig 9.4(c) shows the shift in the wavelength of peak intensity. Except for the case of $10^{12}/cc$ where a slight blueshift is seen, a systematic redshift is observed with increase in pump energy. Maximum changes in the spectral profile, like the reduction in FWHM, increase in the emission intensity and the change in the wavelength of emission, are observed at the pump energy of about $5\mu J$ as is obvious from the figure. Thus, this system starts lasing at around $5\mu J$. The addition of particles reduces this threshold only slightly, but the reduction is noticeable from figure 9.4(a). Furthermore, at any concentration of the scatterers, the emissions are always monomodal, with a single peak appearing at the threshold of lasing.

These observations are quite typical of randomised liquid amplifying media, and our system seems to perform well under conditions of evanescent wave pumping. The notable differences here are that, at sub-diffusive scattering, i.e., at the scatterer density of $10^{10}/cc$ where $l^* >>$ L, there is no effect of the disorder on the emissions of the gain medium. In earlier reports of random amplifying media pumped by a propagating beam, sub-diffusive scatterings did enhance the emissions of the pure amplifying medium. This enhancement was attributed to the high gain in the system, that rendered these weak-scattering events important[3]. The fact that sub-diffusive scatterings could not alter the emissions of the system implies that the gain inside the system was quite low. This is totally due to the evanescent character of the pumping mechanism. Upon addition of more scatteres, though, we achieved random lasing. We have, thus, confirmed that even at low gain levels, the phenomenon of non-resonant feedback leads to random lasing even under weak excitations provided by evanescent waves.

These effects are more pronounced when the dye concentration is 10^{-3}M, as seen from figure 9.5. The two subplots in figure 9.5 show the variation of the FWHM and peak intensity of emission, with increasing pump energy. Here, the threshold reduction

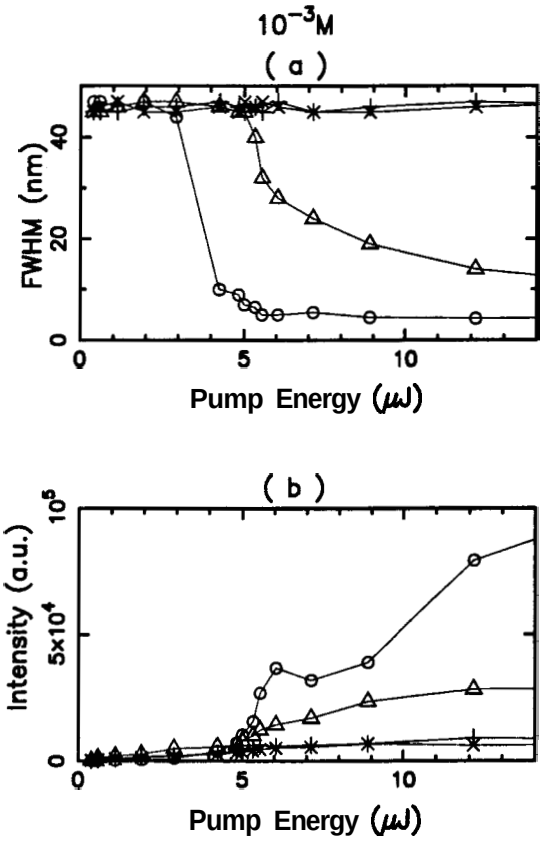

Figure *9.5: Variation of (a)* FWHM *and* ***(b)*** *Peak Intensity of emission with pump energy at a dye concentration of* $10^{-3}M$. *x : Pure dye,* + : $10^{10}/cc$, △ : $10^{11}/cc$, ∘ : $10^{12}/cc$.

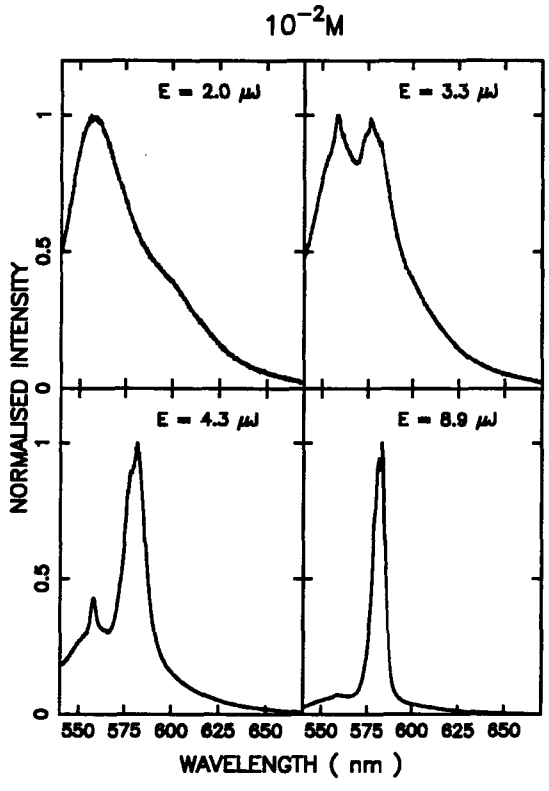

Figure 9.6: *Bichromatic emission seen from the pure dye of concentration* $10^{-2}M$ *at various pump energies, under evanescent pumping. The system lases in the red mode* $(583nm)$ *at high pump energies.*

with increase in scattering strength is very clearly seen from the variation of the FWHM. The emissions from the pure dye and from the weakly scattering amplifying medium are broadband, and low intensity, in contrast to the emissions from medium at higher scatterer concentrations. Clearly, the system does not lase at sub-diffusive regimes of scattering.

Despite the significant shifts in the wavelength of peak emission in both the above dye concentrations, the emission is primarily monomodal. At any given pump energy or scatterer concentration, a single peak is seen, in contrast to the two peaks seen for a higher dye concentration of 10^{-2}M. Fig 9.6 shows the emission spectrum of the pure dye at the concentration of 10^{-2}M at different pump energies. The spectrum peaks at 557nm at a low pump energy of 2/15. A second peak develops at $582nm$, which we call the red mode hereafter, at around $3\mu J$, which grows at the cost of the first peak, which we call the

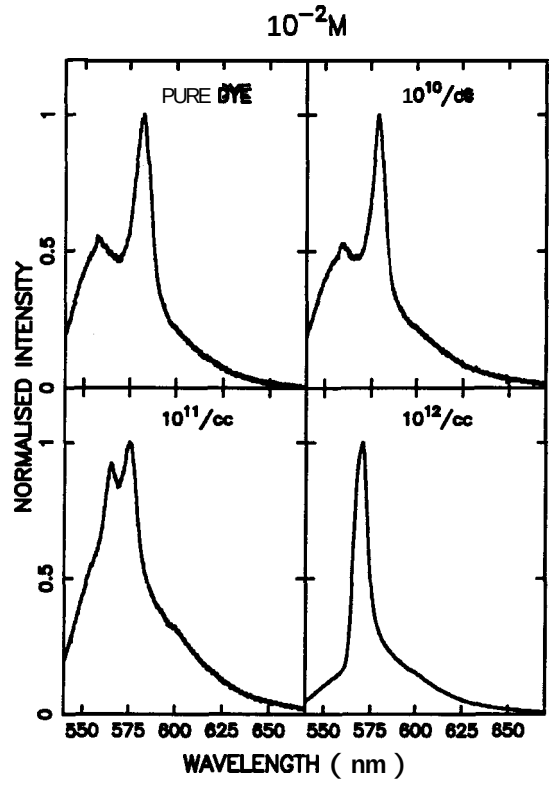

Figure 9.7: *Normalised emission spectra from a dye of concentration* $10^{-2}M$ *with various scatterer concentrations at a pump energy of* $4\mu J$*, under evanescent pumping. Clearly, the addition of scatterers forces the system to lase in the yellow mode* $(570nm)$*, in contrast to the emission from the pure dye shown in figure 9.6. The competition between the two modes upon addition of disorder is obvious from the four subplots.*

yellow mode, hereafter. Eventually, at high pump energies, the red mode narrows down, and the yellow mode is completely suppressed and the system lases at $583nm$.

The addition of scatterers alters the spectrum, as seen from figure 9.7, which shows the emissions for different scatterer densities, for the same pump energy of $4\mu J$. With increasing scatterer concentration, the red mode slowly loses energy to the yellow mode, and eventually the system lases at $570nm$ at a particle concentration of $10^{12}/cc$. The FWHM of the two modes is shown in the figure 9.8. Each subplot shows the FWHM of the red mode (shown by $\triangle$) and the yellow mode (shown by $\circ$). It can be seen that the red mode lases only when the scattering is absent or is weak, and a stronger scattering environment favours the yellow mode. The red mode is totally absent in the presence of

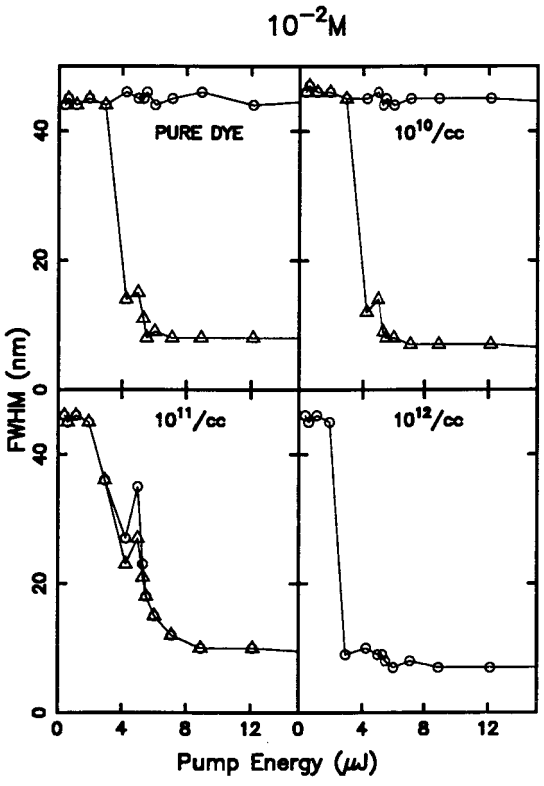

Figure 9.8: *Variation of FWHM, as a function of pump energy, of the two modes (∘ : yellow mode , △ : red mode) of the bichromatic emission seen from the dye of concentration* $10^{-2}M$. *The effect of addition of scatterers is evident from the four subplots. The broadband yellow mode starts narrowing as scatterers are added. The red mode is absent in the case of scatterer concentration of* $10^{12}/cc$, *and the system lases in the yellow mode.*

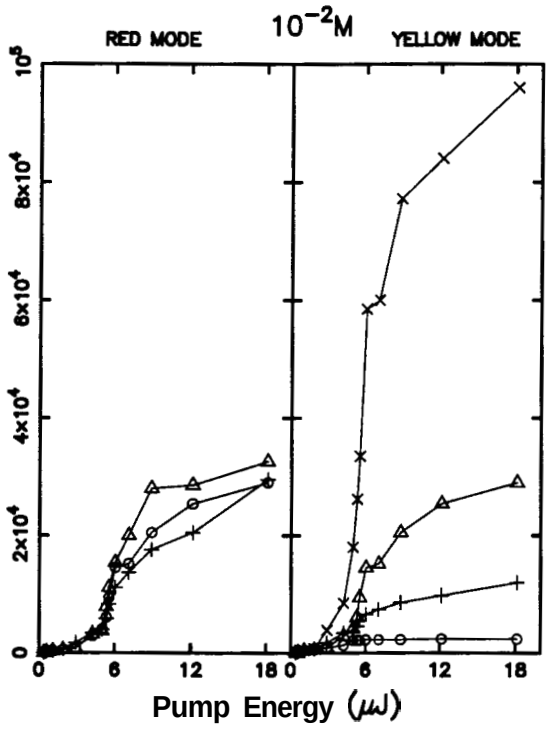

Figure **9.9:** *Peak intensity of emission in the two modes as a function of pump energy at various scatterer concentrations.* o : *Pure dye,* + : $10^{10}/cc$, △ : $10^{11}/cc$, x : $10^{12}/cc$.

stronger scattering, i.e., at a scatterer concentration of $10^{12}/cc$. Figure 9.9 showing the peak intensity, illustrates the mode competition between the two modes in the presence of disorder. One can see that, in a weak scattering environment, the red mode grows in energy upon increase in the pump energy, while in the presence of strong scattering, the yellow mode takes over. The system always lases in the red mode for the pure dye and the dye with a scattering concentration of $10^{10}/cc$. Only at higher concentration of scatterers does the dye lase in the yellow mode. This again underlines the fact that sub-diffusive scatterings are unable to affect the emissions of the dye.

To compare the emissions of the same system with a three-dimensional pumping, we excited the dye (10^{-2}M) doped with the same scatterers in a conventional setup as described elsewhere[4]. The emission spectra at four different scattering strengths is shown in figure 9.10.

On comparison with figure 9.7, it is obvious that the two systems behave quite differently. In the 3D case, only monomodal emission is observed, and the system lases at $572nm$ (in the yellow mode) at a scattering strength of $10^{12}/cc$. At no pump energy or scatterer concentration was bichromaticity seen in this setup. (We had observed bichromatic emission from the dye at such concentrations, when the solvent was water. However, in this case, where ethanol is the solvent, the dye did not show bichromatic emissions, because the fluorescence efficiency increases in ethanol, and stimulated emission is dominant even at low pump energies). Figure 9.11(a) shows the variation of the FWHM of the emission from the 2D system, and (b) shows the same in case of the 3D system. The 2D system is seen to cross the threshold at very low pump energies, even in the presence of no or weak scattering. It lases in various modes, depending upon the degree of disorder. It is of importance to note that the pump energy had to be increased from $20\mu J$ to $600\mu J$ till notable differences were observed in the emission spectra in the 3D case. In comparison, over a range of $20\mu J$, the 2D system exhibited similar, and more interesting, features in the spectral profiles.

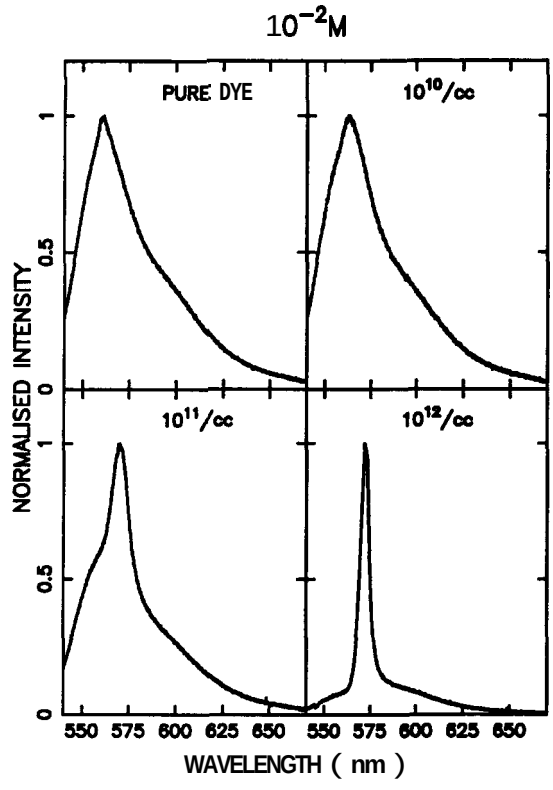

Figure *9.10: Emissions from the same dye at a concentration of* $10^{-2}M$ *under three-dimensional pumping conditions, at various scatterer concentrations at a pump energy of* $600\ \mu J$. *The emission is always monomodal.*

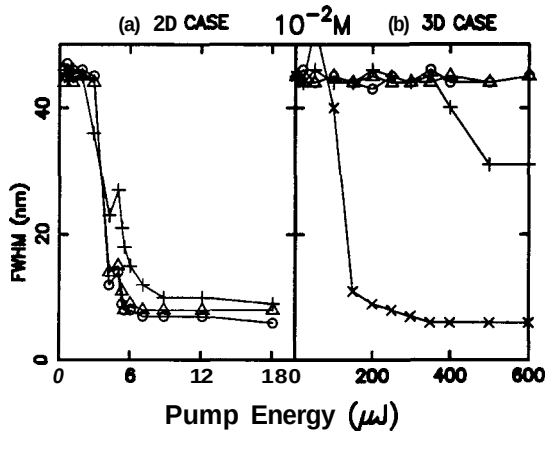

Figure *9.11:* FWHM *of the emission spectrum as a function of pump energy of (a) the 2D pumped system and (b) the 3D pumped system.* o : *Pure dye,* $\triangle$: $10^{10}/cc$, + : $10^{11}/cc$, x : $10^{12}/cc$. *Note that the X-axis is scaled differently in the two cases.*

9.4 Two-dimensional nature of the system

The above results conclusively prove that the random amplifier performs very well and possibly more efficiently even under evanescent pumping. At minimal pump energies, the mechanism of light amplification with nonresonant feedback within a two-dimensional amplifying region has been shown to produce random lasing. We may consider the amplifying medium to be two dimensional, as pumping is most efficient in the green, which, in this case, is present only as an evanescent wave. Thus, even though a volume of dye is present, only the two dimensional surface in contact with the prism is amplifying. It may be argued that the emission from this layer may pump the layers above, but this is very weak. In the case of the dye concentration of 10^{-2}M, the absorption of the primary emission from the slab is strong, and the unexcited dye above the pumped slab can be considered as an absorbing boundary. The wavelengths at which absorption cross-section of the dye molecules is large are absorbed by the unpumped dye, which re-emits fluorescence back to the active region. Thus, the interface between the strongly absorbing medium and the pumped medium, which has a large mismatch in the imaginary refractive index, acts as a reflecting boundary. In that case, both the pumped region and the emitting region become two-dimensional. The light received by the detector apparatus is that which is emitted, amplified and scattered within the two-dimensional region, before leaking out of the partially reflecting boundary, in this case, the prism surface. Indeed, it is the case of the 10^{-2}M dye that the most interesting effects were seen, that deviated from the 3D pumping.

There are two important factors contributing to the lowering of the threshold of lasing. The evanescent wave does not propagate into the medium beyond a certain depth, and so as the intensity of the pump is increased, the evanescent field strength increases in the same two-dimensional region of space. It just leads to larger population inversion in the same region of the dye. Consequently, the amplification length reduces rapidly with increase in the pump intensity. As a result, the system tends to amplify the spontaneous

emission at lower pump energies. Therefore, even in the absence of scattering or in the weak-scattering limit, the amplifying medium exhibits narrowband emission. This is seen in the dye of concentration 5×10^{-4}M. The addition of scatterers, then, accelerates this process, and reduces the threshold of lasing. In this case, because of the low concentration of the dye, effects arising from self-absorption are negligible. When population inversion is strong, self-absorption is suppressed by stimulated emission, which favours the yellow wavelengths. The self-absorption effects are, however, not negligible in the case of the dye concentration of 10^{-3}M, where, lasing is not observed in the case of pure dye or in the presence of weak scattering. We believe that this system, in conditions of weak scattering does not lase, because of absorption of the yellow wavelengths by the ground state molecules in the unpumped region of the dye. In the presence of strong disorder, i.e., a scatterer concentration of $10^{12}/cc$, random lasing is seen in the yellow wavelengths, suggesting that stimulated emission is responsible for the observed spectral profile.

In the dye with a high concentration, the initial lasing in the red mode is purely due to the self-absorption of the yellow wavelengths emitted inside the two-dimensional region. The fact that the red mode narrows down suggests that these effects are quite strong, and also that the pumping is weak, and cannot generate enough population inversion to suppress the self-absorption. The addition of scatterers, however, drastically changes the emissions, which lase in the yellow mode. The low gain in the system is countered by the effect of the enhanced pathlengths of the photons, between multiple scattering. This happens only in the diffusion regime of disorder.

9.5 Conclusions

In conclusion, we have studied the emissions of random amplifying media, a laser dye into which microparticles were suspended, under excitation by an evanescent wave created at a glass-alcohol interface. This is equivalent to two-dimensional pumping since the evanescent field decays inside the amplifying medium within a distance of the order of a

wavelength. Further, assuming that the region above the slab of excited dye provided a reflecting interface due to the large mismatch in the imaginary part of the refractive index, we infer that the fluorescence detected from the system was emitted from and confined within, a two-dimensional slab. We observed enhanced effects of multiple scattering in the case of the evanescent pump. We examined effects like linewidth narrowing, threshold reduction and shift in the wavelength of peak intensity, as a function of both the degree of disorder and the pump intensity. The pump energy required to make the system lase was found to be much lower than that required in the case of a three-dimensional system, all other parameters remaining identical. This could be possibly attributed to the fact that evanescence is non-propagating, and increase in the pump energy only increases the inversion in a specific volume of the random amplifier. Consequently, the effective gain lengths of the photons are greatly reduced. That the gain was low in our setup was proved by the fact that sub-diffusive scatterings were not made effective, unlike the case of a propagating pump. In the presence of sufficiently strong disorder, drastic changes were seen in the emissions from the random amplifying medium, particularly in the high concentration dye.

An important tunable parameter that is provided by this setup is the thickness of the pumping region. The extent upto which the evanescent pump light exists inside the medium depends upon the angle of incidence. Thus, this thickness of the amplifying region can be increased or decreased by changing the angle of incidence, whilst still maintaining its quasi two-dimensional character. In that case, the emission characteristics can be controlled by the angle of incidence, which is much simpler than tuning the degree of disorder or changing the concentration. Such a parameter for tuning is not available in a three-dimensional system.

These results are of importance since they underline the ease of pumping of the random amplifying media which are now accepted as the new robust laser media. As opposed to various conventional laser systems, where the pumping requirements are large, our results show that random amplifying media can be pumped by minimal energies, and

yet, narrow linewidths with high intensities can be obtained. Indeed, the effects of longer pathlengths of photons was evident even in a weak scattering system like ours, and the use of stronger scatterers (eg, GaAs crystals) could easily take the photon transport into strong localisation regime. In that case, we believe, extreme narrow linewidths, as observed in the random lasing using semiconductor powder can be achieved at even lower thresholds. In another application, using the evanescence generated outside the core of optical fibres, it could be possible to amplify the signal carried within the fibre. Our results do not incorporate the coherence of the pump anywhere as a necessary condition of pumping, implying thereby that similar conditions can be achieved by incoherent pumping, provided adequate energies are obtained.

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Chapter 10

Conclusions

In this thesis, I have described my experimental and numerical investigations on various aspects of light scattering in passive and active random media. In the case of passive media, the emphasis was on the aspect of polarisation of scattered light. I investigated a recently developed technique of imaging of opaque objects through scattering media, using polarisation discrimination. The technique involved modulation of the polarisation of input light, and detecting the modulation of the output light. The scattered light changed its polarisation state (the depolarisation being proportional to the angle of scattering) and hence lost the memory of the modulation. The ballistic light, however, exited the scattering medium with the same modulation of polarisation. Upon detecting this modulation, and quantifying the amplitude of modulation through fourier transforming methods, the amount of ballistic light in the transmitted light could be determined. After obtaining two-dimensional shadowgrams from turbid media, I used the concept of stereography to make three-dimensional images so as to get depth information of the hidden object.

The coherent backscattered (CBS) cone from multiply scattering samples is quite difficult to see, especially in the case of weak scattering. However, noting that CBS is polarisation preserving, the method of polarisation gating was applied to coherent backscattering, and I obtained a marked increase in the CBS signal to noise ratio.

Upon imaging an object in a scattering medium of optical thickness of about $30l^*$, I

proceeded to study the dependence of depth of imaging on the scattering anisotropy of the sample. I found that it was easy to image deeper in a sample of isotropic scatterers. The method of imaging by fourier spatial filtering was inherent in the technique described above. Thus, it enabled me to compare the efficiency of imaging by polarisation gating and fourier spatial gating, and to an extent, temporal gating. It was found that polarisation gating results in images with a better contrast and resolution, especially in the case of isotropic scatterers. In the case of anisotropic scatterers, the three processes result in similar images. This was owing to the fact that, the method of polarisation gating selected the ballistic light out of the total forward scattered light as the signal. Ballistic light exists upto deeper optical depths in samples with isotropic scatterers because of the larger scattering mean free path. Hence, it was easier to image in isotropic samples. Further, the average angle of scattering, and hence the depolarisation, is larger in case of isotropic scatterers. So the process of polarisation discrimination discarded even the weakly scattered light as noise resulting in cleaner images. As the anisotropy increased, the depolarisation upon scattering decreased, and the process could not distinguish between the ballistic and the weakly scattered snake light. Therefore the images extracted in anisotropic samples were low in contrast and resolution.

The experiments on active random media involved the study of the spectral properties of a randomised laser dye. I present experimental results that are typical of such "random lasers". Based upon these results, a model was put forth, and verified numerically through extensive Monte Carlo simulations. The model was general and could incorporate different scattering strengths, concentrations, pump energies etc. Without making any a priori assumption about the scattering strength or the gain, the model explained the experimental observations. The behaviour of the unpumped region of the amplifying scattering medium was also included in explaining the spectral features. In particular, the model explained phenomena like threshold reduction, linewidth narrowing, bichromatic emission and its dependence on concentration, competition between the two modes in bichromatic emission and shift of wavelength of peak emission at high pump powers. The model emphasised

the fact that two phenomena that occur simultaneously inside the random medium, *viz* stimulated emission and self-absorption, are responsible for the spectral characteristics of the emission. The Monte-Carlo code that I developed can predict the emission spectrum, given a dye-scatterer system.

I then used the above model to design and construct a device that had an exponential gain profile along its length. Upon excitation, the two processes of stimulated emission and self-absorption acted simultaneously but in opposite directions. As a result, the emission was anisotropic, and a certain band of wavelengths appeared to be forbidden from propagating in one direction, but was enhanced in the opposite direction. Thus, for this particular band of wavelengths, the device acted as an optical diode. This band of wavelengths was the same as the one which has an overlap of the absorption and emission spectra of the amplifying medium.

Further, having realised that disorder enhances random lasing by reducing the threshold of lasing, I studied the behaviour of the random laser under evanescent excitation. I found that the random laser behaved well even under extremely weak excitations realised by evanescent waves. When the concentration of the dye was large enough to assume that the emitting medium was two-dimensional, I found that the emissions from this two-dimensional region were drastically different from a three-dimensional system with the same concentration and strength of disorder. Lasing at large wavelengths was seen in the emissions in the weakly scattering samples. Upon addition of scatterers, the system switched to lasing at shorter wavelengths at very low pump energies. This proved that self-absorption was rapidly suppressed by the stimulated emission even at low excitation energies. This could be attributed to the rapid shortening of the gain length of a photon inside the amplifying medium, because of reflection of light from the strongly absorbing boundary back into the pumped region.

To sum up, the results presented in this thesis provide insight into the polarisation aspects of light transport through passive random media, and explain the physical princi-

ples underlying the phenomenon of random lasing in active random media. Some practical applications of these processes are also demonstrated.