Some Investigations of Light Scattering
in
Active and Passive
Random Media

by

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DECLARATION

I hereby declare that this thesis is composed independently by me at the Raman Research Institute, Bangalore, under the supervision of Dr. Hema Ramachandran. The subject matter presented in this thesis has not previously formed the basis of the award of any degree, diploma, associateship, fellowship or any other similar title.

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CERTIFICATE

This is to certify that the thesis entitled **Some Investigations of Light Scattering in Active and Passive Random Media** submitted by Sushil Mujumdar for the award of the degree of DOCTOR OF PHILOSOPHY of Jawaharlal Nehru University is his original work. This has not been published or submitted to any other university for any other degree or diploma.

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Contents

Preface

The propagation of light in random media is an interesting phenomenon because multiple scattering of light scrambles the direction of propagation of light and increases its dwell time inside the medium. This can have many interesting consequences, some of which are dealt with in this thesis. This thesis is broadly classified into two parts, one concerning multiple scattering within passive random media, and the other concerning scattering within active random media.

Passive media are those that have negligible absorption at the wavelength of interest. Consequently, the amplitude of a wave does not change during transport through a random medium. Our investigations concerning passive media pertained to the change in polarisation of light upon recurrent scattering inside a disordered medium. These studies have direct bearing on the problem of "imaging through turbid media". This is a long-standing problem in biomedical optics that is aimed at developing techniques to image objects hidden in turbid media using radiation at optical wavelengths. Our technique of imaging opaque objects in turbid media uses the depolarisation of light upon scattering as a means to distinguish between scattered and unscattered light. In the subsequent experimental and computational studies, we focus our efforts on the phenomenon of depolarisation of polarised light, upon multiple scattering and elucidate its bearing to imaging.

Active media are those that have a strong absorption or gain at the wavelengths of interest. It is known that a fluorescent system shows drastic differences in its emission when it also contains disorder. At high gain levels achieved through excitation by high power lasers, a homogeneous high-efficiency medium emits narrowband light with high intensity. When it is made disordered, similar characteristics of emission are observed at much lower gain levels. In that case, the narrowband, high intensity emission from the active random medium is called random lasing, and such media are called random lasers. We study a particular class of random lasers, that which are obtained by adding colloidal microspheres to laser dyes. The experiments and simulations described in this thesis provide significant insight into the physical phenomena that govern random lasing.

The first chapter of the thesis starts with an introduction of the basic concepts relevant to single scattering of light by small particles, and is followed by the essentials of the theory of multiple scattering of light. Depending upon the strength of scattering, various theories are applied to analyse wave propagation through the disordered medium. This chapter discusses the most extensively used diffusion theory and its applicability.

In the second chapter, we proceed, to introduce the concept of polarisation of light in scattering. The calculation of Stokes vectors of a given polarisation state of light is explained through a series of thought experiments. Since there is a fair bit of numerical work in the thesis regarding the evolution of polarisation in a scattering medium, we introduce the concept of Mueller matrices that are used to calculate the change in polarisation upon interaction with a linear medium. Scattering off a particle is such an interaction, and we shall describe the scattering particle Mueller matrix that has been used in subsequent chapters.

The third chapter of the thesis deals with the topic of imaging of opaque objects through scattering media. Following a brief introduction to various techniques used hitherto in imaging, the chapter discusses in detail the technique of polarisation discrimination to extract two dimensional images from random media using the polarisation memory of unscattered light. The technique has been extended to three-dimensional imaging using the process of stereographic imaging to obtain depth information of objects immersed in scattering media.

Since the process of polarisation discrimination picks out the polarisation preserving light from the total scattered light, it has been employed to enhance the signal to noise ratio in coherent backscattering experiments. The fourth chapter introduces the phenomenon of coherent backscattering of light from scattering media, and describes our experiments involving polarisation discrimination on backscattered light.

The fifth chapter, which is the last in this part of the thesis, deals with the experimental and numerical investigations of the depolarisation of forward scattered light through random media. This forward scattered light is used as a signal by the conventional imaging systems, and we show, through our numerical simulations, that our system selects only the ballistic content contained in the forward scattered light, and hence results in cleaner images. Experimental studies prove that it is easier to image through an isotropically scattering medium than in an anisotropically scattering medium. Extensive Monte Carlo simulations shed light on the process of depolarisation, and explain the experimental findings. The results also suggest that polarisation gating and temporal gating together should provide a means of studying the snake photons alone, that regime of scattering which remains elusive to most experimental methods.

The sixth chapter forms the introduction to the essentials of light scattering through active random media. This chapter familiarises the reader with the concepts of random lasing, its differences from conventional lasing, 'and the diffusion of light within a gain medium. The random laser system studied in this thesis is a laser dye into which colloidal microparticles are suspended. Details of typical dye molecule energy states, and the radiative transitions are discussed. This provides an understanding of the overlap of the typical absorption and emission spectra of dye molecules. As is shown later in the thesis, this overlap turns out to be a crucial factor in determining the lasing characteristics of such a random laser.

In the seventh chapter, we present experimental results that are typical of such active random media. A model is put forth, and verified numerically through extensive Monte Carlo simulations. These explain the experimental results very well. The model is general and can be applied to any kind of a dye-scatterer medium, with different scattering strength, concentration, pump energy etc. Thus, the model does not make apriori assumptions about whether the scattering is diffuse, or whether the gain is high. The behaviour of the unpumped region of the amplifying scattering medium is also included in explaining the spectral features. In particular, the model explains phenomena like threshold reduction, linewidth narrowing, bichromatic emission and its dependence on concentration, competition between the two modes in bichromatic emission, shift of wavelength of peak emission at high pump powers etc. Clearly, most of the experimental results are reproduced by the model in our numerical simulation. Ultimately, it becomes obvious from this study that the two phenomena that occur simultaneously inside the random medium, *viz* stimulated emission and self-absorption, govern the spectral characteristics of the emission.

In the eighth chapter, we put our model to practical use by designing a configuration of the gain medium that forces the two processes to act in opposite directions. A certain frequency-band of light undergoes stimulated emission when it travels in one direction, and suffers from self-absorption when it travels in the opposite direction. As a result, the emissions in the two directions are different. The device that is made using such a configuration of the gain medium acts as an optical diode for that frequency band. We discuss our experimental studies of such a device, and also explain its behaviour through Monte Carlo simulations.

The above analysis of the phenomenon of random lasing proves that multiple scattering within gain media can lead to high-gain narrowband emission even at low gain levels. We study, in the ninth chapter, the behaviour of the random laser system under conditions of extremely weak excitations, provided by evanescent waves. Evanescent wave coupled excitation provides very low gain levels, such that weak subdiffusive scatterings are incapable of reducing the threshold of lasing of the amplifying medium. However, in the presence of diffusive scattering, random lasing is clearly observed even at such low gain levels. Further, on the virtue of the non-propagating character of evanescent waves, we show that the pumped region and the emitting region are two-dimensional, especially in the case of high concentration laser dyes. In this case, we compare the emissions of the evanescent wave pumped system with the emissions from a system pumped with a propagating beam. We show that the significant differences between the two are a consequence of the two-dimensional character of the active medium.

Finally, in the tenth chapter, the conclusions obtained from this research work are listed out.

Apart from the introductory chapters on each part, each chapter carries enough introductory details to enable the reader to read and understand it independently. References to each chapter are given at the end of the chapter.

The following publications contain the reports of most of the research presented in this thesis.

1. Spectral features of emissions from random amplifying media.
   Sushil Mujumdar and Hema Ramachandran
   Optics Communications, 176, 31 (2000)

2. Use of a graded-gain random amplifier as an optical diode.
   Sushil Mujumdar and Hema Ramachandran
   Optics Letters, 26, 929, (2001)

3. Optical diode using a graded-gain random amplifier.
   Sushil Mujumdar and Hema Ramachandran
   Conference Digest, CLEO-Europe 2000, Nice, France.

4. Stereographic imaging of opaque objects in turbid media.
   Sushil Mujumdar and Hema Ramachandran
   Conference Digest, International Conference on Optics and Optoelectronics
   Dehradun, India. (December 9-12, 1998.)

5. Enhancement of image resolution and contrast using polarisation modulation : Dependence on the scattering anisotropy.
   Sushil Mujumdar and Hema Ramachandran
   Communicated to Applied Optics.

6. Evanescent wave pumping of random amplifying media.
   Sushil Mujumdar, Hema Ramachandran and N. Kumar
   To be communicated.

Part I

PASSIVE MEDIA