Abstract

Light scattering in active disordered media has stirred vast interest towards the unique phenomenon of random lasing. In a random laser (RL), feedback for light amplification is provided by the trapping of light via multiple scattering. In the weakly scattering random lasers, there exists strong leakage of photons from the system along with amplification. In such cases, the reason for the appearance of coherent resonant modes in the emission spectra is still a subject of discussion. However, unique statistical behavior of RL mode intensities and their glassy behavior further enrich the underlying physics responsible for their origin. The extreme sensitivity of RL to subtle morphological changes at nano and microscales adds to their applicability. Despite various proposed applications, RL as a tool to quantify the dynamic behavior of material evolution has not been explored extensively.

In this thesis, studies have been carried out on the characteristics of RL modes in weakly scattering systems by using active polymer waveguides, dye-doped gelatin solutions and whispering gallery mode resonators. A high refractive index film of 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran (DCM) dye doped polyvinyl alcohol (PVA) deposited on a silica substrate acts as a disordered active planar waveguide (DCM-PVA) and exhibits RL when pumped optically. In order to investigate the possible origin of scattering centers in this polymeric film due to the formation of dye aggregates, time correlated single photon counting (TCSPC) measurements at different dye concentrations were carried out. These aggregates are responsible for multiple scattering and the gain characteristics of the active polymer waveguide. The polycrystalline nature of the DCM-PVA waveguides have also been confirmed by X-ray diffraction studies. The gain coefficient of the waveguide has been determined by variable stripe length method. Finally, the RL characteristics have been studied under circular and stripe excitation configurations, thereby completely characterizing the origin of scattering and gain in the DCM-PVA waveguides.

Later, statistical analyses involving cross-correlation and survival function studies of RL from a solvent rich DCM-PVA waveguide were carried out under a constant heating condition. The slow evaporation of high boiling point solvent used to form the waveguide creates morphological changes in the film. Here, RL is shown to be an effective tool to probe such minute changes in the polymer waveguide. The statistical analyses of random lasing spectra were further employed to investigate the microscopic changes occurring during the sol-gel transition in a Rhodamine 6G(R6G) dye-doped gelatin solution. Finally, we have investigated the statistical properties of intensity fluctuations and glassy behavior of light in three different lasing cavities namely: DCM-PVA planar waveguide open cavity, DCM-PVA bottle whispering gallery mode resonator (WGMR) and DCM-PVA spherical WGMR. The complex behavior of mode interactions have been captured using the distribution of intensity fluctuations, covariance and replica symmetry breaking analyses. These studies reveal the uniqueness of RL open cavity in comparision to conventional laser cavities with weak disorder. The work presented in the thesis mainly focuses on the glassy and statistical behavior of modes in open RL systems and highlights the potential of this tool to monitor the microscopic changes occurring within the RL system.

Keywords: Random laser, Disordered media, Dye-doped polymers, Intensity fluctuations, Replica symmetry breaking, Whispering Gallery Mode Resonator, Bottle microresonator, Spherical microresonator