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Structural, Morphological and Random Laser Action for Dye-ZnO Nanoparticles in Polymer Films

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ABSTRACT

Laser dye Rhodamine 6G (R6G) was incorporated in Poly MethyleMetha Acrylate (PMMA) polymer at different particles density from zinc oxide (ZnO) as (nanowire, nanoflower) synthesized using the hydrothermal method as random laser active medium. The thickness and emission spectra of the films were characterized by Scanning Electron Microscopy (SEM) and Fluorescence spectrum. The optimum results at the Width Half-Maximum (FWHM) are about (13, 11) nm for both ZnO wires and flower respectively at high pumping energy equal (20, 16) mJ respectively with optimum concentrations are 10-4mol/l .While Amplified Spontaneous Emission (ASE) was observe at low pumping intensity using Second Harmonic Generation(SHG) Nd-YAG pulse laser working at wavelength 532 nm. The homogeneous thickness of the films equal (14.6 –15) µm was used. Finally, the performances of ZnO random laser active medium for both shapes (wires and flowers) are determined via the relationship between the emission line and FWHM. From the results, appears that the maximum emission intensity depends on the particle shape, concentration and pumping intensity.

Keywords: Random Laser Dye R6G, PMMA, Zinc Oxide, Hydrothermal.

1. INTRODUCTION

Laser action happens when an energetic medium dispersed with scattering particles or when there is a pulverized active material powder [1,2]Random laser consequences can be seen in a variety organic or inorganic powders on solid state luminescent or laser crystals [3,4] liquid laser dyes with scatters [5] polymeric films including or not including scatters that introduced intentionally [6] ZnO scattering films and nanoclusters dye and infiltrated opals porous media are infiltrated with liquid crystal including dyes and other things [7-11]. Random lasers are powerfully scattering media to strengthen light. There are notable similarities between the traditional lasers and these systems based over obtain media enclosed of a cavity with two mirrors in order to offer optical feedback, one example for this is the threshold for lasing action and the frequent reduction in random lasers. Random lasers optical properties are completely different than those properties of conventional lasers in that the propagation pump and the fluorescence light is diffusive. Cavity systems in contrast with scattering are useful. The random laser threshold is lowered by a stronger scattering as the efficiency of shorter transport since multiple scattering provides the feedback. In random lasers, threshold is radically reduced if the photon transport means free path reaches emission wavelength that has been stimulated [12].
The present work studied the influence of the shapes and nanostructures on the amplifying (as active medium) random laser performance and to determine the lasing threshold by change the dye concentration and Nd:YAG pumping energies with fixed ZnO nanoparticles concentration.

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2. EXPERIMENTAL WORK

2.1 Chemicals and Methods

ZnO NWs and NFs were prepared on a quartz substrate by hydrothermal technique [14,16]. R6G laser dye ($C_{28}H_{31}N_{203}Cl$) with molecular weight 479.02 g/mol supplied by Lambda Physics LC (5900): Methanol alcohol (CH_3OH), CH_2CL_2 grade purity supplied by Gainland Chemical Company and PMMA with chemical forms ($C_5O_2H_8$) supplied by Inter chimiques SA France and used as a host for laser dye and nanoparticles .The solutions of laser dye are prepared by dissolving the required amount of the dye in methanol. The concentrations of dye solutions were: $5x10^{-3}$, 10-3, $5x10^{-4}$,10^{-4} and $5x10^{-5}$ mol/l. The powder of ZnO was suspended in dye at particles density ($0.184x10^{13}$) cm-3.The mixing volume ratio of R6G (10^{-3} - 10^{-5}) mol/l PMMA ($5x10^{-2}$ mol/l) is 1: 0.5 ml for the samples in the liquid phase. Spin coating technique was used to prepare the thin films for morphological and spectral emission.

2.2 Characterization and Experimental Setup

Figure 1 illustrated a schematic diagram of the optical elements arrangement for the emission spectrum measurement. The films are pumped by second harmonic generation (SHG) Nd:YAG pulsed laser working at a wavelength 532 nm and frequency repetition rate of 2 Hz. The pump beam was focused on the thin film surface by a spherical lens with a 20 mm focal length at an angle of about 30°. The emissions light from the films are detected by a spectrometer (Ocean Optics HR4000 CG-UV-NIR) pass during optical fiber. The emission spectra were recorded at different gradually-increasing pumping energies.

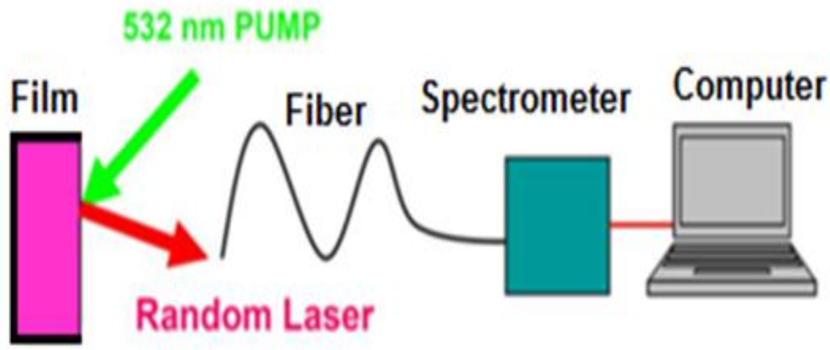


Figure 1. Schematic of the experimental arrangement for random laser emission measurement.

The RL performance was investigated for different films of dyes and ZnO particles. Series of emission spectra for each combination were recorded for increasing levels of pumping. RL characterization was done by measuring lasing threshold, peak emission wavelength and spectral line width of emission spectra as a function of pump energies.

3. RESULTS AND DISCUSSION

Figure 2 (a,c) shows a Scanning Electron Microscope (SEM) morphology images for the ZnO as different shapes prepared with different reduced agents [14] . The mean particle size of ZnO suspension is (100-329 and 89-120) nm for nanowire and nanoflower respectively. Also, the Figure 2 (b, d) explain profile (side view) images for the thickness measurement in different regions of the film surface was approximately average value equal (14.6 –15) µm .Also, notes from the figure the homogeneity thickness along the prepared film.

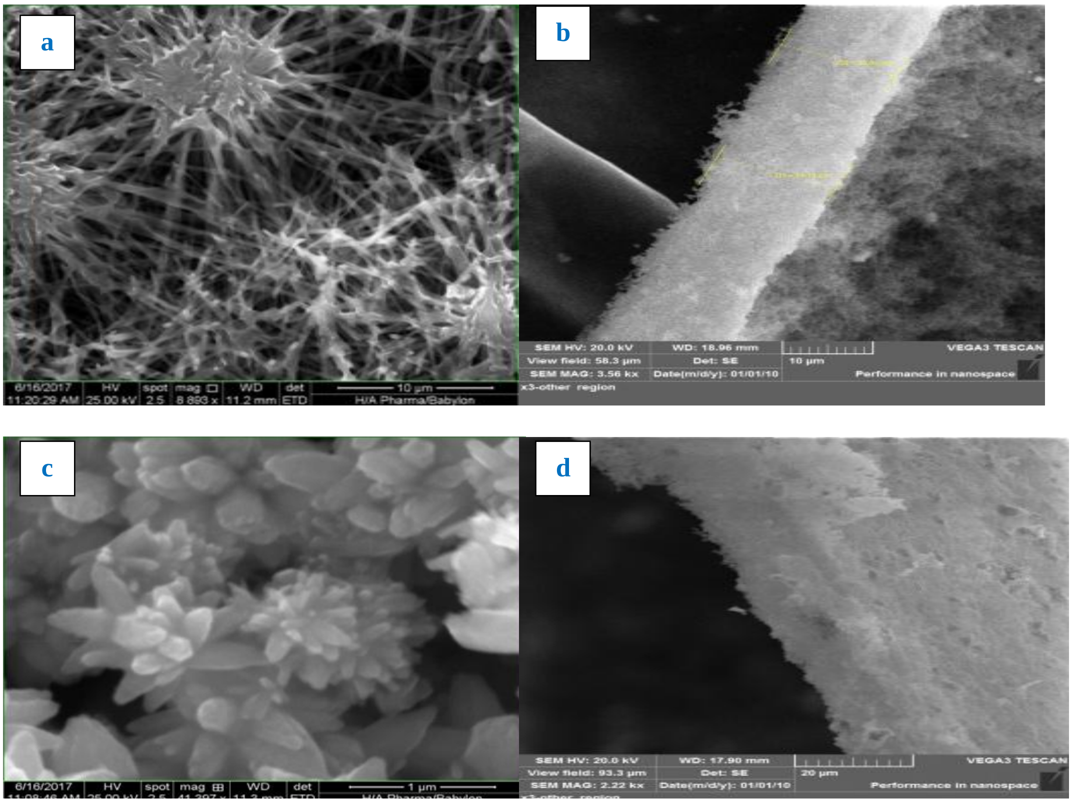


Figure 2. Illustrate SEM images for morphology and thickness measuring of films (a, b) topography images and side view for ZnO nanowire (c, d) topography images and side view for ZnO nanoflower.

The fluorescence spectra of the films at 0.184 x 10^{13} cm-3 for ZnO nanowire or nanoflower and various concentrations of R6G are shown in Figure 3. It shows that the peak wavelength occurs at 567 nm without shift. It was observed that a ZnO concentration of 10^{-4} mol /l of R6G dye hosted by PMMA had the highest intensity and the narrowest bandwidth and at 0.184 x 10^{13} cm-3 ZnO nanowire or nanoflower gives the optimum results agreement with reference [13]. Figure 3 shows the maximum intensity with shortening duration (bandwidth) at FWHM (25, 23) nm compared with more than 40 nm in the case of without ZnO in accordance to the inset of Figure 3. Similarly, a narrow bandwidth of 26 nm was recorded at FWHM for 10-5 mol / l of R6G with zinc oxide. The last value is less than 45 nm in the case of without zinc oxide, this is consistent with reference [13].

According to these preliminary results indicate that concentrations R6G and zinc oxide in the film samples are an optimized amplification of multiple scattering processes in this kind of random laser. It gives a narrow spectral line width that these processes take place in same time without a remarkable any influence on the dye response. Thus, achieve one of the significant systems of real conditions.

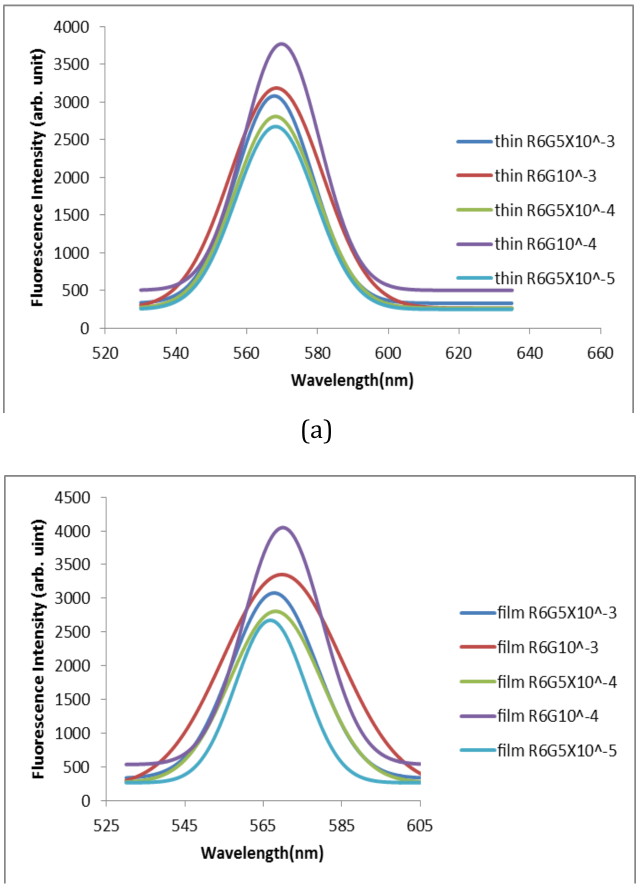
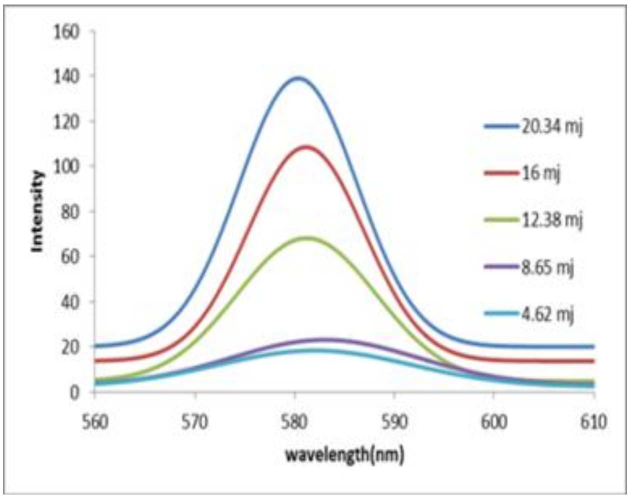


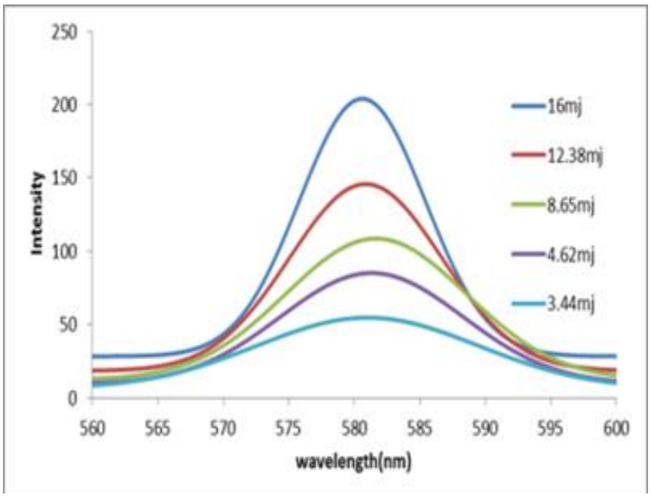
Figure 3. Illustrated the fluorescence spectra of samples with various concentrations of R6G dye hosted by PMMA and at 0.184x10^{13} cm-3 ZnO (a) nanowire (b) nanoflower.

Figure 4 illustrates the emission spectra obtained from the random laser arrangement, at different pumping energies for 10-4 mol/l of R6G dye hosted by PMMA and at 0.184 x 10^{13} cm-3 of ZnO nanowire or nanoflower. Maximum transition focused on the wavelength of 567 nm, as can be seen from the natural fluorescence in Figure 3, the density of a small pump with power pump equal (4.62 and 3.33) mJ, and can be observed only amplification of spontaneous emission which is measured by the peak intensity of bandwidth spectral value in the wavelength equal to 580 nm at FWHM about (23 and 20) nm zinc oxide wire and flower respectively.

When raising the pumping energy above to (20.34 and 16) mJ a much shorter peak at approximately the same peak wavelength is equal to be (13and 11) nm at FWHM, these results are consistent with the results of the research [13], [16], [17].



(a)



(b)

Figure 4. Emission spectra of the film sample 10^{-4}mole/liter of R6G and $0.184x10^{13}cm^{-3}$partical density of ZnO doped PMMA for different pumping energies at 2HZ repetition rate (a) nanowire (b) nanoflower.

4. CONCLUSION

The type of random laser was manufacture via hydrothermal method. From the results obtained from the dye concentrations R6G and all of ZnO (NW's & NF's) as scatter centers studied to give an indication of the concentrations required for optimal shape of nanoparticles. The results of random laser measurements show that the narrowing bandwidth at FWHM is found to be approximately (13,11) nm for ZnO wire and ZnO flower respectively.

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