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Random laser in ZnO powder pumped by picosecond pulses

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Abstract. We report our studies of random laser from ZnO powder at room temperature. The powder of ZnO colloidal spheres was synthesized by hydrolysis of Zinc acetate dehydrate. The random lasing around 380nm was produced from a layer of the ZnO powder pumped by the third harmonics (355nm) of a picosecond mode locked Nd:YAG laser (PL2143B - Ekspla). Above a pump threshold, discrete lasing modes of random laser action with resonant feedback appeared in the emission spectrum. Our experimental results show that random lasing can be produced from a layer of ZnO spherical nanoparticle powder on glass substrate.

Key words: Random laser, Resonant feedback, Monodisperse colloidal spheres, Hydrolysis.

1. Introduction

The random laser action in disordered medium was demonstrated experimentally [1-3]. The physical mechanism of optical confinement is based on the Anderson localization of light in a random medium of micro-scale [4]. When the scattering mean free path becomes equal or less than the wavelength, photon may return to the scatter from which it was scattered before and it may results in a closed loop that serves as a resonator. If the amplification along such a loop path exceeds the loss, lasing action could occur. This kind of a laser is called random laser. The requirement of the phase shift along the loop being equal a multiple of 2π determines the oscillation frequencies. In random lasers there are not "well-defined" cavities but ''self-formed'' cavities due to strong optical scattering in gain medium. The recurrent light scattering events (or closed loop paths for light) provide resonant feedback for lasing. Above a pump threshold, discrete lasing modes appear in the emission spectrum in addition to a drastic increase of emission intensity. The fabrication of random microlasers is much easier and cheaper than that of most convention microlasers which requires expensive facilities [4].

Various observations of random laser action with resonant feedback in semiconductor powder were reported. The scattering strength can be enhanced in disordered medium by reducing the particle size [4]. Morphology of particles also takes an important role to produce random laser with resonant feedback.

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Zinc oxide, ZnO, is a wide-gap semiconductor material, it has a large exciton binding energy (60meV) and a small Bohr radius (18Å), which in principle could allow efficient excitonic gain for lasing at room temperature [5,6]. ZnO random laser actions of various forms have been extensively studied. The lasing media include bulk single-crystal, thin films fabricated by different methods, phosphor powder and pellets. Many different techniques such as sputtering, reactive thermal evaporation, spray pyrolysis, pulsed laser deposition, MOCVD and MBE have been used in the preparation of ZnO thin films. UV laser action was observed in ZnO polycrystalline films [3], in ZnO powder film [4], in ZnO thin-film [7]. In general, single-crystalline films or polycrystalline films grown or deposited on substrates by various methods require expensive equipments.

In this paper, we report an experimental result of recurrent light scattering enhance for lasing in a layer of ZnO powder formed by ZnO colloidal spheres of 100-300nm diameter. Our research focus on laser action in ZnO layer painted on glass substrate.

2. Experimental

The powder of ZnO colloidal spheres was produced by a reaction similar to that described by Seelig et al [8]. ZnO was synthesized by hydrolysis of Zinc acetate dehydrate. This technique employs a two-step reaction process that allows close and predictable control of the size of the spheres. Crystalline structure of the ZnO particles synthesized was verified by XRD (SIEMENS D5005). Average size and morphology of the ZnO particles were characterized by FE-SEM (HITACHI S-4800). A ZnO colloidal sphere powder layer of about 400μm thickness was prepared on glass substrate.

We used the third harmonics (355nm) of a picosecond mode locked Nd:YAG laser (PL2143B-Ekspla) with pulse duration of 30ps and repetition rate of 10Hz as an optical pump to observe lasing emission from the ZnO layer. The block diagram of measurement and the detection system are shown in Fig.1. The pump beam was focused to a spot of 3mm diameter on the ZnO layer of the synthesized powder with an incidence angle of 45°. The emission spectrum from the ZnO layer was collected in around normal direction of the layer surface by a double grating monochromator with 0.2nm spectral resolutions. We choose this optical scheme to avoid pump light reflection from layer surface to monochromator. The spectrum signal was obtained by an acquisition system which consists of a PMT (R585-S), Amplifier, Boxca Averager SR250, A/D converter and Computer interface Module SR245.

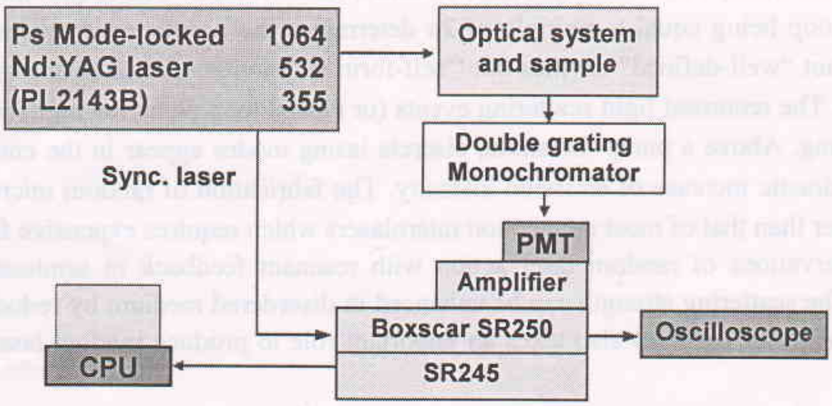

Fig. 1. Experimental arrangement.

3. Result and discussion

Fig.2 shows XRD pattern of the ZnO particles which confirms the wurtzite structure of the product. The FE-SEM image of the ZnO powder was shown in Fig.3. The ZnO particles are nearly spherical with the size distribution of 100-300nm and the spheres are made up of numerous nanocrystallites.

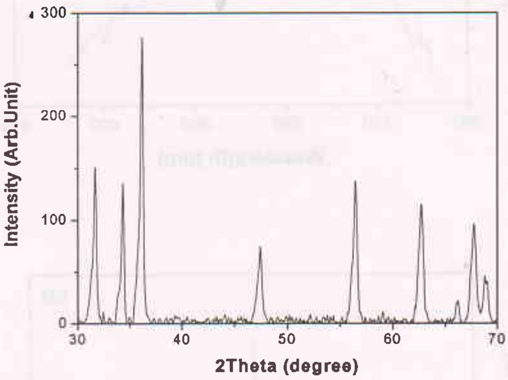

Fig. 2. XRD pattern of the ZnO samples.

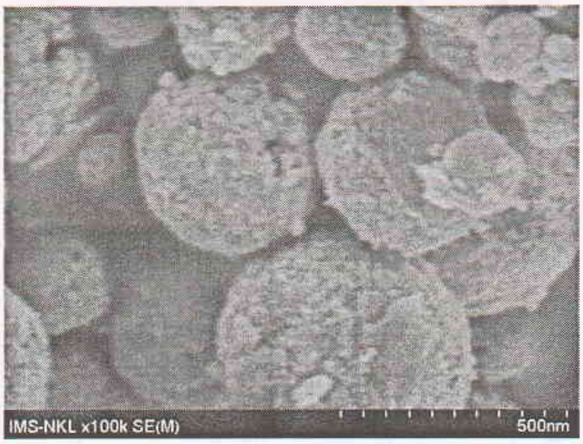

Fig. 3. The FE-SEM image of the ZnO sample.

The emission spectrum from a layer of the ZnO powder is shown in Fig.4. At low pump intensity, the spectrum consists of a single broad spontaneous emission peak (Fig.4a). When the pumping pulse energy exceeds 5μJ, we obtained the discrete peaks on the emission spectrum of 375-390nm (Fig.4b). The peak intensity increases drastically with the pump pulse energy (Fig.4c, 4d). The discrete peaks-emission spectrum with different patterns was also obtained in different directions from ZnO layer surface.

Because the ZnO layer is not parallel-plane, the possibility of lasing in the vertical cavity formed by the layer surface and layer - substrate interface can be neglected. The lasing emission spectrum with random spikes can be considered as a superposition of the equidistant discrete lasing modes which have origin from recurrent light scattering in a three-dimensional gain medium. The average size of ZnO colloidal spheres (100-300nm) is smaller than the emission band center wavelength of ZnO (380nm). According to the theory of random laser, it is suitable for recurrent light scattering to provide resonant feedback. In addition, the spherical shape of ZnO nanoparticles can enhance this recurrent light scattering [8].

The recombination of excitons in ZnO gives radiation in the region around 380nm with the intrinsic lifetime shorter than 200ps. The third harmonics (355nm) of a picosecond mode locked Nd: YAG laser (PL2143B-Ekspla) with pulse duration of 30ps is suitable to excite the ZnO powder for lasing.

In this experiment, we did not focus pump beam on ZnO layer. The pumped active medium area is a spot of about 3mm diameter which results in a low lasing threshold of 5μJ pulse energy.

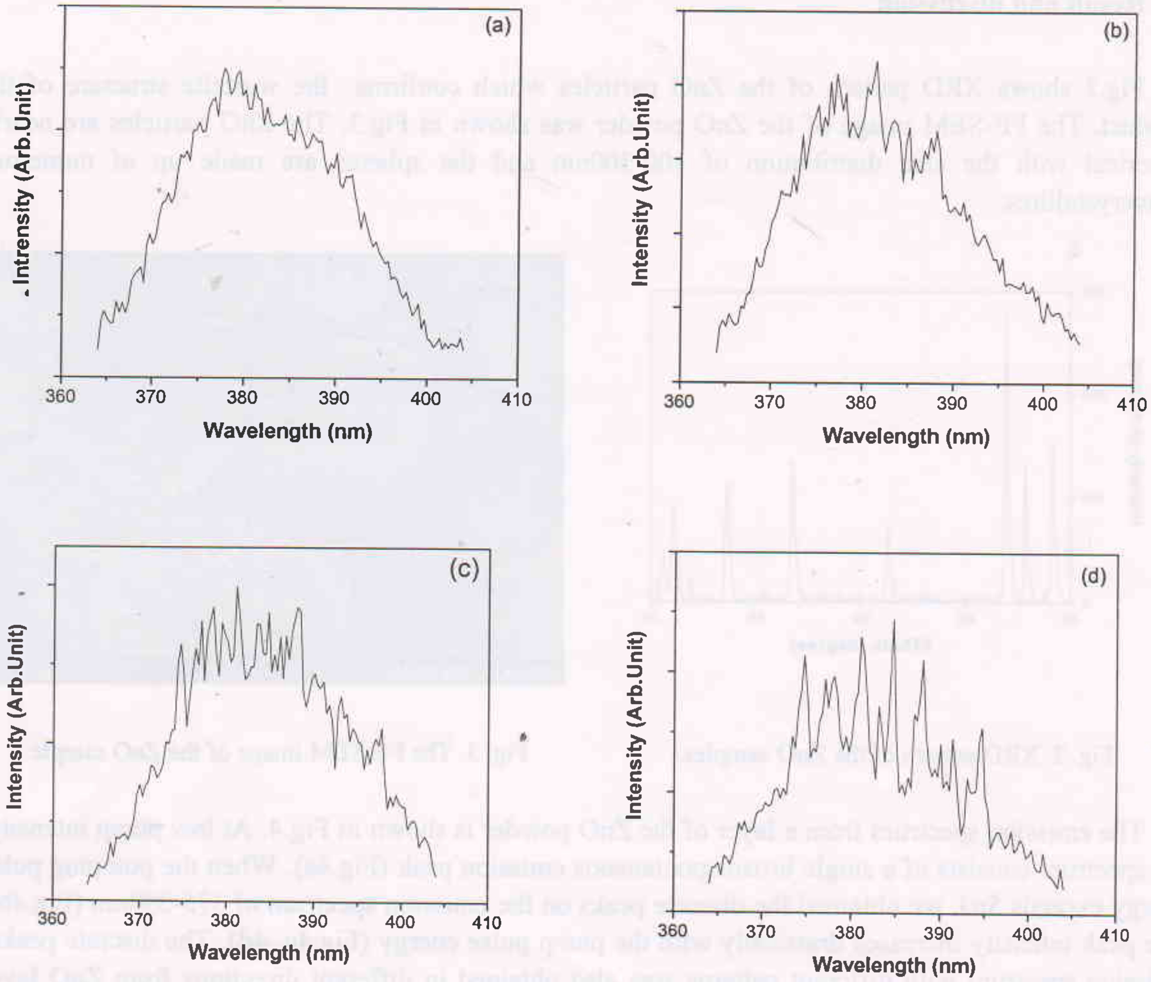

Fig. 4. Emission spectra collected from the layer of ZnO powder pumped by picosecond laser pulses with increasing pulse energy: a) 3μJ; b) 5μJ; c) 10μJ; d) 15μJ.

4. Conclusion

Powder of ZnO monodisperse spheres of 100-300nm diameter was synthesized by hydrolysis of Zinc Acetate dehydrates. Using the third harmonics (355nm) of a picosecond mode locked Nd: YAG laser (30ps pulse width and 10 Hz repetition rate) as an optical pump we obtained random lasing from a layer of the synthesized ZnO powder. Discrete peaks appear on the emission spectrum (around 380nm) and peak intensity increase drastically with pump pulse energy. It suggests that ZnO monodisperse colloidal spheres enhance recurrent light scattering in the layer of ZnO powder. There was a superposition of laser oscillations so that the laser spectrum with equidistant peaks was replaced by a spectrum with random spikes. The powder of ZnO colloidal spheres produced by hydrolysis of

Zinc Acetate dehydrate can be used as a random laser active medium. It may be a promising candidate for laser paint application.

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