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RANDOM LASING FROM A LAYER OF ZNO POWDER PAINTED ON GLASS SUBSTRATE UNDER EXCITATION OF NANOSECOND AND PICOSECOND LASER PULSES

NGUYEN THE BINH AND PHAM VAN THIN
Hanoi University of Science, Vietnam National University, Hanoi
PHAM THU NGA
Institude of Materials Science, VAST
NGUYEN ANH TUAN
Japan Advanced Institute of Science and Technology

Abstract. *The ZnO powder which consists of monodisperse colloidal spheres with average diameter of 100-300 nm was synthesized by hydrolysis of Zinc acetate dehydrate. The random lasing around 380 nm was produced from a layer of the ZnO powder painted on a glass substrate and optically pumped at 355 nm. Our experimental results show that random laser action of a layer of ZnO spherical nanoparticles and the characteristics of these random lasers under excitation of nanosecond and picosecond laser pulses.*

I. INTRODUCTION

The demonstration of random lasers opens up the possibility of using disordered medium as alternative sources of microlasers. In random lasers there are not well-defined cavities but self-formed cavities due to strong optical scattering in gain medium. It was demonstrated that recurrent light scattering can provide resonant feedback for lasing in a three-dimensional random medium [1]. 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. 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 [2].

Among several semiconductor materials, ZnO is a wide-gap material which has attracted great interest because of its potential to produce efficient blue and violet-light-emitting lasers. Zinc oxide has a large exciton binding energy (60 meV) and a small Bohr radius (18 $\mathring{A}$), which in principle could allow efficient excitonic gain to operate at room temperature [3]. Exciton-exciton scattering-induced stimulated emission is very important for the realization of low-threshold lasers at the room temperature. Random laser action in ZnO has been extensively studied in various forms. 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 powder [4–6], in ZnO polycrystalline films [7]. 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 [1]. Morphology of ZnO nanoparticles takes an important role to produce random laser with resonant feedback.

In this paper, we report the random laser action from a layer of ZnO powder formed by ZnO monodisperse colloidal spheres of 100-300 nm diameter. Interestingly, such a layer of ZnO powder can be painted on any substrate.

II. 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) and TEM (JEM 1010). A layer of the synthesized ZnO powder was simply performed on a glass substrate by mixture with purified water. The dried ZnO powder layer has a thickness of about 300 μm. The random laser action of ZnO nanostructure was studied under optical excitation by nanosecond and picosecond laser pulses.

Nanosecond pulse pump was provided from the third harmonics (355 nm) of a Nd:YAG laser (Quanta-Ray Pro 230 Newport, USA) with pulse duration of 5 ns and repetition rate of 10 Hz. Random laser emission was observed by a MS-257 spectrograph with a CCD IntraSpec (Oriel-Newport, USA). The experimental arrangement was shown in Fig.1a. The pump beam was focused to a spot of 3 mm diameter on the ZnO layer at an accidence angle of 45^0. The emission spectrum from the ZnO layer was collected around the direction normal to the layer surface.

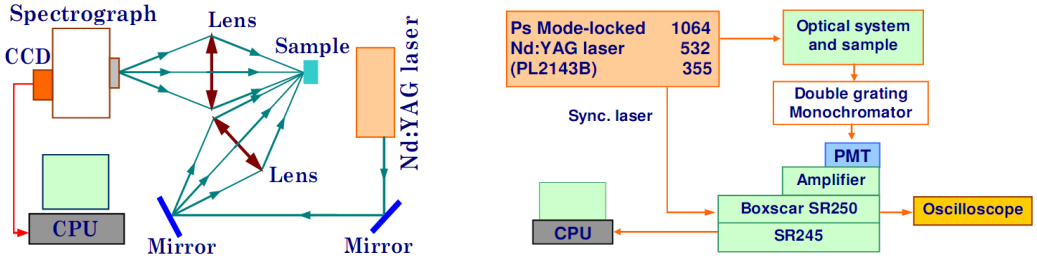


Fig. 1. Experimental arrangement

Picosecond pulse pump was carried out by the third harmonics (355 nm) of a picosecond mode locked Nd:YAG laser (PL2143B-Ekspla) with pulse duration of 30 ps and

repetition rate of 10 Hz. Emission spectra were analyzed by a double grating monochromator with 0.2 nm spectral resolutions. Then the spectrum signal was detected by an acquisition system which consists of a PMT, Boxca Averager SR250, A/D converter and Computer interface Module SR245. The block diagram of measurement and the detection system were given in Fig. 1(b).

III. DISCUSSION

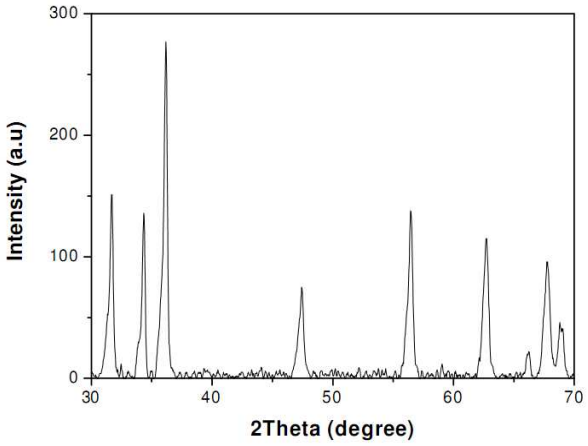
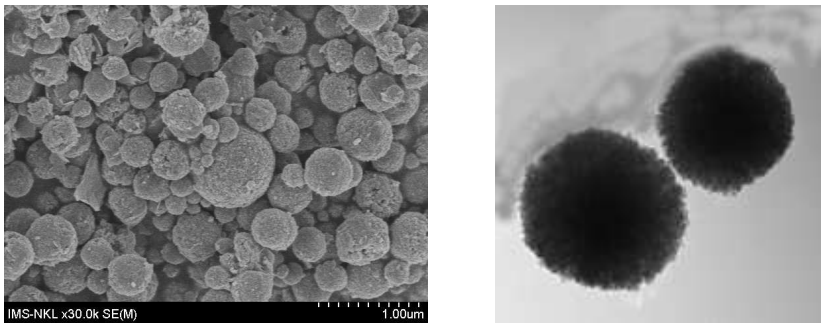


Fig. 2. XRD pattern of the ZnO samples

XRD pattern of our ZnO samples is shown in Fig. 2 which confirms the wurtzite structure of ZnO crystal. SEM images of the ZnO powder were given in Fig. 3(a). The ZnO particles are nearly spherical with the size distribution from 100 nm to 300 nm. TEM images (Fig.3b) demonstrate that the spheres are made up of numerous nanocrystallites.



(a) (b)

Fig. 3. The FE-SEM image (a) and TEM image (b) of the ZnO sample

Using MS-257 spectrograph with CCD detection we study random laser spectrum from the layer of synthesized ZnO powder pumped by nanosecond laser pulses. In multiple pulse registration, the emission spectrum did not show disrete peaks which are a characteristic feature of laser emission. In single pulse registration, the emission spectra showed some interesting features as shown in Fig.4. At low pumping intensity, a single spontaneous emission band that describes the transition from the conduction band to the valence band of ZnO (FWHM is about 20 nm) was observed. When the pumping pulse energy exceeds a threshold level (8 mJ) some spikes appear on the spectrum profile and the emission spectrum band width becomes narrower (FWHM is about 5 nm). The inset Fig. 4 shows the integrated emission intensity as a function of excitation intensity.

As we know, the recombination of excitons in ZnO gives radiation in the region around 380 nm with the intrinsic lifetime shorter than 200 ps. In the case of nanosecond pumping, a quasi-continuous lasing action could be expected. Lasing process composes several laser oscillations with different frequency modes. The laser spectrum under single shot nanosecond pumping is an enough constructive superposition of several laser oscillations so that we can observe some spikes that appear on the spectrum profile but quite different from that under picosecond pumping. Meanwhile, it is a deconstructive superposition in case of multiple pulse registration so that any spike appears.

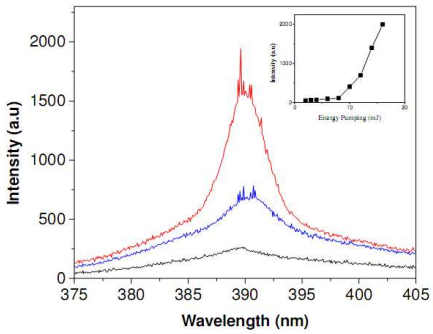


Fig. 4. The emission spectra in single shot registration under excitation of nanosecond pulse

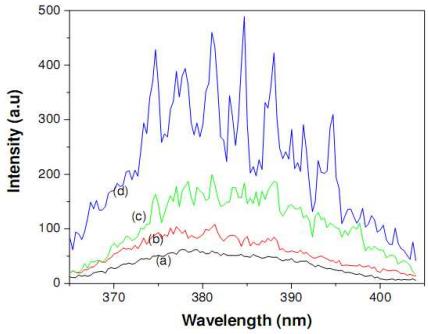


Fig. 5. The emission spectra under excitation of picosecond pulse with increasing pulse energy: a) 3 μJ; b) 5 μJ; c) 10 μJ; d) 15 μJ

In order to observe better random laser spectra from ZnO podwer we need picosecond laser pumping. The emission spectrum from a layer of the synthesized ZnO powder excited by picosecond laser pulses was shown in Fig. 5. At low pump intensity, the spectrum consists of a single broad spontaneous emission band (Fig. 5(a)). When the pumping pulse energy exceeds 5 μJ we obtained the discrete peaks on the emission spectrum of 375-390 nm (Fig.5b). The peak intensity increases drastically with the pump pulse energy (Fig. 5(c), Fig. 5(d)). The discrete peak-emission spectrum with different patterns was also obtained in different directions from the ZnO layer surface.

Because the ZnO layer is not parallel-plane, the lasing possibility in the vertical cavity formed by the layer surface and substrate surface can be neglected. Our experimental results suggest that the discrete peak-emission spectrum belongs to a constructive superposition of the discrete lasing modes which have origin from recurrent light scattering in

a three-dimensional gain medium. The powder of ZnO colloidal spheres of 100-300 nm diameter may enhance recurrent light scattering to provide resonant feedback so that the laser threshold is expected to be enough. The third harmonics (355 nm) of a picosecond mode locked Nd:YAG laser (PL2143B-Ekspla) with pulse duration of 30 ps is suitable to excite the ZnO powder for lasing. In fact, we can obtain lasing action without focusing laser pump beam. In the case, the lasing threshold is around 5 μJ when the pumped spot is 3 mm diameter.

IV. CONCLUSION

Powder of ZnO monodisperse spheres of 100-300 nm diameter was synthesized by hydrolysis of Zinc acetate dehydrates and used as active material of random microcavity laser. Using the third harmonics (355 nm) of a picosecond mode locked Nd: YAG laser (30 ps pulse width and 10 Hz repetition rate) as an optical pump source we obtained random lasing from a layer of the synthesized ZnO powder. Discrete peaks appear on the emission spectrum (around 380 nm) 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 random laser oscillations so that the laser spectrum with equidistant peaks was replaced by a emision spectrum of random peaks under picosecond pulse pumping. A constructive superposition of random laser oscillations with some spikes on spectrum profile was also obtained under single-shot nanosecond pulse pumping.

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