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Ultraviolet Random Laser Action of Nano-structured Zinc Oxide

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1. Introduction

Zinc oxide has been traditionally used in transparent conducting films, piezoelectric transducers, optical waveguides, varistors, acoustic wave devices and gas sensors. Especially, the large exciton binding energy at room temperature (~ 60 meV), which corresponds to 2.4 times the effective thermal energy at room temperature, ensures efficient ultraviolet (UV) emission from the excitons. The significant binding energy can also be changed by changing structures. The stability of the exciton makes ZnO a promising material for the realization of excitonic laser gain at room temperature. The strong excitonic effect in ZnO is expected to play an important role in the stimulated emission and gain processes. ZnO is widely recognized as an ideal material for room-temperature excitonic devices in optoelectronic areas.[1]

2. Experiments

The details of the preparation method are similar to those described in earlier literatures.[2,3] Both x-ray diffraction (XRD) and transmission electron microscopy (TEM) measurements were carried out for structural and morphological characterization. For pulsed pumping, we used the third harmonic of Nd:YVO₄ yttrium orthovanadate laser (3.51 eV photon energy) with a pulse width of ~ 500 ps and repetition rate of 1 kHz. The excitation laser beam was directed normally and focused onto the sample surface with power being varied with an optical attenuator. Spontaneous and stimulated emissions were collected by a fiber bundle and coupled into a 32-cm focal-length monochromator with a 1200 lines/mm grating, and then detected by either an electrically cooled charge-coupled device (CCD) detector. All of the experiments were performed at room temperature.

X-ray diffraction (XRD, Siemens D5000) analyses were conducted using Cu-K α radiation to determine the crystallinity of the ZnO samples. The spectra reveal the samples are preferentially orientated along (002) plane under suitable growth conditions, respectively. In addition, the diffraction peaks become sharper due to the increased particle size as well as the enhanced crystallinity. Figure 1 shows the TEM images of highly disordered ZnO nanoparticles and well-aligned one dimensional (1-D) nanorods via chemical solution and vapor transport technique, respec-

tively. A selective area electron diffraction pattern (SAED, shown in inserts of Fig. 1) of the same nanorod with zone axis confirmed that it was a single-crystalline wurtzite structured ZnO crystal. The TEM results are consistent with those of XRD measurement, and this also demonstrates that the nanostructures are single-crystal wurtzite ZnO.

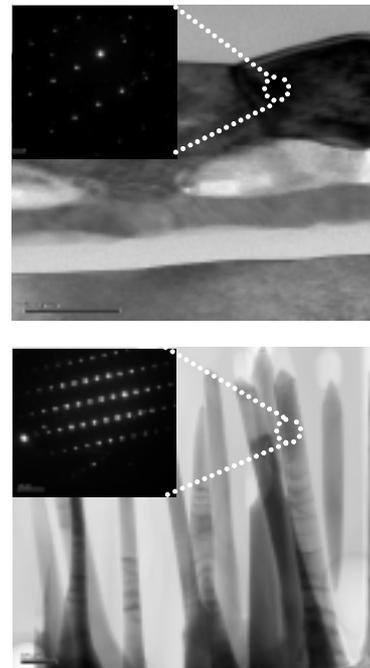


Fig. 1 TEM images of (a) highly disordered ZnO nanoparticles and (b) 1D nanorods, respectively. The insets show the SAED patterns.

Room-temperature optical characteristics of chemical-derived ZnO nanoparticles and nanorods were studied under an optical excitation (setup was shown in Fig. 2). The chemical-solution-derived samples showed only amplified spontaneous emission spectra with a weak wavelength of about 390 nm even at a pump intensity of 1.0 MW/cm². On the other hand, only the sample annealed at typical temperature showed a “kink” in the light-light curve, which is characteristic of laser action. In addition, the high-quality 1-D ZnO nanorods showed similar lasing

characteristic. At low pumping intensity, we observed a single broad spontaneous-emission peak. As the pumping intensity was increased, the peak became sharper. When the excitation intensity exceeded a threshold, the emission spectra consisted of a large number of narrow peaks, as shown in Fig. 3 indicating the onset of laser action in our nano-structured single-crystalline samples. The threshold intensity for the lasing was determined from superlinear behavior on optical power versus luminescence intensity.[4] Conventional lasing due to the Fabry-Perot cavities between the top and bottom natural facets or the lateral hexagonal microcrystallinities unlikely to occur in our samples. This is because the small thickness and the rough surface and boundaries of our ZnO nano-structured samples. Hence, random lasing mechanism is the only possibility for our ZnO nanostructures as the corresponding high-gain media and scattering mean-free path along the lateral direction might satisfy the requirements of random laser action, which has been observed in different forms of zinc oxide.

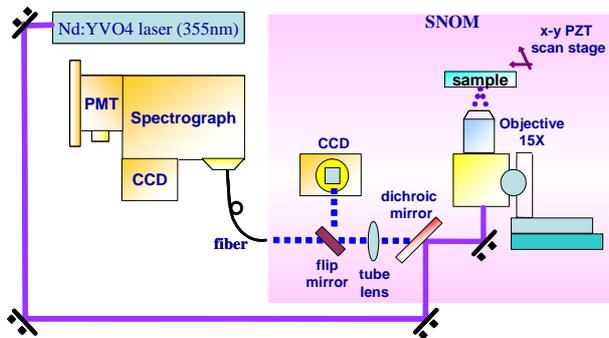


Fig. 2 Schematic illustration of optical-pumping setup.

In addition, it has been well-known that the light-scattering strength inside the annealed ZnO polycrystalline films is proportional to the corresponding average grain size. From our experimental results, it is noted that all ZnO samples annealed at low temperatures by chemical solution method exhibited no lasing. Thus, this can be attributed to the worse crystallinity and small ZnO grains annealed under lower temperatures.

3. Conclusions

Particularly, room-temperature UV random lasing of nano-structured ZnO samples has been demonstrated. It is shown that the nano-structured ZnO samples can exhibit random laser action depending on the growth condition. The threshold intensity for the lasing is comparable to earlier reported data. These results indicate that polycrystalline ZnO nanostructures prepared by chemical technique may be a promising material for further photonic devices. Further investigation to clarify the lasing mechanism between the nano-structured ZnO is still underway.

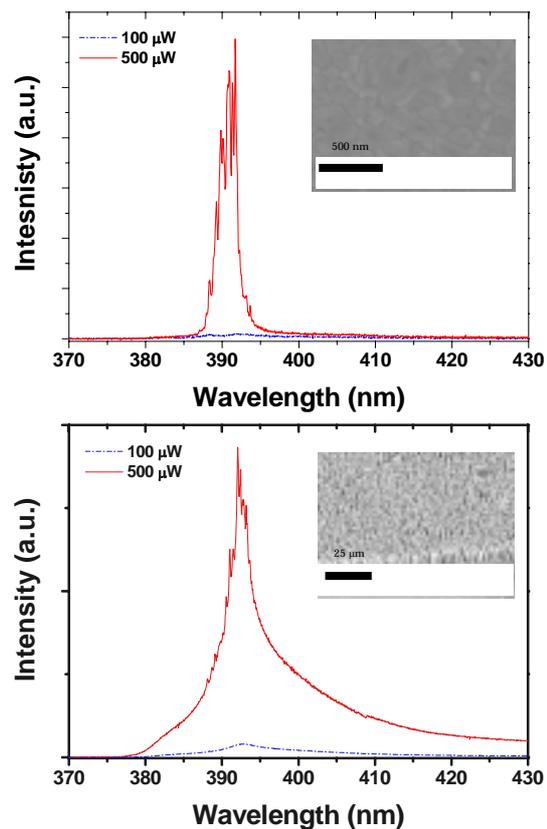


Fig. 3 The corresponding spontaneous and stimulated emission spectra under pumping intensities of (a) ZnO nanoparticles and (b) 1D nanorods.

References

- [1] Q. X. Yu, B. Xu, Q. H. Wu, Y. Liao, G. Z. Wang, R. C. Fang, H. Y. Lee, and C. T. Lee, *Appl. Phys. Lett.* **83** (2003) 4713.
- [2] S. Y. Kuo, W. C. Chen, and C. P. Cheng, *Superlattices and Microstructures* **39** (2006) 162.
- [3] H. C. Hsu, C. Y. Wu and W. F. Hsieh, *J. Appl. Phys.* **97** (2005) 64315.
- [4] X. Q. Zhang, I. Suemune, H. Kumano, J. Wang and S. H. Huang, *J. Appl. Phys.* **96** (2004) 3733.