

## L-2

## Growth of P-type ZnO Films by Pulsed Laser Deposition Infrared Light Assist

\*Hiroki Namai<sup>1</sup>, Toshihiro Hiraide<sup>2</sup>, Satoshi Kurumi<sup>3</sup> and Kaoru Suzuki<sup>3,4</sup>

Abstract: Zinc Oxide (ZnO) materials are the device of blue light-emitting-diodes (LEDs) that are alternate the gallium nitride (GaN). The ZnO films are grown on sapphire substrate (0001) by infrared-light-assist (IRA) pulsed laser deposition (PLD) using a focused Nd: YAG laser (wave length : 355 nm). After deposition, the Nd : YAG laser irradiates to the ZnO films for annealing. In this paper, we investigate the conditions of pulsed laser annealing for improvement crystallization of the ZnO films. The annealing process implements under following conditions: the wave length is at 266, 355, 532, and 1064 nm, respectively. The X-ray diffraction (XRD) spectra yield the ZnO films with a (002) and c-axis preferred orientation by the laser annealing below 532 nm of wave length. By 532 nm annealing, exciton emission spectra is caused by photoluminescence (PL) measurement.

## 1. Introduction

Zinc oxide (ZnO) materials are expected light-emitting diodes (LEDs) of alternate gallium nitride (GaN). The characteristics of ZnO are wurtzite crystal structure and wide gap energy of 3.37 eV that similar to GaN. The exciton binding energy of ZnO (60 meV) is larger than that of GaN (28 meV) at room temperature, giving advantage over GaN for the exciton-related device applications.<sup>[1]</sup> The ZnO is n-type semiconductor by oxide (O<sub>2</sub>) defects. A p-type ZnO is obtained by displacing nitrogen on oxygen parts. For growth the p-type ZnO films, we choose a infrared-light-assisted-pulsed-laser-deposition (IRA-PLD). Our previous report, the p-type ZnO films were obtained by IRA-PLD.<sup>[2]</sup> However, the reproduction error of the film comes out. Post-annealing is necessary to improve the crystalline of ZnO films with reproduction of p-type ZnO films.

In this article, we investigate the dependent on the laser annealing wave length for crystalline of ZnO films.

## 2. Experiment

## 2.1 ZnO films grown by IRA-PLD

ZnO films were grown on sapphire (Al<sub>2</sub>O<sub>3</sub>) (0001) substrates by IRA-PLD using a focused Nd: YAG laser (LOTIS TEE LS 2147, wave length = 355 nm, fluence = 1.2 J/cm<sup>2</sup>, pulse width = 20 ns, repetition frequency = 10 Hz ) for 60 min. Laser target ZnO bulks are obtained from ZnO powders (99.99[%]), pressing at 80 kN for 20 min. The distance between the target bulk and the substrate was 20 mm. Atmosphere pressure is adjusted at 40 mTorr by vacuum and flowing nitrogen gas. The substrate temperature was adjusted to 473 K under IR irradiation.

## 2.2 Pulsed laser annealing of ZnO films

After growth, a focused Nd: YAG laser (energy= 500 mJ/cm<sup>2</sup>) irradiate to ZnO films for annealing under the air as shown in Fig. 1. The laser wave length was changed at 266, 355, 532 and 1064 nm, then the number of shots were 2 shots, using single shot mode. The character of the ZnO films were investigated by X-ray-diffraction (XRD) measurement (RIGAKU RINT-2000), photoluminescence (PL) measurement using He-Cd laser (KIMMON KR1801C, 325 nm) and transmittance measurement using spectra analyze r measurement (EPP2000-UVN-SR-50)

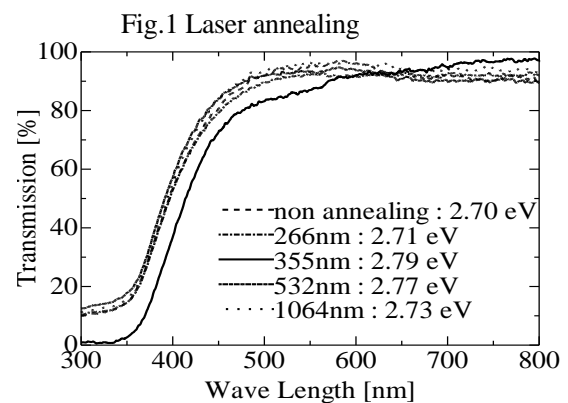
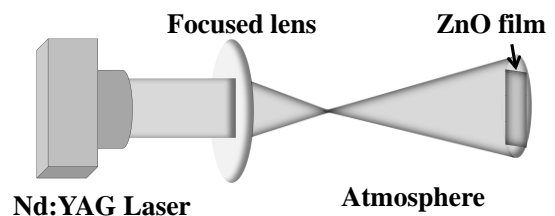


Fig. 2 Optical transmittance spectra of ZnO films

1: Student, Department of Electrical Engineering, CST, Nihon-u. 2: Graduate student of Electrical Engineering, CST, Nihon-U. 3: Department of Electrical Engineering, CST, Nihon-U. 4: Advanced Materials Science Center and Center for Creative Materials Research.

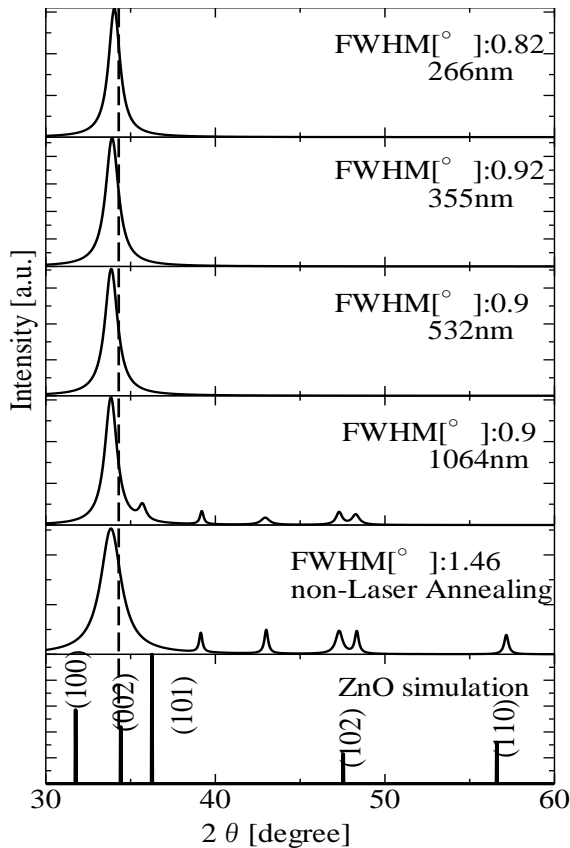


Fig.3 XRD spectra of ZnO films

### 3. Experimented result and discussion

The optical transmission spectra of the ZnO films are shown in Fig. 2. There is little dependence between transmittance of films and annealing wave length. All ZnO films transmit visible light area above 80 %, and ultraviolet area below 20 %. The energy gap of ZnO films are changed from 2.70 eV to 2.79 eV. The XRD spectra of wave length ZnO films in Fig.3 indicate a strong orientation of (002) axis peak below annealing wave length of 532 nm. The 266 nm laser annealing obtains narrower full width half maximum (FWHM) of (002) axis peak and shifts nearly ideal peak than others wave length. Because of the high photon energy, short annealing wave length improves the crystalline of the ZnO films. The PL emission properties of annealing ZnO films shown in Fig. 4. The film of annealed at 532 nm has high exciton emission and low O<sub>2</sub> defects emission .

Energy gap of O<sub>2</sub> defects is from 2.2 eV to 2.8 eV. The energy of 532 nm lie between the values. Therefore, the photon energy of the annealing wave length improves O<sub>2</sub> defects emission. With that exciton emission becomes intense.

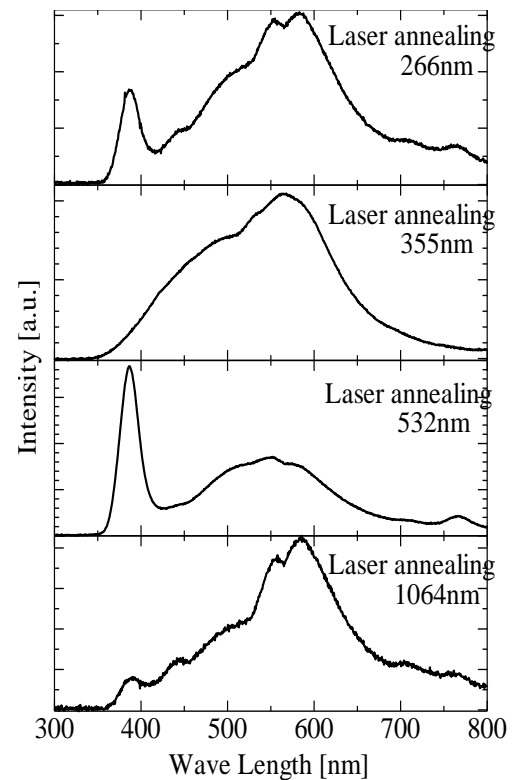


Fig.4 PL spectra of ZnO films

### 4. Conclusion

Crystalline and emission characteristic of the growth ZnO films are improved by laser annealing. Among them, the best annealing wave length of improvement crystallization is 266 nm, the wave length of emission characteristic is 532 nm.

### Acknowledgement

A part of this study has been financially supported by the “Toward Zero-Emission Energy” at Institute of Advanced Energy, Kyoto University, and Institute of Quantum Science, Nihon University.

### Reference

- [1] F.K. Shan, B.C. Shin, S.C. Kim, Y.S. Yu Ceramic Society, **24** 1861-1864 (2004)
- [2] T. Hiraide, S. Kurumi, K. Takase and K. Suzuki, Appl. Phys. A, DOI 10.1007/s00339-012-7228-4
- [3] A. Tsukazaki, M. Kubota, A. Ohtomo, T. Onuma, K. Ohtani, H. Ohno, S.F. Chichibu, M. Kawasaki, Jpn. J. Appl. Phys. **44**, 643–645 (2005)