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The Lasing Action Observed in Aligned ZnO Nanowires

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Abstract: Aligned ZnO nanowires (NWs) were obtained by hydrothermal method using annealed ZnO film as seed layer. Besides Si(400), only ZnO(002) diffraction peak was observed in X-ray diffraction (XRD) pattern. The lasing emissions were observed in optically pumped aligned ZnO NWs. Multi-mode emission peaks emerged when the excitation power density exceed the threshold. The integrated intensity of the spectra increases nonlinearly with the excitation power density, and the transition point from the spontaneous emission to lasing at about 96 kW/cm² can be clearly observed.

Key words: lasing; ZnO; aligned nanowires

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1 Introduction

ZnO is an attractive candidate material for UV optoelectronics devices due to its direct band gap of 3.37 eV at room temperature and a high exciton binding energy of 60 meV. ZnO has been used in the UV light emitting diodes (LEDs), the UV laser diodes (LDs), the UV photodetectors (PDs)¹⁻⁶, and other optoelectronic devices. The cavity of the ZnO lasers could be formed by the parallel surfaces such as top and bottom-surfaces or the lateral surfaces of the hexagonal shaped ZnO. Besides the narrowing emission line and the enhanced nonlinear emission intensity, the clear laser cavity mode is a definitive character for lasers. From the results

reported by Tang⁷ and Yang⁸, we could conclude that ZnO hexagonal structure could form the natural Fabry-Pérot (FP) lasing cavities for the close-packed ZnO microcrystallite thin films or separated ZnO nanorods. As we know, the sidewall of the ZnO hexagonal structure could provide lateral confinement⁹. Herein, we proposed that the cone shaped end ZnO NWs surfaces provided laser cavities, with an optical fiber placed at 58° relative to the normal direction of incident laser beam, the lasing action was observed.

2 Experiments

The ZnO thin film was prepared by radio-frequency (RF) magnetron sputtering method using

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a 99.999% pure ZnO target under a power of 130 W. After sputtering ZnO thin film on Si substrate, the rapid annealing process was explored to deal with the thin film at 1 000 °C for 30 min in air. Then, a hydrothermal method was explored to grown ZnO NWs on thin film using $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ and $\text{C}_6\text{H}_{12}\text{N}_4$ as reactant source.

The morphology of the samples were investigated by field-emission scanning electron microscopy (FESEM; Hitachi S4800 microscope) equipped with energy-dispersive X-ray (EDX). The crystal structure of the sample was studied by XRD Bruker D8GADDS X-ray diffractometer using Cu K α radiation. A mode-locked femtosecond Ti:sapphire laser with an optical parametric amplifier (OPA) were employed for the stimulated emission measurement of the ZnO NWs.

3 Results and Discussion

Fig. 1 (a) shows the FESEM image of ZnO NWs synthesized on the Si (100) substrate by inducing the ZnO-assisted layer. We can see that nearly all the ZnO NWs grow vertically on the substrate surface. The synthesized ZnO NWs are more than 100 nm in diameter. The XRD pattern of the NWs is displayed in Fig. 1 (b). The dominant diffraction peak of the sample can be attributed to wurtzite hexagonal ZnO. For the NWs, only one (002) diffractive peak can be observed, which indicates that NWs are of perfect *c*-axis orientation. The peak position for NWs is located at 34.5°, which equals to that of bulk ZnO. The full width at half-maximum (FWHM) for NWs is 0.11°, and the magnified pattern (002) shown in the inset comprises two peaks, which implies a good crystallinity of ZnO NWs.

The optically pumped emission measurements were performed using an optical parametric amplifier (OPA) in an active passive mode-locked femtosecond Ti:sapphire laser. The excited wavelength of the output laser was 350 nm. Fig. 2(a) shows the optically pumped emission spectra with the laser beam normal to the sample surface while the emission spectra were detected with an optical fiber placed at 58° relative to the incident laser beam. As the excitation

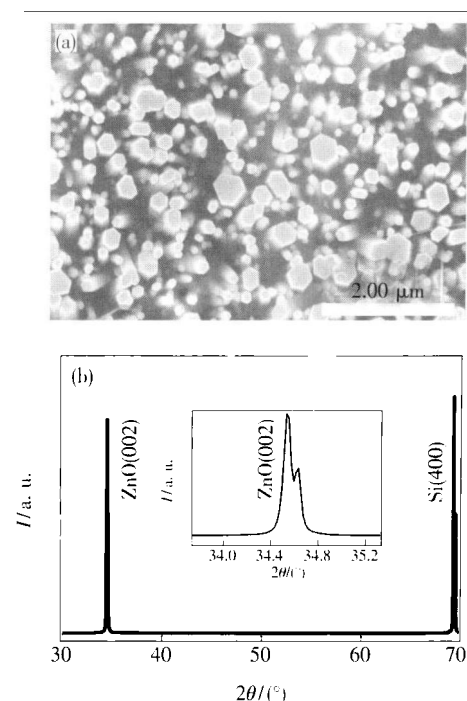


Fig. 1 (a) SEM image of the aligned ZnO nanowires; (b) XRD pattern of the ZnO nanowires grown on Si substrate.

densities changing from 20.8 to 57.6 $\text{kW} \cdot \text{cm}^{-2}$, only a broad spontaneous emission that the main emission peak shifted from 3.3 eV to 3.2 eV is observed with the emission intensities increasing linearly. When the excited laser power density exceeds the threshold level of about 96 $\text{kW} \cdot \text{cm}^{-2}$, two sharp peaks with less than 0.006 eV emerged on the broad spontaneous emission band. As the excitation density increasing to 160 $\text{kW} \cdot \text{cm}^{-2}$, as shown in Fig. 2(b), three obvious sharp peaks were observed accompanied with further red-shift for the lasing modes. As the pump power density increases, the gain becomes larger enough to enable cavity modes with higher loss to start lasing. As shown in the Fig. 2(c), the integrated intensity of the spectra increases nonlinearly with the excitation power density, and the transitions from the spontaneous emission to lasing at about 96 $\text{kW} \cdot \text{cm}^{-2}$ can be clearly observed.

Due to the easily formed cone shaped morphology of the ZnO nanowires during the final growth process, and the easily change of the direction of the laser light, with an optical fiber placed at 58° relative to the incident laser beam, the lasing action was observed.

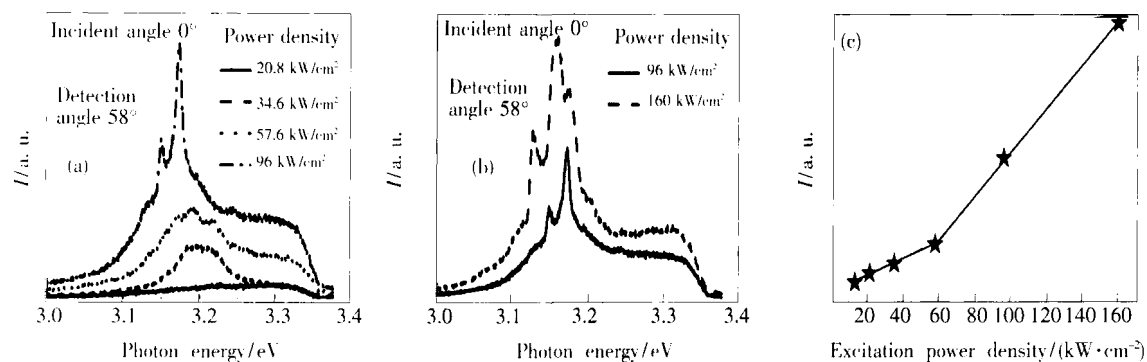


Fig. 2 (a,b) The evolution of the RT PL spectra in the ZnO nanowires as the excitation density increases from 20.8 to 160 kW/cm² by a mode-locked femtosecond Ti:sapphire laser; (c) The corresponding integrated intensity of the stimulated emission as a function of the excitation power density.

4 Conclusion

In conclusion, aligned ZnO NWs was fabricated by the low temperature hydrothermal method using annealed ZnO film. The optical pumped emission

spectra confirmed that the lasing action take place in the aligned NWs. Based on our experiment, we believe that direction controlled laser will be realized in the ZnO NWs with perfect structure.

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垂直氧化锌纳米线中的激射现象

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摘要: 通过水热的方法以退火 0.5 h 的 ZnO 薄膜作为籽晶, 得到垂直的 ZnO 纳米线。在 X 射线衍射谱中, 除了 Si 的(400)衍射峰以外, 只观察到了 ZnO 的(002)衍射峰。室温光致发光谱中出现了强的紫外发射峰, 同时也伴随着弱的缺陷相关的发射。这些数据表明垂直的 ZnO 纳米线序列有着较好的晶体质量。同时, 通过光泵浦也观察到了 ZnO 纳米线中的激光发射。当激发功率密度超过阈值且进一步增加时, 出现了多模发射峰, 其积分强度随着激发功率密度的增大呈非线性的增长, 并且在 $96 \text{ kW} \cdot \text{cm}^{-2}$ 处能清晰地观察到从自发发射到激射的转变。

关 键 词: 激光; ZnO; 垂直纳米线

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