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Optically Pumped Lasing from ZnO Nanorods Using Au as Reflection Mirror

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Abstract: ZnO nanorods were grown on a thin Au layer by hydrothermal method. In the photoluminescence (PL) spectrum, a strong UV emission and a weak deep-level emission were observed. The optically pumped lasing could be detected in ZnO nanorods. The integrated intensity of the spectra increases nonlinearly with the excitation power density, which indicated there exists the stimulated emission. And multi-mode emission peaks emerged when the excitation power density exceeded the threshold. Using Au layer as mirror, the light loss was further decreased, which resulted in the decrease of the laser threshold to 31.2 kW/cm^2.

Key words: lasing; ZnO; nanorods
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利用金作为反射镜的氧化锌纳米棒光泵激光

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摘要: 通过水热法在溅射了一层金的 Si 片上生长了 ZnO 纳米棒。实验观察到 ZnO 纳米棒的室温光致发光谱中出现了强的紫外发射峰,同时还伴随有弱的缺陷相关的发射,这表明通过该种方法生长的 ZnO 纳米棒晶体质量较好。同时,通过光泵浦也观察到了 ZnO 纳米棒中的激光发射。当激发光功率密度超过阈值,且进一步增加时则出现多个发射峰,其积分强度随着激发功率密度的增大呈非线性增长,进一步表明存在受激发射。利用金属层作为反射镜可以进一步降低损耗,从而达到降低阈值的目的。

关 键 词: 激光; ZnO; 纳米棒

1 Introduction

As one of the wide band gap semiconductors, ZnO has a high exciton binding energy of 60 meV. It has been used in the UV light emitting diodes (LEDs), the UV laser diodes (LDs), the UV

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photodetectors (PDs)[1-6], and other optoelectronic devices. The optically pumped laser has already observed in ZnO thin films and nanowires with a low threshold, which is much lower than that of GaN[7]. Due to its single crystalline structure and inherent resonant cavity, one dimensional (1D) ZnO nanostructures attracted more attentions of researchers in realizing its nanolaser applications. The hydrothermal method is considered to be a simple, convenient, inexpensive, and environmental- friendly for ZnO gerowth at low temperature. The low growth temperature will keep the morphology of the substrate unchanged during the growth process. To further reduce the threshold, the optical loss in the structures must be decreased. Because the size of 1D ZnO nanostructure is much smaller than the emission wavelength, the optical loss is significant. Therefore, to increase the reflectivity, one end facet is necessary to lower the threshold. One easy way in ZnO nanostructures to obtain better reflectivity is to synthetize ZnO nanostructures directly on a metal thin film. In this paper, we fabricated ZnO nanorods directly on a thin metal layer (Au). The thin metal layer took the role of a reflective mirror and reduced the optical losses. Then, the optical pumped lasers threshold could be further lowered.

2 Experiments

First, the Si substrate was cleaned by organic solvents and rinsed by de-ionized water to remove the contaminations. The Au thin film with the thickness of about 75 nm was deposited on a Si substrate by an ion sputtering method using a 99.99% pure Au target under an ion current of 10 mA. Then, the ZnO nanorods were grown by a hydrothermal method using zinc acetate[$Zn(CH_3COO)_2 \cdot 2H_2O$] and hexamethylenetetramine($C_6H_{12}N_4$) as reactant sources. 0.02 mol/L [$Zn(CH_3COO)_2 \cdot 2H_2O$] and 0.02 mol/L $C_6H_{12}N_4$ were dissolved in de-ionized water to form a 50 mL solution. Then, the 30 mL solution was moved to a reaction kettle of 50 mL capacity. The substrate was put into the solution. The reaction kettle was preserved at 90 ℃ for 16 h. After the reaction, the samples were taken out of the reaction kettle, cleaned with de-ionized water and dried in air at 60 ℃ for 2 h.

The morphology of the sample was investigated by a field-emission scanning electron microscopy (FESEM: Hitachi S4800 microscope) equipped with energy-dispersive X-ray (EDX). The photoluminescence (PL) measurement was performed using a JY-630 micro-Raman spectrometer with the 325 nm line of He-Cd laser as excitation source. A mode-locked femtosecond Ti: sapphire laser with an optical parametric amplifier (OPA) was employed for the stimulated emission measurement of the ZnO nanorods.

3 Results and Discussion

Fig. 1 shows the FESEM images of the ZnO nanorods. As shown in the Fig. 1, ZnO nanorods are about 5 μm in lengths and around 200 ~ 300 nm in diameters. The inset of Fig. 1 shows the cross-sectional FESEM image of the Au thin film on the Si substrate. The obtained Au thin film is about 75 nm thick.

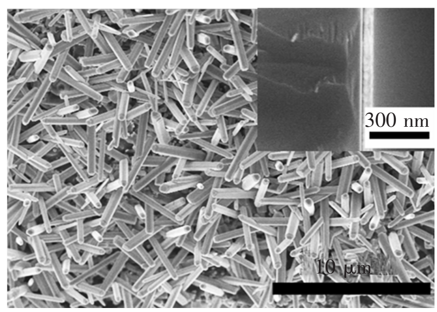

Fig. 1　SEM images of the ZnO nanorods grown on Si with Au layer about 75 nm. The inset of Fig. 1 shows the corresponding cross-sectional SEM images for the Si and Au layer.

Fig. 2 shows the room temperature PL spectrum of the ZnO nanorods. A strong UV emission located at 380 nm with a very weak visible emission located around 550 nm. The UV emission is considered as a near-band free exciton related emission and the weak visible emission usually results in the radiacive recombination of a photo-generated hole with an electron occupying the oxygen vacancy or surface state. The high aspect ratio of the UV/visible emission indicates the high quality of the ZnO nanorods.

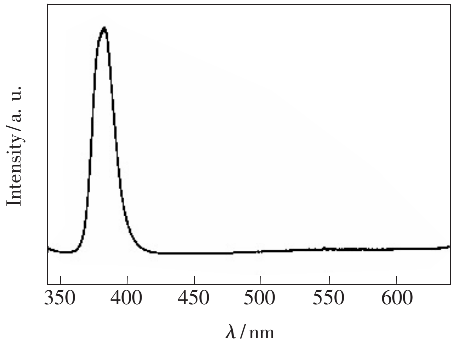

Fig. 2 The room temperature PL spectrum of the ZnO nanorods

In order to explore the stimulated emission in ZnO nanorods, the excitation-power-density dependent emission experiment was performed by using an optical parametric amplifier (OPA) equipped active passive mode-locked femtosecond Ti: sapphire laser with the excited wavelength of 350 nm at room temperature. The pump beam was focused on the ZnO nanorods with an incident angle of 60° to the normal direction of the sample, and the emission signals were collected in normal direction of the sample. In Fig. 3(a), it could be clearly observed three sharp emission peaks at the power density of 163 kW/cm^2. Moreover, the PL emission peak was slightly shifted to the low energy side while increasing the excitation power density from 35 kW/cm^2 to 163 kW/cm^2. This redshift could be induced by the band-gap renormalization due to Coulomb interactions among amplified free carriers at the band edge through the electron-hole plasma (EHP) process.

The room temperature excitation power density dependent PL emission spectra are shown in Fig. 3(b). The emission intensity is increases linearly when the excitation power density is raised from 7 kW/cm^2 to 21 kW/cm^2. But with the further increasing power density from 35 kW/cm^2 to 163 kW/cm^2, the emission intensity increased superlinearly. Such nonlinear increase of the emission intensity and the narrowed FWHM indicate the stimulated emission occurs in ZnO nanorods. And the threshold could be determined at 31.2 kW/cm^2 as shown in Fig. 3(b).

It is worthwhile to elucidate the reason for the low lasing threshold of the ZnO nanorods. From the results reported by Tang *et al.*[8] and Yang *et al.*[9], we could conclude that the faces of the ZnO

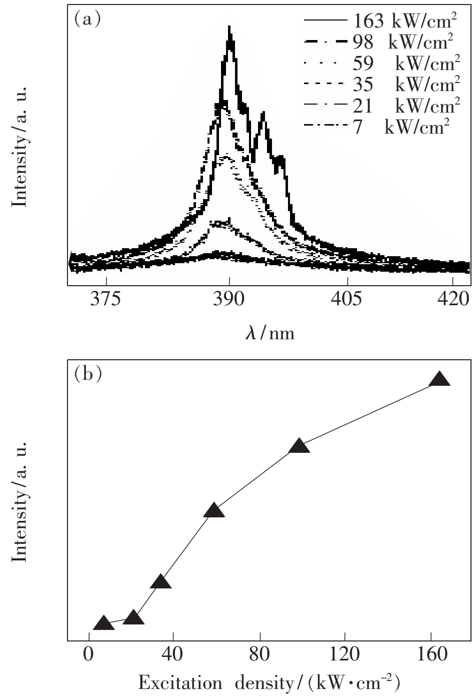

Fig. 3 (a) The room temperature PL spectra of the ZnO nanorods grown on Au layer excited by the mode-locked femtosecond Ti: sapphire laser at different excitation power densities. (b) The intensity of the stimulated emission as a function of the excitation power density.

nanorods formed the natural Febry-Perot (F-P) lasing cavities. Since the refractive index of ZnO is larger than that of air and Au, the top surface of the small diameter ZnO nanorods serves as a mirror to define an optical microcavity. The threshold gain g_{th} of the ZnO nanorods laser could be expressed as the following equation:

$$g_{th} = \alpha + \frac{1}{2L}\ln\frac{1}{R_1R_2}, \tag{1}$$

where, α is the loss along the length, L is the ZnO nanorods length, R is the end facet reflection[10]. According to this formula the threshold gain is a strong function of the length and the facet reflection. For a given cavity length, the high reflectivity would lead to lower threshold gain. Thus, the low threshold for the stimulated emission could be achieved on Au/Si substrate.

4 Conclusion

In conclusion, the optically pumped lasing was realized in the ZnO nanorods by growing ZnO on Au coated Si substrate. The threshold was 31.2 kW/cm^2, which was much lower than previous reports. The reason for low threshold lies in the formation of microcavities in the ZnO nanorods, which caused by the introduction of high reflectivity Au layer on Si substrate.

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