Local Nonlinearities of ZnO Nanostructures

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Abstract— The results of two-photon luminescence studies of zinc oxide (ZnO) microstructures consisting of microrods and microplanes are presented in this paper. When ZnO microstructure is excited by a femtosecond laser with a wavelength of 730 nm, two areas of luminescence are registered: the excitonic and impurity induced. The luminescence spectra of microstructures as function of temperature as well as the laser spot position along the structure are reported.

1. INTRODUCTION

Zinc oxide (ZnO) nano- and micro-structures have attracted enhanced interest of researchers due to their unique optical properties. ZnO posesses excitonic luminescence in the ultraviolet range that makes it very perspective cheap material for applications in optoelectronic devices [1, 2]. Recently semiconductor nanolasers based on nanorods of GaN, CdS and ZnO as well were realized [3]. The typical diameter of the rods is in the range of 50–200 nm and their length is about 2–40 microns, which makes them one of the smallest lasers today. It has been shown that even a single nanorod can work as a laser cavity with one or more modes of lasing [4]. A large refractive index contrast of ZnO nanorod regarding to the ambient air increases the quality factor of the laser cavity. In this paper, we report the radiation properties of the ZnO nanorods and nanoplanes ensembles.

2. EXPERIMENT AND RESULTS

ZnO microstructures were obtained by the MOCVD growth in a horizontal double furnace MOCVD set-up analogously to [5]. Fig. 1 shows SEM images of the obtained structures. They consist of arrays of microrods and microplanes randomly oriented along the axial axis of the whole structure.

Two-photon luminescence (TPL) experiments were performed with Ti: sapphire laser radiation (fundamental wavelength $\lambda = 730$ nm, pulse width 100 fs) focused onto a spot of about 100 μm perpendicular to the structure axial axis (see inset in Fig. 2). The sample was put in the Nitrogen cryostat in order to provide temperature measurements. Luminescence radiation from the sample was collected onto the entrance aperture of the waveguide, through which light transfer in a monochromator with small losses. The output luminescence intensity was measured by photomultiplier tube.

Figure 2 shows excitonic part of the luminescence spectrum (from 370 nm to 450 nm) of ZnO microstructure measured at different positions of the laser spot (see inset in Fig. 2). Excitation wavelength was 730 nm, the measurements were carried out at room temperature. Phonon side bands could not be resolved at room temperature although they are clearly seen at low temperature (Fig. 4). Both intensity (Fig. 2(b)) and spectrum shape are changed depending on the laser

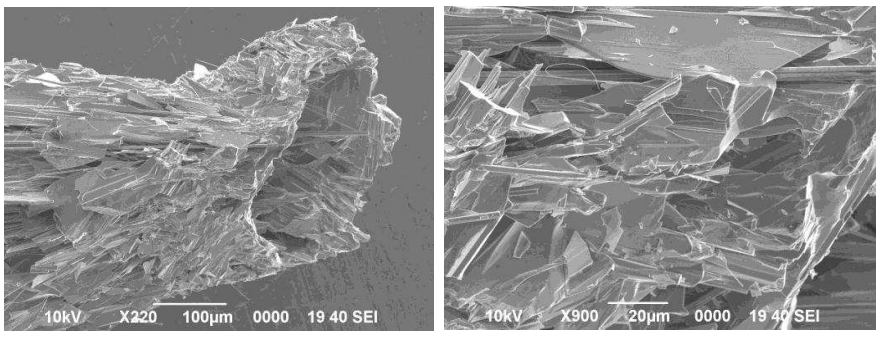

Figure 1: SEM micrographs of ZnO microstructures at different scales.

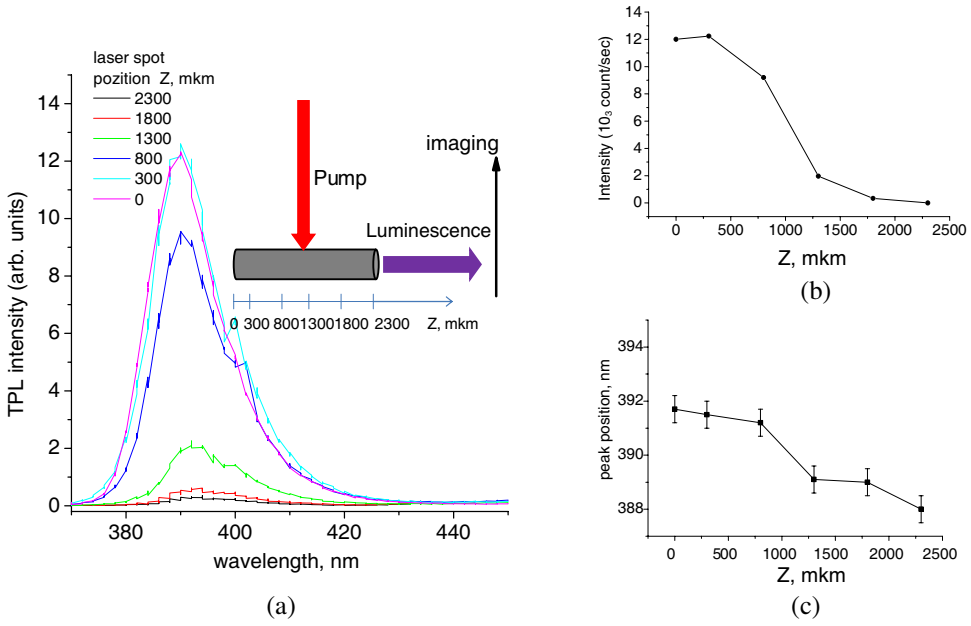

Figure 2: TPL spectra of ZnO microstructure at (a) different laser spot positions; (b) TPL intensity and (c) peak position of the center of excitonic peak as function of laser spot position; inset: geometry of TPL experiment.

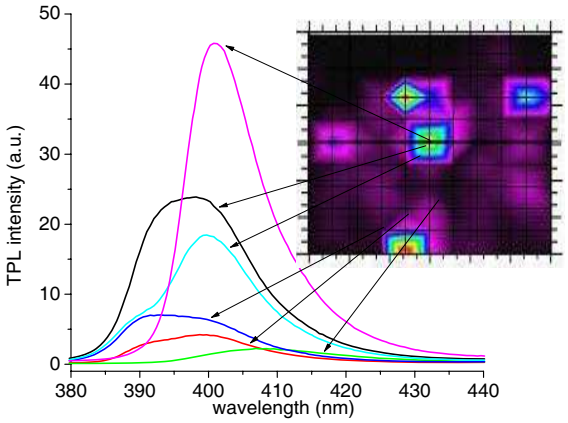

Figure 3: Scanning microscope TPL image of the sample surface.

spot position, the latter due to the different focusing conditions, the former due to the sample inhomogeniety and interplay between phonon side-bands. The same inhomogeneity which results in a quite strong change of the excitonic maximum shape is observed in TPL scanning microscopy image (Fig. 3). These studies show of inhomogeneity of the luminescence signal and hence inhomogeneoty of the composition along the sample.

Complete luminescence spectra were measured at different temperatures (Fig. 4). They consist of two parts — blue excitonic part with well pronounced phonon repetitions and wide shapeless red part. Generally the red part is associated with the presence of impurities [6]. The temperature dependence of excitonic part follows ordinary exponential law (Fig. 4(a)).

The graph shows two peaks, a narrow peak corresponds to the recombination of electron-hole pairs, and the broader-transitions at impurity levels. Reduce the temperature leads primarily to an increase in the total luminescence intensity (Figs. 4(b), (c)) and the spectral resolution of the excitonic lines.

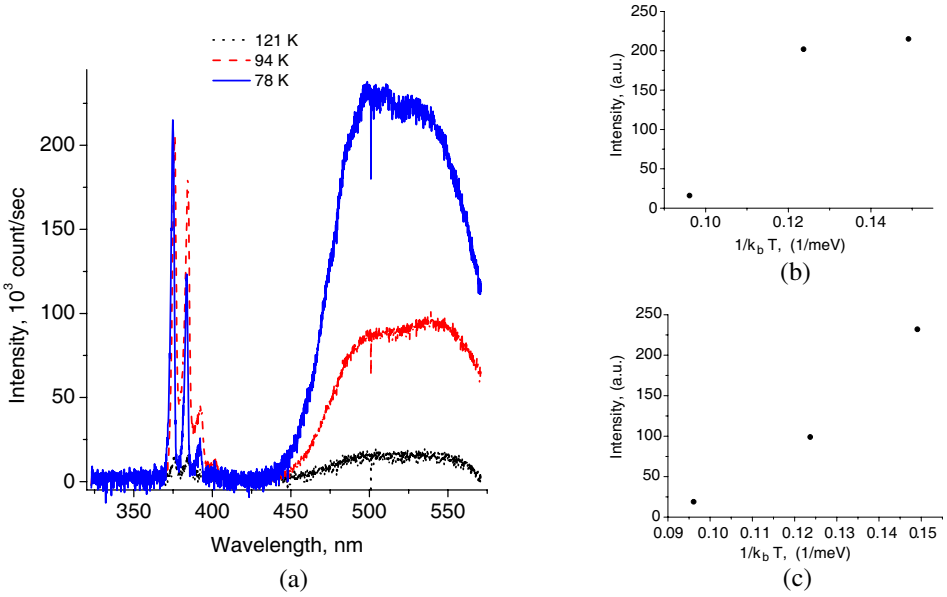

Figure 4: (a) Luminescence spectra of structures based on zinc oxide at different temperatures; Intensity of impurity peak dependence on temperature with theoretical fitting curve. (b) TPL intensity dependences on the temperature for the main excitonic peak and (c) impurity peak.

3. CONCLUSIONS

The results show that the intensity of the excitonic luminescence of zinc oxide microrods increases when the incident radiation falls close to the very edge of sample. Peak position dependence on the laser spot coordinate proves the fact of heterogeneous composition of the sample. Also showed, that a decrease of temperature leads to an increase of the TPL intensity and to the improvement of spectral resolution of the excitonic lines.

ACKNOWLEDGMENT

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