Lasing in ZnO Nanowires is Electron-Hole Plasma Lasing

Marijn A.M. Versteegh, Daniël Vanmaekelbergh, and Jaap I. Dijkhuis
Debye Institute for Nanomaterials Science, Utrecht University, Princetonplein 1, 3584 CC Utrecht, The Netherlands
e-mail: j.i.dijkhuis@uu.nl

Abstract: Lasing in ZnO nanowires is often interpreted as exciton lasing. However, our experiments and theoretical calculations on the laser threshold and the emission spectrum show that ZnO nanowire lasing is electron-hole plasma lasing.
OCIS codes: (140.3610) ultraviolet lasers; (140.5960) semiconductor lasers

1. Introduction

ZnO nanowires, with diameters in the order of 200 nm and lengths in the order of 10 μm, are promising materials for applications in miniature optoelectronic devices. They show subwavelength waveguiding and are among the smallest known lasers. A ZnO nanowire acts as a Fabry-Pérot cavity: the laser emission originates mainly from the ends of the wire and the spectrum shows laser peaks corresponding to standing waves in the wire [1].

Since Huang et al. [2] reported room temperature lasing in ZnO nanowires for the first time, ZnO nanowire lasing is usually described as excitonic, at least, if excitation is not too far above laser threshold. The idea behind this claim is that excitons in ZnO survive at room temperature, because their binding energy is 60 meV, considerably larger than $k_B T$ (25 meV).

We demonstrate, by experiments and theoretical calculations, that excitons are in fact not involved in room temperature laser action in ZnO nanowires. Even at the laser threshold excitons cannot exist. Rather, laser emission is the result of stimulated emission from recombining electrons and holes in an electron-hole plasma. The properties of laser action in ZnO nanowires, being the laser threshold, photon energy and spacing between the Fabry-Pérot laser modes, are well described by many-body electron-hole plasma lasing theory.

2. Threshold carrier density and Mott density

In literature one can distinguish three arguments to justify the claim of excitonic lasing in ZnO nanowires. The first argument for exciton lasing is that the threshold carrier density is below the Mott density. For increasing density, the Coulomb attraction that binds the electron and hole together into an exciton is more and more screened. If the density is higher than a critical density, the Mott density n_M, excitons cannot exist, and carriers form an electron-hole plasma.

Our calculation [3] of the Mott density gives $n_M = 1.5\cdot10^{24}$ m^{-3}. This value for the Mott density has to be compared with the actual carrier density in a ZnO nanowire at laser threshold. The usual method of exciting a ZnO nanowire laser is by a pump laser pulse. We computed the carrier densities at the laser threshold from the pump intensities and fluences given in literature. We find that the vast majority of reported laser thresholds actually occur above the Mott density, between $1.5\cdot10^{24}$ m^{-3} and $1.5\cdot10^{26}$ m^{-3}. We conclude that exciton lasing can be excluded for the vast majority of reported lasing ZnO nanowires at room temperature.

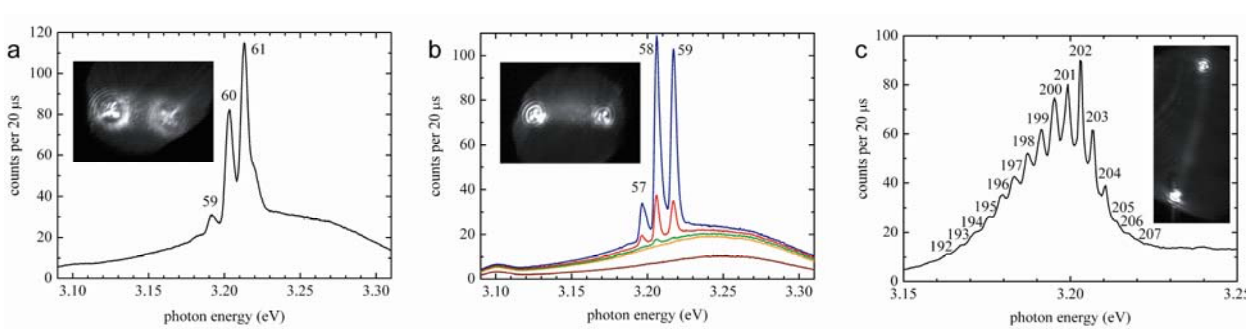

Fig. 1. Laser action in three nanowires. (a) 7.6 μm long nanowire. Excitation: 120-fs 800-nm pulses at F = 1585 J/m^2 (three-photon absorption). (b) 8.9 μm long nanowire. Excitation: 120-fs 800-nm pulses at F = 1619, 1904, 1940, 1974, and 2050 J/m^2 (three-photon absorption). (c) 23.5 μm long nanowire. Excitation: 120-fs 267-nm pulses at 25 J/m^2 (the laser threshold is at 11 J/m^2). Insets show UV images of the lasing nanowires. Fabry-Pérot mode numbers are indicated.

In order to determine the carrier density at the laser threshold as certain as possible, we excited ZnO nanowires in three different ways: by 5-ns 355-nm pulses from a 50-Hz Nd:YAG laser, and by 120-fs 267-nm and 120-fs 800-nm pulses from an amplified 1-kHz Ti:sapphire laser. The UV emission from the excited nanowires was collected by an objective and imaged on a CCD camera. Simultaneously the emission spectrum was measured by a spectrometer, coupled to a liquid-nitrogen-cooled CCD camera. We performed extensive measurements on seventeen nanowires. All threshold densities found were above $5\cdot10^{25}$ m^{-3}, clearly far above the Mott density. As an example, Figure 1 shows images and spectra for three nanowires.

3. Emission photon energy and band gap renormalization

The second argument sometimes given for excitonic lasing is that the emission photon energy is below the band gap, typically at about 3.2 eV, while the band gap of unexcited ZnO is 3.37 eV at room temperature. In order to check this argument, the band gap renormalization must be taken into consideration. The results of Ref. 3 indicate that at a density of $2.6\cdot10^{25}$ m^{-3} the band gap of room temperature ZnO has decreased from 3.37 eV to 3.20 eV. For all our nanowires the measured threshold density was higher than this value. Consequently, nanowire lasing takes place above the band gap.

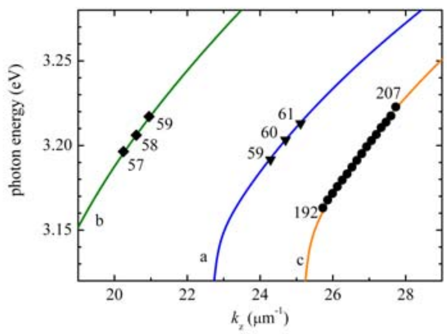

Fig. 2. The measured Fabry-Pérot modes of the three ZnO nanowires of figure 1 fitted onto theoretical dispersion relations at the densities (a) $6\cdot10^{25}$ m^{-3}, (b) $1.3\cdot10^{25}$ m^{-3} , and (c) $6\cdot10^{25}$ m^{-3}. Fabry-Pérot mode numbers are indicated.

4. Fabry-Pérot modes and dispersion relations

The third argument for excitonic lasing is that the spacing between the observed laser peaks is consistent with the exciton-polariton dispersion relation. We computed the refractive-index spectra of highly excited ZnO, using many-body quantum theory [3]. From these density-dependent refractive-index spectra we calculated the dispersion relations of light inside ZnO nanowires, taking the confinement of light due to the small nanowire dimensions into account. In Figure 2 our experimental data for the nanowires of figure 1 are fitted onto the theoretical dispersion relations. The measured Fabry-Pérot laser modes are in excellent agreement with the theoretical dispersion relations at carrier densities of more than an order of magnitude above the Mott density.

5. Conclusion

Room temperature lasing in ZnO nanowires is often described as excitonic lasing, or exciton-polariton lasing. In virtually all cases however, one deals with electron-hole plasma lasing. We have studied laser action in ZnO nanowires excited by nanosecond and femtosecond pulses. The laser thresholds and laser spectra found are in excellent agreement with many-body electron-hole plasma theory. Theoretical dispersion relations excellently describe the spacing between the Fabry-Pérot laser modes.

[1] L.K. van Vugt, S. Rühle, and D. Vanmaekelbergh, "Phase-Correlated Nondirectional Laser Emission from ZnO Nanowires", *Nano Lett.* **6,** 2707 (2006).

[2] M.H. Huang, S. Mao, H. Feick, H. Yan, Y. Wu, H. Kind, E. Weber, R. Russo, and P. Yang, "Room-Temperature Ultraviolet Nanowire Nanolasers", *Science* **292,** 1897 (2001).

[3] M.A.M. Versteegh, T. Kuis, H.T.C. Stoof, and J.I. Dijkhuis, "Ultrafast screening and carrier dynamics in ZnO: theory and experiment", to be published.