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Stimulated emission of disordered media based on crystalline ZnO powders

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***Abstract*. Low-threshold lasing ($E_{\rm th} = 0.94$ mJ cm^{-2}) is obtained in crystalline ZnO powders at room temperature. The stimulated emission spectra exhibit a distinct mode structure. The powders consisting of nanocrystals with the tetrapod-shaped morphology were synthesised by high-temperature pyrolysis of organic zinc salts.**

Keywords: *zinc oxide, powder laser, lasing in disordered media.*

The possibility of lasing in disordered objects has been theoretically substantiated by Letokhov in the 1960s [1]. Powder (or 'random') lasers are the lasers of a fundamentally new type. Laser action in them takes place in the absence of an external resonator inherent in a conventional laser and feedback is provided due to multiple scattering of light. The observation of efficient low-threshold UV lasing in polycrystalline zinc oxide films [2] stimulated extensive studies of objects based on ZnO. By now lasing has been obtained in materials of a high optical quality such as films, nanostructures, and nanocomposites [3 – 7]. In [8], laser action was achieved in polycrystalline clusters of size $\sim$1 μm. However, there are only few papers on lasing in crystalline ZnO powders, which are typical representatives of disordered media.

Zinc oxide is a wide-gap semiconductor ($\Delta E \simeq 3.37$ eV) with a unique set of mechanical, electric, and luminescent properties. It has the highest exciton binding energy ($\sim$60 meV) among binary semiconductors, which provides the existence of the UV luminescence band caused by the recombination of excitons at temperatures up to 500 K. It was pointed out that ZnO is a very promising material for obtaining UV lasing at room temperature [9].

Microcrystalline ZnO powders were synthesised by high-temperature pyrolysis of organic zinc salts. The microcrystals were regular concretions in the form of tetrapods (the length of a linear fragment was up to 10 μm and its width was up to 300 nm, Fig. 1).

Stimulated emission in ZnO powders was excited by 355-nm, 4-ns third-harmonic pulses from a Nd^{3+}:YAG laser with a pulse repetition rate of 1 kHz. The diameter of the laser beam was $\sim$1 mm. The exciting laser beam was directed normally to the surface of a powder sample. Emission from the sample surface was directed via a flexible multi-core silica fibre to the entrance slit of an MS-300 spectrometer equipped with a CCD array (the spectral resolution was 0.1 nm). The fibre was oriented at the certain angle θ to the sample surface.

Figure 2 shows the emission spectra of a zinc oxide powder recorded at room temperature at different excitation levels for $\theta \approx 15°$. For excitation levels lower than 0.94 mJ cm^{-2}, the emission spectra consisted of one spontaneous luminescence band of zinc oxide at $\lambda \sim 389$ nm with the half-width $\Delta\lambda \sim 12$ nm. For excitation levels exceeding 0.94 mJ cm^{-2}, the equidistant narrow lines of half-width $\Delta\lambda \sim 0.2$ nm spaced by $\sim$0.58 nm appeared against this band background, the intensity of these bands drastically increasing with the pump level (Fig. 3). One can see from Fig. 2 that when the excitation level exceeds the threshold by approximately a factor of 1.4, the lasing lines dominate in the emission spectrum.

The dependence of the 389.9-nm lasing line intensity on the excitation level is shown in Fig. 3. One can see that the line intensity begins to increase rapidly above the threshold. The threshold nature of the change in the emission spectra (the appearance of new narrow lines and a drastic increase in their intensity with increasing the pump level) and a distinct mode structure of these lines confirm the observation of lasing in zinc oxide powders.

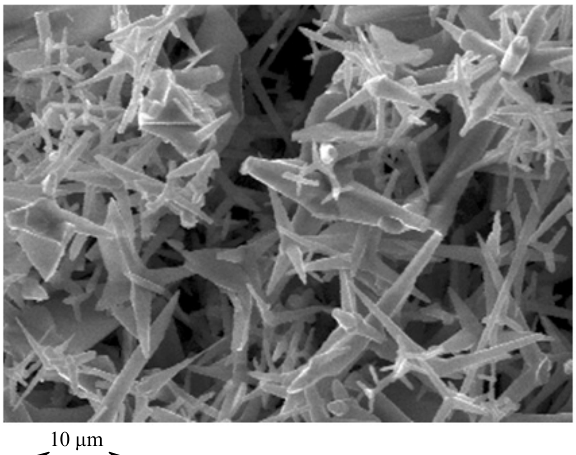

Figure 1. Morphology of ZnO powder crystallites.

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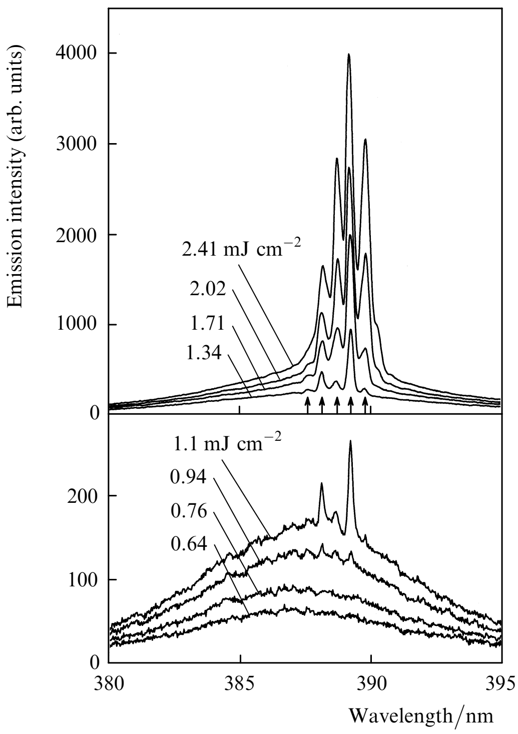

Figure 2. Emission spectra of a ZnO powder at different optical excitation levels. The arrows indicate the positions of equidistant stimulated emission lines.

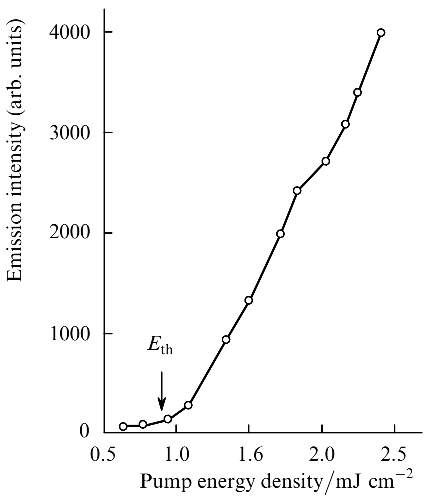

Figure 3. Dependence of the 389.9-nm lasing line intensity on the pump level.

The lasing spectrum was stable at a constant excitation level in our experiments. The type of the lasing spectrum, the lasing threshold, and the mode structure depended on the observation point on the powder surface and the observation angle θ, which can be explained by the spatial disorder of a powder sample and a random nature of closed resonator structures formed in it. These peculiarities of the lasing spectra will be discussed in detail elsewhere.

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