EMISSION FROM ZnS SUBJECTED TO TWO-PHOTON OPTICAL EXCITATION WITH THE SECOND HARMONIC OF A NEODYMIUM LASER

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Submitted May 30, 1967

Zh. Eksp. Teor. Fiz. 53, 1293–1295 (October, 1967)

Stimulated ultraviolet radiation from ZnS single crystals cooled to liquid nitrogen temperature was excited by two-photon pumping with the second harmonic of a neodymium laser. The emission wavelength was $\lambda = 0.33\ \mu$, and the line width was ≈ 15 Å for excitation power densities of ≈ 150 MW/cm^2.

STIMULATED ultraviolet emission (at a wavelength $\lambda = 0.33\ \mu$) was observed in hexagonal single crystals of zinc sulfide. ZnS crystals were excited by the two-photon absorption of the second harmonic of a neodymium laser ($\lambda = 0.53\ \mu$).

Up to now, laser emission in the ultraviolet range has been observed in semiconductor single crystals of ZnO[1] and ZnS[2, 3] by exciting them with an electron beam. The optical two-photon excitation of semiconductors has been limited to the use of ruby or neodymium lasers.[4–6] Development of powerful sources of optical harmonics and their use as two-photon pumping sources for wide-gap semiconductors has made it possible to obtain stimulated emission in the ultraviolet region.

Figure 1 shows the experimental arrangement used. Radiation from a Q-switched neodymium laser (pulse energy ≈ 1 J, pulse duration ≈ 30 nsec) was applied to an amplifier. The amplified radiation (radiation power density at the amplifier output $S_\omega = 150$ MW/cm^2, $\lambda = 1.06\ \mu$) was transformed in a potassium dihydrogen phosphate (KDP) crystal into the second harmonic ($S_{2\omega} \approx 25$ MW/cm^2, $\lambda = 0.53\ \mu$). The length of the KDP crystal, cut at a frequency locking angle, was 3 cm. To increase the intensity of the second harmonic, we used a cylindrical lens with a focal length F = 150 mm.

We investigated single crystals measuring $4 \times 4 \times 5$ mm, the distance between the faces forming the optical resonator being 5 mm. A crystal was placed on top of a copper cold duct cooled to liquid nitrogen temperature. The luminescence spectrum of a ZnS crystal was photographed through a ZMR-3 monochromator. A lens of focal length F = 150 mm was used to project one of the optical resonator faces onto the monochromator slit.

When the excitation power density was $S_{2\omega} \approx 50$ MW/cm^2, a strong luminescence, with a maximum in the region of 0.36 μ, was observed. The intensity of this luminescence increased quadratically when the pumping power was increased; this indicated that the excitation was of the two-photon kind.

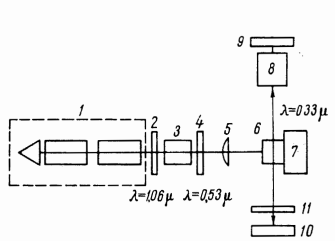

FIG. 1. Experimental layout: 1) neodymium-glass laser with amplifier; 2), 4), 11) optical filters; 3) KDP crystal; 5) cylindrical lens; 6) ZnS sample; 7) cryostat cold duct; 8) ZMR-3 monochromator; 9) photographic camera; 10) screen or photographic plate.

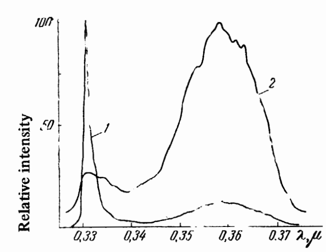

FIG. 2. Luminescence spectrum for $S_{2\omega} = 150$ MW/cm^2 (curve 1) and $S_{2\omega} = 120$ MW/cm^2 (curve 2); the ordinate scale for curve 2 is 10 times that for curve 1.

Figure 2 shows the luminescence spectrum of a ZnS crystal for excitation power densities of $S_{2\omega} = 120$ MW/cm^2 (curve 2) and $S_{2\omega} = 150$ MW/cm^2 (curve 1). When $S_{2\omega}$ was increased to 150 MW/cm^2, the width of the spectral line decreased to 15 Å and its spectral intensity increased considerably. At the same time, the divergence of the emitted radiation decreased.

At $S_{2\omega} \approx 100-150$ MW/cm^2, the intensity of the excitation should decrease rapidly along the length of a crystal due to the two-photon absorption. The intensity of the pumping radiation, after passing a layer of thickness x, in which two-photon absorption takes place, is given by the equation[5, 7]

$$S_{2\omega}{}^x = \frac{S_{2\omega}(1-r)e^{-\alpha x}}{1+\beta\alpha^{-1}(1-r)S_{2\omega}[1-e^{-\alpha x}]}$$

where α is the linear absorption coefficient in this layer; r is the reflection coefficient; β is the two-photon absorption coefficient. The intensity of the exciting beam should decrease to half in a ZnS crystal about 1 mm long (assuming that the two-photon absorption coefficient is $\beta \approx 0.1$ cm/MW [7]). This can explain why the stimulated radiation is not observed over the whole thickness of a crystal but in a layer next to the face illuminated with the pumping radiation. The high excitation threshold of the stimulated line is evidently due to imperfections in the available crystals.

It should be mentioned that the third and fourth harmonics of the laser radiation can also be used as multiphoton pumping sources for dielectrics.[8] KI single crystals, having a large quantum yield at liquid nitrogen temperature, are very promising in this respect.[9]

In conclusion, the authors thank L. A. Sysoev for supplying the zinc sulfide crystals, and R. V. Khokhlov,

S. A. Akhmanov, and D. N. Klyshko for discussing the results.

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Translated by A. Tybulewicz
149