

# AN OPTICAL CdS OSCILLATOR UNDER TWO-PHOTON EXCITATION BY A RUBY LASER

PHYSICS

1965

SovietRxiv

---

View the original and related papers at <https://sovietrxiv.org/items/ru-196501.35978>

Source: Math-Net.Ru and CyberLeninka. Machine translation. Verify with the original.

**Abstract**

**Full Text**

UDC 535.89

*PHYSICS*

V. K. KONYUKHOV, L. A. KULEVSKII,

Corresponding Member of the Academy of Sciences of the USSR A. M. PROKHOROV

## **AN OPTICAL CdS OSCILLATOR UNDER TWO-PHOTON EXCITATION BY A RUBY LASER**

The proposal to create an optical oscillator on a CdS crystal with optical excitation was made in 1962 <sup>(1)</sup>. The principal difficulty that must be overcome in realizing this proposal is that the probability of nonradiative transitions is large in comparison with the probability of radiative recombination. This difficulty is characteristic of most semiconductor materials. To maintain an inverted population, excitation sources are needed that provide a high rate of generation of electron-hole pairs in the volume of the semiconductor. Along with an electron beam <sup>(2-5)</sup>, laser radiation can serve as such an excitation source <sup>(6,7)</sup>, and the photon energy may be smaller than the forbidden-band width <sup>(8,9,16)</sup>, since at high photon densities two-photon interband absorption occurs.

In our paper we report the main characteristics of an optical oscillator on a CdS single crystal under two-photon excitation.

The CdS single crystals on which the experiments were carried out were grown from the gas phase by the Piper method and had no natural faces. To determine the characteristics of the material, reflection and luminescence spectra ( $T = 77^\circ \text{ K}$ ) were investigated under excitation by a mercury lamp with excitation-light wavelength  $\lambda = 366 \text{ m}$ . It is known that under these conditions the reflection spectrum contains the lines of two free excitons  $A$  ( $\lambda_A = 4870 \text{ \AA}$ ) and  $B$  ( $\lambda_B = 4840 \text{ \AA}$ ), while in luminescence only the line of exciton  $A$  appears with sufficient intensity <sup>(10-12)</sup>. In our material we observed two lines in the reflection spectrum, and one line in luminescence. The distance  $\Delta\lambda = 30 \text{ \AA}$  between the lines in the reflection spectrum remained unchanged from sample to sample, but their position on the wavelength scale changed from sample to sample. The lines in the reflection spectrum were always shifted toward the blue, and the magnitude of the shift reached  $40 \text{ \AA}$ . On the basis of additional considerations concerning the polarization dependence of both lines, we believe that the two lines in the reflection spectrum belong to excitons  $A$  and  $B$ , and that the single line in the luminescence spectrum belongs to exciton  $A$ . In the reflection spectrum the long-wavelength line is observed only for  $E \perp c$ , while

Fig. 1. Luminescence spectrum of a CdS crystal under the action of a ruby laser. A sample of size  $5 \times 3 \times 3$  mm has two plane-parallel faces. Exciting-flux density: 1–20 MW/cm<sup>2</sup>, 2–40 MW/cm<sup>2</sup>, 3–200 MW/cm<sup>2</sup>, 4–500 MW/cm<sup>2</sup>. CdS radiation is polarized with vector  $E \perp c$ .

Figure 1: Fig. 1. Luminescence spectrum of a CdS crystal under the action of a ruby laser. A sample of size  $5 \times 3 \times 3$  mm has two plane-parallel faces. Exciting-flux density: 1–20 MW/cm<sup>2</sup>, 2–40 MW/cm<sup>2</sup>, 3–200 MW/cm<sup>2</sup>, 4–500 MW/cm<sup>2</sup>. CdS radiation is polarized with vector  $E \perp c$ .

the short-wavelength line is observed for  $E \perp c$  and  $E \parallel c$ , which agrees with the behavior of the lines of excitons  $A$  and  $B$ , respectively. In the luminescence spectrum the radiation is polarized with  $E \perp c$ , which is characteristic of the emission of exciton  $A$ . Variations in the energy of the free excitons indicate variability of the forbidden-band width in CdS crystals, namely an increase of it by  $\sim 0.01 \div 0.02$  eV, which apparently can be explained either by deviation from stoichiometric composition or by the influence of foreign impurities.

The emission of CdS crystals ( $T = 77^\circ$  K) under the action of a ruby laser with a power of  $\sim 50$  MW/cm<sup>2</sup> is located in that part of the spectrum which is usually occupied by bound excitons, i.e., between the frequencies of the free excitons and edge emission. At low excitation levels,

when there is no narrowing of the spectrum due to regeneration, the luminescence spectrum is rather broad,  $\sim 0.04$  eV. Its position on the wavelength scale undergoes the same changes in passing from sample to sample as does the spectrum of free excitons. On CdS single-crystal samples the coefficient of two-photon absorption ( $T = 300^\circ$  K) was measured at different flux densities of the incident radiation ( $\lambda = 694$  m $\mu$ ). The results of the measurements are collected in Table 1.

**Table 1**

Flux density of the incident radiation, MW/cm <sup>2</sup>	55	25	10
Absorption coefficient, cm <sup>-1</sup>	1.1	0.5	0.2

As follows from the table, the coefficient of two-photon absorption is proportional to the light-flux density and in absolute magnitude agrees with theoretical estimates<sup>(8,13)</sup>. If the flux density is increased by focusing the laser beam to 500 MW/cm<sup>2</sup>, then the absorption coefficient increases to 10 cm<sup>-1</sup>; therefore over a length of  $\sim 0.1$  cm the energy of the laser beam will be absorbed almost completely. Under these conditions the laser provides a generation rate of electron-hole pairs of  $\sim 10^{27} \div 10^{28}$  pairs/sec  $\cdot$  cm<sup>3</sup>, which is comparable with the rate of pair generation under electron excitation<sup>(4,14)</sup> and makes it possible to obtain population inversion in CdS.

Fig. 2. Oscillograms of the exciting radiation (a) and luminescence of the CdS crystal (b). Sweep speed  $100 \cdot 10^{-9}$  sec/cm

Figure 2: Fig. 2. Oscillograms of the exciting radiation (a) and luminescence of the CdS crystal (b). Sweep speed  $100 \cdot 10^{-9}$  sec/cm

**Fig. 1.** Luminescence spectrum of a CdS crystal under the action of a ruby laser. A sample of size  $5 \times 3 \times 3$  mm has two plane-parallel faces. Exciting-flux density: 1–20 MW/cm<sup>2</sup>, 2–40 MW/cm<sup>2</sup>, 3–200 MW/cm<sup>2</sup>, 4–500 MW/cm<sup>2</sup>. CdS radiation is polarized with vector  $E \perp c$ .

We investigated the luminescence of CdS crystals ( $T = 88 \pm 1^\circ\text{K}$ ), with two plane-parallel faces, at flux densities up to 500 MW/cm<sup>2</sup>, and observed a change in the shape of the spectrum as the flux density increased (see Fig. 1). The narrowing of the spectrum indicates the appearance of population inversion and amplification of light waves in the crystal. Simultaneously, on the screen upon which the radiation from the crystal falls, a clear interference pattern appears, and the radiation becomes directional (beam divergence angle  $\sim 10^\circ$ ). Both phenomena point to spatial coherence of the radiation.

An oscillographic observation was carried out of the light pulses from CdS crystals ( $T = 88 \pm 1^\circ\text{K}$ ) in the spectral region  $490 \div 500$  m $\mu$ ,

which showed that the luminescence of the crystal in time repeats the shape of the pulse ( $\tau = 40$   $\mu$ ) of the exciting light at all excitation levels. The pulse has a bell-like shape with a duration 50% shorter than the duration of the ruby-laser pulse (see Fig. 2). The shortening of the pulse from the CdS crystal is explained by the fact that the absorbed power depends quadratically on the incident power. The identical shape of the exciting pulse and of the light pulse from the CdS crystal indicates that the luminescence process proceeds in a steady-state manner even under spontaneous emission.

On the basis of the available data, let us estimate the inversion population and the probability of nonradiative transitions. The magnitude of the inversion population can be determined from the condition that the amplification in the medium compensates the light losses due to incomplete reflection of the light wave from the end surfaces of the crystal:

**Fig. 2.** Oscillograms of the exciting radiation (a) and luminescence of the CdS crystal (b). Sweep speed  $100 \cdot 10^{-9}$  sec/cm

$$e^{\alpha l r} \sim 1,$$

where  $l = 0.5$  cm is the length of the crystal,  $\alpha$  is the gain coefficient,  $r = 0.2$  is the reflection coefficient from the end face (inactive absorption in the

crystal is not taken into account). The gain coefficient is related to the inversion population  $N$  by the relation

$$fN = \frac{cm}{\pi e^2} \alpha \Delta\nu,$$

where  $f$  is the oscillator strength of the transition,  $\Delta\nu = 10^{13} \text{ sec}^{-1}$  is the width of the luminescent-emission spectrum.

If the value  $f = 2.5 \cdot 10^{-3}$  [1] is used, then  $N = 5 \cdot 10^{17} \text{ cm}^{-3}$ . This value, in order of magnitude, coincides with the number of impurities in the crystal [15] and indicates that light generation occurs on bound excitons.

The probability  $w$  of nonradiative transitions ( $T = 90^\circ\text{K}$ ) can be determined by comparing the rate  $Q$  of generation of electron-hole pairs with the magnitude of the inversion population:

$$Q \sim Nw.$$

If one takes  $Q = 10^{27} \div 10^{28} \text{ pairs/sec} \cdot \text{cm}^3$ , as was discussed above, then

$$1/w = 10^{-9} \div 10^{-10} \text{ sec}.$$

The short lifetime of the exciton in the excited state explains the low quantum yield  $\eta$  of the luminescent emission, since

$$\eta = 1/w\tau_p,$$

where  $\tau_p$  is the radiative lifetime of the exciton. If  $\tau_p$  is calculated from the relation:

$$f\tau_p = \frac{3mc^3}{8\pi^2 e^2 \nu^2 n^2},$$

where  $n = 2.5$  is the refractive index of the CdS crystal,  $1/w\tau_p$  proves to be equal to  $10^{-3} \div 10^{-4}$ , which agrees with the measured quantum yield  $\sim 10^{-4}$ .

In conclusion, the authors express their gratitude to V. V. Osiko and V. S. Orlov for providing CdS single crystals, to V. S. Vavilov, E. A. Konorova, and V. B. Stopachinskii for useful discussions, and to I. V. Matrosov for assistance in the work.

Lebedev Physical Institute  
Academy of Sciences of the USSR

Received  
11 VIII 1965

## REFERENCES

1. D. G. Thomas, J. J. Hopfield, J. Appl. Phys., **33**, 3243 (1962).
2. N. G. Basov, O. V. Bogdankevich, A. G. Devyatkov, DAN, **155**, 783 (1964).
3. S. Benoit la Guillaume, J. M. Deberer, Congrès intern. sur la physique des Semiconducteurs, Paris, 1964.
4. V. S. Vavilov, E. A. Nolle, V. D. Egorov, Fiz. tverd. tela, **7**, 934 (1965).
5. C. E. Hurwitz, R. J. Keyes, Appl. Phys. Lett., **5**, 139 (1964).
6. N. G. Basov, A. Z. Grasyuk, V. A. Katulin, DAN, **161**, 1306 (1965).
7. R. J. Phelan, R. H. Rediker, Appl. Phys. Lett., **6**, 70 (1965).
8. R. Braunstein, N. Ockman, Phys. Rev., **134**, A 499 (1964).
9. N. G. Basov, A. Z. Grasyuk, I. G. Zubarev, V. A. Katulin, JETP Letters to the Editor, **1**, no. 4, 29 (1965).
10. D. G. Thomas, J. J. Hopfield, Phys. Rev., **116**, 573 (1959).
11. B. S. Razbirin, Dissertation, Leningrad, 1964.
12. C. E. Bleil, J. Broser, Congrès intern. sur la physique des Semiconducteurs, Paris, 1964, p. 13.
13. I. V. Keldysh, JETP, **47**, 1945 (1964).
14. N. G. Basov, O. O. Bogdankevich, A. G. Devyatkov, JETP, **47**, 1588 (1964).
15. D. G. Thomas, J. J. Hopfield, Phys. Rev., **128**, 2135 (1962).
16. N. G. Basov, Report at the Conference on Quantum Electronics in Puerto Rico, June, 1965.

*Note: Figure translations are in progress. See original paper for figures.*

*Source: Math-Net.Ru and CyberLeninka. Machine translation. Verify with the original.*