

the self-focusing beam breaks up into individual thinner beams as a result of the inhomogeneity of the wave front.

Thus, irradiation and breakdown of crystals that are transparent in the infrared region by means of a powerful pulse from a CO₂ laser has made it possible to observe directly thermal self-focusing of 10- μ radiation in solids having $dn/dT < 0$.

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BOSE-EINSTEIN CONDENSATION OF EXCITONS IN A CdSe CRYSTAL

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The purpose of this investigation was an experimental search for the phenomenon of Bose condensation of excitons.¹⁾

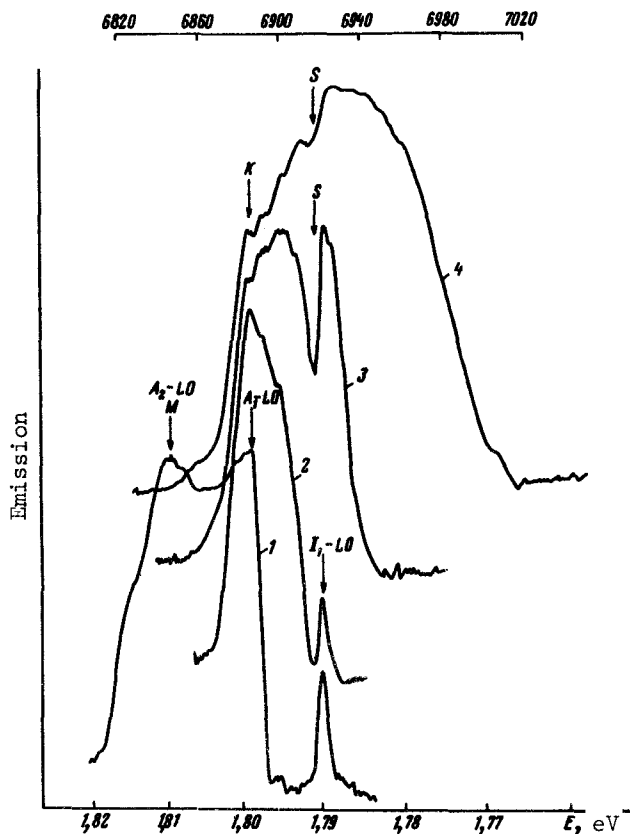
The CdSe crystal was chosen because the excitons in it are predominantly repelled from one another [2], and not attracted, and this, as is well known [1, 3], is the condition for Bose condensation of excitons.

The authors have focused their attention on the emission spectrum of CdSe in the LO band of interaction of the excitons with the optical phonons [4]. As was noted by one of the authors²⁾, this band can serve as a good indication of the onset of the Bose condensation of excitons, owing to the appearance of a narrow line (peak) on the long-wave boundary of this band, due to the vanishing of the momentum of the excitons following the Bose condensation.

The investigations were carried out at 4.2°K. The excitation source was the second harmonic of neodymium laser ($\lambda_{exc} = 5300 \text{ \AA}$). The pulse duration

¹⁾A detailed bibliography on Bose-Einstein condensation of excitons is given in Moskalenko's monograph [1].

²⁾E.F. Gross, Paper delivered at the Institute of Semiconductors, USSR Academy of Sciences, 2 December 1969.



Microphotographs of the emission spectrum of a CdSe crystal at $T = 4.2^\circ\text{K}$ following excitation with a laser at 5300 \AA and at different power levels (W). Curve 1 - $W = 55 \text{ kW/cm}^2$; curve 2 - $W = 150 \text{ kW/cm}^2$; curve 3 - $W = 200 \text{ kW/cm}^2$; curve 4 - $W = 500 \text{ kW/cm}^2$.

a new emission line, $\lambda = 6887 \text{ \AA}$, with width $2 - 3 \text{ \AA}$, appears on its long-wave edge, we shall designate it by K.

Simultaneously, a continuous radiation background appears on the long-wave side of the line K. With increasing excitation, this background becomes stronger and propagates in the long-wave side. At an excitation $W = 1500 \text{ kW/cm}^2$ the background reaches the wavelength region $\sim 7000 \text{ \AA}$.

On this background there are observed on the long-wave side of the K line three new emission lines γ_1 , γ_2 , and γ_3 (see the table), forming together with the line K an equidistant sequence with an approximate period 8 \AA (2 meV). The

did not exceed $5 \times 10^{-8} \text{ sec}$. The excitation power ranged from 6 to 1500 kW/cm^2 . According to rough estimates, the exciton concentration reached $10^{17} - 10^{18} \text{ cm}^{-3}$. The emission spectra were photographed with an instrument having a dispersion of 30.5 \AA/mm in the investigated region.

At excitation powers 6 - 50 kW/cm^2 , the luminescence spectrum of CdSe is similar to the spectrum described in [2], where the CdSe crystal was likewise excited with a neodymium laser. Further, at an excitation power $W = 55 \text{ kW/cm}^2$, the spectrum of the crystal still has many features in common with the spectrum obtained by excitation with a DRSh mercury lamp [5] (see the figure). Curve 1 shows clearly the first phonon replica of the free exciton $n = 1$ of the series $\Gamma_9 - \Gamma_7$ ($A_1 - \text{LO}$); the first phonon replica of the free exciton $n = 2$ ($A_2 - \text{LO}$) [5] (according to another interpretation - the band M due to exciton collision [2]), and the line of the bound exciton $I_1 - \text{LO}$. The resonance emission lines of the excitons are exceedingly weak. We note that the $A_1 - \text{LO}$ band has, at an excitation $W = 55 \text{ kW/cm}^2$, an asymmetrical Maxwell-like shape, due to the fact that the excitons have kinetic energy [4].

With further increase of the excitation, at $W \approx 150 \text{ kW/cm}^2$, a sharp change takes place in the emission spectrum of the crystal (see the figure).

The short wave part of the $A_1 - \text{LO}$ band decreases in intensity, and

Spectral position of the lines and S band observed
in the case of strong excitation with a laser.

Designation	Wavelength, Å	Energy, eV	Distance between lines, eV
K	6887	1,7999	
λ_1	6896	1,7976	0,0023
λ_2	6904	1,7955	0,0021
λ_3	6912	1,7934	0,0021
S ¹⁾	{ 6917 6921	{ 1,7921 1,7911	0,0010

¹⁾The end points and width are indicated for the band S.

width of these lines is about 3 Å. Some of the lines have a doublet structure³⁾.

In addition, on the long-wave side of the new lines there is observed one more singularity, namely a break in the continuous spectrum, in the form of a narrow band⁴⁾ (which we designate by S), with approximate width 4 Å and with end points 6917 - 6921 Å⁵⁾ (see the figure). On the long-wave side of the band S, the radiation background can be suspected of having a complicated structure. The spectral position and the widths of the new lines and of the band S did not vary in the entire investigated excitation interval (150 - 1500 kW/cm²). All that took place was a redistribution of the intensity among the lines, and the spreading of the continuous background in the long-wave direction. It is also remarkable that within the limits of the measurement error (± 1 Å) we observed no temperature shift of the spectrum, a shift that might have been observable in the case of strong excitation.

When the excitation increased above 150 kW/cm², the band A₂ - LO (or M) weakens and narrows down considerably. In some crystals it disappears completely.

It is difficult to explain the described experimental results within the framework of the usual concepts, and we propose that the observed phenomena are due to Bose-Einstein condensation of excitons:

1. The appearance of the narrow K line and the simultaneous radical changes in the distribution of the intensity of the exciton-phonon band are

³⁾All the new lines, as well as the line K, are not modes of the coherent emission, since they are invariably observed in crystals of different thickness, and do not depend on the geometry of the experiment. However, in the case of low excitation power (DRSh lamp) there exist three phonon-replica lines of bound excitons with wavelengths 6894, 6902.5, and 6913 Å. We do not believe, however, that laser excitation produces just these bound-exciton lines, since they have somewhat different wavelengths. In addition, the emission of bound-exciton lines usually is not amplified with increasing excitation.

⁴⁾An analysis of the experimental data leads us to the conclusion that this break in the radiation background cannot be due to either light absorption or interference phenomena in the CdSe [6].

⁵⁾The line I₁ - LO, with $\lambda = 6922.5$ Å, hinders the determination of the exact value of the long-wave end point.

characteristic of the onset of Bose condensation of excitons, and agree with theoretical investigations of V.A. Gergel'⁶⁾ and of Kazarinov and Suris [7].

The presence of the exciton-annihilation line K shows that the excitons are not destroyed on going over into the condensate (they do not dissociate into electrons and holes), and consequently there is no "metallization" of the excitons in our experiments with the CdSe crystal.

2. The intense quasicontinuous emission spectrum on the long-wave side of the line K can be naturally connected with acoustic oscillations in the exciton condensate when the annihilating excitons interact in the Bose condensate both with the quanta of the exciton sound and with other excitons of the condensate, when they acquire a momentum $p \neq 0$; this also agrees with the theoretical investigations of V.A. Gergel'.

3. The narrow break in the continuous emission spectrum (the band S) should possibly be regarded as a spectroscopic manifestation of the energy "gap" Δ in the spectrum of the electron excitations in the Bose condensate of the excitons. This gap was predicted many times in theoretical papers [8].

4. It is remarkable that the distance between the lines λ_{1-3} (2 meV) is equal to double the width of the gap of the band S in the background (1 meV). Since it follows from the theoretical papers [9] that double the gap width equals the binding energy of the electron-hole pair in the condensate, we consider this coincidence not to be accidental. It can be assumed that the lines λ_{1-3} are the result of the breaking of the electron-hole pairs (breaking energy 2 meV) at the expense of the exciton annihilation energy.

5. Account must be taken of the possible existence of plasma oscillations in the exciton condensate (due to the exciton annihilation energy); these oscillations can lead to the appearance in the spectrum of individual lines that fall in the region of the continuous spectrum observed by us.

6. The presence of a jumpwise change in the spectrum of the CdSe crystal with increasing excitation is also evidence in favor of the Bose condensation of the excitons.

7. The absence of a dependence (or perhaps the weak dependence) of the distance between the individual lines and the width of the band S on the excitation intensity can be understood if it is assumed that with increasing excitation and with increasing number of excitons the Bose condensate increases its volume in the crystal lattice, retaining the same density and other physical properties, until it fills the entire crystal.

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THREE-PHOTON IONIZATION OF A HELIUM ATOM IN THE EXCITED 2S STATE

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Multiphoton ionization of atoms in the ground state has been under diligent study during the last few years. We have obtained experimental data on multiphoton ionization of the helium atom in an excited state. Observation of such a process was reported in [1]. Besides being of independent interest, an investigation of the multiphoton ionization of an atom in the excited state may contribute greatly to our understanding of the process of multiphoton ionization of an atom from the ground state via a resonant intermediate excited state. In a large number of cases, the transition from the intermediate state to the continuous spectrum has a multiphoton character.

The experiment consisted of the following. The He atoms of a discharge-afterglow plasma, in the excited metastable states 2^1S and 2^3S , were exposed to focused radiation of a Q-switched ruby laser (Fig. 1). The ions produced in the plasma by the laser radiation were registered with a probe [1]. For ionization from the two states, it is necessary to absorb three quanta of ruby-laser radiation. It can be assumed, however, that the probability of ionization from the state 2^1S is much higher, since the energy of the two quanta is close to the energy of the 6^1S level (frequency deviation ~ 30 cm^{-1}). In the case of ionization from the 2^3S state, the frequency deviations are of the order of several hundred cm^{-1} .

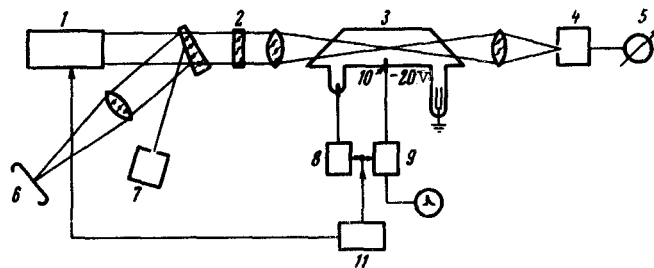


Fig. 1. Experimental setup: 1 - laser, 2 - radiation attenuator, 3 - discharge tube, 4 - calorimeter, 5 - galvanometer, 6 - photographic film; 7 - photocell, 8 - circuit producing the discharge in the tube, 9 - amplifier, 10 - probe, 11 - circuit for synchronizing the discharge shutoff with the laser pulse.

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