THE SPECTRUM OF TWO-PHOTON ABSORPTION IN CdS NEAR THE FUNDAMENTAL ABSORPTION EDGE

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Abstract

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THE SPECTRUM OF TWO-PHOTON ABSORPTION IN CdS NEAR THE FUNDAMENTAL ABSORPTION EDGE

The creation of lasers made it possible to observe experimentally a number of two- and multiphoton processes in matter, and also stimulated the appearance of numerous theoretical works in this direction. The work of Hopfield, Worlock, and Park ([1]), in which two-photon absorption was first observed in KJ crystals using a ruby laser and a source of ultraviolet radiation with a continuous spectrum, marked the beginning of two-photon spectroscopy. In semiconductors the two-photon absorption coefficient was first calculated in the work of Braunstein ([2]), and later, in the work of Braunstein and Ockman ([3]), luminescence in CdS was observed and investigated by means of two-photon excitation with a ruby laser. The two-photon absorption coefficient at the frequency of a ruby laser was first measured in the work of Konyukhov, Kulevskii, and Prokhorov ([4]). Finally, with the aid of two-photon excitation it proved possible to obtain generation in some semiconductors ([4–6]).

The character of the frequency dependence of the two-photon absorption coefficient in semiconductors is determined by the symmetry properties of the bands between which the transitions occur, i.e., by the selection rules for transitions from the valence band to the intermediate band and from the intermediate band to the conduction band. By investigating the frequency dependence of the two-photon absorption coefficient, one can determine the character of the two-photon transitions, which are divided into allowed–allowed, allowed–forbidden, and forbidden–forbidden. The frequency dependence of the two-photon absorption coefficient for each of these types of transitions was calculated in the work of Braunstein and Ockman ([3]).

In CdS, which does not possess a center of inversion, there is no parity prohibition for the transitions valence band—intermediate band and intermediate band —conduction band, so that both transitions may prove to be allowed simultaneously, which is impossible for crystals with a center of inversion.

If interband transitions at the value of the quasi-momentum $k = 0$ are allowed, then near $k = 0$ the matrix elements of the transitions remain approximately constant. In this case the frequency dependence of the absorption coefficient is

Fig. 1. Scheme of the experimental setup

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determined only by the density of states, i.e., by a factor of the form $(\hbar\omega_1 + \hbar\omega_2 - E_g)^{1/2}$. Therefore one may expect that in CdS, of the three possible cases considered in ([3]), the dependence of the two-photon absorption coefficient K_2 on the frequency ω_2 will be of the form

$$K_2(\omega_2) \sim N_1(\hbar\omega_1 + \hbar\omega_2 - E_g)^{1/2},$$

where N_1 is the density of photons $\hbar\omega_1$, and E_g is the band-gap width.

In the present work we investigated the frequency dependence of the two-photon absorption coefficient in CdS at room temperature. Two light sources were used, one of which gave monochromatic radiation, while the other emitted a continuous spectrum.

The source of monochromatic radiation was a Q-switched laser on neodymium-activated glass ($\hbar\omega_1 = 1.17$ eV) with an output energy of 1.5 J and a pulse duration

30 nsec. A pulsed xenon lamp IFR-2000 was used as the second light source; a capacitor bank with an energy of 1 kJ was discharged through it, and the pulse duration in this case was 250 μsec. The scheme of the experimental setup is shown in Fig. 1 and is analogous to the scheme used in the work of Hopfield, Worlock, and Park (^1).

With the aid of objective O_1, the pulsed lamp was imaged with a magnification of ~ 1 onto the CdS crystal, which in turn, with the aid of objective O_2, was also imaged with a magnification of ~ 1 onto the entrance slit of a double-prism monochromator DMR-4. The thickness of the CdS crystal in the direction of the light beam from the lamp was 0.6 cm. The laser beam passed through the CdS crystal in a direction perpendicular to the lamp light beam.

Fig. 1. Scheme of the experimental setup

After passing through the monochromator, the light from the pulsed lamp fell on a photomultiplier, the signal from which was fed to the input of the DÉO-1 and S1-11 oscillographs. The frequency interval ω_2 within which it was possible to measure the two-photon absorption coefficient was bounded on the low-frequency side by the decrease in the spectral sensitivity of the photomultiplier, and on the high-frequency side by the decrease, due to one-photon absorption, of the intensity of the lamp light that had passed through the CdS crystal. The absorption of the lamp light that had passed through the crystal was observed at the moment when the laser light pulse passed through the crystal; the magnitude of the light attenuation was recorded with the aid of the S1-11 oscillograph (Fig. 2b).

Fig. 2

Figure 2: Fig. 2

The average intensity of the lamp light does not change over a time equal to the duration of the laser pulse; however, the presence of intensity fluctuations lowers the accuracy of the measurements and imposes a limit on the minimum value of the two-photon absorption coefficient that can be measured.

Synchronization of the lamp and laser flashes was carried out so that the laser pulse coincided with the maximum of the lamp intensity. Oscillographic recording of the lamp intensity was performed with the aid of the DÉO-1 oscillograph (see Fig. 2a). By determining from the oscillograms the intensity of the light I_2 that had passed through the crystal and its attenuation ΔI_2 at the moment when the laser pulse passed, one can measure the value of the two-photon absorption coefficient for a specified photon energy $\hbar\omega_2$.

By selecting, with the aid of the monochromator, a certain frequency from the continuous spectrum of the lamp radiation, one can obtain the two-photon absorption spectrum, which is bounded on the low-frequency side by the condition $\hbar\omega_1 + \hbar\omega_2 = E_g$, i.e., by the law of conservation of energy, and on the high-frequency side by the condition $\hbar\omega_2 = E_g$, i.e., by the onset of one-photon absorption of light quanta $\hbar\omega_2$. The two-photon absorption coefficient at the frequency ω_2 does not depend on the photon density N_2 of the lamp radiation (photon energy

$\hbar\omega_2$) and is proportional to the photon density N_1 in the laser beam (photon energy $\hbar\omega_1$), which, on passing through the CdS crystal, practically did not change. The latter circumstance is due to the fact that the total (over the entire spectrum) photon density of the lamp radiation incident on the crystal is approximately three orders of magnitude smaller than the photon density in the laser beam; consequently, the two-photon absorption coefficient for the laser beam is smaller by the same factor. In this case, the law of attenuation of the lamp-light intensity on passing through the crystal will be exponential, and not hyperbolic, as in the case of absorption of two photons from one and the same source (7).

Fig. 2. *a* —oscillogram of the intensity of the IFK-2000 pulsed lamp. Sweep duration 250 μsec/cm; *b* —oscillogram of the intensity of the lamp light that has passed through the CdS crystal. Sweep duration 250 μsec/cm. The absorption signal is visible during passage of the laser pulse through the crystal (an upward deflection corresponds to a decrease in intensity).

The two-photon absorption coefficient was calculated by the formula

$$K_2 = \frac{1}{l} \ln \frac{I_2}{I_2 - \Delta I_2},$$

Fig. 3 and Fig. 4 plots

Figure 3: Fig. 3 and Fig. 4 plots

where $l = 0.6$ cm is the thickness of the crystal; I_2 is the intensity of the lamp light after passage through the crystal in the absence of the laser pulse; ΔI_2 is the attenuation of the lamp-light intensity at the moment when the laser pulse passes through the crystal.

The spectrum of two-photon absorption in CdS, obtained at a laser-radiation power of 50 MW/cm^2 ($\hbar\omega_1 = 1.17$ eV), is presented in Fig. 3. The fairly large scatter of the experimental points is associated with the nonuniform distribution of the intensity over the cross section of the laser beam, which, moreover, can vary from flash to flash (8), with instability of the laser output power, and also with fluctuations in the lamp intensity.

In order to establish the law of variation of the two-photon absorption coefficient with frequency ω_2, we processed the experimental data to find the exponent n in the expression

$$K_2 \sim (\hbar\omega_1 + \hbar\omega_2 - E_g)^n.$$

The processing was carried out by the least-squares method on an M-20 electronic computer. In Fig. 3, the solid line shows a parabola, po-

obtained as a result of this processing, the exponent n of this parabola turned out to be 0.57 ± 0.05. Thus, the dependence of the two-photon absorption coefficient on frequency is described quite well by the expression

$$K_2 \sim N_1(\hbar\omega_1 + \hbar\omega_2 - E_g)^{1/2},$$

which corresponds to an allowed–allowed two-photon transition.

At the frequency $\hbar\omega_2 = 1.77$ eV, the dependence of the two-photon absorption coefficient on the laser power was measured (for three power values); it is presented in Fig. 4. The linearity of this dependence confirms the two-photon character of the absorption. The absolute value of the absorption coefficient at the frequency $\hbar\omega_2 = 1.77$ eV at a laser power $I_0 = 50$ MW/cm^2 is

Fig. 3. Spectrum of two-photon absorption in CdS. The circles denote experimental points. The solid curve is a parabola with exponent 0.57, obtained by the method of least squares.

Fig. 4. Dependence of the two-photon absorption coefficient on laser power. $I_0 = 50$ MW/cm^2. The measurements were made at the lamp radiation frequency $\hbar\omega_2 = 1.77$ eV.

~ 1 cm^{-1}.

At the frequency $\hbar\omega_2 = (2 \cdot 1.78 - 1.17)$ eV $= 2.39$ eV, the two-photon absorption coefficient is about 1.5 cm^{-1}. These values are in agreement with the data of Ref. ([4]).

It is of interest to investigate the two-photon absorption spectrum at low temperatures on a spectral instrument with high dispersion, in order to measure the two-photon absorption coefficient in the exciton lines. In connection with the theoretical calculations of Inoue and Toyozawa ([9]), it is of interest to carry out polarization measurements, which will provide information on the band structure in CdS. Experiments in this direction are being carried out, and the results will be published.

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