

# Appearance of Fano antiresonance in excited exciton states in CdS

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The spectral and magneto-optical studies have shown for the first time that the doublet form of the absorption bands observed at low temperatures in very thin single crystals of CdS in the region of excitons with a principal quantum number  $n = 2$  is due to antiresonance interaction of  $S$  and  $P$  states analogous to the Fano effect.

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The doublet structure of the  $A$  and  $B$  exciton bands for states with  $n = 2$  was first detected in Ref. 1 in low temperature absorption spectra of very thin single crystals of CdS. The physical nature of this structure has remained unknown until now.

The point is that the nearly identical doublet structure was observed for three absorption bands:  $A$  exciton in E $\perp$ C polarization and  $B$  exciton in both polarizations. The selection rule for these bands allow transitions to the excitonic  $S$  state (as to the lowest states with principal quantum number  $n = 1$ ). According to Elliott's classification,<sup>2</sup> they are direct allowed transitions and their intensity exceeds by two orders of magnitude the intensity of transitions to the  $P$  state, so that in the case that these intensities add, it is precisely the  $S$  state that must be responsible for the complicated form of the corresponding bands. The proposition in Ref. 1 that the effect is due to terms linear in  $K$  in the conduction band (loop of extrema) was not subsequently confirmed.

To reveal the origin of the fine structure, we performed additional spectral and magneto-optical investigations, whose results are presented in this paper. These investigations permitted interpreting for the first time the complex form of these bands based on Fano's theory.<sup>3</sup>

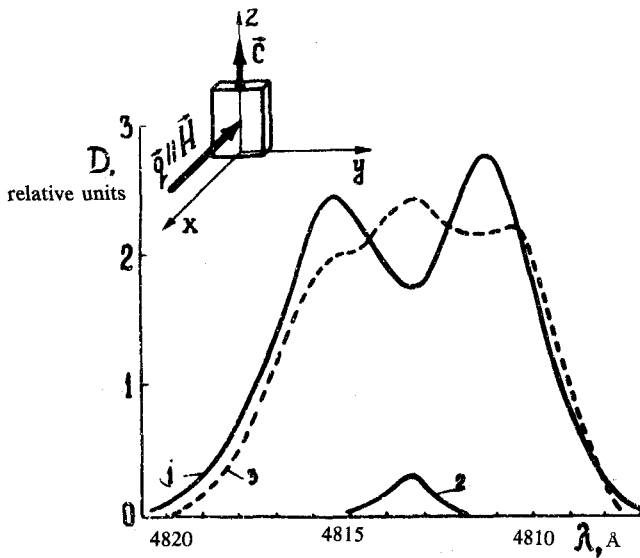


FIG. 1. CdS absorption spectra in the region of the  $A$  exciton,  $n = 2$  with  $H = 0$  ( $E \parallel C$  for curve 1;  $E \parallel C$  for curve 2) and  $H = 31$  kOe ( $E \parallel C$  for curve 3).

Curve 1 in Fig. 1 shows the absorption spectrum of a single crystal of CdS with thickness  $\sim 0.8 \mu\text{m}$  in the region of the  $A$  exciton  $n = 2$ . It was recorded in  $E \parallel C$  polarization on a DFS-12 spectrometer with spectral slit width  $0.05 \text{ \AA}$  at  $4.2 \text{ K}$ . The geometry of the experiment, shown in the insert in Fig. 1, is such that  $C \parallel z, q \parallel x$ . Here  $E$  and  $q$  are the electric and wave vectors of the light wave, and  $C$  is the optical axis of the crystal. It is evident that curve 1 has the same double hump shape as described in Ref. 1. The selection rules in this polarization allow, in addition to transitions the  $S$  state, transitions to the  $P_z$  state as well, which, as already noted, must be two orders of magnitude less intense.<sup>2</sup>

Curve 2 in Fig. 1 represents the absorption in  $E \parallel C$  polarization, where the transition only to the  $P_{xy}$  exciton state is allowed. This spectrum was measured with a thicker specimen ( $\sim 2 \mu\text{m}$ ), and, as is evident, describes the quite narrow and quite intense absorption band. This circumstance confirms experimentally the theoretical assertion<sup>2</sup> that the oscillator strength of transitions to the  $P$  states is really much smaller than to the  $S$  states. Thicker (up to several tens of  $\mu\text{m}$ ) crystals which have been polarized also reveal a transition to the  $P_z$  state forbidden by the selection rules. For specimen thicknesses used by us, the corresponding line was missing.

A comparison of the absorption spectra showed that the position of the dip in curve 1 always coincides with the peak in curve 2 in all crystals studied. This coincidence is apparently not accidental. The observed phenomenon is analogous to that described in Fano's theory,<sup>3</sup> where the interaction of discrete absorption bands with a continuous background is examined. In the case of a complete "antiresonance," i.e., an antiphase excitation, a dip should be observed at the location of the discrete level in

the continuous spectrum. This suggested to us that the double hump form of curve 1 is due to antiresonant interaction of  $S$  and  $P$  exciton states. The present situation is slightly more complicated than Ref. 3, since here the continuous background is formed by the wide absorption band of the  $S$  state, while the narrow band of the  $P$  state forms the discrete level.

The action of the external magnetic field  $H$  on the shape and energy position of the dip in curve 1, within the framework of the assumption made here, must be due to the well-known effect of splitting and shift of the  $P$  states of the  $A$  exciton with  $H \neq 0$ , which was studied in detail in Ref. 4 for  $E \parallel C$  polarization.

Curve 3 in Fig. 1 indicates the absorption spectrum recorded in a magnetic field  $H_x = 31$  kOe and  $H_y = H_z = 0$  for  $E \perp C, q \parallel H$ . The two dips clearly evident in curve 3 occur at the locations of the spectrum where, according to Ref. 4, the  $P_y$  and  $P_z$  states of the  $A$  exciton with  $n=2$  split by the magnetic field should be located. It was established from traces of absorption spectra with different values of the field from 0 to 31 kOe that as the field increases, the main dip is displaced toward short wavelengths, the dip situated in the long-wavelength wing of curve 3 appears for  $H > 20$  kOe, and its amplitude increases as the field increases from  $\sim 20$  to 30 kOe.

Measurements of the magnetic field dependences of the shift of the bottoms of the dips shown in curve 3 are represented by points in Fig. 2. The solid and dashed lines indicate the field dependence of the spectral position of  $P_y$ ,  $P_z$  and  $P_x$  exciton states obtained in Ref. 4. It is evident in Fig. 2 that the main dip behaves just like the  $P_y$  component. The additional dip appears when the  $P_z$  component is enhanced in  $E \parallel C$  polarization due to mixing by the field with the  $P_y$  component.<sup>4</sup> The dip corresponding to the  $P_x$  state is missing. This is probably due to the fact that in the geometry of our experiment ( $q \parallel x$ ), the electric vector of the light wave does not excite this state.

Thus the experimental data obtained indicate convincingly the fact that the position of both dips in the absorption band with  $E \parallel C$  correlate with the position of the

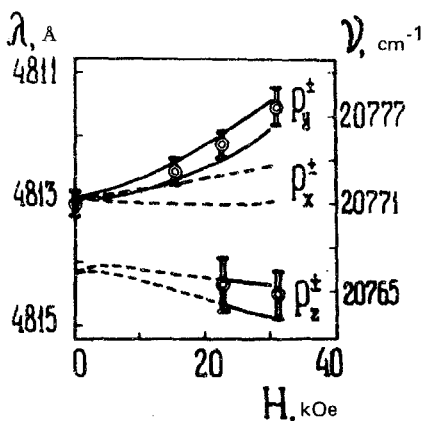


FIG. 2. Effect of a magnetic field on the spectral position of antiresonance dips in the absorption curve of  $A$  exciton,  $n=2$ ,  $E \perp C$  (points) and split  $P$  components of exciton with  $E \parallel C$  according to Ref. 4 (lines).

split  $P$  states of the  $A$  exciton with  $\mathbf{E} \parallel \mathbf{C}$  and confirm our assumptions concerning their antiresonant nature. On this basis, we assume that the doublet form of the absorption band of the  $A$  exciton in the state  $n = 2$  can be interpreted as the appearance of a Fano antiresonance due to the interaction of  $S$  and  $P$  states of the exciton.

This effect, judging from available measured spectra, is a general effect for all absorption bands corresponding to excitation of states with  $n = 2$ .

A nontrivial result, from the point of view of Fano's theory, is the fact that states which are allowed in orthogonal polarizations  $\mathbf{E} \perp \mathbf{C}$  and  $\mathbf{E} \parallel \mathbf{C}$  interact. This can apparently be explained if it is assumed that the perturbation leading to interaction of these states is the wave vector of the moving exciton.<sup>4</sup>

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<sup>1</sup>M. S. Brodin and M. I. Strashnikova, Proceedings of the 9th International Conference on the Physics of Semiconductors, Nauka, Moscow 1969, Vol. 1, p. 489.

<sup>2</sup>R. J. Elliott, Phys. Rev. **108**, 1384 (1957).

<sup>3</sup>U. Fano, Phys. Rev. **124**, 1866 (1961).

<sup>4</sup>J. J. Hopfield and D. G. Thomas, Phys. Rev. **122**, 35 (1961).