Effect of spatial dispersion on the change of the phase of reflected light in CdS and CdSe crystals

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An analysis is presented of the phase change produced when light is reflected from CdS and CdSe, based on the theory of spatial dispersion with allowance for the damping and for the surface layer. The existence of normal waves of optical and exciton type is observed experimentally.

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A number of recent communications have demonstrated the important role of spatial dispersion in the exciton region. Observations of this type were made in investigations of interference patterns in thin crystal plates, [1,2] and in crystal reflection[3,4] and absorption[5] spectra. It is obvious that complete information on the optical properties of substances can be obtained by comprehensive investigations. In particular, in the study of the reflection spectra it is necessary to know the corresponding phase characteristics.

This communication is devoted to an investigation of the spectral dependence of the change of phase when light is reflected from single-crystal cadmium sulfide or selenide. The choice of the object is dictated by the need for using a number of constants to calculate the phase characteristics. Most attention is paid to fully polarized band head lines of the exciton series A. In this case only one component of the polarized light undergoes a phase change that depends on the frequency of the light. Interference patterns were obtained for different light incidence angles in the temperature interval from 4.2 to 77 °K. An analysis of the ellipticity of the reflected light was carried out with the aid of a Babinet compensator, that introduces a phase difference that is linear in the height of the spectrograph slit. The spectrum was recorded photographically. The phase shift was determined from the shift of the interference fringes. The distance between neighboring fringes corresponds to a phase shift $\delta = 2\pi$.

The obtained interference patterns (Fig. 1) are characterized by the following essential peculiarties: a) the phase of the reflected light increases monotonically from the long-wave side, passing through a resonance ω_T; b) in contrast to the classical behavior, the phase varies not up to π, but up to 2π; c) at sufficiently high crystal temperatures, two pictures are observed simultaneously on the interference patterns, one similar to the low-temperature one and decreasing in intensity with rising temperature, while the other demonstrates the region of the anomalous behavior of the phase (Fig. 1b). A similar picture is observed also at low temperatures for high-energy excitonic states.

DISCUSSION OF RESULTS

The observed spectra can be obtained only by resorting to the concepts of spatial dispersion in the exciton region. According to Pekar, [6] the refractive
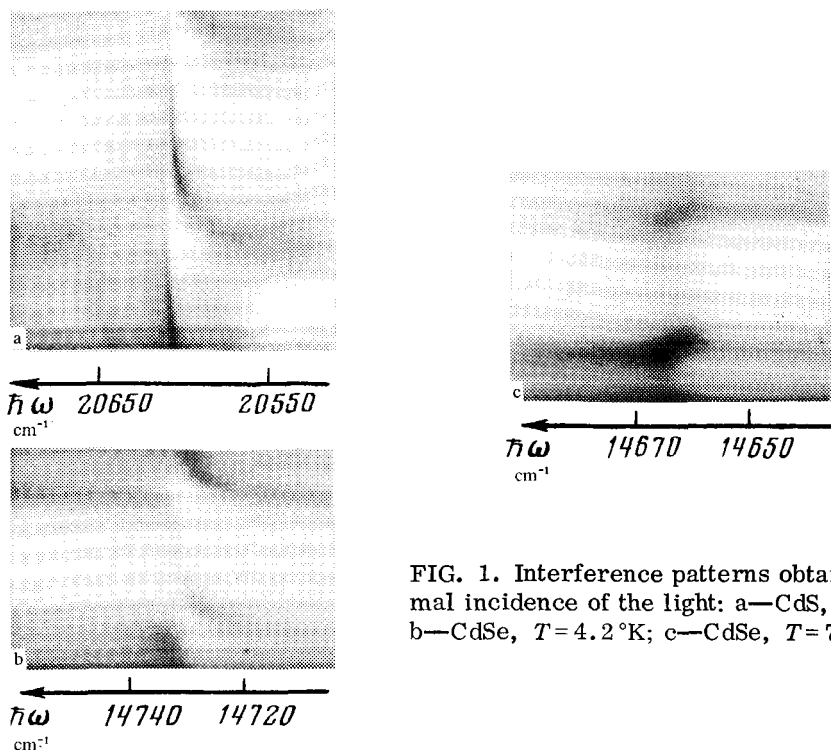
FIG. 1. Interference patterns obtained at normal incidence of the light: a—CdS, $T = 4.2\,°K$; b—CdSe, $T = 4.2\,°K$; c—CdSe, $T = 77\,°K$.

indices can be expressed in the form

$$n_{\pm}^2 = \frac{1}{2}\left\{\left[\epsilon + \frac{2mc^2}{\hbar\omega_T^2}(\omega - \omega_T)\right] \pm \left[\left(\epsilon - \frac{2mc^2}{\hbar\omega_T^2}(\omega - \omega_T)\right)^2 + \frac{8mc^2}{\hbar\omega_T^2}\epsilon\omega_{LT}\right]^{1/2}\right\},$$

(1)

where m is the exciton mass, ϵ is the background dielectric constant, $\hbar\omega_T$ the energy of the transverse exciton, and ω_{LT} is the longitudinal-transverse splitting. In analogy with[7], a theoretical analysis of the change of phase calls for allowance for the relative contributions of waves of type n_+ and n_-

$$\widetilde{n}^* = \frac{n_*}{1 - q} + \frac{n_-}{1 - 1/q}\;;\quad \widetilde{n}^* \equiv n^* + l\kappa^*\;,\quad \text{where}\quad q = \frac{\epsilon - n_+^2}{\epsilon - n_-^2}\;.$$

(2)

Using the generalized Fresnel equation for the reflection of light

$$r = \frac{R}{A} = \frac{\widetilde{n}^* - 1}{\widetilde{n}^* + 1}\;,$$

(3)

where R is the reflected wave and A is the incident wave, we obtain an equation for the phase shift δ of the light upon reflection:

$$tg\,\delta = \frac{2\kappa^*}{n^{*2} + \kappa^{*2} - 1} \quad . \tag{4}$$

Substituting in (4) the values of the exciton constants from[1,5], we obtain the curve shown in Fig. 2a. The maximum value is $\delta = \pi$, contradicting the experimental value 2π. To explain the true behavior of the phase difference in the vicinity of ω_L it is necessary to introduce an exciton-poor surface layer.[8] The change of phase due to reflection, with allowance for the layer, can be estimated from the formula

$$tg\,\delta = \frac{r_{23}(1 - r_{12}^2)\sin 2\beta}{r_{12}(1 + r_{23}^2) + r_{23}(1 + r_{12}^2)\cos 2\beta} \quad , \tag{5}$$

where the subscripts 1, 2, and 3 pertain to the waves in the vacuum, in the layer, and in the crystal; 2β is the phase shift following double passage through the layer. Formula (5) can be generalized to include the case of arbitrary light-incidence angles. The results of the reduction of the experimental data are
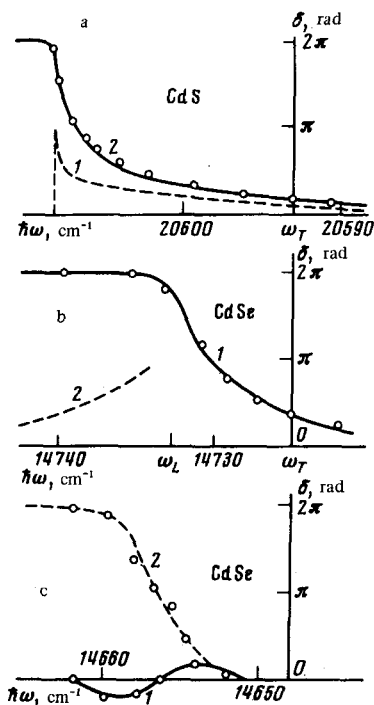

FIG. 2. a—CdS: 1—theoretical $\delta = \delta(\omega)$ curve without allowance for the surface layer, 2—with allowance for a layer of thickness $l = 80$ Å; attenuation $\hbar\Gamma = 0$; b—CdSe: 1—$l = 50$ Å, $h = 0.4$ cm^{-1}; 2—behavior of $\delta = \delta(\omega)$ at an incidence angle $\alpha = 70°'$; c—CdSe: $T = 77$°K. The circles mark the experimental data of Fig. 1.

shown in Fig. 2, and for cadmium selenide it was necessary to introduce in (1) the damping of the opto-exciton wave. The difference between the pictures observed near ω_L for the cadmium sulfide and selenide is due to the different surface qualitities of the crystals, a fact that manifests itself also in the usual reflection spectra.

A very interesting feature is possessed also by the phase interference patterns at incidence angles corresponding to the Brewster angle for pure light waves in the spectral region $\omega > \omega_L$. The phase decreases monotonically in this case from $\sim \pi$ to zero, as shown dashed in Fig. 2b; this indicates that only waves of the opto-exciton type take part in the reflection. Away from the Brewster angle, in either direction, the interference patterns in this spectral region have the usual form of horizontal fringes.

The interference patterns obtained at relatively high crystal temperatures can apparently be explained on the basis of the theory of Davydov and Myasnikov, [9] as a result of the manifestation of waves of the optical and of the excitonic type. At crystal temperatures above 77 °K, only one plot of the phase is observed (Fig. 2c, curve 1). For high-energy excitons, this phenomenon is observed at lower crystal temperatures.

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