Effect of surface potential on the phase of an exciton wave

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The method developed by us [Phys. Stat. Sol. (b) **72**, 161 (1975); Soviet Physics-Solid State **19**, No. 8 (1977)] for the investigation of exciton interference spectra is used to study the phase of an exciton wave. A strong dependence of the phase on the wave vector is observed for the first time. This behavior of the phase is due to the influence of the surface potential.

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The problem of the influence of a surface potential on the exciton states of crystals has long attracted the attention of researchers.[3–5] Unfortunately, however, the available experimental data on the shape of the excitonic light-reflection bands have so far not been analyzed with use of the realistic potential models proposed in[3,4]. This is attributed both to the difficulty of such an analysis and to the fact that the singularities of the reflection lines can be qualitatively explained by using a very simple potential, that of a hard wall.[5] The problem of the exciton surface potential has therefore remained open. A study of the dependence of the exciton-wave phase shift δ on the length of the wave vector k when an exciton is reflected from the surface of a crystal has uncovered new possibilities of finding the surface potential. Such an investigation is made possible by the appearance, in the spectra of thin crystals, of interference between supplementary waves[1] or of size-effect quantization of the excitons.[2] We report here on the behavior of $\delta(k)$ at exciton kinetic energies in the interval ~ 0.2—0.7 of its binding energy.

We investigated the reflection and transmission spectra of thin high-grade CdSe crystals grown from the gas phase. The region of the exciton A was investigated at normal incidence of the light. The sample temperature was 1.6 K. The spectra were recorded with an instrument having a dispersion 1.9 Å/mm.

Figure 1 shows the reflection spectra of three crystals (a—c), with close albeit somewhat different thicknesses (~ 0.8 μm), in the energy interval indicated above. The structure due to the interference of the supplementary waves[1] (small oscillations) is clearly seen against the background of the Fabry-Perot interference of a light-like wave (large-scale oscillations). We note two important features of the spectra. 1) In the regions $\lambda \approx 6772$ Å and $\lambda \approx 6758$—6755 Å there is no supplementary structure, and the spectral positions of these "empty" sections almost coincide for the different samples and are independent of the positions of the interference singularities. 2) Starting with $\lambda \approx 6778$ Å, the period of the supplementary oscillations increases somewhat more slowly than predicted by a theory[1,6] that disregards the phase of the exciton wave (curve d).

Both features can be explained from a single point of view, namely, they can

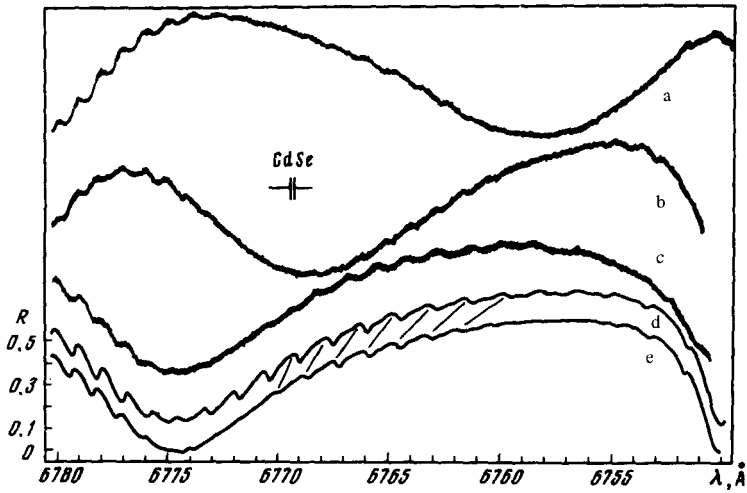

FIG. 1. Reflection spectra of CdSe in the region between A excitons, $n=1$ and $n=2$: a–c) experiment at $T=1.6$ K and $E \perp c$; d) theoretical calculation without allowance for $\delta(\omega)$; e) the same with allowance for $\delta(\omega)$.

be attributed to a definite $\delta(k)$ dependence. Curve (e) in Fig. 1 was obtained as a result of taking this dependence into account within the framework of a phenomenological theory[6,7] with supplementary boundary conditions of the form[1)]

$$\left(P(z,\omega) + \gamma(\omega)\frac{c}{\omega}P'(z,\omega)\right)\Big|_{z=\pm a} = 0, \tag{1}$$

where P is the exciton contribution to the polarization, $z=\pm a$ corresponds to the faces of the crystal, and $\gamma(\omega)$ is a phenomenological function of the frequency and is connected with $\delta(\omega)$ by the relation

$$\gamma(\omega) = \frac{\omega}{ck}\operatorname{tg}\delta(\omega). \tag{2}$$

If $\delta=\pm\pi N$ $(N=0,1,2,\cdots)$, then $\gamma=0$, and the strongest supplementary oscillations[1] are observed in this case. If $\delta=\pm\pi(N+\frac{1}{2})$, then $\gamma=\pm\infty$ and there are no oscillations.[1] Using this fact, we can choose $\delta(\omega)$ such that the theoretical spectrum (curve e) has "empty" sections in the very places where they are experimentally observed. It is obvious that δ should change in the considered wavelength interval by $\sim(3/2)\pi$. It can furthermore be stated that δ should increase with increasing ω, for only in this case does the spectrum become compressed. To plot curve e, we used the approximation

$$\delta/_k = (2.67(\omega-\omega_0)-53.9)\text{Å}, \tag{3}$$

where ω_0 is the resonant frequency (in cm^{-1}). The remaining parameters needed for the calculation are close to those cited earlier,[1,2] but for the discussed part of the spectrum it was essential to take into account the influence of the higher-lying state on the dispersion of light-like waves.

Thus, an investigation of the singularities of the interference spectra of

thin crystals permits assessment of the phase of the exciton waves in the crystal and consequently of the manifestation of the surface potential.

[1)]A more rigorous theory will be developed in a detailed article.

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