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DIRECT OBSERVATION OF HYPERBOLIC EXCITONS

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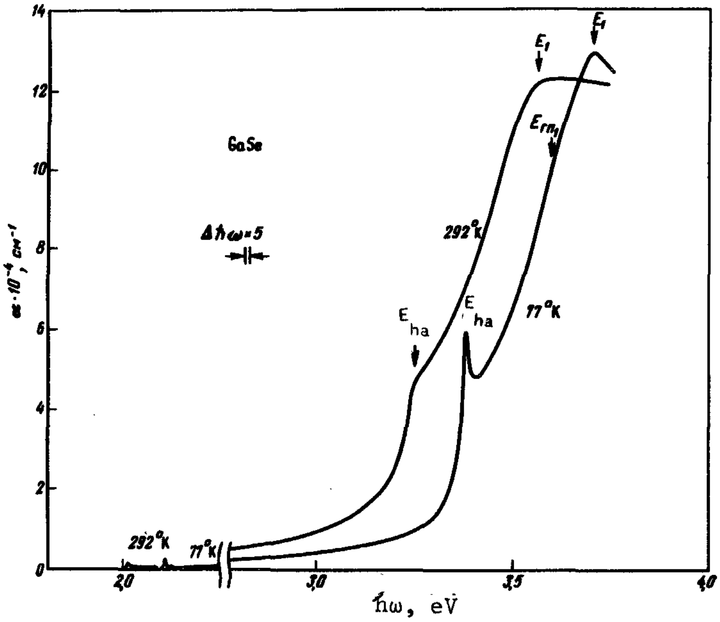
Phillips [1] proposed that electrons and holes can be bound into excitons not only at energies close to the energies of the absolute extrema, but also near the saddle points M_1 and M_2. Phillips called such excitons saddle-point excitons (hyperbolic excitons). He assumed that the existence of hyperbolic excitons can lead to the appearance of a distinct structure neat the points M_1 in the optical spectra. Velicky and Sak [2] and Kane [3] found that the Coulomb effects at the saddle critical points M_1 should become clearly pronounced in the optical spectra. Duke and Segall [4], on the other hand, theoretically refuted the existence of hyperbolic excitons. By measuring the transmission of thin samples of indium antimonide and cadmium telluride at low temperatures, Cardona and Harbeke [5] observed a structure which they ascribed to the exciton nature of the $L_3 \rightarrow L_1$ transition. Comparing the absorption coefficient above the energies of the first exciton peak with Elliott's formula [6] for allowed direct transitions, they obtained for the exciton binding energy values of 0.04 and 0.035 eV for InSb and CdTe, respectively. However, as indicated by Marple and Ehrenreich [7], another explanation of the structure is also possible. Shaklee, Rowe, and Cardona [8] confirmed, on the basis of the differential reflection spectrum of InSb near the critical point M_1, the existence of hyperbolic excitons in this material. Obviously the problem of the possible existence of bound states near saddle points and their manifestation in optical spectra is not yet ultimately solved.



Absorption spectra of GaSe above the edge of the fundamental absorption.

In this paper we present a clearcut proof favoring the existence of hyperbolic excitons with GaSe as an example.

GaSe is an hexagonal layered structure. Its absorption spectrum near the intrinsic absorption edge was investigated in a number of papers [9].

This material is particularly favorable for the observation of hyperbolic excitons, since the absorption in it above the fundamental edge is small and the layered structure leads to a clearcut display of the critical points.

To measure the transmission of GaSe in the region of larger absorption, we used thin samples (1 μ and 0.55 μ), split off from GaSe crystals. The sample thicknesses were determined from an analysis of the interference pattern of the transmission spectra. The figure shows the absorption spectra of GaSe at room temperature and at 77°K. At 77°K one sees clearly an absorption peak at (3.378 ± 0.002) eV and a saddle point M_1 at (3.696 ± 0.002) eV. One can note a complete analogy between the experimental curve and the theoretical curve given by Phillips [1] in the case of hyperbolic excitons. The peak depends strongly on the temperature. At 292°K it vanishes and there remains a steep rise on the absorption curve. Such a strong temperature dependence is one of the proofs of the fact that the peak is connected with excitons. The hyperbolic character of these excitons follows from the fact that it is formed near a critical point of the M_1 type. The latter is confirmed by the course of the absorption curve obtained by us and by the dependence of the reflection coefficient in this region of the energy, and also agrees with the theoretically calculated band structure for GaSe [9]. The exciton binding energy is equal to 0.318 ± 0.006 eV (77°K).

From the temperature shift of the spectra we determine

$$\Delta E_{\mathrm{ha}}/\Delta T = -5{,}9\cdot 10^{-4}\ \mathrm{ev/deg} \quad \text{and} \quad \Delta E_1/\Delta T = -5{,}8\cdot 10^{-4}\ \mathrm{eV/deg},$$

which agrees with the result obtained by Akhundov et al. [10] from the reflection spectrum.

Compared with the half-width of the line of the principal exciton peak at the fundamental absorption edge ($\Gamma \simeq 0.01$ eV at 77°K), the half-width of the exciton line at M_1 is larger ($\Gamma \simeq 0.02$ eV).

Thus, the lifetime of the hyperbolic excitons is approximately half as large, and this may be connected with their dynamic instability.

It is interesting to note that the growth rate α decreases noticeably near $h\nu = 3.6$ eV at 77°K. It is natural to assume that it is connected with the excited state E_{hai} of the hyperbolic excitons.

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