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Polariton Effects in the Exciton Absorption of GaSe

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An anomalous temperature and thickness dependence in the optical absorption of the lowest direct exciton of GaSe is reported. The effect, which is interpreted as a result of the competition between exciton-photon interaction and exciton dissipation mechanisms (phonon and surface scattering), provides strong experimental evidence for the polariton description of the exciton absorption process in semiconductors.

Hopfield has shown that the creation of an exciton by a photon in a crystal does not in itself lead to any absorption of light, unless the exciton undergoes a scattering process.[1] A very direct consequence of this result, which, however, has not received much attention so far, is the possibility of a substantial reduction of the exciton absorption in comparison with the corresponding value of the semiclassical theory.[2] In fact, this has to be expected when the "polariton" frequency f (i.e., the rate of exchange of energy back and forth between the excitonlike and photonlike states of the polariton excitation) is fast compared with the frequency γ which characterizes phenomenologically the exciton (polariton) scattering processes. In this Letter we present and discuss some data on the optical absorption of the lowest direct exciton in GaSe, which provide strong experimental evidence for the above effect.

GaSe is an indirect gap semiconductor: There is, however, a direct transition only ~50 meV higher, to which the lowest direct exciton is associated.[3] Since when the electric field $\vec{E}$ of the radiation is perpendicular to the $\vec{c}$ axis both the direct and indirect transitions are allowed only if the spin-orbit interaction is taken into account, this exciton has a relatively low absorption strength, and is therefore well suited for experimental investigation. We have performed a detailed study of the temperature dependence of its optical absorption, on several samples of vari-

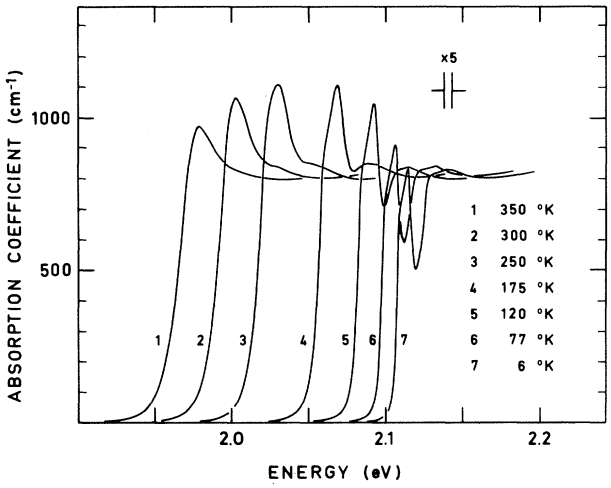

FIG. 1. Absorption spectra of a sample 97 μm thick, at the temperatures indicated. The spectral resolution is also shown.

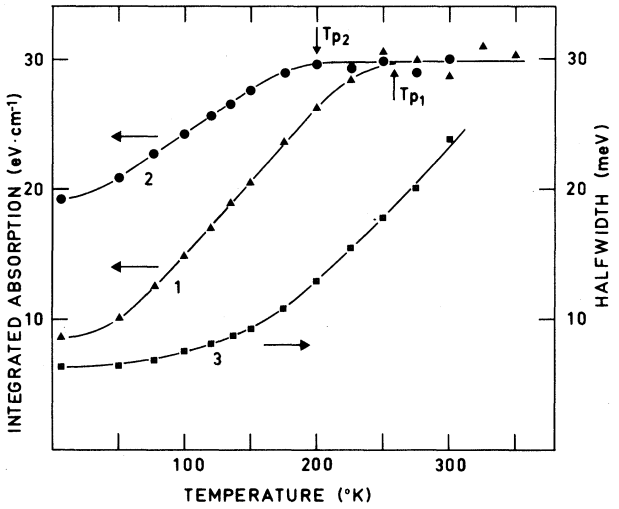

FIG. 2. Temperature dependence of the integrated absorption for samples of thickness 97 μm (curve 1) and 26 μm (curve 2). Temperature dependence of the experimental half width for the sample 26 μm thick (curve 3).

ous thickness, ranging from 26 to 150 μm. The transmission spectra were taken at normal incidence ($\vec{E} \perp \vec{c}$) on strain-free mounted samples. Particular care has been taken in increasing the sensitivity of the optical apparatus by screening stray light and using phase-sensitive-detection techniques: With a series of calibrated filters we have checked that reliable data, without distortion or saturation effects, can be taken up to an optical density of ~ 6. The transmission data have finally been corrected for reflection losses according to standard procedures to get the absorption coefficient α.

The data we report refer to a set of crystals, all cleaved from the same ingot, having p-type carriers with concentration $n_p = 2.1 \times 10^{14}$ cm^{-3} at 300°K. They are, however, representative of results obtained on many series of samples grown under different conditions. The absorption spectrum of a sample 97 μm thick, at several temperatures T between 6 and 350°K, is reported in Fig. 1. The striking feature of this figure is the strong reduction of the exciton absorption at low temperatures; correspondingly (Fig. 2, curve 1), the integrated absorption A (i.e., the area under the exciton curve) is also strongly reduced when the temperature is lowered below the value T_{p1}, whereas it remains constant (within the experimental error) at the value $A = 29.8$ eV cm^{-1} for $T > T_{p1}$. The decrease of A below T_{p1} is in sharp contrast with the classical prediction according to which A, being related to the oscillator strength of the exciton transition,[2,4] is independent of T. Qualitatively similar results have been obtained for all the samples we have investigated. Moreover, from the quantitative viewpoint, a thickness dependence of A is also observed. In particular, whereas at high temperatures A is the same for all the samples, both the reduction of A and the temperature T_p decrease as the thickness d of the samples decreases. As an example, curve 2 of Fig. 2 shows A versus T for a sample 26 μm thick. Also the thickness dependence of α is dramatically shown in Fig. 3, where the absorption spectrum at 77°K for samples of thick-

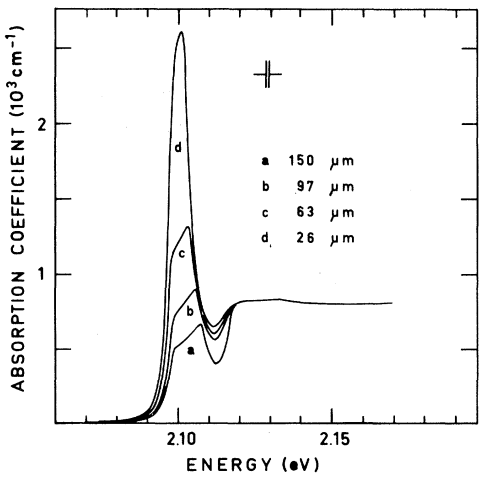

FIG. 3. Absorption spectra at 77°K for the samples with the thickness indicated. The spectral resolution is also shown.

ness 26, 63, 97, and 150 μm is reported. This result is, of course, in sharp contrast with the classical theory of the optical absorption, where α is, by definition, independent of d. It is important to notice that the reduction of α concerns *only* the exciton curve; the "continuum" above the exciton region is, in fact, independent of both temperature and thickness (Figs. 1 and 3), in perfect agreement with the classical theory.

The anomalous behavior presented above cannot be explained by invoking a contribution to the exciton absorption from the underlying indirect transition. In fact, whereas this mechanism could in principle explain a temperature reduction of A, it cannot lead to a thickness dependence. Moreover, it can be ruled out on the basis of the orders of magnitude involved. The contribution to the optical absorption in the exciton region from the indirect transition can indeed be estimated, up to 300°K, to be not higher than 6 cm^{-1}, even in the allowed configuration $\vec{E} \| \vec{c}$.[5] It is expected to be considerably lower for $\vec{E} \perp \vec{c}$, and anyway it is unable to explain, for example, the decrease in α of nearly 300 cm^{-1}, passing from 175 to 6°K, for the sample of Fig. 1. Further evidence against the relevance of the indirect transitions for the observed effects comes from the lack of any T dependence of both the absorption coefficient α in the "continuum" and the integrated absorption A for temperatures above T_p.

Exciton luminescence could at first explain the anomalies in the results, since it can cause an apparent decrease in α, with the correct qualitative trend for both the T and d dependences. Apart from the quantitative difficulties of this explanation, however, we have excluded it on an experimental ground: In fact, by monitoring a possible emission simultaneously with the absorption measurements at a small angle from the transmission direction, we have detected no signal, even with sensitivities higher than the one used for the transmitted beam.

A unified explanation for both the temperature and thickness dependence of the exciton absorption reported in Figs. 1–3 is directly provided by the polariton character of the exciton "absorption" process, and by the fact that any *real* exciton absorption of light is indeed due to exciton (polariton) scattering processes.[1] Since a polariton is scattered only when in an excitonlike state, it is evident that when $f > \gamma$ the quantum oscillations of the polariton energy between excitonlike and photonlike states are too fast for the dissipation mechanisms to act with efficiency; the light can therefore pass through the crystal with reduced attenuation. On the contrary, when $f < \gamma$, the energy can be fully dissipated when in the excitonlike state before it returns to the photonlike state, and the proportionality of the semiclassical theory between A and the oscillator strength F of the exciton transition is thus full recovered. The dissipation frequency γ is temperature and thickness dependent. The T dependence is brought about by the exciton-phonon scattering processes (for example, in the high-temperature limit, exciton–acoustic-phonon scattering depends linearly on T)[6]; the d dependence is due to surface scattering processes, which in turn depend on the surface-to-volume ratio of the samples.[1] Since the polariton frequency f is practically constant for a given crystal, by varying T and by using samples of different d, one can vary $\gamma(d,T)$ and, possibly, pass from the classical region $\gamma > f$ to the "polariton" region $\gamma < f$, thus seeing the absorption vary from the classical behavior towards the region of polariton-induced transparency.

In the case of GaSe, f can be estimated from the approximate relation[1]

$$f \simeq \omega_0 (\pi\beta)^{1/2}, \tag{1}$$

where ω_0 is the exciton frequency and β is related to the plasma frequency ω_p and the oscillator strength F by

$$\beta = (\omega_p^2/\omega_0^2)(F/4\pi\epsilon). \tag{2}$$

Here ϵ is the dielectric constant not including the contribution from the exciton resonance. Substituting (2) in (1), one gets

$$f \simeq \omega_p (F/4\epsilon)^{1/2}. \tag{3}$$

Using formula (9.15) of Ref. 2, with the high-temperature value of A, and assuming $m_e^* = 0.17 m_e$[7] and $\epsilon = 7.95$, one gets finally $f = 2.12 \times 10^{13}$ sec^{-1}. The frequency γ is usually evaluated from the experimental half width Γ of the exciton curve. In the presence of the polariton effects, however, this procedure can be strongly deceptive, as suggested by Fig. 3. One can therefore make use of it only in the classical region above T_p. Evaluating γ at T_{p2} from Fig. 2, curve 3, we find $\gamma = 1.98 \times 10^{13}$ sec^{-1}: At this temperature the condition $\gamma \lesssim f$ for the onset of the polariton effects begins to be satisfied, in confirmation of the proposed explanation.

An analysis of light propagation in crystals, taking the exciton-photon interaction into full account, has recently been given by Davydov and

Serikov.[8] This work provides an approximate formula for the absorption coefficient in terms of f and γ, from which we get the integrated absorption (in eV cm^{-1}):

$$A = (\pi h f^2 \gamma / 2c)(\gamma^2 + f^2/4)^{-1/2}. \quad (4)$$

As can be easily seen, in the limit $f < \gamma$, one gets from (4) the semiclassical result that A is proportional to f^2 and therefore, from (3), to F, whereas, for $f > \gamma$

$$A \simeq \pi h f \gamma(d, T)/c, \quad (5)$$

which contains a temperature and thickness dependence of A. The quantitative application of formula (4) to our results will be considered elsewhere.[9]

Further confirmation of the polariton nature of the reported effects on the exciton curve come from their absence on the "continuum" above the exciton region[1] and from the exciton line shape itself whose peculiar asymmetry can be related to the polariton dispersion curve.[9]

In our opinion the phenomena we have observed in GaSe would have to be rather general, and ought to be observed whenever an exciton peak is sufficiently resolved from the absorption continuum and its line shape and integrated absorption are carefully investigated. Indeed, a temperature dependence of A has recently been seen also in CdS[10] and Cu_2O[11] and some scattered evidence exists even for GaSe,[12] though it has always been overlooked; on the other hand, the thickness dependence due to polariton effects is here reported for the first time. (We note that this dependence is not oscillatory, and is observed at temperatures too high to be ascribed to spatial dispersion effects.[13])

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Supernova Explosions, the New Leptons, and Right-Handed Neutrinos

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New leptons and right-handed neutrinos, created by weak neutral currents in stellar interiors, will do much to produce a supernova explosion. I study their production and absorption mechanisms, and note that angular momentum carried by neutrinos might affect the rotation of the subsequent neutron star.

The theory that neutrinos initiate supernova explosions has recently been vigorously investigated after Freedman[1] discussed coherent scattering via the weak neutral current. Preliminary calculations by Wilson[2] suggested that the neutrino pressure, within the Weinberg-Salam model,[3] was not large enough to blow off the envelope. More extensive calculations[4] continue to show