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THE EFFECT OF SPATIAL DISPERSION ON THE ATTENUATION OF EXCITON POLARITONS IN SEMICONDUCTING FILMS

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We calculate the effective light absorption coefficient $K(\omega)$ and the frequency-integrated absorption $I = \int K(\omega)\, d\omega$ caused by spatially dispersive excitons in a semiconductor film. A sharp decrease of I is found for small masses and long lifetimes of the exciton. Oscillations of I as a function of thickness are also reported.

1. INTRODUCTION

IT IS WELL KNOWN that exciton polaritons, i.e. excitations of mixed exciton–photon character, are generated when light interacts with excitons in semiconductors. According to Pekar [1] and Hopfield [2], conversion of photons into polaritons (a process taking place at the crystal surface) should not in itself be regarded as a "photon absorption" event. An actual loss of energy flowing with the beam only occurs when the polariton is scattered away from the beam. This can happen due to agents such as phonons or imperfections that cause the polariton to change its state within a mean lifetime τ. Each scattering event attenuates the polariton beam which then converts back into an attenuated light beam at the outgoing crystal surface. Pekar [1] and Hopfield [2] have argued that the frequency-integrated light attenuation must vanish in the zero-scattering limit, $T = 1/\tau \to 0$.

From a different point of view, Loudon [3] has investigated the propagation of electromagnetic energy through a classical absorbing dielectric with a single resonant frequency ω_T. The dielectric function is

$$\epsilon(\omega) = \epsilon_0 + \frac{4\pi\beta\omega_T^2}{\omega_T^2 - \omega^2 - i\omega\Gamma} \tag{1}$$

where ϵ_0 is a (real) background dielectric constant, $4\pi\beta$ is the oscillator strength and $T = 1/\tau$ is again the inverse oscillator lifetime. The energy velocity V_E, defined as the ratio between Poynting vector and energy density, tends to zero linearly as Γ is let to zero for all frequencies in the "stop band", $\omega_T \leqslant \omega \leqslant \omega_L = \omega_T(1 + 4\pi\beta/\epsilon_0)^{1/2}$. It follows that the light absorption coefficient, given by $K = V_E\tau$, does not vanish in this limit. In particular, the frequency-integrated absorption $I = \int K(\omega)\, d\omega$ remains constant and finite as $T \to 0$ and equals

$$I_0 = 2\pi^2\beta\omega_T^2/c\epsilon_0^{1/2}. \tag{2}$$

The apparent contradiction between the two points of view summarized above is the main concern of this note. We show by explicit calculation that when a finite exciton mass M is allowed for, that is when the classical dielectric function equation (1) is replaced by the "spatially dispersive" form

$$\epsilon(\omega, q) = \epsilon_0 + \frac{4\pi\beta\omega_T^2}{\omega_T^2 + \dfrac{\hbar\omega_T q^2}{M} - \omega^2 - i\omega\Gamma} \tag{3}$$

then a properly defined integrated absorption coefficient does vanish as $\Gamma \to 0$.

Technically, there is some ambiguity in the definition of an absorption coefficient when the exciton mass is finite as in (3) because of the well-known partition of the incident energy into two modes [4, 5], each

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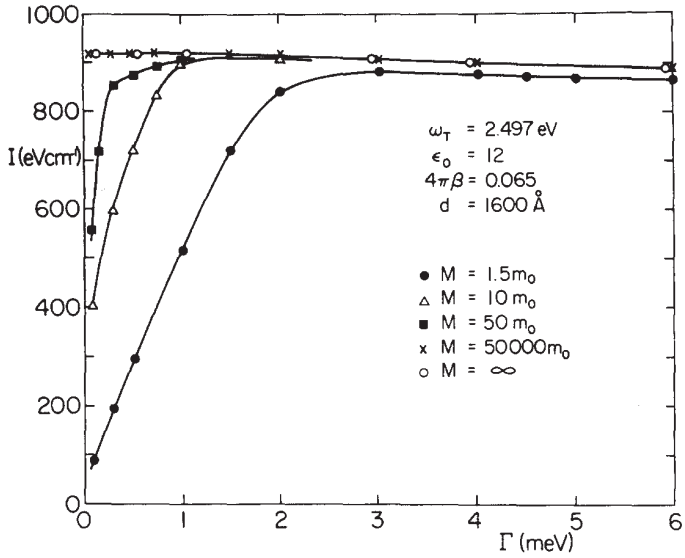

Fig. 1. Calculated integrated effective absorption coefficient of an exciton polariton in a film as a function of damping. The parameters are arbitrarily chosen to model the 1*s* exciton of PbI_2. The slight deviations of all curves from the classical value as well as from one another for large Γ are an artefact due to our finite integration domain.

with a different spatial damping. We have avoided the difficulty by defining an effective absorption coefficient K [6] in terms of observable quantities, the reflectivity R and the transmission T in a film geometry. The relevant formulae for R and T are standard [1], and many calculations illustrating their features as due to spatial dispersion are available for this geometry [6–9]. We want to emphasize that our attention here is focussed on the behaviour of light attenuation with Γ while we wish to keep everything else (choice of additional boundary condition, detailed form of damping process, etc.) as simple and unsophisticated as possible.

2. CALCULATIONS AND RESULTS

We consider a semiconductor film of thickness d, described by the dielectric function (3). Excitons have mass M and damping

$$\Gamma(q) = \Gamma + cq^2 \tag{4}$$

where cq^2 has the form required by, e.g. acoustic phonon scattering [10]. We will, however, usually present results for constant Γ ($c = 0$) since they are (except for lineshape of $K(\omega)$ [6]) very similar to those for $c \neq 0$. The normal incidence reflectivity R and transmission T of the film are calculated with Pekar's method [1] which relates the incident, reflected and transmitted fields E_0, E_R, E_T, respectively, to the fields of the two polariton modes inside the film, namely E_1, E_2 (propagating forward) and E_1', E_2' (propagating backward). The additional boundary condition (*a.b.c*) chosen for our purposes is Pekar's [4] requirement that the exciton polarization should vanish at both surfaces. We do not concern ourselves here with the many intricacies of the *a.b.c* problem [11]; Pekar's condition is simple, it does conserve energy [12] and it has been shown to work well enough in many practical cases [5, 13, 14].

The resulting $R(\omega, d)$ and $T(\omega, d)$ are similar to those reported by several authors [7–9] and will not be reproduced here. We then proceed to fit at each ω the calculated R and T values with R' and T' obtained for a *classical* (non-spatially dispersive) film [15] of the same thickness d. The two parameters of the best fit – which is very non-linear and is therefore carried out numerically [16] – are the real and imaginary part of an effective refractive index $\tilde{n} = n + ik$. We finally obtain the "effective absorption coefficient" [6] of the semiconductor in the film as $K(\omega, d) = 2\omega k/c$. Thus K is operationally defined as the absorption coefficient of a film of classical material producing the same optical response as that of the actual spatially dispersive film. The reason why K so defined now depends on the thickness d – whereas it is of course thickness independent in a truly classical case – is that the two polariton fields E_1, E_2 get attenuated with distance very differently from one another, whereby the total energy is not a simple exponential with distance (except when the thickness is much larger than the decay length of one of the two modes). The ω-dependence of the effective absorption coefficient exhibits a characteristic lineshape. Typical examples are those of Fig. 1 in [6]. The classically sharp asymmetric peak between ω_T and ω_L becomes "truncated" when M is decreased and there is a strong

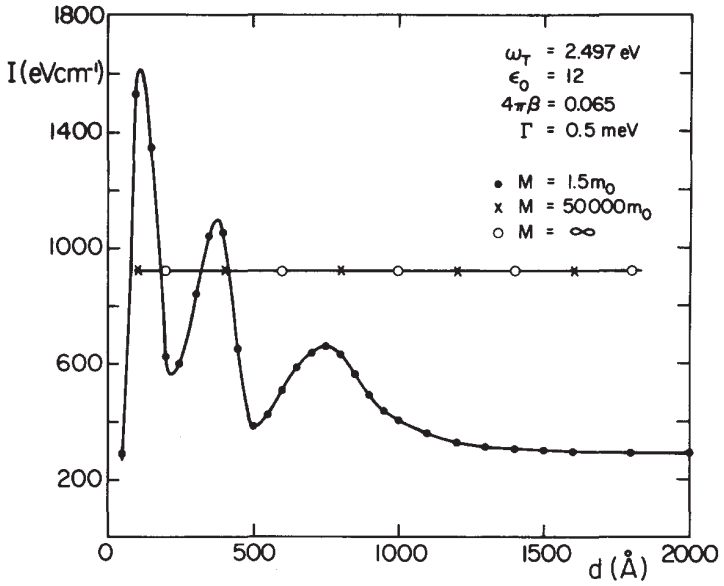

Fig. 2. Thickness dependence of the integrated effective absorption coefficient. The oscillations are attributable to interference between the two polariton modes.

reduction of absorption at each frequency in the stop-band.

This reduction is well reflected in the integrated absorption $I(d) = \int d\omega\, K(\omega, d)$ of Fig. 1. For any given mass one can identify a (broadly defined) critical value of Γ below which I drops from a value very close to the classical limit (2) down toward zero. However, we find that the critical value of damping is very mass-dependent. Our calculations show that it decreases steadily when the mass is increased, as illustrated by Fig. 1. (We attribute the absence of a drop for $M = 5 \times 10^4 m_e$ to our inability, for numerical reasons, to go below $\Gamma =$ 0.1 meV.) This behaviour strongly suggests that $I_\infty = I(M = \infty)$ eventually becomes a non-analytic function of Γ, i.e. $I_\infty(\Gamma \to 0) = I_0$ in agreement with Loudon [3] and only for Γ identically zero does I_0 ever vanish.

The constancy of I when $\Gamma \to 0$ found by Loudon [3] can then be ascribed to the infinite mass assumption and the consequent lack of polariton propagation.

For a finite mass, the occurrence of transparency for $\Gamma \to 0$ confirms Pekar's and Hopfield's idea that undamped polaritons do not cause optical absorption. This situation is reminiscent of a one dimensional version of the well-known optical theorem of scattering theory [17]: the total scattering cross-section and the imaginary part of the forward scattering amplitude are proportional to one another. In our case, the total cross-section for polaritons to scatter into any other state is proportional to Γ while the absorptive forward amplitude is related to K since the beam propagates across the film like $e^{(iK)iz}$.

We have attempted to make contact with an earlier treatment by Davydov and Serikov [18]. Starting with a microscopic approach, which deals with both the photon field and the exciton–phonon field quantum-mechanically, the authors also arrived at the conclusion that a critical value of damping exists below which the absorption drops. However, their critical value is set by the mode-mode gap at $q = \omega_T c^{-1} \epsilon_0^{1/2}$, roughly given by

$$f \cong \omega_T (4\pi\beta/\epsilon_0)^{1/2}. \tag{5}$$

With the parameters adopted for Fig. 1 this is about 0.18 eV, i.e. two orders of magnitude larger than our numerical results for Γ_{critical}, given by the knee of I vs Γ in Fig. 1. Furthermore, the result (5) is mass-independent while ours is very critically dependent on M. At present, the reasons for this disagreement are not clear to us. Phenomenological and microscopic approaches should, in principle, agree, particularly since quantum effects (e.g. stimulated emission) are not expected to be important in this case. (The main difference between atoms and free excitons is in fact that inversion in one well-defined quantum state is much harder to achieve for the latter.) We can only point out that a critical damping as high as (5) should imply that practically all known optically allowed excitons are narrow enough to be well inside the anomalous regime which does not seem to be the case.

It is also of interest to examine the dependence of the integrated effective absorption I upon the film thickness d. We see from Fig. 2 that while I is low and constant for large enough d, it generally increases and

picks up an oscillating behaviour as the thickness decreases. The oscillations, clearly attributable to interference of the two polariton modes in the film, cease when the thickness exceeds the ω-averaged decay length of at least one of the two modes. As Fig. 2 shows, this is of order 1000 Å in the present case.

3. POSSIBILITY OF EXPERIMENTAL OBSERVATION

Our formulation in terms of an effective absorption coefficient $K(\omega, d)$ and integrated absorption $I(d)$ has the advantage that it can be compared directly with experiment. In fact, most authors extract the optical constants of their films using the classical formulae [15] so that their reported absorption coefficient is directly comparable with our results.

The strong damping-dependence (that is, in practice, temperature dependence) of absorption and integrated absorption demonstrated by Fig. 1, as well as the thickness dependence of Fig. 2 should be observable in all cases where Γ falls below its critical value at low T. The present numerical investigation has been arbitrarily carried out with parameters $\omega_T = 2.497$ eV, $\epsilon_0 = 12$, $4\pi\beta = 0.065$ and $M = 1.5\, m_0$ modelling the 1s exciton of PbI_2 [14]. They were obtained by fitting normal incidence reflectivity spectra of cleaved PbI_2 crystals to reflectivity curves calculated from equation (3) and using Pekar's boundary condition. Oblique incidence reflectivity lineshapes for p-polarized polarization under large angles of incidence ϕ turn out to have a sharp reflectivity minimum (see, e.g. Fig. 8 of [14]) which is very sensitive to the value of Γ and rather insensitive to the choice of M. We were able to derive the temperature dependence of Γ by such measurements and appropriate fitting procedures for $\phi = 78°$ as a function of temperature. It turns out that the damping increases from $\Gamma = 0.5$ meV at 4.2 K to about 2 meV at 60 K. From Fig. 1 we therefore expect that the integrated absorption of a PbI_2 film of appropriate thickness should remain constant and classical down to about 60 K and then drop reaching one third of the classical value at 4.2 K. The experimental problem for the verification of this prediction consists mainly in the preparation of thin crystalline samples with Γ-values comparable to those obtained in single crystals of a fairly high degree of perfection. Other materials such as CdS [13] have lower M-values and also smaller low-temperature Γ-values than PbI_2 and indeed qualitative indications of a behaviour of the integrated absorption as shown in Fig. 1 have been reported for CdS [19] and also for Cu_2O [20].

4. CONCLUSIONS

We have presented here a calculation of effective light absorption coefficients and of integrated absorption of a semiconductor film which explicitly exhibits the behaviour to be expected when spatial dispersion of the exciton is important. Our main result is contained in Fig. 1 which shows how the integrated absorption drops when the exciton lifetime exceeds a critical value. This critical damping in turn decreases with the mass in a way which connects well with Loudon's classical calculation. Our numerical values for the critical damping, however, are much smaller than those suggested by Davydov and Serikov. An experimental test of our results, including an oscillatory thickness-dependence of the integrated absorption, is in principle possible in PbI_2 as well as in other materials which are presently being considered. Finally, the dependence of light attenuation on mass and damping described here can also be of importance in other phenomena such as photoconductivity, photoluminescence and resonant light scattering.

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