# patial Dispersion Effects in Resonant Polariton Scattering. II. Resonant Brilloui Scattering\*

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Inelastic scattering of light near an exciton resonance is considered, using a polari scription of the scattering process. We obtain the following results: for incident frequency approaching the resonance from below, the Brillouin shift increases; very near and above approacning the resonance from below, the Brillouin shift increase:<br>resonance, extra lines are predicted with intensities comparable to form an octet instead of the familiar Brillouin doublet. Numerical calculations of the Brillouin scattering for CdS are given: The shifts and extra lines seem to be observable.

## I. INTRODUCTION

The eigenmodes of a system consisting of a crystal and a radiation field have been denoted polaritons. For a system consisting of a semi-infinite crystal (for  $z > 0$ ) and a vacuum ( $z < 0$ ), a polariton consists of the sum of incident plus reflected light field (for  $z < 0$ ), plus polarization waves (for  $z > 0$ ). The relative amplitudes of the constituents of this entire mode are determined by the asymptotic conditions (at  $z = -\infty$ ), and by the full set of boundary conditions [Maxwell plus additional boundary conditions (ABC)].<sup>1</sup> Hence a complete description of an elastic or inelastic scattering process involves calculation of a transition from one such polariton to another.

The dispersion curves inside the infinite or semi-infinite' crystal for the case of an exciton in dispersion) are well known and are shown schemat a 1s internal state with a finite total mass (spatial ically in Fig. 1. To describe inelastic scattering of polaritons by photons, we must satisfy the kinematic requirements of conservation of polariton pseudomomentum inside the crystal

$$
\vec{k} - \vec{k}' = \vec{q}
$$

and energy

$$
\omega - \omega' = \pm \omega(q) \tag{1.2}
$$

In these equations  $\vec{k}$  is an initial and  $\vec{k}'$  a scattered polariton wave vector, both inside the crystal;  $\omega(q)$  is the phonon frequency, and  $\pm$  denotes Stokes and anti-Stokes processes.

Figure 1 illustrates the kinematics of backward Brillouin (Stokes) scattering. For incident frequency below the exciton frequency  $\omega_{ls}$ , a single shift is observed which increases as the frequency approaches  $\omega_{1s}$  (corresponding to the lowest transition  $\vec{k}_2 + \vec{k}'_2$  in the case of backward scattering). Sufficiently close to or above resonance, we predict three additional Brillouin (Stokes) lines. They correspond to scattering processes from the initial state I to the final states  $I'$ -IV'. A transition to

transition. Including the anti-Stokes components one should observe an octet instead of the Brillouin doublet in that frequency region. In this paper we give two calculations of the relative amplitudes of the members of the multiplet: a semiclassical one and a quantum mechanical one; the results agree in essence. When coupling coefficients and other parameters appropriate to CdS

one of these final states can occur if momentum conservation (1. 1) and energy conservation (1.2) are satisfied for one pair of momenta  $(\vec{k}_i, \vec{k}'_i)$ . The acoustic phonon dispersion "tunes" the permitted

the new lines may be observable. The calculation are inserted, the resulting calculation shows that has been carried out for the ABC of the previous paper, <sup>1</sup> as well as for two other sets of ABC. The predicted Brillouin scattering differs quantitatively in these cases; hence observation of the new members of the multiplet may resolve the long-



FIG. 1. Dispersion curves of polaritons inside the crystal for  $k$  and  $-k$  directions (schematically). The igure illustrates the kinematics of backward Brillouin scattering. I is the initial state, I'-IV' are the four possible final states. The pair of momenta  $(k_i, k_j)$  which satisfy the momentum and energy conservation are joined by an arrow.

 $6\overline{6}$ 

 $(1, 1)$ 

standing disagreement regarding the correct ABC, i.e., the structure of the eigenmode. Experimental determination of the shift and the width of the Brillouin lines would permit the complete determination of the complex refractive indices of the polariton branches in the crystal. In particular one would obtain directly the total exciton mass in a prescribed  $\vec{k}$  direction.

## II. SEMICLASSICAL TREATMENT

Maxwell's equation for the Fourier component of the scattered electromagnetic wave with frequency  $\omega' = c k_h$  is

$$
-\left[\nabla^2 + k_0^{\prime 2} (1 + 4\pi \chi_{k_0^{\prime}})\right] E' = 4\pi k_0^{\prime 2} (\delta \chi E_{k_0})_{k_0^{\prime}}.
$$
 (2. 1)

The operator on the left-hand side is the inverse of the polariton propagator at frequency  $\omega'$ . In Eq.  $(2. 1)$  the  $\tilde{r}$  arguments are not written explicitly to simplify the notation. The susceptibility  $\chi$ is given by the integral operator in Eq.  $(2, 1)$  of Ref. 1 with  $\chi_0 = 0$ . The right-hand side of  $(2.1)$  involves a source term proportional to the electric field  $E_{\vec{k}_0}$ of the incident polariton.  $\vec{k}_0$  denotes the momentum of the incident photon outside the crystal.  $\delta \chi$ is the change in the susceptibility due to density fluctuations  $\delta n(\vec{r}, t)$  produced by acoustical phonons:

$$
\delta \chi = \frac{\partial \chi}{\partial n} \, \delta n = - \chi \, \frac{\partial \chi^{-1}}{\partial n} \, \chi \delta n \quad . \tag{2.2a}
$$

In the frequency region near one resonance and very far from all the other resonances,  $\partial \chi^{-1}/\partial n$  can be considered as frequency independent. In the deformation-potential approximation it is also considered to be independent of all  $\tilde{r}$  arguments, and we write

$$
\frac{\partial \chi^{-1}}{\partial n} = C \tag{2.2b}
$$

Since the incoming light wave has frequency  $\omega \neq \omega'$ ,  $E'$  has only scattered waves. The formal solution of (2. 1) is

$$
E' = -\frac{4\pi k_0^2}{\nabla^2 + k_0^2 (1 + 4\pi \chi_{k_0^*})} \chi_{k_0^*} C(\delta n P_{\vec{k}_0})_{k_0^*} .
$$
 (2.3)

Here we used the relation  $P_{\mathbf{k}_0} = \chi E_{\mathbf{k}_0}$ , where we label the polarization and electric field by the external wave vector. Introducing the complete, orthonormal set of eigensolutions  $E_{\mathbf{k}^{\bullet}}^{\mathbf{i} \mathbf{n}}(\vec{\mathbf{r}})$  of the denominator of  $(2, 3)$  with incoming scattered waves, <sup>2</sup> one obtains

$$
E' = -4\pi k_0^2 \sum_{\vec{k}_0'} \frac{1}{\nabla^2 + k_0'^2 (1 + 4\pi \chi_{k_0'})} E_{\vec{k}_0'}^{\text{in}} T_{\vec{k}_0'} r_{\vec{k}_0}
$$
 (2.4)

with the scattering amplitude

$$
T_{\vec{k}_0'}^* \vec{k}_0 = C \int d\vec{r} d\vec{r}' dt E_{\vec{k}_0'}^{\frac{1}{2n}}(\vec{r})^*
$$
  
 
$$
\times \chi_{k'_0}(\vec{r}, \vec{r}') P_{\vec{k}_0}(\vec{r}') \delta n(\vec{r}', t) e^{-i (\omega - \omega')t} . \quad (2.5)
$$

(The superscript "in" always refers to the states satisfying incoming wave conditions.) From Eq. (2.4) we obtain for the electric field  $E_{\vec{k}_0}^{\prime}$  scattere in the direction  $\vec{k}'_0$  at a distance R from the crysta

$$
E'_{\vec{k}_0'} = -4\pi k_0'^2 \frac{1}{(2\pi)^3} \int d\Omega_{\vec{r}} \int_0^\infty dk_0'^{\prime} k_0''^2
$$

$$
\times \frac{e^{ik_0'^2} R \cos \varphi}{k_0'^2 - k_0'^{\prime 2}} T_{\vec{k}_0' \vec{k}_0}
$$

$$
= (k_0'^2 / R) e^{ik_0'^2} T_{\vec{k}_0' \vec{k}_0} . \qquad (2.6)
$$

The thermally averaged scattered energy flux The thermally averaged scattered energy rids<br> $d^2I_s/d\Omega d\omega'$  into the solid angle  $d\Omega_{\vec{k}\cdot\vec{k}}^* \equiv d\Omega$  and the frequency interval  $d\omega'$  is

(2. 2a) 
$$
\frac{d^2I_s}{d\Omega d\omega'} = \frac{C}{8\pi} k_0'^4 \langle |T^*_{\mathbf{k}_0'} \mathbf{k}_0|^2 \rangle, \qquad (2. 7)
$$

where  $\langle \cdots \rangle$  means a thermal average over the density fluctuations.

For the sake of simplicity we consider backward scattering along the z direction for the calculation of the  $T$  matrix element. Let us introduce the polarization fields $P^{\bf in}_{\bf k\ell}$  =  $\chi E^{\bf in}_{\bf k\ell}$  of the scattered wave: inside the crystal. According to Ref. 1,  $P_{\vec{k}_0}(\vec{r})$  and  $P_{\vec{k}_0}^{\text{in}}(\vec{r})$  can be decomposed inside the crystal into two polariton components:

$$
P_{\vec{k}_0}(\vec{r}) = P_1 e^{ik_1 z} + P_2 e^{ik_2 z} \quad , \tag{2.8a}
$$

$$
P_{\nu}^{1n}(\mathbf{\vec{r}})=P_1'e^{-ik_1^2z}+P_2'e^{-ik_2^2z}.
$$
 (2.8b)

The wave numbers  $k_i$  and  $k'_j$  are solutions of the dispersion relation  $(3, 9)$  of Ref. 1. In general,  $k_i$ and  $k_i$  are complex owing to the finite exciton lifetime, and we write them as  $k_i + i\kappa_i$  and  $k'_i + i\kappa'_i$ . Inserting Eq.  $(2.8)$  into  $(2.5)$  yields

$$
T_{k_0}^* \dot{F}_0 = CF \sum_{i,j} P_i P'_j \int dt \int_0^{\infty} dz \exp[i(k_i + k'_j)z - (\kappa_i + \kappa'_j)z - i(\omega - \omega')t] \delta n(z, t) , \qquad (2.9)
$$

where  $F$  denotes the transverse area exposed to the incident radiation. Equation (2. 7) can now be written in the form

$$
\frac{d^{2}I_{s}}{d\Omega d\omega'} = \frac{C}{8\pi} k_{0}^{\prime 4} F C^{2} \sum_{i,j} |P_{i}|^{2} |P_{j}'|^{2} \frac{1}{2\pi}
$$

$$
\times \int_{-\infty}^{\infty} dq \frac{1}{(k_i + k'_j - q)^2 + (k_i + k'_j)^2} \, \delta(q, \, \omega - \omega') \, .
$$
\n(2.10)

Here we have neglected interference terms between the components of the multiplet, which is justified for sufficiently small damping of the polaritons. Moreover, the correlation function for the density fluctuations is assumed to be translationally invariant. That is, one assumes the crystal is infinite for phonons, while taking the boundary into account for excitons. The function

$$
\varepsilon(q,\,\omega) = \int_{-\infty}^{\infty} dz\,dt\,\langle \delta n(z,\,t)\,\delta n(0,\,0)\rangle\,e^{\,i(\alpha z - \omega\,t)}\qquad \quad (2.\;11)
$$

is the van Hove spectral distribution of the thermal density fluctuations. <sup>3</sup> Using the explicit form for the density fluctuations in terms of phonon creation and destruction operators  $c_{\,\pmb{\cdot}}^{\,\dagger}$  and  $c_{\,\pmb{\cdot}\,}^{\,\dagger}$ 

$$
\delta n(z) = i \int dq \, e^{iqz} \, \left( \frac{\hbar |q|}{4 \pi \rho c_s} \right)^{1/2} c_{q+1} \, c_{q+1} \, c_{q+1} \, (2.12)
$$

we obtain

$$
\delta(q,\omega) = \frac{\hbar|q|}{2\rho c_s} \left\{ [1 + n(\omega)] \delta(\omega - c_s|q|) + n(\omega) \delta(\omega + c_s|q|) \right\}. \quad (2.13)
$$

Here  $\rho$  denotes the mass density,  $c_s$  the longitudinal sound velocity, and  $n$  the Bose-Einstein distribution

$$
n(\omega) = 1/(e^{\hbar \omega/kT} - 1)
$$
 (2. 14)

Finally we must relate the polarization amplitudes

 $\sim$  10  $\pm$ 

 $P_i$  and  $P'_i$  in (2.10) to the incident and reflected field amplitudes  $E_{\text{in}}$  and  $E_{\text{re}}$ . Using Maxwell's boundary conditions as well as the ABC from Ref. 1 and a background dielectric constant  $\epsilon_0$ , one has

$$
E_{1n} + E_{r0} = E_1 + E_2 = \left(\frac{1}{n_1^2 - \epsilon_0} - \frac{1}{n_2^2 - \epsilon_0}\right) 4\pi P_1,
$$
\n(2. 15a)  
\n
$$
E_{1n} - E_{r0} = n_1 E_1 + n_2 E_2 = \left(\frac{n_1}{n_1^2 - \epsilon_0} - \frac{n_2}{n_2^2 - \epsilon_0}\right) 4\pi P_1.
$$
\n(2. 15b)

Solving  $(2. 15a)$  and  $(2. 15b)$  for  $P_1$ , we get

$$
|P_1|^2 = |P_2|^2 = \frac{1}{4\pi^2} \frac{|n_1^2 - \epsilon_0|^2 |n_2^2 - \epsilon_0|^2}{|n_1 - n_2|^2 |\epsilon_0 + n_1 + n_2 + n_1 n_2|^2} |E_{1n}|^2.
$$
\n(2.16)

The primed (scattered) fields obey the boundary conditions of incoming waves [see Eq.  $(2, 4)$ ]. An immediate consequence is that they also satisfy Eqs. (2. 15a), (2. 15b), and (2. 16) if each amplitude and refractive index is primed and  $E'_{1n}$  is set equal to 1 due to the normalization of the states  $E_{\text{K}}^{\text{in}}(\tilde{\mathbf{r}})$ . in Eq. (2.4). The incident energy flux is  $I_1 = (c/8\pi)$  $|E_{\text{in}}|^2$ . We thus obtain for the differential cross section for backward scattering, taking  $E_{in} = 1$  in Eq. (2. 16):

$$
\frac{d^2\sigma}{d\Omega d\omega'} = \frac{d^2I_s}{d\Omega d\omega'} / I_I = \frac{k_0'^4 C^2}{4\rho c_s^2} \sum_{i,j=1}^2 |P_i|^2 |P'_j|^2 \frac{F}{\pi c_s}
$$
\n
$$
\times \left( \frac{[1 + n(\omega - \omega')] \hbar |\omega - \omega'|}{[k_i + k_j' - (\omega - \omega')/c_s]^2 + (k_i + k_j')^2} + \frac{n(\omega - \omega') \hbar |\omega - \omega'|}{[k_i + k_j' + (\omega - \omega')/c_s]^2 + (k_i + k_j')^2} \right). \quad (2.17)
$$

## III. QUANTUM-MECHANICAL TREATMENT

The Hamiltonian  $H$  of our system can be decomposed as

$$
H = H_0 + \delta H \tag{3.1}
$$

 $H_0$  contains the free fields for the photons, excitons, and phonons for a half-infinite crystal and also the bilinear interaction between the photons and the excitons.  $H_0$  can be diagonalized by a linear transformation and then has the form

$$
H_0 = \sum_{\vec{k}_0} \hbar c \, |\vec{k}_0| B_{\vec{k}_0}^{\text{tout}} B_{\vec{k}_0}^{\text{out}} = \sum_{\vec{k}_0} \hbar c \, |\vec{k}_0| B_{\vec{k}_0}^{\text{tin}} B_{\vec{k}_0}^{\text{in}}.
$$
 (3.2)

The operator  $B_{k_0}^{\frac{1}{2}}$  creates a polariton which consists of an incident photon with momentum  $\vec{k}_0$  and a reflected and transmitted photon outside of the crystal. Inside the crystal this polariton, in general, contains two polarization waves with different wave numbers. Moreover, the different parts satisfy Maxwell's boundary conditions and the ABC at the surface. The labels "out" and "in" denote retarded and advanced boundary conditions, respectively. The ground state  $|0\rangle$  of  $H_0$  is defined by  $B_{\vec{k}_0}^{\text{in}}|0\rangle = 0$  (or equivalently  $B_{\vec{k}_0}^{\text{out}}|0\rangle = 0$ ) for all  $\vec{k}_0$ .

 $\delta H$  denotes the interaction between the polarization fields and acoustic phonons and has the form

$$
\delta H = C \int P'(\vec{r}) \delta n(\vec{r}) P(\vec{r}) d^3 r . \qquad (3.3)
$$

Classically (3. 3) leads to the source terms on the right-hand side of Eq.  $(2.1)$ . In this section  $P(\vec{r})$ denotes the associated operator, however.

The differential cross section for scattering of a photon with wave number  $\bar{k}_0$  (outside the crystal) into a scattered photon with wave number  $\vec{k}'_0$  (outside the crystal) exciting an acoustic phonon with wave number  $\tilde{q}$  in the crystal is

$$
\frac{d^2\sigma}{d\Omega d\omega'} = \left(\frac{k_0'}{2\pi\hbar c}\right)^2 \sum_{\vec{\mathbf{q}}} \left|T_{\vec{\mathbf{k}}_0\vec{\mathbf{q}},\vec{\mathbf{k}}_0}\right|^2 \delta(\omega - \omega' - c_s \left|\vec{\mathbf{q}}\right|)
$$
\n(3.4)

with

$$
T_{\vec{k}_0}^{\bullet} \vec{a}, \vec{k} = \langle 0 \left| B_{\vec{k}_0}^{\text{in}} C_{\vec{a}} \delta H B_{\vec{k}_0}^{\text{tout}} \right| 0 \rangle \; . \tag{3.5}
$$

Using the fact that

$$
P_{\vec{k}_0}^{\text{out}}(\vec{r}) = [P(\vec{r}), B_{\vec{k}_0}^{\text{out}}], \qquad (3.6a)
$$

$$
P_{\vec{k}_0}^{\text{in}}(\vec{r}) = \left[ B_{\vec{k}_0}^{\text{in}}, \ P(\vec{r}) \right] \tag{3.6b}
$$

are no longer operators but just  $c$  numbers,  $(3, 5)$ 

can be written as

$$
T_{\vec{k}_0^t \vec{a}, \vec{k}_0} = C \int d\vec{r} P_{\vec{k}_0^t}^{in} (\vec{r})^* \langle 0 | \delta n(\vec{r}) | \vec{q} \rangle P_{\vec{k}_0}^{out} (\vec{r}) . (3.7)
$$

It is also convenient to consider the two polariton<br>wave fields  $E_{\vec{k}_0}^{\text{in}}$  and  $E_{\vec{k}_0}^{\text{out}}$  which are constructed<br>similarly to (3.6a) and (3.6b) with the operator  $P(\vec{r})$  replaced by the operator  $E(\vec{r})$ :

$$
E_{\mathbf{k}_0}^{\text{out}}(\mathbf{\vec{r}}) = [E(\mathbf{\vec{r}}), B_{\mathbf{k}_0}^{\text{out}}], \qquad (3.8a)
$$

$$
E_{\vec{k}\ell}^{\text{in}}(\vec{r}) = [B_{\vec{k}\ell}^{\text{in}}, E(\vec{r})]. \tag{3.8b}
$$

The polariton wave fields on the left-hand side of Eqs.  $(3, 6)$  and  $(3, 8)$  obey the classical Eqs.  $(2, 1)$ and  $(3.1)$  of Ref. 1. This can be easily derived from (3. 2) and the fact that the operator  $E(\vec{r})$  obeys the classical Maxwell equation. Thus all the results of the semiclassical treatment can be taken over. The quantum-mechanical treatment only fixes the asymptotic values of the wave fields outside the crystal, namely,

$$
E_{\mathbf{k}_0}^{\text{out}}(\mathbf{\vec{r}})=E_{\text{in}}e^{ik_0z}+E_{\text{re}}e^{-ik_0z}, \quad z<0 \qquad (3.9a)
$$

$$
E_{\vec{k}_0}^{\text{in}}(\vec{r}) = E'_{\text{in}} e^{-ik_0^*z} + E'_{\text{re}} e^{ik_0z}, \quad z < 0. \quad (3.9b)
$$

The amplitudes  $E_{in}$  and  $E'_{in}$  are determined by the normalization condition that in the asymptotic limit the incident photon wave has the amplitude 1. A direct evaluation of (3.8) leads to

$$
|E_{1n}|^2 = 2\pi c \hbar k_0, \quad |E'_{1n}|^2 = 2\pi c \hbar k'_0. \quad (3.9c)
$$

The evaluation of  $(3, 5)$  and  $(3, 4)$  is very similar to that of  $(2, 5)$  and  $(2, 10)$  in the semiclassical case. One obtains the expression  $(2, 17)$  again with the slight difference that the factor  $k_0^{\prime 4}$  in (2.17) is replaced by  $k_0^3 k_0$ . The difference is due to different definitions of the cross section: Classically it was defined in terms of the scattered energy flux, and quantum mechanically in terms of scattered photon quanta.



FIG. 2. Refractive indices  $n_i + i\nu_i$  of the two polaritons vs frequency near the 1s exciton in CdS. The halfwidth of the exciton is 10 cm<sup>-1</sup>. The solid line represents the real part  $n_i$ , the dashed line the imaginary part  $\nu_i$ .



FIG. 3. Brillouin shifts (lower part of the figure) and relative widths  $\Delta$  (upper part of the figure) of backward scattered photons in CdS vs frequency for the geometry  $\mathbf{E}_I \parallel \mathbf{E}_s \perp \mathbf{k} \perp \mathbf{c}$ .  $\mathbf{E}_I(\mathbf{E}_s)$  is the electric field vector of the incident (scattered) field and  $\bar{c}$  the c axis. For simplicity only excitations of LA phonons have been considered.

## IV. DISCUSSION: RESULTS FOR CdS

The different factors in the expression (2, 17) for the cross section have a simple physical meaning. The first is slowly varying with frequency and exhibits the familiar  $\omega'^4$  law. The polarization amplitudes  $P_i$  and  $P'_i$  are determined by Eq. (2.16). Both show a strong frequency dependence near the resonance because of the frequency dependence of the transmission coefficient for the incident and outgoing wave, and because of the resonance enhancement of the susceptibility. The first term in large parentheses in Eq.  $(2, 17)$  describes Stokes processes, and the second anti-Stokes processes. each weighted with thermal factors. In the classical limit  $(kT \gg h | \omega - \omega' |)$  we have

$$
[1 + n(\omega - \omega')] \hbar |\omega - \omega' | \approx n(\omega - \omega') \hbar |\omega - \omega' | \approx kT
$$
\n(4.1)

For the process  $i + j$  the scattered light produces a Lorentzian line shape with maximum at

$$
\omega' = \omega \pm c_s \left| k_i + k'_j \right| \tag{4.2a}
$$

and a relative linewidth of

$$
\Delta = 2 \frac{k_i + k'_j}{k_i + k'_j} \tag{4.2b}
$$

If one integrates the cross section over  $\omega'$ , the

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FIG. 4. Total efficiency ij for backward scattering in CdS vs frequency for  $\mathbf{E}_I \parallel \mathbf{E}_S \perp \mathbf{k} \perp \mathbf{c}$ . The label ij means scattering from branch  $i$  to branch  $j$  in the nomenclature of Fig. 2. Total efficiency is the scattering cross section [Eq.  $(2.17)$ ] for  $F = 1$  cm<sup>2</sup>.

factors following the polarization amplitudes simplify to give the effective scattering volume  $F/$  $(\kappa_i + \kappa'_i)$  (except for a thermal factor). The relations  $(4. 2a)$  and  $(4. 2b)$  were recently used by Sandercock<sup>4</sup> to determine the complex refractive index of Si and Ge, for incident frequencies in the continuum.

We made numerical estimates for CdS for the different quantities in Eq. (2. 17). The following crystal parameters were used:  $\omega_{1s}$  = 20700 cm<sup>-1</sup>,  $4\pi\omega_p^2 = 4.16 \times 10^6$  cm<sup>-25</sup> (corresponding to  $A = 0.0047$ in the notation of Ref. 5, p. 236),  $\Gamma = 10 \text{ cm}^{-1}$ ,  $\epsilon_0$ In the notation of Ref. 5, p. 250),  $\mathbf{I} = 10 \text{ cm}$ ,<br>= 8,  $M_{\perp} = 0.9 m_e$ ,  $^6 c_s = 4.48 \times 10^5 \text{ cm/sec}$ , and c = 8,  $M_1$ = 0.9 $m_e$ ,  $c_s$  = 4.48×10° cm/sec, and c<br>= - 1.925×10<sup>3</sup>.<sup>7</sup><sup>a</sup> Here  $M_1$  is the exciton mass and  $c_s$ the longitudinal sound velocity for a direction perpendicular to the  $c$  axis. The most uncertain values are the oscillator strength  $4\pi\omega_p^2$  and the phenomenological damping constant  $\Gamma = 2\eta$  [see Eq. (2.8) of Ref. 1]. In Ref. 5 the values  $A = 0.0625$  and  $\Gamma$  < 1 cm<sup>-1</sup> were used in explaining reflection data. Our values for  $A$  and  $\Gamma$  correspond to a lower bound for the effects of spatial dispersion and cross sections. Our  $\Gamma$  value is a compromise between Refs. 5 and 8. CdS was chosen because it has already been investigated in the opaque region below the

gap by Brillouin scattering.<sup>9</sup> Moreover, it is known<sup>9,10</sup> that in this crystal the acoustic phonon and the excitons are mainly coupled by a deformation potential which is of the form of our interaction Hamiltonian (3.3). For simplicity we consider only longitudinal acoustic phonons in the following though the transverse acoustic phonons can equally well couple to the excitons. The cross sections are always calculated in the classical limit (4.1).

Figure 2 shows the two solutions for the complex refractive index  $n + i\nu = k/k_0$  near the resonance for  $\vec{E} \perp \vec{k} \perp c$ . Near the 1s resonance both polaritons are strongly damped. For higher frequencies the photonlike polariton is much less damped than the excitonlike polariton.

The frequency shifts and widths of the scattered lines are shown in Fig. 3 for backward scattering. The usual Brillouin line corresponds to scattering on the photonlike parts of the polariton branches. This means scattering on the branch 22 (that is  $i = 2$ ,  $j = 2$ ) below the resonance, and 11 above the resonance. Typical values for the acoustic phonons are 1-2 cm<sup>-1</sup>. When  $\omega$  approaches  $\omega_{1s}$  very closely from below, the predicted Brillouin frequency shift  $\omega_B$  from branch 22 is seen to increase sharply. When  $\omega$  approaches  $\omega_{1s}$  from above,  $\omega_B$  (from branch 11) decreases sharply. The other branches correspond to the other scattering possibilities with typical values for the acoustic phonons of





Figure 4 shows the frequency dependence of the four total efficiencies. The total efficiency is the scattering cross section calculated with the exposed area,  $F = 1$  cm<sup>2</sup>, in Eq. (2.17). Below the resonance only the 22 cross section is essentially nonzero, and strongly increases with frequency in spite of the decreasing penetration depth  $1/(n_2+n'_2)$ . The two maxima in the total efficiencies  $12 \approx 21$  and <sup>22</sup> correspond to "in" and "out" resonances. For  $\omega \gg \omega_{1s}$  the 11 scattering on the lightlike polariton branch dominates. Near  $\omega_{1s}$  all four processes are comparable.

In Fig. 5 we show the effect of different  $ABC$ on the cross section 22. Three curves are shown 1, 2, and 3, for each of which one ABC was used:

 $(P_1 - \chi_0 E_1) + (P_2 - \chi_0 E_2) = 0,$  ABC 1  $n_1(P_1 - \chi_0 E_1) + n_2(P_1 - \chi_0 E_2) = 0$ , ABC 2

Supported in part by the National Science Foundation. ~On leave of absence from Max Planck Institut fur

Festkörperforschung, Stuttgart, and Physikdepartment der TU, München.

fOn leave of absence from the Physikdepartment der TU, München. Supported in part by Deutsche Forschungsgemeinschaft.

<sup>§</sup>Supported in part by AROD.

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$$
\frac{P_1}{(n_1 - n_e)(n_1^2 - 1)} + \frac{P_2}{(n_2 - n_e)(n_2^2 - 1)} = 0, \quad \text{ABC } 3.
$$

From the derivation in Ref. 1 we believe that ABC 1 properly allows for exciton reflection in contrast to ABC 3 derived in Refs. 11 and 12. The ABC 2 is a limiting case of the type proposed in Ref. 5. All three curves show "in" and "out" resonances with the 1s exciton. To distinguish between the three curves note on 1 the slow fall off above  $\omega_{1s}$ , on 2 the sharp fall off above  $\omega_{1s}$ , and on 3 the additional resonance at about 15 cm<sup>-1</sup> above  $\omega_{1s}$ , due distinguished distinguished as to the extra factor in the denominator in ABC 3. Hence measurement of the cross section could decide the correct ABC experimentally.

#### ACKNOWLEDGMENT

Wilhem Brenig and Roland Zeyher are grateful to the members of the Physics Department of New York University for their kind hospitality.

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