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RESONANT BRILLOUIN SCATTERING OF EXCITONIC POLARITONS IN CuBr

Yasuo Oka[+] and Duy-Phach Vu

Max-Planck-Institut für Festkörperforschung
7000 Stuttgart 80
Federal Republic of Germany

The dispersion relation of the excitonic polaritons in CuBr has been studied up to $k \sim 10^7$ cm^{-1} by resonant Brillouin scattering (RBS). The polariton parameters given by the invariant expansion formulation are determined from these measurements. The RBS is found to take place predominantly via piezoelectric coupling.

I. Introduction

Excitons in cuprous halides have interesting properties owing to the large electron-hole exchange interaction. This interaction causes the splitting of the singlet (Γ_5)- and the triplet ($\Gamma_{3,4}$)-states and the longitudinal-transverse (L-T) splitting of the Γ_5 state as well as additional splitting and shift of the exciton ground state at finite k(center of mass momentum of the exciton).

In this paper we study the dispersion relation of excitonic polaritons in CuBr by RBS. This method is particularly useful for cuprous halides due to the large exchange effects: Brillouin scattering enhanced by polariton resonance is expected to arise mostly around the region between the T- and L-exciton energies [1]. The L-T splitting Δ(LT) in CuBr is 12 meV while Δ(LT) = 0.08 meV in GaAs [2] and 1.9 meV in CdS [3]. Therefore we can investigate by RBS the dispersion relation of the polaritons for CuBr up to a k-value more than one order of magnitude larger than for the latter materials.

II. Experimental results

Figure (1) shows the Brillouin spectra obtained in back-scattering configuration for the (111) surface. In the bottom spectrum, where the excitation photon energy 2.9644 eV is about 1 meV higher than the triplet exciton state, Brillouin lines are seen on both Stokes and anti-Stokes sides, overlapping with the exciton luminescence. As the excitation energy increases the energy shifts of these lines increase and several additional lines appear. In the top spectrum, where the excitation energy is higher than the L-level, a number of further Brillouin lines can be observed. The energy shifts of the observed Brillouin lines are plotted in Fig.(2) as a function of the incident photon energy. The solid circles indicate the strong lines while the open circles show the weak lines. The crosses represent the luminescence lines due to the triplet exciton and the bound excitons: Their energies are independent of the excitation photon energy. Similar Brillouin scattering is seen for the (110)- and (001)-surfaces.

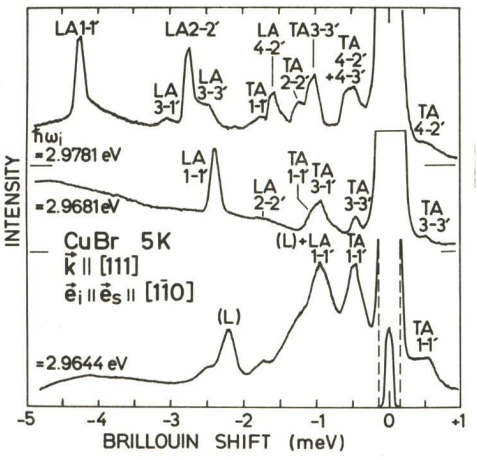

Fig. 1 Resonant Brillouin scattering spectra: (L) indicates the luminescence band

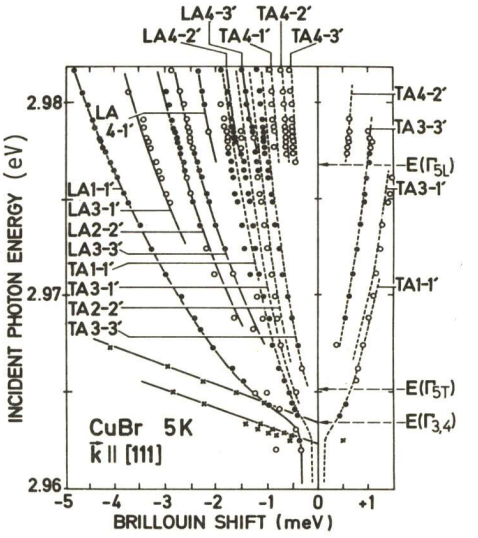

Fig. 2 Observed Brillouin shifts for various incident photon energies: The crosses indicate the luminescence lines. The solid and dashed lines are calculated.

III. Discussion

The exciton states of zinc blende crystals have been described recently by Hönerlage et al. [4], extending the invariant expansion formulation of Cho [5]. The exciton Hamiltonian is given by

$$H = H^{o} + H^{1}(k) + H^{2}(k), \tag{1}$$

where H^{o} describes the splitting of the exciton ground state at k=o: The singlet-triplet as well as the L-T splitting. $H^{1}(k)$ represents the k-dependent part which is diagonal with respect to the electron states:

$$\begin{aligned} H^{1}(k) = {} & \{C_k[k_x\{J_x, J_y^2-J_z^2\} + \text{c.p.}] + G_1 k^2 \mathbb{1}_h \\ & + G_2[k_x^2(J_x^2-\tfrac{1}{3}J^2) + \text{c.p.}] + 2G_3[k_y k_z\{J_y, J_z\} + \text{c.p.}]\} \otimes \mathbb{1}_e. \end{aligned} \tag{2}$$

Here $\mathbb{1}_e$ and $\mathbb{1}_h$ are unit matrices of dimension 2 and 4 respectively and J_i are the angular momentum matrices for holes. $\{A, B\} = (AB+BA)/2$. Each term of $H^{1}(k)$ corresponds to a term of the effective mass Hamiltonian: C_k is the k-linear term, G_1 describes the isotropic mass and G_2 and G_3 indicate the splitting and warping of the exciton bands.

The k-dependent exchange interaction is expanded as follows:

$$\begin{aligned} H^{2}(k) = {} & \delta_1 k^2 \underline{\sigma}.\underline{J} + \delta_2[k_x^2(\sigma_x J_x - \tfrac{1}{3}\underline{\sigma}.\underline{J}) + \text{c.p.}] \\ & + 2\delta_3[k_x k_y \tfrac{1}{2}(\sigma_x J_y + \sigma_y J_x) + \text{c.p.}] \end{aligned} \tag{3}$$

where σ_i are the Pauli spin matrices representing the electron spin state. The k-dependent exchange terms consist of the isotropic and anisotropic parts, i.e., δ_1 and δ_2, δ_3. The k-dependent Hamiltonians mix the exciton states of k=o in the finite k-region, and strongly affect the dispersion of the exciton. The polariton dispersion is

derived from the exciton states given by Eqs. (1-3) using the multiple oscillator model for the exciton-photon coupling [4].

The scattering of the polariton by an acoustic phonon takes place under conservation of crystal momentum and energies:

$$\underline{k}_i = \underline{k}_s + \underline{q} \text{ and } E_m(\underline{k}_i) = E_n(\underline{k}_s) \pm \hbar v q,$$

where the initial polariton in the m-th branch of energy $E_m(\underline{k}_i)$ is scattered to the $E_n(\underline{k}_s)$ state by an acoustic phonon whose momentum and group velocity are q and v. The velocities of sound are calculated, from the known elastic constants [6], e.g. 3.33×10^5 cm/sec for the LA phonon and 1.25×10^5 cm/sec for the TA phonon in the direction of $\underline{q}||[111]$. In RBS of GaAs and CdS [2,3], $q\sim6\times10^5$ cm^{-1} while the Brillouin shift of -4 meV in Fig.(2) yields $q\sim2\times10^7$ cm^{-1} for the case of CuBr. Therefore we can determine the dispersion relation up to the region of $k\sim10^7$ cm^{-1} which corresponds to 1/5 of the distance from the center to the Brillouin zone edge.

With an iterative procedure we calculated the energy shifts for the LA- and TA-phonon scatterings by the polariton branches derived from Eqs. (1-3) using the known sound velocities. The full curves of Fig.(2) are the results of this calculation in which we have used the value of the exciton parameters listed in Table 1. The assignment of LA 1-1' indicates the scattering by the LA phonon from the polariton branch 1 to the branch 1' which is the same branch as 1 but for backwardly-propagating polaritons. The optically active polariton branches are shown in Fig.(3) for the three principal k-directions. For the [001]-direction the polariton mode consists of the lower two branches due to the heavy- and light-hole excitons and the upper photon-like branch, while the middle branch shows two-fold splitting in the [111]-direction. In the [110]-direction the L-exciton mode couples with photons through the k-dependent perturbations (branch 4) when the polarization vector is along the [001]-direction.

Table 1

	present work	TPRS [4]	TPA [7]
$G_1^{(a)}$	0.65±0.01	0.69	0.69
G_2	-0.10±0.03	0	-0.25
G_3	-0.08±0.01	0	-0.71
$\delta_1^{(a)}$	0.1 ±0.05	0.24±0.02	
δ_2	0	0	
δ_3	0	0	
$C_k^{(b)}$	$(50\pm6)\times10^{-8}$	$(73\pm4)\times10^{-8}$(c)	

(a) units of $\hbar^2/2m_o$
(b) units of meV.cm
(c) 70×10^{-8}, Ref.[9]
$E(\Gamma_{3,4}) = 2.9633$eV
$E(\Gamma_{5T}) = 2.9650$eV
$E(\Gamma_{5L}) = 2.9766$eV

The lowest polariton branch in Fig.(3) is considerably isotropic, owing to the small values of G_2 and G_3 compared with G_1. Table 1 also shows those values obtained by Hönerlage et al. with two photon Raman scattering (TPRS) [4] and by Mattausch and Uihlein with two photon absorption (TPA) [7]. The Luttinger parameters deduced from TPA measurements were converted to the exciton parameters [4,8]. The parameters obtained by TPA correspond to strongly anisotropic dispersion curves: For the [110]- and [111]-directions the lowest branch shows negative curvature while for the [001]-direction the curvature is positive. In contrast to this, we have obtained, in all three principal directions, almost equal Brillouin shifts for the 1-1' scattering, which are well fitted with our nearly isotropic parameters. Therefore the di-

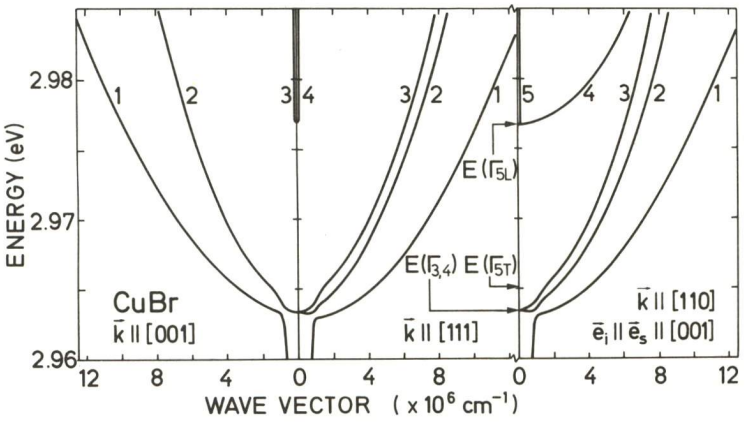

Fig. 3 Polariton dispersion relation for the three k-directions

rect observation by RBS indicates that the anisotropy of the excitonic polariton states is not as strong as that derived from the TPA studies. Further, our parameters account well for the dispersion of the polaritons obtained from the TPRS experiment which correspond to $k<15\times10^5$ cm^{-1}, although they are numerically very different from those given by Hönerlage et al. As we have obtained the Brillouin shifts due to different types of scattering between polariton branches with high accuracy, we believe we can derive from these results highly reliable values of the polariton parameters.

Finally we note that CuBr is a highly piezoelectric material: The piezoelectric constant e_{14} = 0.27 C/m^2[6]. Therefore the interaction between the excitonic polaritons and acoustic phonons should be dominated by piezoelectric coupling rather than by deformation potential coupling. The piezoelectric coupling in zinc blende crystals is allowed along the [111]-direction for LA phonons and along the [110]-direction for TA phonons polarized parallel to [001]. For phonons along the [001]-direction, the piezoelectric coupling does not arise. The experimental results show that the piezo-active phonons give rise to strong Brillouin lines as can be seen in Fig.(1) for the [111]-direction. Hence, the RBS of excitonic polaritons in CuBr takes place dominantly through piezoelectric coupling.

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+Alexander von Humboldt fellow. Permanent address: Research Institute for Scientific Measurements, Tohoku University, Sendai, Japan.

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