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Piezomodulation of excitonic reflectance spectra in CuCl

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We have investigated the piezoreflectance spectra of the 1s Z_3 and Z_{12} excitons in single crystals of copper chloride CuCl at 95 K with linearly polarized light. The spectra were studied for the applied low pressure $\boldsymbol{p}$ ($\boldsymbol{p}$ lower than 1 MPa) directed along the [001], [111] and [110] axis with the wave vector $\boldsymbol{k}$ of the incident light parallel to the $[1\bar{1}0]$ direction. A strong piezo-optical response is observed for the Z_3 exciton to regard to the one observed for Z_{12}. From the stress-induced shifts and splitting of the excitonic levels, we have been able to deduce the shear excitonic deformation potentials: $b = (-0.18 \pm 0.02)$ eV and $d = (0.30 \pm 0.03)$ eV.

1. Introduction

A large number of linear and non-linear stress-optical experiments on copper halides CuBr, CuCl and CuI have been reported [1–10]. In copper chloride CuCl the theoretical fine structures of the 1s excitons associated with the Γ_6 and Γ_7 bands (Z_3 excitons) and with the Γ_6 and Γ_8 bands (Z_{12} excitons) in unstressed crystals are well known. The electron-exchange interaction is particularly important because the excitonic Rydberg is large ($R_{\mathrm{exc}} = 189$ meV) [11] and the exciton radius is small ($a_{\mathrm{exc}} \approx 0.7$ nm) [11]. Accordingly, the short-range part of the electron-hole exchange interaction splits the singlet state Γ_5 (dipole allowed) and the triplet state Γ_2 (Z_3) or $\Gamma_3 + \Gamma_4$ (Z_{12}) (dipole forbidden) whereas the long range part splits further the singlet state into transverse optically active states $\Gamma_{5\mathrm{T}}$ and a longitudinal inactive state $\Gamma_{5\mathrm{L}}$. On the other hand, these singlet and triplet states are mixed at $k \neq 0$ by the presence of the k-linear term, which results from the lack of the inversion centre in zincblende materials such as CuCl. The application of uniaxial stress to semiconductors produces changes in the lattice constant and symmetry of the crystal, and, as a consequence, these cause important changes in the electronic properties, which manifest themselves in the optical ones.

The aim of this paper is to report an original piezoreflectivity study of CuCl in the excitonic range at the temperature of the 95 K and evaluate by piezomodulation methods the important electronic parameters characterising this material as deformation potentials.

2. Experimental

Samples having a parallelepiped shape, of about 5×3 mm^2 reflecting area and 0.5 mm thickness, were used. They were cut from single crystal blocks of CuCl grown by the melting zone method. The sample surfaces were oriented by X-ray Laue transmission measurements. The obtained cleavage planes were of high optical quality. The piezomodulation of the reflectivity spectrum of CuCl was performed by mounting it on a piezoelectric transducer of lead zirconate-titanate type. The transducer was driven by a sinusoidal electric field of 200 V/mm and an alternating stress of about 1000 Hz (different to the resonance frequency) was respectively applied along the [001], [111] and [110] crystal directions.

The monochromatic light was obtained from a high resolution 1 m JOBIN YVON HR single grating monochromator fitted with an OSRAM XBO 75 Watt high pressure xenon arc as the light source. The linear dispersion of the monochromator was 1.35 nm/mm in the first order. The wavelength bandwidth of the monochromator was of about 0.1 nm. The resulting alternating signal was detected and amplified, with a good signal to noise ratio by an E.G.G. model 5208 lock-in amplifier. The optical signals were detected with a P21 photomultiplier tube. The apparatus was entirely automated. A computer drove both the stepping motor of the monochromator and the lock-in amplifier via BUS IEEE lines and displayed the ratio ($\Delta R/R$).

The measurements were made with the wave vector $\boldsymbol{k}$ of the incident light perpendicular to $(1\bar{1}0)$ cleaved surface and the pressure **p** oriented parallel to one of the three meaning crystallographic directions. The electric vector ε of the reflected light was set either parallel (π component) or perpendicular (σ component) to the direction of the applied stress. A polarizer of GLAN type was used.

3. Experimental results

We report in Figs 1–3 the piezoreflectance spectra $\Delta R/R$ measured at 95 K for the three configurations. The lock-in amplifier is tuned to the fundamental frequency ω of the applied stress. For all measurements, the piezoelectric transducer was driven by electric field of 200 V/mm. In the absence of any means to deduce directly the exact stress at the sample surfaces, we attempt to estimate the magnitude of the applied pressure **p** from transducer characteristics and elastically properties of CuCl. The estimated values in [001], [111] and [110] crystal directions are respectively $p_{001} = 1$ MPa, $p_{111} = 0.75$ MPa and $p_{110} = 0.85$ MPa. These pressures are at the rear sample's surfaces that are in the side of the transducer. Taking into account of the thickness 0.5 mm of the samples, the pressures at the reflecting surfaces are lower than the ones at the rear surfaces. Each presented piezoreflectivity spectrum is the result of an average over several measurements. The zero-lines are determined from signal levels at low photon energy where no absorption and therefore no piezoreflectivity response is detected. The positions of piezoreflectivity structures are given with a maximum error of ±1 meV.

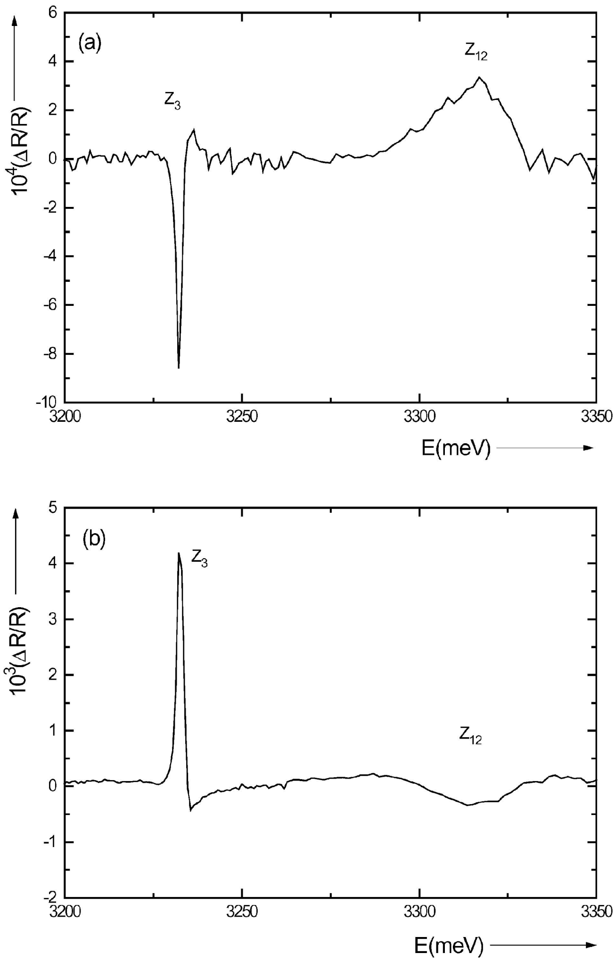

Figure 1 Piezoreflectance spectra of the 1s Z_3 and Z_{12} excitons in CuCl at 95 K: (a) *p*//[001], *ε*//[001] and (b) *p*//[001], *ε* ⊥ [001].

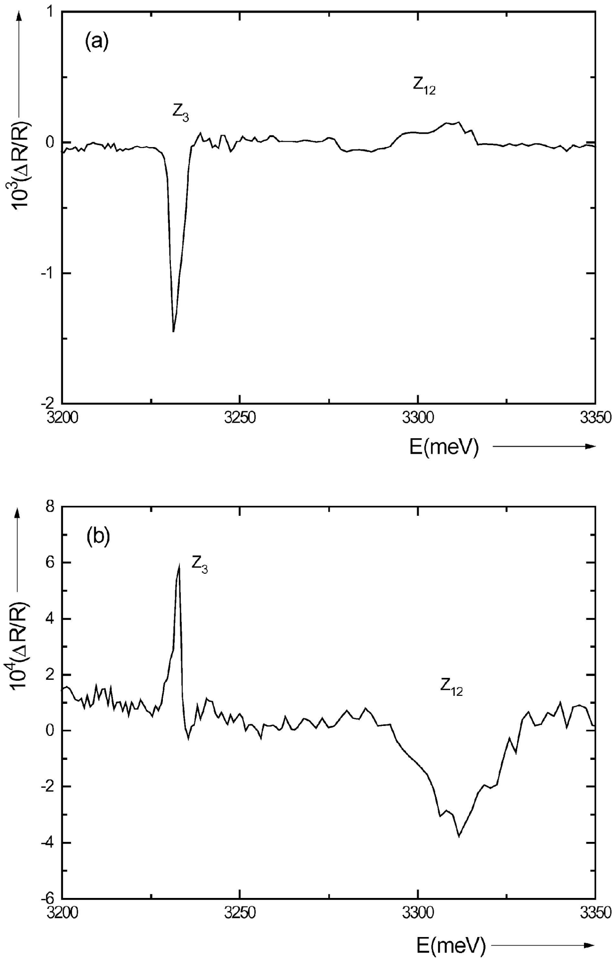

Figure 2 Piezoreflectance spectra of the 1s Z_3 and Z_{12} excitons in CuCl at 95 K: (a) **p**//[111], *ε* //[111] and (b) **p**//[111], *ε* ⊥ [111].

For all configurations the spectrum in polarization $\varepsilon//\boldsymbol{p}$ consists of a negative sharp peak Z_3: (3232 ± 1) meV and a positive broad structure Z_{12}: (3314 ± 2) meV. In the other polarization $\varepsilon\perp \boldsymbol{p}$ the spectrum exhibits a positive sharp peak Z_3 and a negative broad structure Z_{12}. In each polarization $\varepsilon//\boldsymbol{p}$ or $\varepsilon\perp\boldsymbol{p}$ the amplitude of the sharp peak is more stronger than the broad one.

4. Discussion

Under normal conditions CuCl crystallize in the zincblende structure. Taking into account the spin-orbit interaction, the lowest conduction band is characterised by Γ_6 symmetry, the uppermost valence band by Γ_7 and the second valence band by Γ_8 [12–15]. The direct exciton states associated with the Γ_7 and Γ_8 valence bands are called the Z_3 and Z_{12} excitons, respectively, by Cardona [12]. The separation between the 1s Z_3 and 1s Z_{12} excitons from our piezoreflectance measurements at 95 K is $|\Delta| = 84 \pm 1$ meV.

The Z_3 exciton is built up with Γ_6 electron and Γ_7 hole. The total symmetry of this fourfold degenerated

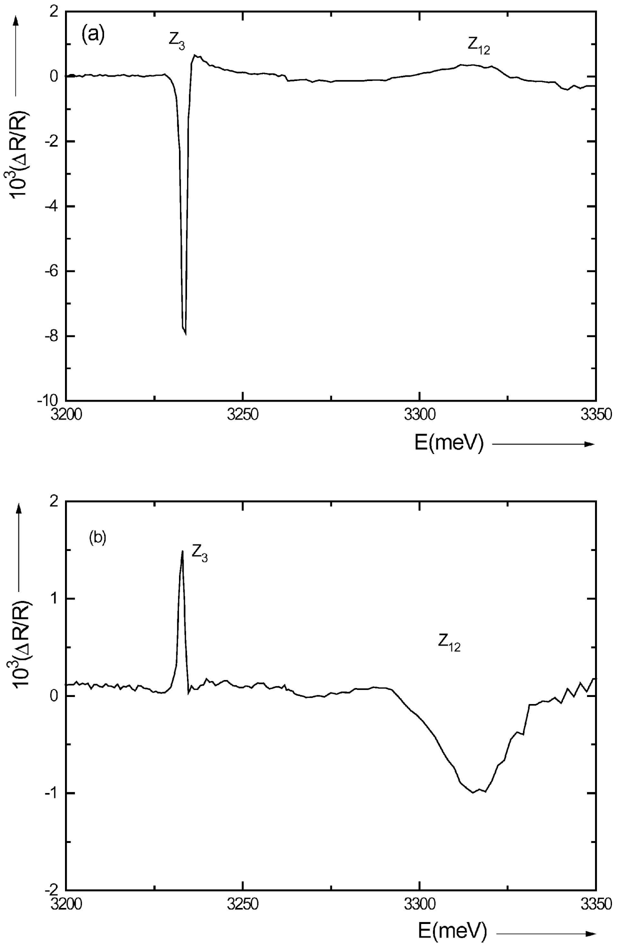

Figure 3 Piezoreflectance spectra of the 1s Z_3 and Z_{12} excitons in CuCl at 95 K: (a) ***p***//[110], ε //[110] and (b) ***p***//[110], $\varepsilon\perp$[110].

exciton is:

$$\Gamma_1 \times \Gamma_6 \times \Gamma_7 = \Gamma_2 + \Gamma_5 \qquad (1)$$

Taking into account both the short and the long-range parts of the electron-hole exchange interactions, the degeneracy of the Z_3 exciton is partially lifted [16]. The singlet triplet separation ($\Gamma_2 - \Gamma_5$) is due to the short (analytic) exchange interaction and the transverse-longitudinal splitting ($\Gamma_{5T} - \Gamma_{5L}$) is due to the long (nonanalytic) exchange interaction [15].

The application of a stress splits the transverse optically active states Γ_{5T} in two polarized components. In all studied configurations $p//[001]$, $p//[111]$ and $p//[110]$, the π components are shifted to lower energies and the σ components to higher ones. The main peak observed in the 1s Z_3 exciton for a given configuration corresponds to one of these components. The calculate difference between the zero stress reflectivity spectrum and the same spectrum shifted towards higher or lower energies gives the sign and the shape of the measured piezoreflectance spectra. In Z_3 the important observed piezo-optical response is due probably to the little spin orbit separation ($\Delta \approx 80$ meV) in CuCl. In the stressed crystal the 1s Z_3 and 1s Z_{12} states are coupled and can be described consequently by the same 1S envelope function.

The Z_{12} exciton is built up with Γ_6 electron and Γ_8 hole. This excitonic state is eightfold degenerate and have a total symmetry.

$$\Gamma_1 \times \Gamma_6 \times \Gamma_8 = \Gamma_3 + \Gamma_4 + \Gamma_5 \qquad (2)$$

As indicate previously, the electron-hole exchange interactions are responsible for the singlet-triplet separation (Δ_{st}) between the ($\Gamma_3 + \Gamma_4$) forbidden optical transition and the (Γ_5) dipole allowed exciton states and of the transverse-longitudinal separation (Δ_{lt}) between ($\Gamma_{5\mathrm{T}}$) and ($\Gamma_{5\mathrm{L}}$) exciton states.

On account of the value of the temperature (95 K) and of the spectral resolution (0.1 nm), we are mainly interested in the effect of the uniaxial stress upon the dipole allowed transverse states $\Gamma_{5\mathrm{T}}$. The uniaxial stress split the exciton $\Gamma_{5\mathrm{T}}$ in two components completely polarized. For the three configurations, the π components are shifted to higher energies and the σ components to lower ones. The main peak observed in the Z_{12} exciton in a piezoreflectivity spectrum for a given geometry corresponds to one of these components.

In all configurations, the stress induced energy shift of Z_{12} is opposite to the one corresponding to Z_3 exciton. This result agrees well with the one obtained in one reflection measurements under large static uniaxial stress [1].

The deformation potentials $(c - a)$, b and d can be directly evaluated from the splitting of the $\Gamma_{5\mathrm{T}}$ exciton in the given configurations.

The stress Hamiltonian H_{p} that describes the splitting of the Z_{12} exciton is as follows [17, 18].

$$\begin{aligned} H_{\mathrm{p}} = {} & H_0 + (c-a)(e_{\mathrm{xx}} + e_{\mathrm{yy}} + e_{\mathrm{zz}})I + b\big(J_x^2 e_{\mathrm{xx}} \\ & + J_y^2 e_{\mathrm{yy}} + J_z^2 e_{\mathrm{zz}}\big) + 2\frac{d}{\sqrt{3}}\left[\left(\frac{J_{\mathrm{x}}J_{\mathrm{y}} + J_{\mathrm{y}}J_{\mathrm{x}}}{2}\right)e_{\mathrm{xy}} \right. \\ & \left. + \left(\frac{J_{\mathrm{y}}J_{\mathrm{z}} + J_{\mathrm{z}}J_{\mathrm{y}}}{2}\right)e_{\mathrm{yz}} + \left(\frac{J_{\mathrm{z}}J_{\mathrm{x}} + J_{\mathrm{x}}J_{\mathrm{z}}}{2}\right)e_{\mathrm{zx}}\right] \end{aligned} \qquad (3)$$

where H_0 is the Hamiltonian of the Z_{12} exciton in the absence of any symmetry breaking effects. J_{x}, J_{y} and J_{z} are the total angular momentum operator ($J = 3/2$) components. e_{ij} are the strain tensor components. c and a are the hydrostatic deformation potentials of the conduction band Γ_6 and the valence band Γ_8 respectively. b and d are the shear deformation potentials of the valence band Γ_8. I is the matrix unit.

The terms describing the effect of a strain on the spin-orbit interaction are preponderant in the case of the Z_{12} exciton ($J = 3/2$). The eingenvalues for the Z_{12} exciton corresponding to the $\Gamma_{5\mathrm{T}}$ states as a function of the stress, with the origin of the energy fixed now upon the $\Gamma_3 + \Gamma_4$ level, are deduced from similar treatment used by Wecker [19] in CuBr. They are given by the following equations:

p // [001]:

$$E_{//} = \Delta_{\mathrm{st}} + [(c-a)(S_{11} + 2S_{12}) - b(S_{11} - S_{12})p_{001}, \qquad (4a)$$

$$E_{\perp} = \Delta_{\mathrm{st}} + \left[(c-a)(S_{11}+2S_{12}) + \frac{b}{2}(S_{11}-S_{12})\right]p_{001}; \tag{4b}$$

p //[111]:

$$E_{//} = \Delta_{\mathrm{st}} + \left[(c-a)(S_{11}+2S_{12}) - \frac{d\sqrt{3}}{6}S_{44}\right]p_{111}, \tag{5a}$$

$$E_{\perp} = \Delta_{\mathrm{st}} + \left[(c-a)(S_{11}+2S_{12}) + \frac{d\sqrt{3}}{12}S_{44}\right]p_{111}; \tag{5b}$$

p // [110]:

$$E_{//} = \Delta_{\mathrm{st}} + \left[(c-a)(S_{11}+2S_{12}) - \frac{b}{4}(S_{11}-S_{12}) - \frac{d\sqrt{3}}{8}S_{44}\right]p_{110}, \tag{6a}$$

$$E_{\perp} = \Delta_{\mathrm{st}} + \left[(c-a)(S_{11}+2S_{12}) + \frac{b}{2}(S_{11}-S_{12})\right]p_{110}; \tag{6b}$$

S_{11}, S_{12} and S_{44} are the elastic constants of the crystal. The energy shift δE of these stress-induced components ($E_{//}$ and $E_{\perp}$) is a linear function of the applied stress $\boldsymbol{p}$. In the case of low pressures, the singlet-triplet separation Δ_{st} is large with regard to stressed term δE. The reflectivity function $R(E_{//})$ or $R(E\perp)$ can be expanded into a power series of the energy E restricted to the first term in the vicinity of Δ_{st}:

$$R(E) = R(E_0) + \delta E\frac{\mathrm{d}R}{\mathrm{d}E}\cos\omega t + (\delta E)^2\left(\frac{\mathrm{d}^2R}{\mathrm{d}E^2}\right)\cos 2\omega t + \cdots \tag{7a}$$

$$\left(\frac{\Delta R}{R}\right)(E) = \delta E\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}\cos\omega t + (\delta E)^2\frac{1}{R}\left(\frac{\mathrm{d}^2R}{\mathrm{d}E^2}\right)\cos 2\omega t + \cdots \tag{7b}$$

where

$$E = \Delta_{\mathrm{st}} + \delta E$$

$R(E_0)$ is the zero stress reflectivity and ω is the fundamental frequency of the applied stress. Therefore the analytical expressions of the piezoreflectance spectra $(\frac{\Delta R}{R})_{//}$ and $(\frac{\Delta R}{R})_{\perp}$ are given by:

p// [001]:

$$\left(\frac{\Delta R}{R}\right)_{//}^{[001]} = [(c-a)(S_{11}+2S_{12}) - b(S_{11}-S_{12})]p_{001}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}, \tag{8a}$$

$$\left(\frac{\Delta R}{R}\right)_{\perp}^{[001]} = \left[(c-a)(S_{11}+2S_{12}) + \frac{b}{2}(S_{11}-S_{12})\right]p_{001}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}; \tag{8b}$$

p// [111]:

$$\left(\frac{\Delta R}{R}\right)_{//}^{[111]} = \left[(c-a)(S_{11}+2S_{12}) - \frac{d\sqrt{3}}{6}S_{44}\right]p_{111}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}, \tag{9a}$$

$$\left(\frac{\Delta R}{R}\right)_{\perp}^{[111]} = \left[(c-a)(S_{11}+2S_{12}) - \frac{d\sqrt{3}}{12}S_{44}\right]p_{111}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}; \tag{9b}$$

p// [110]:

$$\left(\frac{\Delta R}{R}\right)_{//}^{[110]} = \left[(c-a)(S_{11}+2S_{12}) - \frac{b}{4}(S_{11}-S_{12}) - \frac{d\sqrt{3}}{8}S_{44}\right]p_{110}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}, \tag{10a}$$

$$\left(\frac{\Delta R}{R}\right)_{\perp}^{110} = \left[(c-a)(S_{11}+2S_{12}) + \frac{b}{2}(S_{11}-S_{12})\right]p_{110}\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}; \tag{10b}$$

When the applied stress is the same for all directions we can writ:

$$\left(\frac{\Delta R}{R}\right)_{\perp}^{[111]} = \frac{1}{2}\left(\frac{\Delta R}{R}\right)_{//}^{[001]} + \left(\frac{\Delta R}{R}\right)_{\perp}^{[001]} - \frac{1}{2}\left(\frac{\Delta R}{R}\right)_{//}^{[111]}, \tag{11a}$$

$$\left(\frac{\Delta R}{R}\right)_{//}^{[110]} = \frac{1}{4}\left(\frac{\Delta R}{R}\right)_{//}^{[001]} + \frac{3}{4}\left(\frac{\Delta R}{R}\right)_{//}^{[111]}, \tag{11b}$$

$$\left(\frac{\Delta R}{R}\right)_{\perp}^{[001]} = \left(\frac{\Delta R}{R}\right)_{\perp}^{[110]}; \tag{11c}$$

In this way, there are only three independent measurements, $(\frac{\Delta R}{R})_{//}^{[001]}$, $(\frac{\Delta R}{R})_{\perp}^{[001]}$ and $(\frac{\Delta R}{R})_{//}^{[111]}$. On account of the relation (11c), the spectra in Figs 1b and 3b ought to be the same. But the ratio of the piezooptical effects in the region of Z_3 and that of Z_{12} is quite different in Figs 1b and 3b. This difference can be probably

TABLE I

	In this work	Previous work		
b (eV)	-0.18 ± 0.02	1.2	(−1.2)	[1]
		1.34	(−1.34)	[6]
		−0.16	(−1.6)	[22]
d (eV)	0.30 ± 0.03	−2.4	(4.2)	[1]
		0.425	(0.425)	[2]
		−1.65	(2.85)	[6]
		0.75	(0.75)	[22]

attributed to the stress-induced k linear terms effect in Z_3 [1, 4, 18]. Then, we believe that the piezooptical effect in Z_3 in CuCl can be attributed both to the little spin orbit separation ($\Delta \approx 80$ meV) and stress-induced k linear terms effect.

The deformation potentials are related to the absolute value of the applied stress $\boldsymbol{p}$ and the logarithmic derivative of the reflectivity $\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}$. To avoid the uncertainties arising from the derivative spectrum $\frac{1}{R}\frac{\mathrm{d}R}{\mathrm{d}E}$ and the amplitude of the pressure $\boldsymbol{p}$. However, it seems preferable to evaluate the deformation constants b and d from the ratio $(\Delta R/R)_{//}/(\Delta R/R)_{\perp}$ [20].

Using the following values:

$$(c-a) = -0.35 \text{ eV} \quad [2]$$

$$S_{11} = 0.075 \text{ GPa}^{-1} \quad [21]$$

$$S_{12} = -0.03 \text{ GPa}^{-1}$$

$$S_{44} = 0.069 \text{ GPa}^{-1}$$

We have calculated the b coefficient at some wavelength in the explored range of Z_{12} exciton in the direction $p//[001]$. Similarly, we have evaluated the d constant in the $p//[111]$ direction. Each presented value is the result of an average over five treated wavelengths around (3314 meV) with 1 meV apart. The calculated b and d are listed in Table I for comparison with the results of previous measurements.

In this work, we have used $-b$ and $-2d/\sqrt{3}$ in the stress Hamiltonian. In [1] (one photon reflection measurements under large static uniaxial stress) and [2] (one photon absorption measurements under uniaxial static stress up to 3 GPa) the corresponding coefficients are (D_1 and $2D_2$) and ($-3b$ and $-2d\sqrt{3}$) respectively. In [6] non-linear optical measurements under uniaxial stress up to 16 GPa are reported by Fröhlich *et al.* They deduced the coefficients b and d independently of the stress-induced k linear terms and using (b and $2d$) in the stress Hamiltonian and assuming the same 1S envelope function for Z_{12} and Z_3 excitons. In [22], b and d are extracted directly from piezobirefringence measurements. Between brackets we have reported the equivalent values of deformation potentials. The difference between all values is probably due to the difference in experimental conditions, because, samples, temperature, experimental set-up and ranges of applied pressures are different.

5. Conclusion

The piezoreflectance spectra of the 1s Z_3 and Z_{12}excitons of CuCl were measured at the temperature of the 95 K. In the tree studied configurations corresponding to the applied stress $\boldsymbol{p}//[001]$, $\boldsymbol{p}//[111]$ and $\boldsymbol{p}//[110]$, the low stress ($\boldsymbol{p}$ lower than 1 MPa) splits each exciton line into two completely polarized components $\boldsymbol{\varepsilon}\, //\boldsymbol{p}$ and $\varepsilon \perp \boldsymbol{p}$. The stress induced energy shift of Z_3 exciton is opposite to the one corresponding to the Z_{12} exciton. The new data of the deformation potentials b and d were determined by piezomodulation methods independently of the quality of the sample surfaces as usually required for reflectivity measurement under static stress. The present piezooptical measurement will be used for calculating the stress induced variations of refractive constants (Δn and Δk) and dielectric ones ($\Delta\varepsilon_1$ and $\Delta\varepsilon_2$) by Kramers Kronig analysis and will be considered for eventual next paper.

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