

# PIEZOREFLECTIVITY OF Ge AND GaSb IN THE REGION OF $A_3 \rightarrow A_1$ TRANSITIONS

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The relative change of the reflectivity, caused by an oscillating uniaxial stress, has been measured for Ge and GaSb crystals in the region of photon energy between 1.9 eV and 2.6 eV. Making use of the procedure proposed for the theoretical analysis of the results obtained for Ge [1], the quantitative analysis for GaSb has been performed.

It is shown that the optical transitions observed at 2.0 eV and 2.5 eV can be interpreted as  $A_3 \rightarrow A_1$  transitions in the  $A$  direction (or  $L'_3 \rightarrow L_1$  at the  $L$  point).

## 1. Introduction

Investigation of high energy reflectivity spectra provides information about energy gaps at the critical points but does not provide any information about critical points symmetry. Experiments which apply an external perturbation breaking the symmetry of the crystal allow us to obtain information about the location of a given transition in the Brillouin zone. The uniaxial stress may be used as such a perturbation. In our case an *ac* uniaxial stress has been applied and a differential method of measurement has been adopted. This allowed us to increase the sensitivity in comparison with the *dc* method [2, 3, 5, 9, 10, 11]. The theoretical analysis of piezoreflectivity spectra was first performed by Kane [4] and then used by Sell and Kane [1] to prove the identification of the reflectivity structure at 2.1 eV and 2.3 eV in Ge as transitions from the  $A_3(L'_3)$  valence band split by spin-orbit interaction to the  $A_1(L_1)$  conduction band. Our experimental results for Ge, obtained in the same region of wavelength, are presented on Fig. 1 for comparison. The measurements of the piezoreflectivity spectra in GaSb as well as their theoretical analysis are the main purposes of the present paper. We have connected the characteristic shape of  $\Delta R/R$  curves for GaSb with the structure at 2.0 eV and 2.5 eV in the reflectivity  $R$ . This reflectivity structure agrees with the structure in the imaginary part of the dielectric function calculated by Higginbotham *et al.* [7] and interpreted as  $A_3 \rightarrow A_1$  transitions (in the double -groups notation  $A_{4v} + A_{5v} \rightarrow A_{6c}^I$  and  $A_{6v} \rightarrow A_{6c}^I$ ).

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## 2. Basic formulas

The relative change of the reflectivity due to an external uniaxial stress is proportional to the strain  $e$  in the case of small amplitude of stress (both  $\Delta R/R$  and  $e$  are second-rank tensors):

$$\left(\frac{\Delta R}{R}\right)_{ji} = Q_{ijkl}e_{kl}. \quad (1)$$

For crystals of cubic symmetry the fourth-rank tensor  $Q_{ijkl}$  has only three various non-vanishing components [6]. Using the abbreviated notation for fourth-rank tensor, Eq. (1), which represents in fact nine equations, thus can be rewritten in the following form:

$$\begin{aligned} \left(\frac{\Delta R}{R}\right)_{11} &= Q_{11}e_{11} + Q_{12}(e_{22} + e_{33}) \\ \left(\frac{\Delta R}{R}\right)_{12} &= Q_{44}e_{12}. \end{aligned} \quad (2)$$

Other components of  $\left(\frac{\Delta R}{R}\right)_{ij}$  can be obtained by cyclic changes of the subscripts.

Eq. (2) can be rewritten in the symmetry adopted basis [4], [1]:

$$\begin{aligned} \left(\frac{\Delta R}{R}\right)_1 &= Q_1e_1 \\ \left(\frac{\Delta R}{R}\right)_{3\gamma} &= Q_3e_{3\gamma} \quad \gamma = \alpha, \beta \\ \left(\frac{\Delta R}{R}\right)_{5\gamma} &= Q_5e_{5\gamma} \quad \gamma = xy, xz, yz \end{aligned} \quad (3)$$

where all quantities are now symmetrized functions with the symmetry of the crystal.

There are two mechanisms which are responsible for the change of the dielectric function with stress. One of them is the energy band shift and the other one is the change of the wave functions with stress, *i. e.*, the quantities  $Q_1$ ,  $Q_3$  and  $Q_5$  describe these two mechanisms. Each function  $Q_j$  contains a contribution  $Q_j^{\text{shift}}$  from the energy bands shift and a contribution  $Q_j^{\text{mvar}}$  from matrix elements variation. The magnitude of both contributions to the functions  $Q_j$  depends on the symmetry of a given transition. The contributions  $Q_j^{\text{shift}}$  fulfil the following conditions for nondegenerate bands [1]: for the transition at the  $\Gamma$  point one has  $Q_3^{\text{shift}} = Q_5^{\text{shift}} = 0$ , at the  $L$  point or in the  $A$  direction  $Q_3^{\text{shift}} = 0$ , in the  $\Sigma$  direction  $Q_5^{\text{shift}} = 0$ .

It has been found by Sell and Kane [1] that for the transitions considered by them  $Q_1^{\text{mvar}} = 0$ ,  $Q_3^{\text{shift}} = 0$  and  $Q_5$  is a linear combination of  $Q_1$  and  $Q_3$ . We shall use the

relations for  $Q_1$ ,  $Q_3$  and  $Q_5$  which have been given in Ref. [1], to describe the transitions which we assume to be analogous in GaSb. These relations are as follows:

$$Q_1 = -\sqrt{3} D_1^1 \frac{1}{R_0(\omega)} \frac{dR_0(\omega)}{d\omega} \quad (4)$$

$$Q_3 = \sqrt{\frac{3}{2}} D_3^3 / \lambda [\alpha(\epsilon_+^{(1)} - \epsilon_-^{(1)}) + \beta(\epsilon_+^{(2)} - \epsilon_-^{(2)})] \quad (5)$$

$$Q_5 = -\frac{1}{6} \frac{D_1^5}{D_1^1} Q_1 + \frac{2}{3} \frac{D_3^5}{D_3^3} Q_3 \quad (6)$$

where:

$$\begin{aligned} \alpha &= 2A/A^2 + B^2 & \beta &= 2B/A^2 + B^2 \\ A &= n(\epsilon_0^{(1)} - 1) - k\epsilon_0^{(2)} & B &= k(\epsilon_0^{(1)} - 1) + n\epsilon_0^{(2)} \\ \epsilon_+^{(1)} + \epsilon_-^{(1)} &= \epsilon_0^{(1)} & \epsilon_+^{(2)} + \epsilon_-^{(2)} &= \epsilon_0^{(2)} \end{aligned}$$

$2\lambda$  is the spin orbit splitting of the  $A_3$  band,  $D_i^j$  are the deformation potential constants where  $D_1^1$  and  $D_1^5$  describe the bands shift while  $D_3^3$  and  $D_3^5$  describe the matrix elements variation. The quantities  $R_0$ ,  $\epsilon_0^{(1)}$ ,  $\epsilon_0^{(2)}$ ,  $n$  and  $k$  are the reflectivity, the real and imaginary parts of the dielectric function, the refractive index and extinction coefficient for unstrained crystal, respectively.

In the next section we shall try to identify the transitions observed in GaSb as the transitions in the  $A$  direction (or at the  $L$  point) making use of the above formulas. In order to do this we have to show, that  $Q_3$  calculated from experiment using (3) does not contain any contribution arising from the band shifts  $\sim \frac{1}{R} \frac{dR}{d\omega}$  and that  $Q_5$  calculated from experiment does contain this contribution according to (5) and (6).

### 3. Experimental data and analysis

The investigated samples were glued upon a ferrite bar in which the magnetostriction phenomena occurred. Measurements were performed at the resonance point of about 11 kHz. A quartz iodine lamp was used as a light source. An IKS-12 spectrometer was used to obtain the monochromatized light. The reflected light was detected by Zeiss M12 F35 and EMI 6255 SA photomultipliers. Phase-sensitive detection was applied to measure the  $ac$  signal proportional to  $\Delta R$ . The modulated signal  $\Delta I$  and the unmodulated one,  $I$  which was not amplified, were automatically recorded at the same time. The quantity  $\Delta R/R$  was obtained as the ratio of these two signals. The germanium samples were etched in CP4-A and the GaSb samples — in  $\text{NHO}_3 + \text{HF}$  solution at 9:1 ratio. The dimensions of the samples were 2 mm  $\times$  15 mm  $\times$  150  $\mu\text{m}$ . In the remainder of this section we will present the experimental results and quantitative analysis for GaSb.

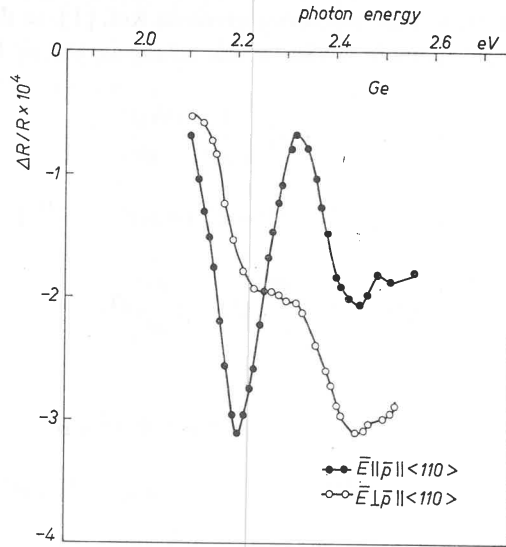


Fig. 1. Relative change of reflectivity of Ge versus photon energy for stress  $\vec{p}$  applied in the  $\langle 110 \rangle$  direction and for two light polarizations ( $\vec{E}$  parallel and perpendicular to the stress direction)

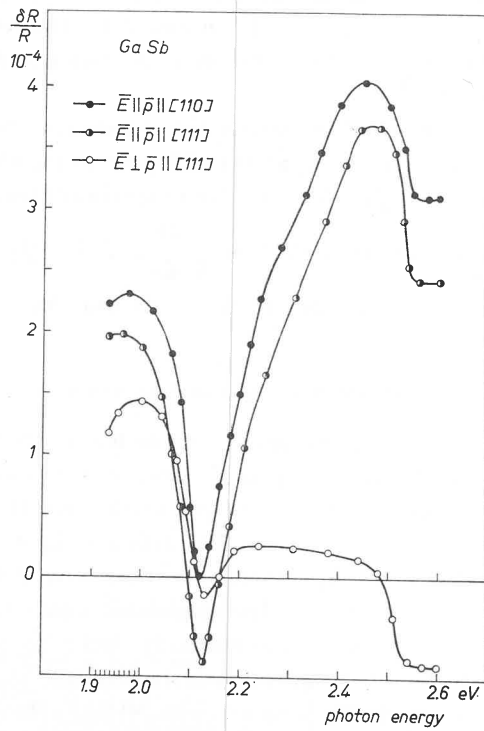


Fig. 2. Relative change of the reflectivity of GaSb versus photon energy. Symbols  $\vec{E}$  and  $\vec{p}$  designate the polarization of light and stress respectively

The measurements of  $\Delta R/R$  were performed for stress in the  $\langle 110 \rangle$  and  $\langle 111 \rangle$  directions using polarized light. The (112) plane was the reflecting plane. The accuracy of the orientation of the crystals was to about 5 deg. The experimental  $\Delta R(\omega)/R(\omega)$  curves are shown in Fig. 2.

The strain tensor  $e_{ij}$  is related with the stress tensor  $\sigma_{kl}$  by the formula

$$e_{ij} = S_{ijkl}\sigma_{kl} \quad (7)$$

where  $S_{ijkl}$  is the elasticity tensor. The stress tensor  $\sigma_{kl}$  is determined by an external uniaxial stress  $\vec{p}$ . Using Eqs (2) and (7) one can express the measured value  $\left(\frac{\Delta R}{R}\right)_{\vec{n}}$  in a given direction  $\vec{n}$  by  $Q_{ij}$  and  $S_{ij}$  ( $S_{ij}$  denotes the simplified fourth-rank tensor  $S_{ijkl}$  by using the same abbreviated notation as in the case of the  $O_{ijkl}$  tensor).

Vector  $\vec{n}$  denotes the direction of the light polarization. Linear combinations of the  $S_{ij}$  and  $Q_{ij}$  tensor components give the symmetrized  $S_j$  and  $Q_j$  functions:

$$\begin{aligned} S_1 &= S_{11} + 2S_{12} \\ S_3 &= S_{11} - S_{12} \\ S_5 &= S_{44} \end{aligned} \quad (8)$$

and analogous relations for  $Q_j$  are valid.

From Eqs (2), (7) and (8) we finally find for our case:

$$\begin{aligned} \left(\frac{\Delta R}{R}\right)_{111}^{111} &= \frac{p}{3}(S_1 Q_1 + 2S_5 Q_5) \\ \left(\frac{\Delta R}{R}\right)_{1\bar{1}0}^{111} &= \frac{p}{3}(S_1 Q_1 - S_5 Q_5) \\ \left(\frac{\Delta R}{R}\right)_{110}^{110} &= \frac{p}{3}(S_1 Q_1 + \frac{1}{2} S_3 Q_3 + \frac{3}{2} S_5 Q_5). \end{aligned} \quad (9)$$

The superscripts design direction of stress  $\vec{p}$  and subscripts — direction of light polarization. From Eqs (9) the functions  $pQ_1(\omega)$ ,  $pQ_3(\omega)$  and  $pQ_5(\omega)$  were calculated after substituting into these equations the measured value of  $\Delta R(\omega)/R(\omega)$  and the following values of  $S_j$  [12]:

$$\begin{aligned} S_1 &= 0.588 \times 10^{-6} \text{ bar}^{-1} \\ S_3 &= 2.076 \times 10^{-7} \text{ bar}^{-1} \\ S_5 &= 2.31 \times 10^{-6} \text{ bar}^{-1}. \end{aligned}$$

We cannot calculate  $Q_1, Q_3$  and  $Q_5$  since the pressure  $p$  has not been measured in our experiment. The functions  $pQ_1$ ,  $pQ_3$  and  $pQ_5$  determined from Eqs (9) are presented in Figs 3, 4 and 5.

If we were able to show that these experimental curves  $pQ_j(\omega)$  are in good agreement with the theoretical  $pQ_j$  calculated from Eqs (4), (5) and (6), the  $\Delta(L)$  origin of the reflectivity structure in GaSb would be proved. For this purpose the theoretical curves described by Eqs (4), (5) and (6) must be fitted to the experimental curves by an appropriate choice of  $D_i^j$ .

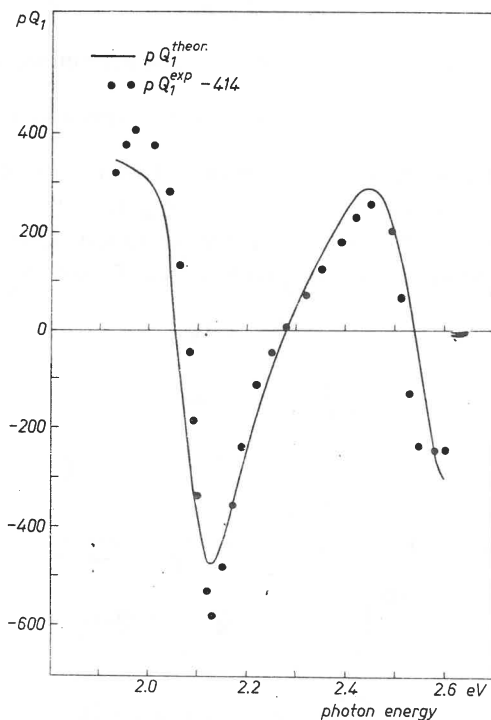


Fig. 3. Experimental  $pQ_1$  function corrected by subtracting the constant  $c_1$  (plotted as points vs photon energy) and the theoretical function  $pQ_1 = 1200 \frac{1}{R_0} \frac{dR_0}{d\omega}$  (solid curve) with  $R_0$  being the reflectivity for the unstrained crystal taken from Ref. [8]

The zero position of the experimental  $\Delta R(\omega)/R(\omega)$  curves and therefore of  $pQ_j(\omega)$  curves can be adulterated in this type of measurement. This zero offset arises from the relative motion of the optical beam and the sample whose surface is not homogeneous.

We have shifted all the experimental curves  $pQ_j(\omega)$  by amounts independent of the wavelength to obtain a good agreement with the theory.

We shall describe now the fitting procedure of each theoretical function  $pQ_j$  to the experimental one:

a) According to Eq. (4) the function

$$a_1 \frac{1}{R_0(\omega)} \frac{dR_0(\omega)}{d\omega} + c_1$$

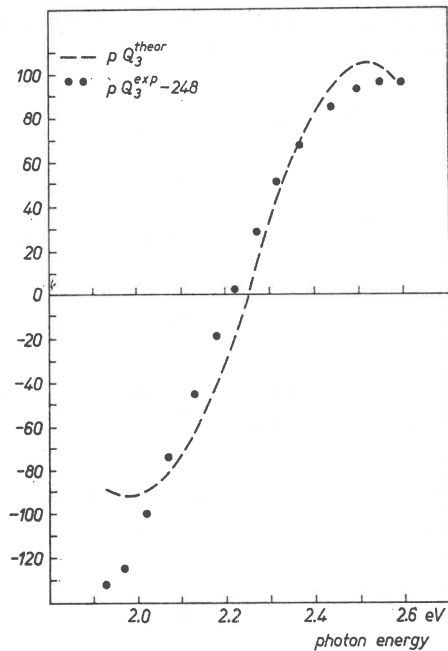


Fig. 4. Experimental  $pQ_3$  function corrected by subtracting the  $c_3$  constant (points) and the fitted theoretical function  $pQ_3 = 600 [\alpha(\epsilon_+^{(1)} - \epsilon_-^{(1)}) + \beta(\epsilon_+^{(2)} - \epsilon_-^{(2)})]$  (dashed curve)

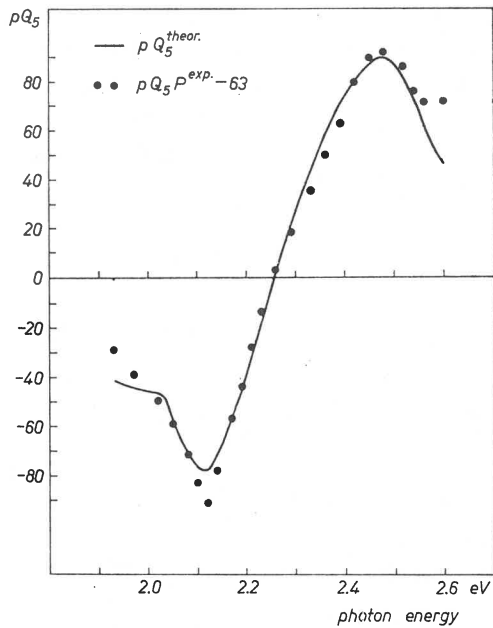


Fig. 5. Fitting of the theoretical function  $pQ_5 = 80 \frac{1}{R_0} \frac{dR_0}{dE} + 430 [\alpha(\epsilon_+^{(1)} - \epsilon_-^{(1)}) + \beta(\epsilon_+^{(2)} - \epsilon_-^{(2)})]$  (solid line) to the experimental function  $pQ_5$  corrected by subtracting  $c_5$  (points)

was fitted to experimental  $pQ_1(\omega)$  where  $a_1 = -\sqrt{3}D_1^1p$  and  $c_1$  is a constant which measures the possible zero offset.

Data of paper [8] were used to obtain  $\frac{1}{R_0(\omega)} \frac{dR_0(\omega)}{d\omega}$ . Both curves, experimental  $pQ_1$  and fitted  $a_1 \frac{1}{R_0} \frac{dR_0}{d\omega}$  are seen in Fig. 3. The curve  $pQ_1(\omega)^{\text{exp}}$  has already been corrected by subtracting  $c_1$  from the experimental value of  $pQ_1$ .

b) The function

$$b_3[\alpha(\varepsilon_+^{(1)} - \varepsilon_-^{(1)}) + \beta(\varepsilon_+^{(2)} - \varepsilon_-^{(2)})] + c_3$$

was fitted to the experimental function  $pQ_3$  according to (5). In order to do this the quantities  $\varepsilon_0^{(1)}$  and  $\varepsilon_0^{(2)}$  must be decomposed into  $\varepsilon_{\pm}^{(1),(2)}$ . We used the following procedure. The contribution to the dielectric function arising from the interband transition between one of the valence band split by spin-orbit interaction  $A_3(L'_3)$  and the conduction band  $A_1(L_1)$  can be written as follows:

$$\varepsilon_{\pm}^{(2)}(\omega) = \frac{1}{\omega^2} \frac{A_{\pm}}{(\omega - E_{\pm})^2 + (\Gamma_{\pm}/2)^2} \quad (10)$$

where  $E_+ = 2.5$  eV,  $E_- = 2.0$  eV.

We have assumed  $\varepsilon_0^{(2)} = \varepsilon_0^{(2)}$  in the vicinity of 2.0 eV. The parameters  $A_-$  and  $\Gamma_-$  were calculated from Eq. (10) for two points near 2.0 eV. Using these constants we were able to calculate  $\varepsilon_-^{(2)}(\omega)$  and  $\varepsilon_+^{(2)}(\omega) = \varepsilon_0^{(2)}(\omega) - \varepsilon_-^{(2)}(\omega)$ . However the function  $\varepsilon_+^{(2)}$  obtained in such a manner did not vanish at 2.0 eV. To improve this, the constant  $A_-$  was modified to obtain a good agreement of  $\varepsilon_-^{(2)} + \varepsilon_+^{(2)}$  with  $\varepsilon_0^{(2)}$  which was taken from Ref. [8]. The decomposition which was obtained, is seen in Fig. 6. In order to decompose  $\varepsilon_0^{(1)}$ , the functions  $\varepsilon_{\pm}^{(1)}$  were written in the form:

$$\varepsilon_{\pm}^{(1)}(\omega) = \frac{1}{\omega^2} f_{\pm}(\omega)$$

where  $f_{\pm}(\omega)$  are Kramers-Kronig dispersion integrals of the functions  $\omega^2 \varepsilon_{\pm}^{(2)}(\omega)$ . Because  $\Gamma_+ \approx \Gamma_-$  we can write:

$$f_+(\omega) = cf_-(\omega - 2\lambda). \quad (11)$$

We have assumed that  $f_+(\omega) = \text{const} = c'$  in the vicinity of 2.0 eV and  $f_-(\omega) = \text{const}$  near 2.5 eV. Using the condition  $\varepsilon_+^{(1)} + \varepsilon_-^{(1)} = \varepsilon_0^{(1)}$  and Eq. (11) the constants  $c$ ,  $c'$ ,  $\varepsilon_+^{(1)}(\omega)$  and  $\varepsilon_-^{(1)}(\omega)$  were successively calculated.

The curve  $pQ_3^{\text{exp}} - c_3$  and fitted curve

$$b_3[\alpha(\varepsilon_+^{(1)} - \varepsilon_-^{(1)}) + \beta(\varepsilon_+^{(2)} - \varepsilon_-^{(2)})]$$

are drawn in Fig. 4.

c) According to (6)  $pQ_5(\omega)$  is to be a linear combination of  $pQ_1$  and  $pQ_3$ .



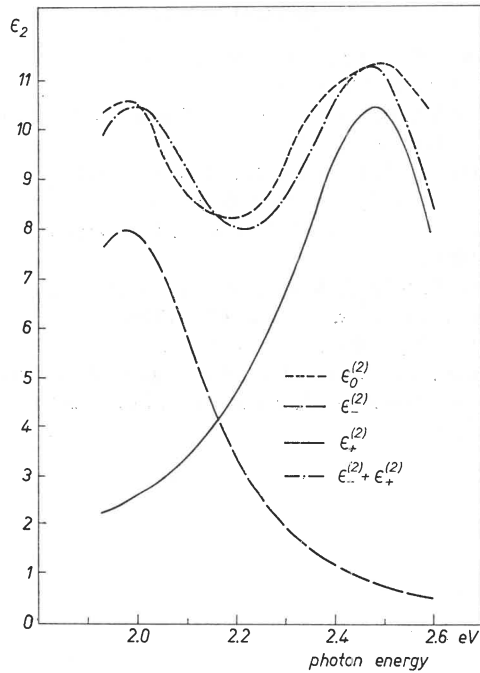


Fig. 6. Decomposition of the imaginary part of the dielectric function  $\epsilon_0^{(2)}$  for the unstrained GaSb crystal

In Fig. 5  $pQ_5^{\text{exp}}(\omega)$  together with the fitted curve

$$a_5 \frac{1}{R_0(\omega)} \frac{dR_0(\omega)}{d\omega} + b_5 [\alpha(\epsilon_+^{(1)} - \epsilon_-^{(1)}) + \beta(\epsilon_+^{(2)} - \epsilon_-^{(2)})]$$

are presented.

The values of fitting parameters are given under the figures. We see in Figs 3, 4 and 5 that agreement between experimental and theoretical curves is good.  $pQ_1$  is in fact the simple derivative of the unstrained  $R(\omega)$ , hence it is due to band shift:  $pQ_1 = pQ_1^{\text{shift}}$ . The  $pQ_3$  function is an excellent proof of the  $A(L)$  symmetry of our transitions even without comparison with the theoretical function. It can be seen that the shape of the experimental  $pQ_3$  is quite different from  $pQ_1$  which is due solely to the energy shift. In the shape of  $pQ_3$  one can not observe any minimum at 2.1 eV, characteristic for  $pQ_1$ .

Hence our  $pQ_3$  is pure  $pQ_3^{\text{mvar}}$  and does not contain the contribution  $pQ_3^{\text{shift}}$  originating from the band shift. The  $pQ_5$  function is in fact a linear combination of  $pQ_1$  and  $pQ_3$  similarly to the case of  $A(L)$  symmetry. It can be then concluded that our results confirm the identification of the transitions in GaSb at 2.0 eV and 2.5 eV as the interband transitions in the  $A$  direction (or at the  $L$  point).

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