PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON THE PHYSICS OF SEMICONDUCTORS, KYOTO, 1966
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 21, SUPPLEMENT, 1966

V-6. Oscillatory Magneto-Absorption in Tellurium

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The fundamental optical absorption and magneto-absorption were investigated on tellurium for light polarized along the c-axis. The experimental results indicate that the interband transition $\varepsilon//c$ is direct-forbidden, with electron-hole interaction. The oscillatory magneto-absorption exhibits a remarkable effect resulting from the lack of the inversion symmetry in tellurium: the intensities of two series of lines are inverted by reversal magnetic field. The magneto-absorption spectrum is quantitatively understood by the contributions of the dipole and quadrupole electric transitions.

§ 1. Introduction

Tellurium exhibits dichroism in the absorption edge: the shape of the absorption curve depends on the radiation polarization with respect to the ternary axis (c-axis) of the crystal.[1] We report experimental results obtained in the absorption edge and magneto-absorption studies carried out with the light polarized parallel to the c-axis. The experiments are performed on very pure single crystals (holes density $10^{13}\mathrm{cm}^{-3}$; mobility $10^5\mathrm{cm}^2\mathrm{V}^{-1}\mathrm{sec}^{-1}$ at 4°K), at liquid helium temperature in a superconducting magnet.

§ 2. Absorption Edge

The experimental results indicate that the interband transition is direct and obeys a selection rule: the transition is allowed for $E\perp c$ and forbidden for $E//c$. Figure 1 shows a plot of the absorption edge for $E//c$. We observe an exciton peak which indicates that the electron-hole interaction has to be taken into account in the absorption process. Beyond the continuum limit, $K(\hbar\omega)$ is satisfactorily fitted with the theoretical expression[2] in the forbidden case:

$$K\propto\pi\alpha(1+\alpha^2)\frac{e^{\pi\alpha}}{\mathrm{sh}\,\pi\alpha}\frac{(\hbar\omega-E_g)^{3/2}}{\hbar\omega}\ ,\quad \alpha^2=\frac{R}{\hbar\omega-E_g}\ .$$

The exciton binding energy R and the energy gap E_g are deduced from this analysis: $R=1.4\pm.2$ meV, $E_g=334.7\pm.2$ meV at 10°K. Extrapolating at zero field the energies of the magnetoabsorption peaks, we obtain $334.8\pm.2$ meV.

Recombination radiation studies performed by C. Benoit à la Guillaume and J. M. Debever[3] are in fair agreement with the absorption data: the spontaneous emission line is polarized perpendicular to the c-axis and the shape of the line indicates that the transition is direct-allowed. Thus we can conclude that the transitions $E\perp c$ and $E//c$ correspond to the same pair of bands. A further proof is provided by magneto-absorption results on the transition $E\perp c$, as we recently

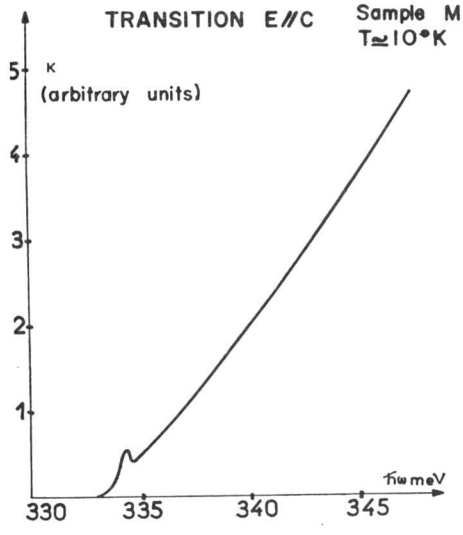

Fig. 1. Absorption coefficient vs. photons energy.

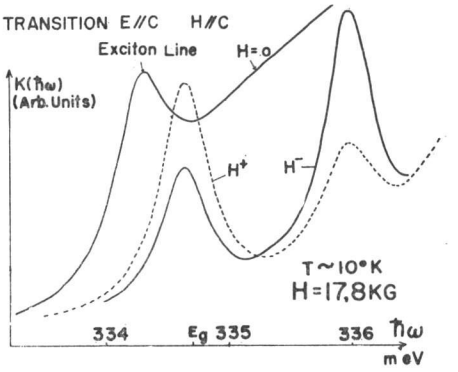

Fig. 2. Detail of the absorption edge near the gap at zero field and for field parallel or antiparallel to c.

observed[4] and obtained the same value of the energy gap.

From group theory, Hulin[5] deduced the selection rules for the electric dipole transitions at different points of the Brillouin zone. The observed selection rule must be valid at the points M and P, which seem the most probable location of the bands extrema. In the case of parabolic bands, the isoenergetic surfaces would consist of two revolution ellipsoids with axes along the *c*-direction, one at the point M and one at the point P, deduced one from the other by symmetry with respect to the center of the Brillouin zone (Kramer's degeneracy). These two ellipsoids correspond to the same energy but to different spin states. Magneto-absorption results are analysed in this model.

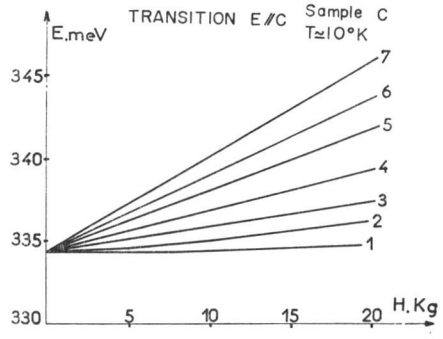

Fig. 3. Magneto-absorption peaks energies vs. magnetic field.

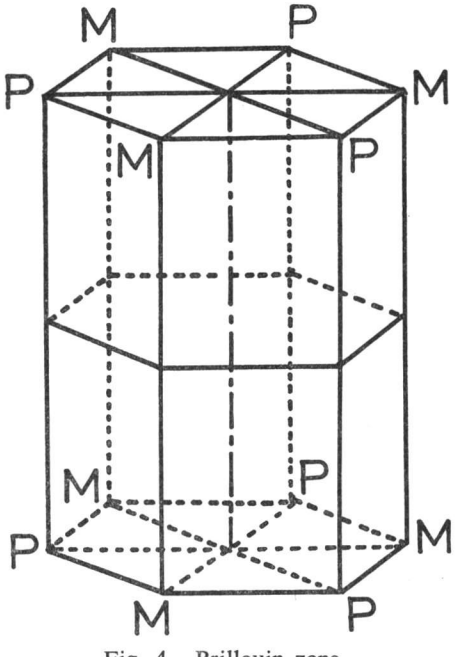

Fig. 4. Brillouin zone.

§ 3. Magneto-Absorption

A. Experimental results

Most of the experiments were performed in the configuration $H//c$, with the photon wave vector $\boldsymbol{q}$ transverse (as indicated in Fig. 6). Using two different orientations for the sample (1 to 4 mm thick), we investigated magneto-absorption for the two possible directions of $\boldsymbol{q}$ ($q//1$: binary axis or $q//2$). In this geometry, a remarkable effect appears, resulting of the lack of inversion symmetry in tellurium: the intensities of pairs of lines are inverted by reversal magnetic field. This effect, observed on the various samples studied and for the two configurations ($q//1$ and $q//2$) of the photon wave vector, is particularly important in low field on the two first lines (Fig. 2), but is observed for the complete spectrum (Fig. 5). At higher field (50 to 60 KG), field reversal interchanges the intensities of weak pairs, but does not affect the stronger peaks.

We show (Fig. 6) that a rotation from 180° of the crystal around the *c*-axis changes one type of spectra into the other. Such an operation is equivalent to the reversal of $\boldsymbol{q}$. Thus we can conclude that the inversion of light propagation gives the same result as the reversal of the field. This indicates that the photon wave vector

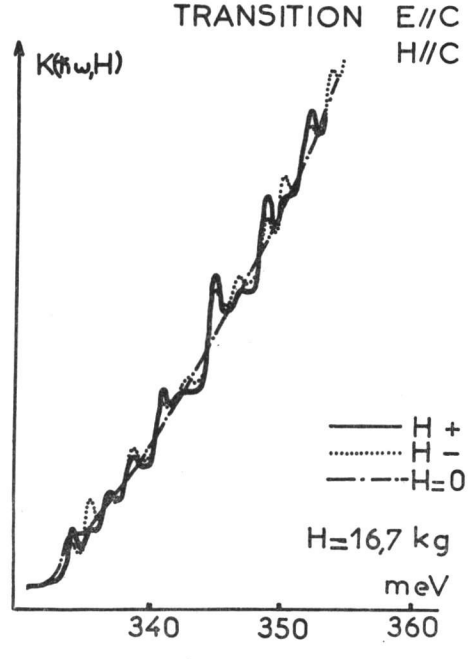

Fig. 5. Magneto-absorption for $E//c//H$ ($H=$ 16.7 KG).

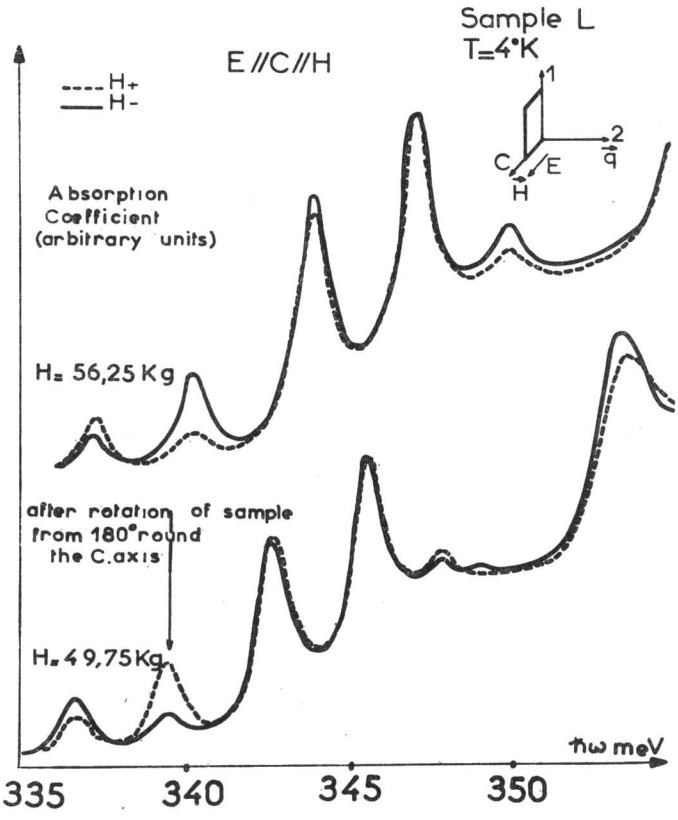

Fig. 6. Magneto-absorption spectrum ($E//c//H$) for the two opposite directions of H along c and after rotation of 180° of the crystal around to the c-axis.

should be taken into account in the interband transition.

In the case $H\perp c$, the energies of the peaks do not depend on the orientation of the magnetic field in the plane perpendicular to the c-axis. This result indicates that the $E(k)=C^{te}$ surfaces must have a revolution axis parallel to the ternary axis.

B. Discussion

The experimental data obtained for $H//c$ are analysed assuming parabolic bands *i.e.* two revolution ellipsoids (one for each spin state) along c. In the case of forbidden transitions, the matrix element:

$$\langle c|\varepsilon e^{iq\cdot r'}\cdot p|v\rangle=[M_{cv}\cdot\pi F_N(r)]_{r=0}$$

neglecting q (dipole electric transition)

$$M_{cv}=\frac{1}{\hbar}[\nabla_k(\varepsilon\cdot p_{cv})]_{k_{\max}}$$

$$=\frac{1}{m}\sum_{\alpha}\left[\frac{p_{c\alpha}(\varepsilon\cdot p_{\alpha v})}{E_c-E_\alpha}+\frac{p_{\alpha v}(\varepsilon\cdot p_{c\alpha})}{E_v-E_\alpha}\right].$$

$F_N(r)$ is the wave function of the relative motion of the electron and hole, $\pi=-i\nabla+(e/2c)H\times r$, $r=r_e-r_H$, $k=k_e=k_H$. The theory predicts oscillatory effects for $H\perp M_{cv}$. From theoretical considerations,[5] we deduce that M_{cv} is perpendicular to $c//H$. Neglecting Coulomb interaction and broadening of levels, the absorption coefficient is given by

$$K\propto\sum_N(N+1)$$

$$\left\{|M^-|^2\left[\hbar\omega-E_g-\hbar\omega_1-\left(N+\frac{1}{2}\right)\hbar\omega^*\right]^{-1/2}\right.$$

$$\left.+|M^+|^2\left[\hbar\omega-E_g-\hbar\omega_2-\left(N+\frac{1}{2}\right)\hbar\omega^*\right]^{-1/2}\right\},$$

$M^\pm=M_x\pm iM_y(H//c//z)$,

$\omega^*=\omega_1+\omega_2$ (ω_1, ω_2: cyclotron frequencies of the electrons and holes).

The selection rules for the dipole electric transitions are $\Delta N=\pm1$ and two series of peaks with intensities increasing as $N+1$ are predicted by the theory.[6,7] The quadrupole electric transitions are depicted by the first order in q of the matrix element. Such transitions correspond to

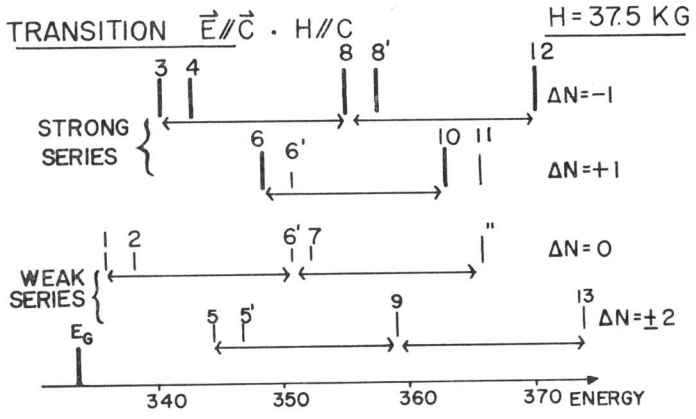

Fig. 7. Energies of the lines observed at 37.5 KG ($H//c$).

the selection rules $\Delta N=0, \pm 2$.

Taken into account spin effects, each series of lines should be split into two components separated by $g^*\mu_B H$ (g^* is a combination of the g factor of the two bands). The energies of the magneto-absorption lines are indicated in Fig. 7 ($H=37.5$ KG): two series of strong lines, with regular spacing, and split into two components (field reversal does not alter the intensity distribution at high field). We assume that these lines correspond to the dipole electric transition $\Delta N=\pm 1$. The spectrum exhibits in addition two series of weaker doublets, the intensities of which are inverted by field reversal. These lines have the expected energies corresponding to the quadrupole electric transitions $\Delta N=0$, $\Delta N=\pm 2$. Magneto-absorption results[4] on the allowed transition $E\perp c$ support this interpretation: the series $\Delta N=0$ are observed (lines 1 and 2) but $\Delta N=\pm 1$ do not occur (lines 3 and 4). Each doublet would result from the spin splitting with the value $g^*=11$. The transverse effective masses for the electrons and holes are deduced from this analysis: $m_{1\perp}=.115\pm.004\, m_0$, $m_{2\perp}=.038\pm.002\, m_0$. The hole transverse effective mass determined by cyclotron resonance[8,9] ($.119\pm.006\, m_0$) is in good agreement with the result obtained for $m_{1\perp}$.

The non-linear dependence (observed on the first lines) for the peaks energies as a function of magnetic field results from the Coulomb interaction.

Elliott and Loudon's calculations[10] developed in the approximation $(\hbar\omega^*/2R)\gg 1$ predict that the main intensity in each magnetic sub-band transition is displaced to the lowest exciton line and the absorption in the continuum is reduced to an insignificant shoulder. The observed peaks correspond probably to the transitions in the bound states, which are shifted below the energies $\{N+(1/2)\}\hbar\omega_c$. The binding energy increases less rapidly than linear with the field and explains qualitatively the non-linearity observed.

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