Sci. Annals. Fac. Phys. & Mathem., Univ. Thessaloniki, **20b**, 137 (1980)

STUDY OF OPTICAL PROPERTIES: DIRECT AND INDIRECT ABSORPTION EDGES

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Abstract. *A method is proposed for the study of optical properties of layered crystals from Fabry-Perot interference fringes. The optical properties of $SiTe_2$, GeS and GeSe have been studied in the vicinity of direct and indirect band gaps. The electron-phonon interactions at the inter band transitions required the participation of LO phonons, whose energies have been estimated.*

1. INTRODUCTION

Considerable interest has recently been raised on the optical properties of layer semiconducting compounds. These compounds crystallize in a layered structure; the bonding between layers is of the Van der Waals type, while the one within each layer is predominantly covalent. The usual method for determining the optical constants of these materials has been a separate determination of the optical absorption constant and the reflectivity. However the anisotropy of these crystals and the accurate determination of the reflectivity introduce difficulties. Nevertheless the discussion of the reflection and absorption spectra in the region of the absorption edge is impossible without the precise knowledge of the extinction coefficient k(λ) and the index of refraction n(λ) especially when the details of the absorption edge are sought.

Therefore the study of the optical properties of layered structure compounds have attracted considerable attention in recent years because of their strong anisotropy along one the crystal axis (usually the c-axis). The band structure of layered compounds, mainly GaS, SaSe has been calculated by Bassani and Pastori Paravicini [1] and also by Kamimura and Nakao [2]. The main feature of these calculations is that they proved the existence of a two-dimensional band structure.

Measurements of the absorption spectrum of these compounds by H. Kamimura et al. [3] and by M. Grandolfo et al. [4] proved the existence of excitons, with limited extension along the c-axis so that they may be consideted as two-dimensional excitons. The compounds MoS_2, $MoSe_2$ and $SnSe_2$ are layered structures too. Their optical properties have been studied extensively by Evans and Young [5], Evans and Hazelwood [6] and Hazelwood [7].

Among the layered compounds with a CdI_2 structure is $SiTe_2$ belonging to IV_a-VI_a group. The optical features of these compounds have been studied by Vennic and Callaerts [8] and Rau and Kannewurf [9]. Experimental results for this compound were obtained [10] by applying interference measurements. The analysis of these results according to the method proposed by Hazelwood [7] led to a more detailed spectrum, that permitted a reassignment of values included in the indirect and direct transition edges.

Tin Sulfide, Tin Selenide, Germanium Sulfide and Germanium Selenide are isomorphous and crystallize as a deformed Sodium Chloride structure. The original structure assignement was made by Hofman [11], who described the unit cell as orthorhombic (D_{2h}^{16}), composed of four molecules. The optical behaviour of the Tin Sulfide, in the vicinity of the absorption edge has been investigated by Albers et al. [12]. Bilenkii et al. [13] investigated the optical properties of thin Tin Sulfide films with natural light. Lambros et al. [14] found that the optical behaviour of this compound and its isomorphous is strongly anisotropic, as its isomorphic Tin Selenide, that was investigated by Arai T. et al. [15].

Thermoelectric Measurements by A. S. Okhotin et al. [16], in Germanium Selenide indicate that the phonon contribution is very important using unpolarised light Kannewurf and Cashman worked on the absorption coefficient of the same compound in Single Crystals. Katti et al. [18] carried out electrical and optical measurements on amorphous films. Asanabe and Okazaki [19] carried out electrical resistivity and Hall coefficient measurements on Germanium Selenide and estimated the energy gap of this material. Vlachos et al. [20] studied the optical properties of this compound in the vicinity of the absorption edge taking into acount the crystal symmetry.

Finally IR Reflectivity measurements have been made by Siapkas et al. [21] in GeSe as by J. M. Chamberlain et al. in SnS [22].

2. DETERMINATION OF OPTICAL CONSTANTS

We present a modification of the method proposed mainly by Hazelwood [7] for the calculation of the optical constants from the modulation of transmitted light due to interference effects. Let us consider a crystal of thickness d and a complex refractive index $N = n + ik$. The transmittance of the crystal e.g. the ratio of the transmitted intensity to the incident, $T = T(\lambda, n, k, d)$. The transmitted intensity is the result of the interference of a large number of coherent rays [23]. So the transmitted energy T is:

$$T = \frac{(1 - R)^2 \left(1 + \frac{k^2}{n^2}\right)}{\left(e^{\frac{4\pi k}{\lambda} d} - R e^{-\frac{4\pi k}{\lambda} d}\right)^2 + 4R\sin^2(\delta + \psi)} \qquad (1)$$

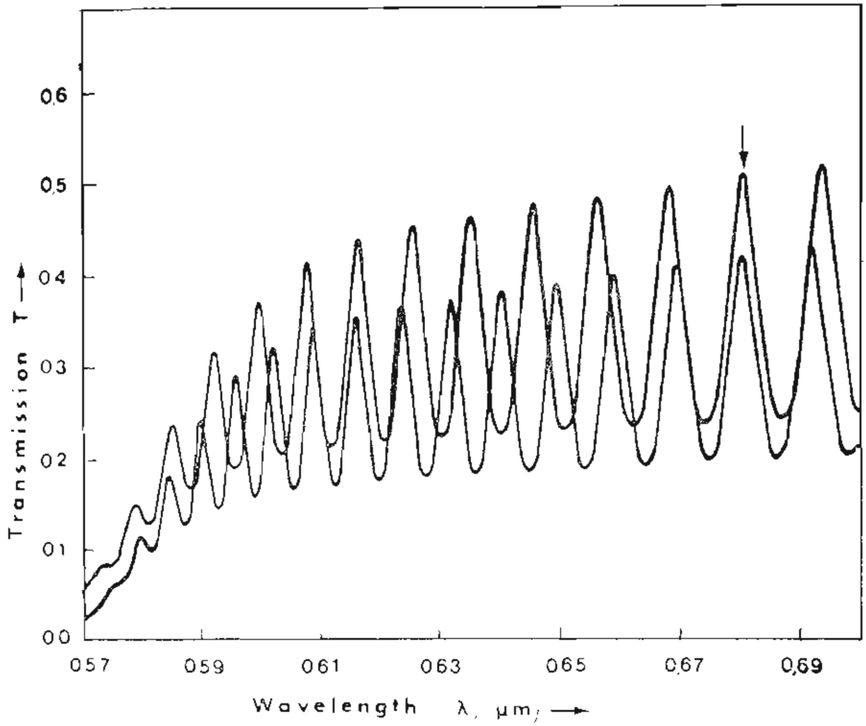

Fig. 1

where the parameters R and Ψ have the values

$$R = \frac{(n-1)^2k^2}{(n-1)^2+k^2} \qquad \Psi = \frac{2k}{n^2+k^2-1} \qquad (2)$$

The parameter R is known as the reflectance of the crystal. The transmittance spectra consists of interference frings (Fig. 1). The condition for a maximum in the transmittance pattern, for normal incidence is:

$$j\lambda = 2nd \qquad (j = 0,\ 1,\ 2,\ \ldots) \qquad (3)$$

W. J. Choyke and L. Patric [24] determine the fringe order j and after that the real part of the refractive index by the equation:

$$j = \frac{\lambda_2}{\lambda_1 - \lambda_2} \qquad (4)$$

Where λ_1, λ_2 are the wavelength of two consequent fringes in the longer wave length part of the spectrum.

Another way to determine the fringes order is to analyse simultaneously the spectra of two crystals. In the spectrum of the first crystal, one fringe is chosen arbitrarily as the zero order one. With his zero order fringe as a reference it is possible to enumerate the consecutive fringes. If, for a certain fringe, the obtained arbitrary order is $J_1(\lambda)$, this would be related to the true order $j_1(\lambda)$ by the following relation:

$$j_1(\lambda) = J_1(\lambda) + A_1 \qquad (5)$$

So that for the fringes in the two spectra the following relation exists between the true and the arbitrary order.

$$\begin{aligned} j_1(\lambda) &= J_1(\lambda) + A_1 = \frac{2d_1n(\lambda)}{\lambda} \\ & \qquad\qquad\qquad\qquad\qquad (6) \\ j_2(\lambda) &= J_2(\lambda) + A_2 = \frac{2d_2n(\lambda)}{\lambda} \end{aligned}$$

where A_1 and A_2 are integral numbers to be determined. Dividing the above equations by parts the result is:

$$\frac{j_1(\lambda)}{j_2(\lambda)} = \frac{J_1(\lambda)+A_1}{J_2(\lambda)+A_2} = \frac{d_1}{d_2} \qquad (7)$$

Let's consider that in the spectra of two crystals A and B, two fringes have maxima at the same wave length. These fringes are assigned as zero order fringes and with these as a reference we enumerate the rest. Since in this case $J_1(\lambda) = J_2(\lambda) = 0$ it follows that:

$$\frac{j_1(\lambda)}{j_2(\lambda)} = \frac{A_1}{A_2} = \frac{d_1}{d_2} \tag{8}$$

If there is a second wavelength at which the maxima of the fringes in the two spectra coincide, we obtain:

$$\frac{j_1(\lambda')}{j_2(\lambda')} = \frac{j_1(\lambda') + A_1}{j_2(\lambda') + A_2} = \frac{d_1}{d_2} = \frac{A_1}{A_2} \tag{9}$$

from which

$$\begin{aligned} J_1(\lambda') &= kA_1 \\ J_2(\lambda') &= kA_2 \end{aligned} \tag{10}$$

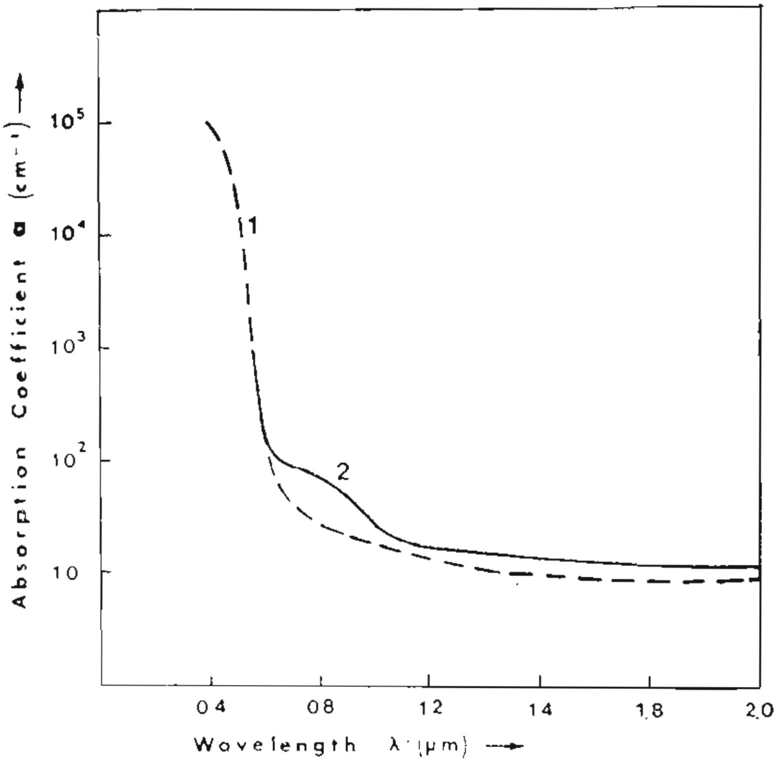

Fig. 2

where k is an integer with a value 0 for the abitrary defined as zero order fringes, for the first coinciding maxima etc.

Thus the real order of the arbitrary is determined as the zero difined fringe.

The transmittance T, expressed by equation (1), is a function of the parameters d, λ, n, k, where d and n are determined independently. So at least the transmittance is a function of the imaginary part of the refractive index, e.q. T = T(k).

In order to determine the value of k we have used a method which is based on the calculation of the real value by successive approximations, using computer processing [25]. In Fig. 2, the results of the absorption coefficient calculated by this method are compared to the results obtained previously by Raw and Kannewurf [9]. The agreement is satisfactory, while as it is easily deducted the fine details of the spectrum are missing from the results of Ran et al., a fact that points out the superiority of the proposed method.

In Fig. 3 we can see the spectral variations of the absorption coefficient which is in a complete agreement with what was obtained by

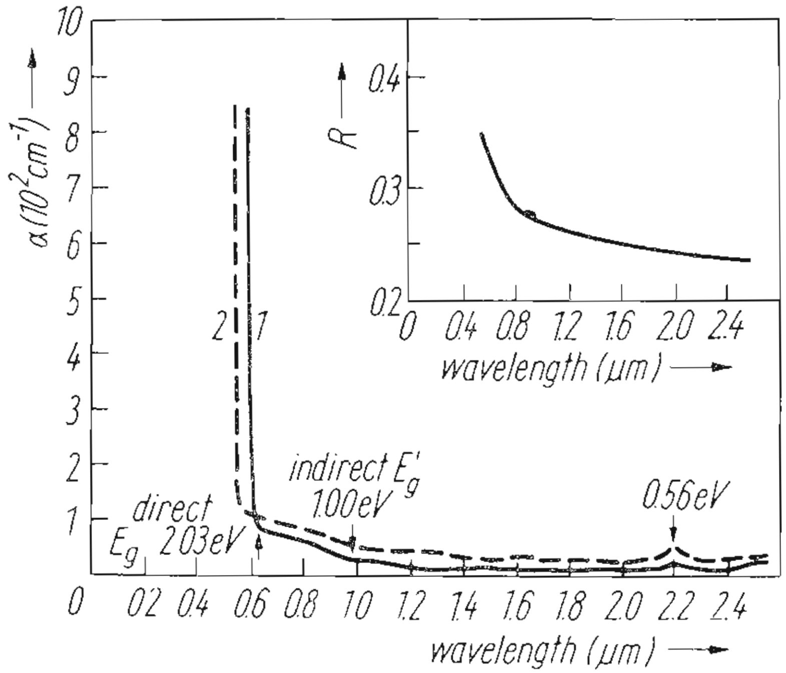

Fig. 3

the Vennic and Callaerts [25] measurements. Also in this spectral we can see the fine details as direct and indirect energy gaps.

The values of the direct energy gap are very close to those of Ran and Kannewurf and also to the obtained by infrared and Raman's study by U. Zwick and K. H. Rieder [27].

3. ANISOTROPIC INDIRECT ABSORPTION EDGE

The IV-VI Semiconductors SnS, SnSe, GeSe, and GeS form an interesting class of isomorphic materials which are characterized by an anisotropic indirect absorption edge. The main absorption mechanism is due to indirect interband electronic transitions with participation of two different phonons. The behaviour of the anisotropic indirect absorption can be accounted by assuming that indirect interband optical transitions are the main absorption mechanism. Thus, Macfarlane et al. [28] proposed that the absorption coefficient in the region of indirect absorption edge could be expressed as a sum of four independent terms as follows:

$$\alpha = \sum_{i=1}^{2} (\alpha_{ai} + \alpha_{ei}) = \alpha_{a_1} + \alpha_{a_2} + \alpha_{e_1} + \alpha_{e_2} \tag{11}$$

Analytically this equation can be written as follows:

$$\alpha = \sum_{i=1}^{2} \left[\frac{\beta_{ai}}{h\nu} \frac{1}{e^{\theta_i/T} - 1} (h\nu - E_g' + k\theta_1)^2 + \frac{\beta_{ei}}{h\nu} \frac{1}{1 - e^{-\theta_i/T}} (h\nu - E_g' - k\theta_i)^2 \right]$$

The terms α_{ai} and α_{ei} are associated with a mechanism involving absorption of phonons and emission of phonons respectively. θ_i is the phonon temperature which is correlated with the coefficients β_{ai} and β_{ei}. So the absorption results can be described by an equation of the form:

$$(\alpha h\nu)^{1/2} = b_0 + b(h\nu - E_1) + \sum_{i=1}^{2} [b_{ai}(h\nu - E_g' + h\omega_i) + b_{ei}(h\nu - E_g' - h\omega_i)] \tag{13}$$

where

$$\frac{\beta_{ai}}{\beta_{ei}} = \rho_i \, e^{\theta_i/T} , \qquad \rho_i = \left(\frac{b_{ai}}{b_{ei}}\right)^2 \tag{14}$$

The energies of the two phonons involved in the four indirect transition mechanism, are obtained from:

$$h\omega_1 = \frac{E_3 - E_0}{2} \quad \text{and} \quad h\omega_2 = \frac{E_2 - E_1}{2} \tag{15}$$

where E_0, E_1, E_2, E_3 are the photon energies at which discontinuities in the curves $(\alpha h\nu)^{1/2}$ against $h\nu$ occur. The exact values of energies at which discontinuities occur can be found from the plotting of $d(\alpha h\nu)^{1/2}/d(h\nu)$ against $h\nu$. The value of the indirect energy gap E_g' is obtained by these experimental curves using the expression:

$$E_g' = \frac{E_0 + E_3}{2} = \frac{E_1 + E_2}{2} \tag{16}$$

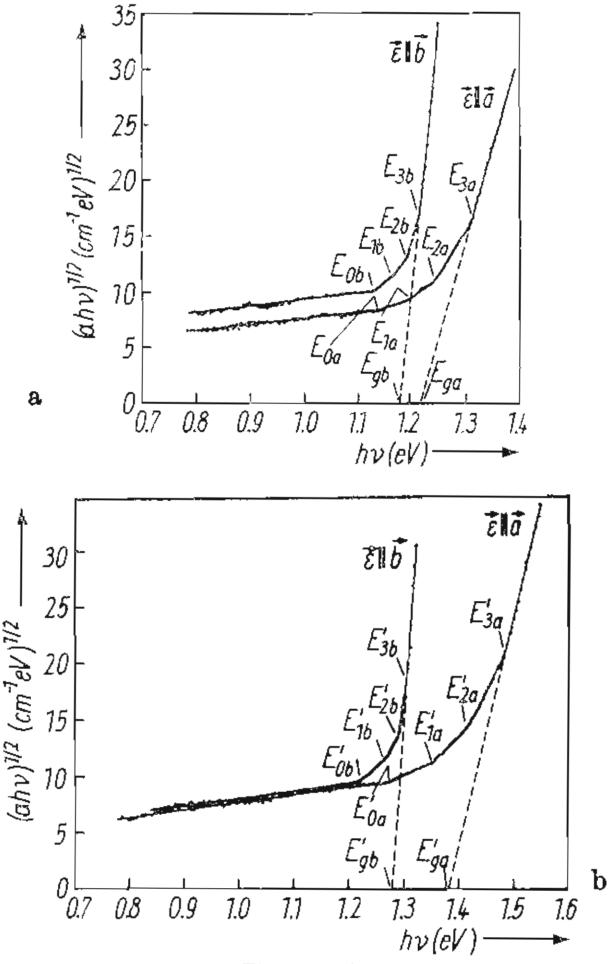

Fig. 4a, 4b

Also the indirect energy gap E_g' is obtained by the intercept with hν axis, in the plott $(\alpha h\nu)^{1/2}$ against hν, of the straight line corresponding to photon energies $h\nu >> E_3$.

In Fig. 4 we can see High-resolution absorption curves for GeSe at 300⁰K and 100⁰K for the two directions of light polarization corresponding to the main axis E || a and E || b.

Every curve decomposites into the five various components corresponding to the terms of equation (13) Fig. 5.

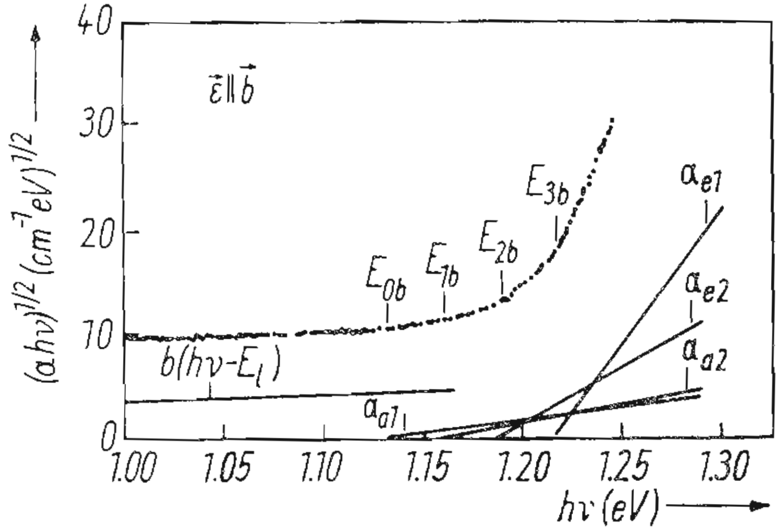

Fig. 5

Also in Fig. 6, using Koskin et al. [29] idea, we obtain plots of quantity $(\alpha h\nu)^{1/2}$ vs pretented energy for two direction of light polarization (E || a, E || b) in SnS at 300⁰K Fig. 6.

A summary of values for the various parameters of GeSe and which are described in this text is presented in Table I.

The present values of indirect band gap of GeSe, at T = 300⁰K for the two directions of light polarization that is $E'_{ga} = 1.22$ and $E'_{gb} =$ 1.18eV compared with the value of 1.2eV obtained by Mande et al. [30] and with the value of 1.16eV obtained by Kannewurf and Cashman [17] using unpolarized light.

In SnS the indirect energy gaps for the two different polarizations are 1.14eV and 1.05eV respectively. These values are in good agreement

TABLE 1

Summary of values for the various parameters of GeSe and SnS in 300⁰ K

Symbols of the parameters	$\varepsilon \parallel a$		$\varepsilon \parallel b$	
	GeSe	SnS	GeSe	SnS
E_o	1.131	1.030	1.133	1.013
E_1	1.195	1.100	1.164	1.060
E_2 (eV)	1.241	1.175	1.190	1.125
E_3	1.308	1.255	1.219	1.175
b_o	5.20	1.34	6.00	2.50
b_{a1}	11.31	11.25	25.76	17.02
b_{a2} $(eV.cm)^{-1/2}$	20.29	25.27	34.98	27.15
b_{e2}	46.54	51.34	108.23	68.47
b_{e1}	84.08	115.96	257.53	145.64
β_{a1}	3.86	7.66	3.86	6.62
β_{a2}	0.67	0.90	0.79	2.80
β_{e2} $x10^{3}(eV.cm)^{-1}$	1.34	1.36	4.61	2.95
β_{e1}	6.84	8.25	53.75	19.60
ϑ_1 (^oK)	1033.00	1311.00	499.00	952.00
ϑ_2	290.00	441.00	150.00	383.00
E'_g	1.22	1.14	1.18	1.09
E'_{g03} (eV)	1.22	1.14	1.18	1.09
E'_{g12}	1.22	1.14	1.18	1.09
$\hbar\omega_1$ (eV)	0.089	0.110	0.043	0.083
$\hbar\omega_2$	0.023	0.038	0.013	0.033

with the values 1.07 and 1.11eV reported by Albers et al. [12], [31].

The values 0.013 and 0.023eV found at room temperature for the low-energy phonons of GeSe are compared favourably with respective LO phonons energies 0.011 and 0.025eV observed in the far infrared spectrum [21].

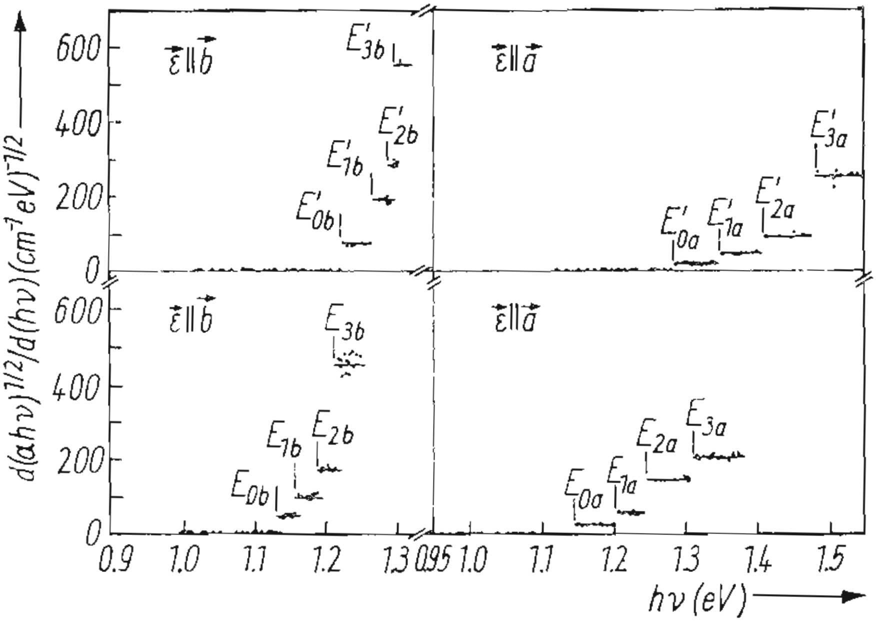

Fig. 6

Haas and Comber [32] reported that the frequency of the longitudinal waves in SnS, is expected to correspond to 320 cm^{-1} which is the value of the low energy phonon involved in the indirect transitions.

4. EXPONENTIAL ABSORPTION EDGES

Recent examination of the detailed behaviour of the absorption coefficient in anisotropic materials by Brada et al. [33] and also by Labros et al. [34] has shown that it varies at the absorption edge in accordance with Urbach's rule [35]

$$\alpha = \alpha_0 \exp \frac{\sigma(h\nu - h\nu_0)}{kT} \tag{17}$$

where α is the absorption coefficient,
h, ν, kT have their usual meaning
α_0 and $h\nu_0$ are material constants
the factor σ which is expressed by the relation

$$\sigma = \sigma_0 (\frac{2kT}{\hbar\omega}) \tanh (\frac{\hbar\omega}{2kT}) \tag{18}$$

changes with direction and depends on the temperature. Sumi and Touozawa [36] reported that σ_0 is a constant describing the ionic character of the material and ω is the angular frequency of the participating phonon. Brad et al. [33] and Vlachos et al. [34] have shown that σ_0 changes at a certain temperature and this is related with a switch-over to a different phonon. Also Vlachos et al. reported that a value for σ_0 smaller than unity indicates the existence of excitonic states lying lower than free exitons. Therefore σ_0 expresses in a way the ionic character of the material.

Extrapolating the straight lines, in the plotting of $\log\alpha$ vs $h\nu$, they converge to a local point ($\log\alpha_0$, $h\nu_0$) which is the same with the local point of the isoabsorption curves. Robert et al. [37] reported that the isoabsorption curves show a linear dependence for temperatures above a certain temperature Tl obeying the relation

$$(h\nu_0)' = (h\nu_0) - \Delta(h\nu) - Te \frac{\partial E_g}{\partial T} \tag{19}$$

where $\Delta(h\nu)$ is the spread of energy values that result from the curvature iso-absorption curves for $T < T\rho \cdot (h\nu_0)'$ is the exciton peak energy and $\partial E_g/\partial T$ the variation of the energy gap with temperature.

Bredner [38] and also Evans and Hazelwood [6] argue that the absorption coefficient in the vicinity of the direct absorption edge layered compounds is independent of the phonon energy. Thus the curve representing the behaviour of a vs $h\nu$, in the region near energy gap, should be a straight line perpendicular to energy axis. This is almost true in the case of $SiTe_2$ only for low temperature data.

In Fig. 7 we can see the application of Urbach's rule in $SiTe_2$ [10] with a good fitting to a straight line. The slopes of the straight lines are temperature dependent. This fact indicates that the absorption in the vicinity of the direct absorption edge is best described by Urbach's rule.

TABLE II

Values of parameters which fit the experimental results of the exponential behaviour of absorption in the vicinity of the dirent energy gap of layered compounds $SiTe_2$, GeSe, GeS.

Symbols of the parameters	$\mathcal{E} \parallel b$			$\mathcal{E} \parallel a$				
	$SiTe_2$	GeS	GeSe	GeS			GeSe	
				$108\sim152\,^{o}K$	$193\sim228\,^{o}K$	$257\sim300\,^{o}K$	$100\sim200\,^{o}K$	$200\sim300\,^{o}K$
$\log \alpha_o$	6.16	5.8	5.0	6.19	5.24	4.54	3.64	3.99
$h\nu_o$	2.48	1.8	1.38	2.50	2.22	2.04	1.62	1.60
$(h\nu_o)'$ eV	2.37	1.75	1.34	2.48	2.19	1.83	1.54	—
$\Delta(h\nu)$	0.11	0.10	0.08	0.19	0.175	—	0.06	—
$T_\ell^{o}\,(^{o}K)$	175	258	237.5	258	258	—	150	—
$\partial E_g/\partial T\,(eV\,^{o}K)^{-1}$	-1.22×10^{-3}	-5.8×10^{-4}	-5.05×10^{-4}		-8.1×10^{-4}			
σ_o	0.56	1.05	1.01	0.65	0.78	0.27	0.27	0.36
$h\nu$ eV	0.027	0.034	0.024	0.044	0.040	0.016	0.022	0.013

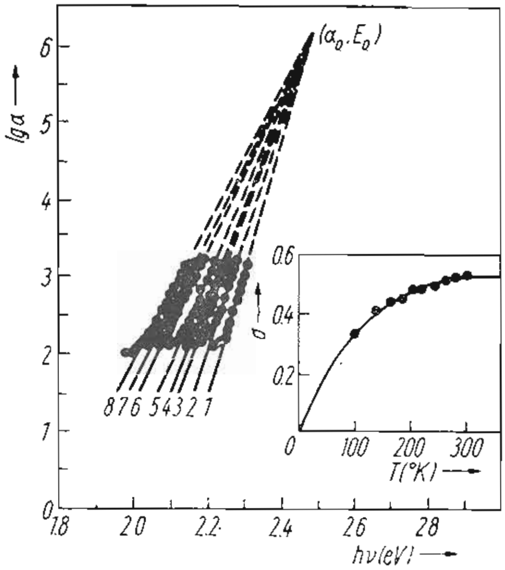
lgα
(α_0, E_0)
0
1
2
3
4
5
6
8 7 6 5 4 3 2 1
1.8
2.0
2.2
2.4
2.6
2.8
hν(eV)
σ
0.2
0.4
0.6
0
100
200
300
T(°K)

Fig. 7a

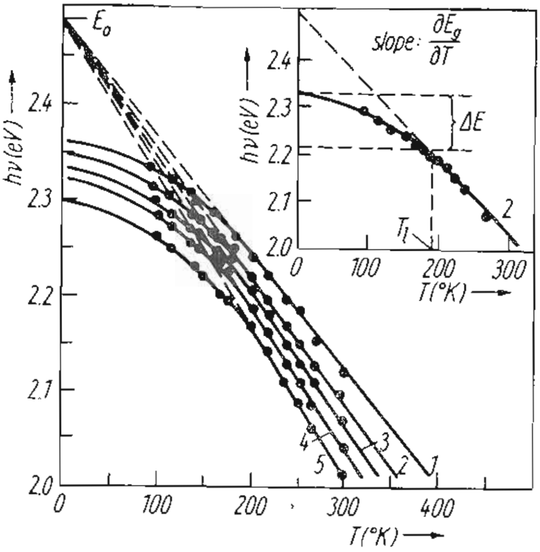
E_0
hν(eV)
2.0
2.1
2.2
2.3
2.4
0
100
200
300
400
T(°K)
1
2
3
4
5
slope: $\frac{\partial E_g}{\partial T}$
ΔE
T_1
2

Fig. 7b

Also in anisotropic semiconductors like GeSe and GeS the semilogarithmic plot of absorption vs energy has a better fitting to a straight line Fig. 8. But there are differences according to light polarization along the a and b axis.

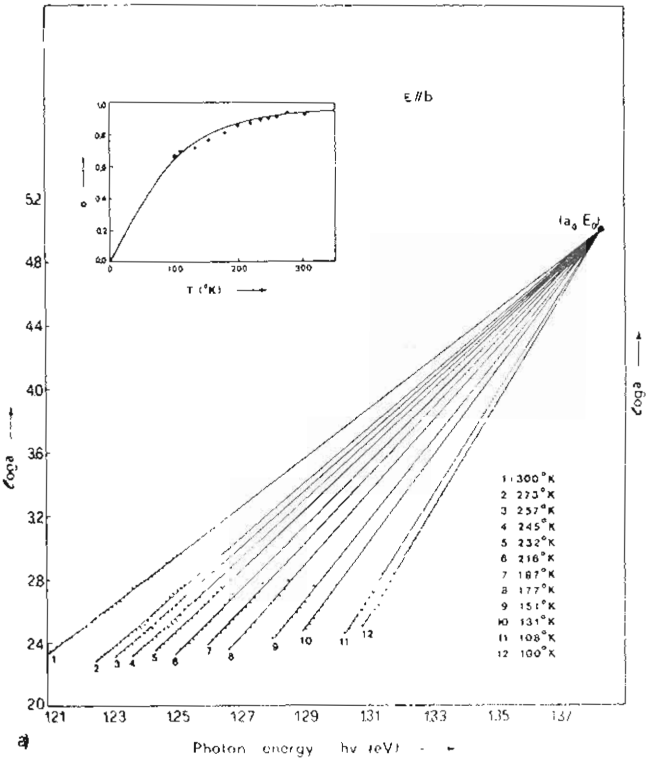

Fig. 8

In the table II we can see the values of the various parameters that are obtained from above, Urbach's rule analysis. These results indicate that for $SiTe_2$ there is an exciton peak energy $(h\nu_0)' = 2.37$eV while the direct energy gap of $SiTe_2$ was found close to 2.03eV at 300°K.

The above results for GeSe indicate that there is a change in the coupling constant indicated by the change in σ_0, and also a switch-over to a different phonon participating in the interaction. Siapkas et al. reported that far infrared studies indicated the existence of three LO phonons along a axis. Situated at 91.179 and 224cm^{-1} respectively.

So the switch-over mechanism in GeSe involved two LO phonons. In the case of GeS [34] three different values of phonon energy are obtained along a axis 0.016, 0.040, 0.044. Also group theoretical analysis indicates that three LO frequency modes are active in the a direction. Wiley et al. [39] have identified one LO phonon (0.015eV or 123.5cm^{-1}) and also observe a second broad peak covering the range 250 — 320cm^{-1}. The calculated LO_2 mode was 0.040eV (325.0cm^{-1}). The three phonons which have a value about 0.044eV justify the view expressed by Wiley et al. that the two phonons are nearly degenarate so that they could not be resolved experimentally in reflection experiments.

So the electron phonon interaction in GeS involves all LO phonon modes expected in the directions studied. Generally the values of the phonons obtained from Urbach's rule application agree with the values obtained in reflection measurements.

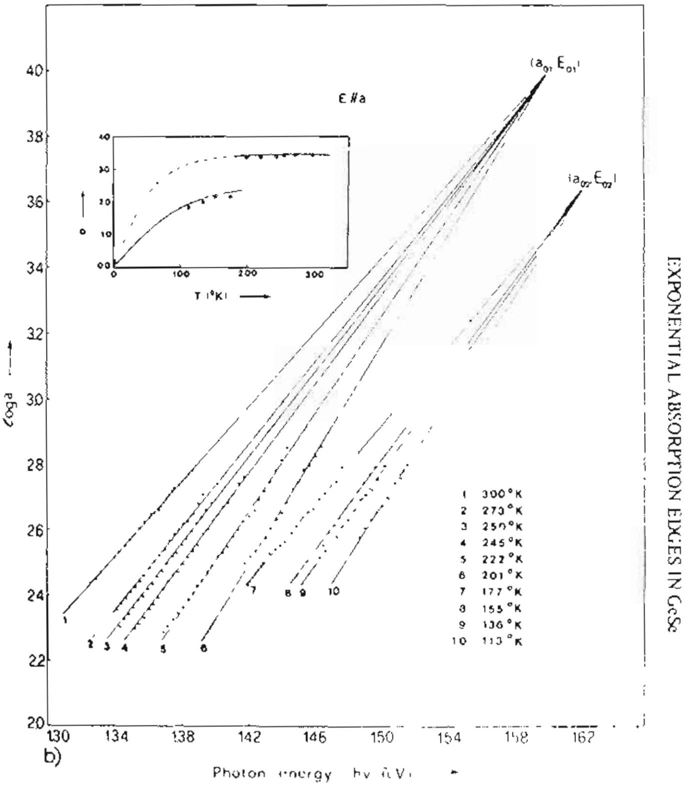

Fig. 8b

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ΠΕΡΙΛΗΨΗ

ΜΕΛΕΤΗ ΟΠΤΪΚΩΝ ἸΔΪΟΤΗΤΩΝ ΑΜΕΣΟ ΚΑΪ ΕΜΜΕΣΟ ΕΝΕΡΓΕΪΑΚΟ ΧΑΣΜΑ

Ὑπὸ

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Γιὰ τὴν μελέτη τῶν ὀπτικῶν ἰδιοτήτων φυλλομόρφων ἡμιαγωγῶν προτείνεται μία μέθοδος ποὺ βασίζεται στοὺς κροσσοὺς συμβολῆς τῶν φασμάτων ὀπτικῆς διαπερατότητος. Μελετήθηκαν οἱ ὀπτικὲς ἰδιότητες τῶν $SiTe_2$, GeS καὶ GeSe στὴν περιοχὴ τοῦ ἄμεσου καὶ ἔμμεσου ἐνεργειακοῦ χάσματος. Οἱ μεταπτώσεις ἠλεκτρονίων μεταξὺ τῶν ζωνῶν γίνονται μὲ τὴν συμμετοχὴ LO φωνονίων τῶν ὁποίων ὑπολογίστηκαν οἱ τιμές.