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Some modern semiconducting materials

1. INTRODUCTION

Although the elemental semiconductor silicon and the compound GaAs continue to dominate in the production of devices, other binary and ternary compounds and alloys are becoming more and more interesting for practical use. The number of semiconducting materials which it is possible to make, is almost unlimited, especially if, besides the inorganic materials, one includes organic semiconductors. Improvements in the technology of producing high quality single crystals in which the concentration of structural defects is very low, has greatly contributed to the possibilities of making devices with semiconducting alloys and compounds. Moreover one can say that binary and ternary semiconductors hold promise for a future technical breakthrough, which should increase their use. With the improvement in the technology of making high quality new materials whose properties should be "ordered" by the user there is a big chance that the cost of these materials will be decreased at the same time so that their production will become economical too.

Several years ago Hall [1] (1974) made large Ge crystals of exceptional perfection and purity whose net concentration of electrically active impurities was below 2×10^{10} cm^{-3}. The properties of these well known material were so improved that it could almost be regarded as a new materials which could be used for making modern detectors for high resolution gamma and X-ray spectrometry. Thus one can say that modern semiconductors need not be quite new materials. They can be quite well known materials such as Ge, which it is now possible to produce at a suitable level of quality for making devices. Some of the so called new materials have not been made before in large enough single crystal shapes for example: SnS, GeS, GeSe, $GeSe_2$ etc. Others like diamond can now be made in a semiconducting form using suitable dopants.

The properties of several groups of "new" and modern semiconducting materials which have been recently studied and are of some promise for making devices in the near future, will now be considered. We will emphasize the material which are most promising, some of which have been investigated in our laboratory.

9

2. ELEMENTAL MATERIALS

Large germanium crystals have been recently made of extremely high purity and perfection containing a carrier concentration of about 2 x10 cm^{-3} and 1200-2000 dislocations per cm^2, (Fig. 1). In addition one chould underline the increasing interest in semiconducting diamond. Although rare natural semiconducting diamond was discovered more than 20 years ago [2] , and synthetic diamond was made soon afterwards [3] , only recently has high quality semiconducting diamond been made. Generally ion implantation has been used [4,5,6] and tne synthesis of SiC on diamond [8,9] . Implantation of diamond with Li$^+$, P$^+$ and C$^+$ leads to "N" type layers and implantation of B$^+$ or Al$^+$ gives "P" type layers.
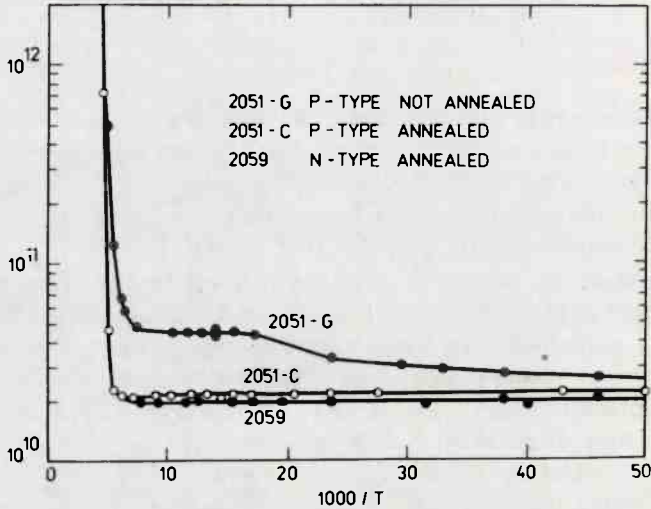

Fig. 1. Temperature dependence of the free carrier concentration in annealed and unannealed Ge (After R.N. Hall [1]).

Implantationt of diamond with Si$^+$ ions leads to SiC synthesis which has been confirmed by photoluminescence analysis. The properties of semiconducting diamond look so promising that Vavilov [10] , concluded at the Twelfth International Conference of the Physics of Semiconductors-Stuttgart 1974-that "before the next millenium diamond must become in indispensible material in solid state electronics".

3. A^{III}B^V COMPOUNDS AND ALLOYS

Many A^{III}B^V compounds such as: GaAs, InSb, InP, GaSb etc, can be used for electronic devices based on the variety of the electronic properties they exhibit.

It appeares important at present to investigate new A^{III}B^V high temperature and energy gap semiconductors like: BN, BP, AlN, AlB$_{12}$, GaN, GaP etc. Even research on the physical properties of these materials has usually been hampered by the successfully grown but not in the form of crystals large enough for the production techniques for normal devices.

10

Boron nitride is a material not found in nature. BN has two main allotropic forms: hexagonal, which resembles graphite in softness and many other physical properties, and cubic, which resembles diamond in its physical properties. Cubic BN can be produced by the simultaneous application of high pressure and high temperature to the hexagonal form of the material [11]. Although BN was first synthesised in 1957, electrical conductivity, reflectivity [12], absorption and luminiscence measurements in cubic BN have been properly done only recently [13].

Boron phosphide has been grown epitaxially on Si substrates by thermal reaction [14]. Research on AlN has been effected also my the small size of the crystals. Reflectivity measurements have been made on 1 mm^3 samples[15]. Experimental results on aluminium dodecaboride AlB$_{12}$ show that it is an analogue of rhombohedral boron in some of its electrical and thermal properties [16]. An anomaly has also reported that its thermal conductivity does not depend on temperature in the low temperature range.

Recent success in the deposition of large single crystal films of GaN has produced renewed experimental interest in this semiconductor [17, 18]. GaN has potential usefulness for UV luminescent devices [19].
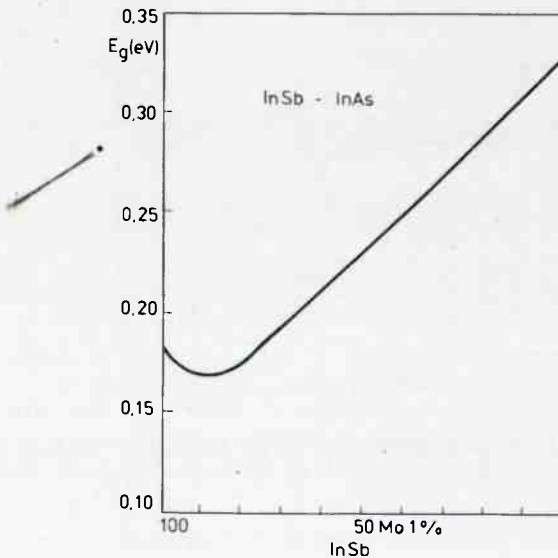

Fig.2. Energy gap (Eg) as a funcion of composition for InSb-InAs alloys.

Some years ago much work was done on A^{III}B^{V} alloys, for instance: InSb-GaSb, InSb-InAs, GaAs-InAs [20,21], but all the samples were polycrystalline, Latteraly with advances in technology interest in these alloys has been renewed especially in GaSb-InSb alloys, because of their bearing on the Gunn effect [22]. Some experimental work has been done on the scattering processes and transport phenomena under pressure of these alloys [23]. Measurement of the mobility when as isotropic strain is applied, with its logarithmic defferential, gives direct data about the scattering processes involved.

11

Recently some new data have also appeared on single and polycristal of
InSb-InAs and GaAs-InAs alloys [24,25] . Alloys of GaAs-InAs with com-
positions nearer to compound GaAs and in the form of large single crystals
could be used for making lasers with freguancies slightly different from pure
GaAs. Also InSb-InAs single crystals are promising materials for making
photodetectors in extending the wavelength towards the further IR range as
compared with pure InSb. In Fig.2 it can be seen that for InSb-InAs alloys
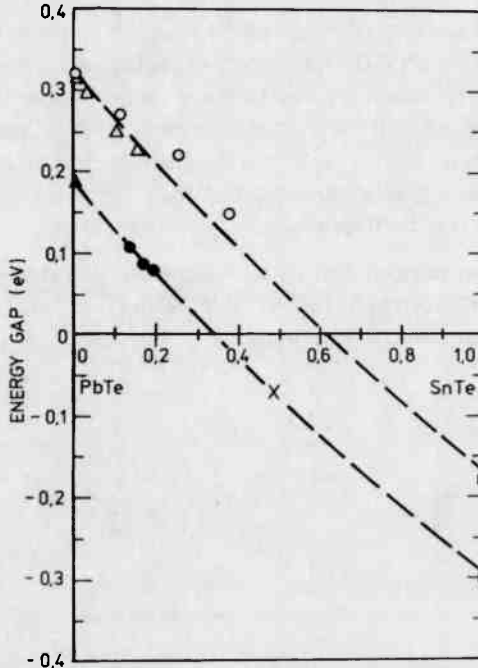the energy gap has a minimum at about 1o mol% InAs.

Fig.3. Variation of energy gap (Eg) with composition and temperature
for Pb₁₋ₓSnₓTe alloys (After Nikolic and Dimmock[27,28,29])

4. LEAD-TIN-TELLURIDE ALLOYS

Since the first obervation of the decrease in the energy gap with composition
of both PbTe-SnTe and PbSe-SnSe alloys [26,27,28] , steady progress has
been made in the area of Pb-Sn salt physics nad applications. According to
the band model of Dimmock et al. [29] for the PbTe-SnTe pseudobinary alloy
system, increasing SnTe composition initially decreases the energy gap
(Fig.3) as the L_6^- and L_6^+ states approach each other (Fig.4).

Crystals of PbTe-SnTe alloys have been made using various techniques with
carrier concentration almost always in the ranoe between 10^{18} and 10^{19} cm^{-3}.
The carrier type and carrier concentration is determined primarily by deviation
from stoichiometry. By teh use of annealing techniques [30,31] , homogeneous
crystals of both "N" type and "P" type material as well as diffused p-n
junctions have been made. Monocrystalline epitaxial layers of PbTe-SnTe have
been produced as photovoltaic devices capable of radiation detection in the
8-14 ,um range [32] . Diode lasers are fabricated from vapor grown diffused
PbTe-SnTe crystals in which the Sn concentration is tailored to produce lasers

12

with various emission wavelengths over the broad range of 6.5 /um to 31.8/urn. The devices can be temperature, pressure or magnetic field turned over small wavelength ranges [33] . The main disadvantage is that the operation of the devices is restricted to low temperatures.
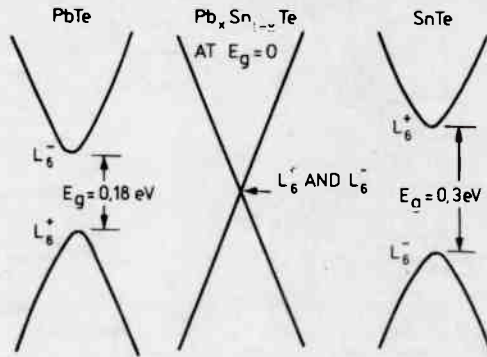

Fig.4. Schematic representation of the valance and conductivity bands at 12K for PbTe, $Pb_{1-x}Sn_xTe$ at $E_g = 0$ and SnTe (After Dimmock [29]).

Recently it has been shown that the carrier concentration in PbTe-SnTe alloys can be reduced by Cd diffusion and also doping with Zn [34] . A significant reduction in carrier concentration to about 10^{16} cm^{-3} followed by an increase in mobility (Fig.5) has been achieved using one zone and two zone
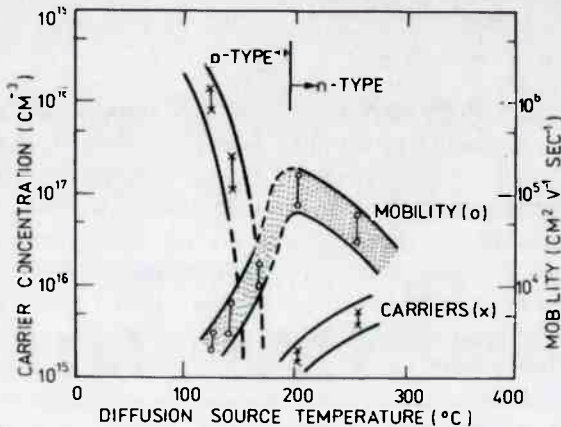

Fig.5. Carrier concentration and mobility as function of diffusion source temperature for PbTe doped with the Cd (Atfer Linden [34]).

annealing techniques for the diffusion of Cd into the vapor-grown crystals of these alloys. This reduction of carrier concentration should make it possible to make photoconductive defectors of much better quality and probably low capacitance photodiodes. Lately it has also been shown that high quality heterojunction photodiodes can be made from layers grown by a llguid phase epitaxy technique [35] . Moreover BaF_2 substrate can be used for making Schottky diodes [36] because BaF_2 can withstand the full photolithographic process of device fabrication, (as is shown in Fig. 6).
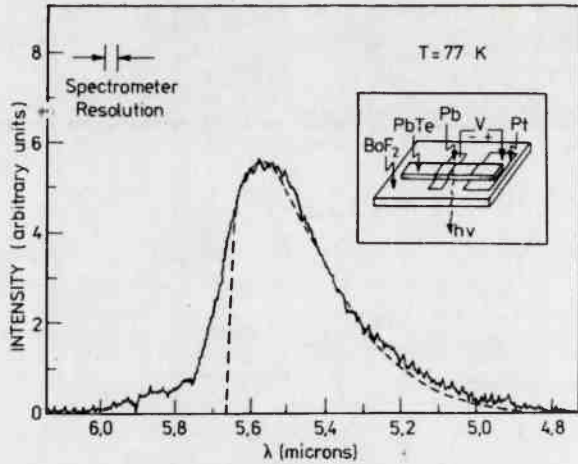
Fig.6. A frequency characteristic for a thin film PbTe Schottky diode on a BaF₂ Substrate (After Chen [36]).

All these examples mean that a number of basic experiments has been done on the properties of the materials and one may expect to fabricate various devices from PbTe-SnTe alloys in the near future. Here one should say that it is less and less regarded as an disadvantage if the mentioned devices should be used only when they are cooled down to cryo temperatures.

5. MAGNETIC SEMICONDUCTORS

As ordinary semiconductors have nonlinear reactions to an applied electric field, so magnetic semiconductors aslo have a nonlinear reaction to an applied magnetic field [37] . An interaction exists between the electronic states and magnetism with the possibility of making electronic devices which would be controlled by an applied magnetic field. In the initial research the main attention was directed to Europium oxides and chalogenides, EuO, EuS and EuSe, which have been studied for several years [38] . More attention was then paid to chromium chalcogenide, $CdCr_2Se_4$, $HgCr_2Se$ etc [39,40] . The conduction electrons from the oxides belong to bands formed primarily from the d orbitals of the cations. The d bands are rather narrow because the ions are relatively far apart which leads to low carrier mibilities (< 1 cm^2/Vs).

The chromium chalcogenide spinels, $CdCr_2Se_4$ and $CdCr_2S_4$ are ferromagnetic semiconductors with Curie temperatures of 84.5°K and 129.5°K respectively. They contain Cr^{3+} ions on the octahedral sites and non magnetic ions on the tetrahedral sites. If there are magnetic ions on the tetrahedral sites, for instance Fe^{2+}, Co^{2+}, the material is ferromagnetically ordered. $CdCr_2Se_4$ is usually a "P" type semiconductor which can be changed to an "N" type by doping with trivalent elements. In "N" type $CdCr_2Se_4$ the conduction electrons interact strongly with the magnetism, so that its electrical properties appear anomalous as a function of temperature [41] , as is shown in Fig.7. In the same temperature range there is also a large shift of the optical

14

energy gap (Fig.8). These observations give strong evidence of the existence of a large interaction between the electronic states and the localized magnetic moments.

Besides chromium chalcogenide spinels another spinel-type semiconductor $CdIn_2S_4$ [43] has appeared recently. For this compound even the band structure has been calculated [44]. It is also a promising material for photoconductivity devices [45]. It has also been shown that $CdIn_2S_4$ has a partial inverse spinel structure [46], where Cd^{2+} ions occupy tetrahedral and octahedral sites equally. Infrared reflectivity spectra measurements below and

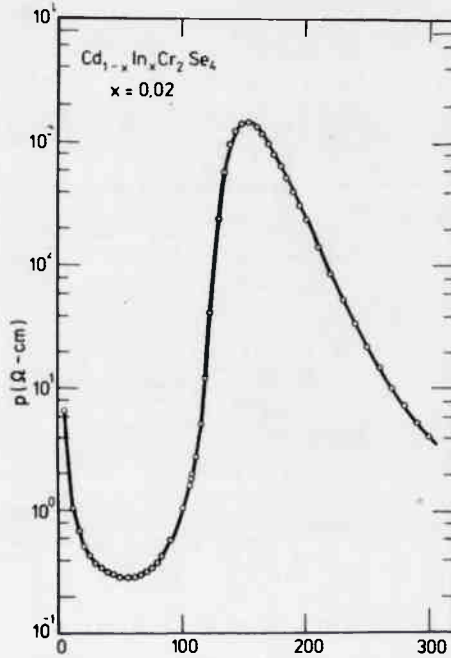Fig.7. Resistivity versus temperature for the n-type $Cd_{1-x}In_xCr_2Se_4$ with x=0,02 (After Amith and Gonssius [41]).
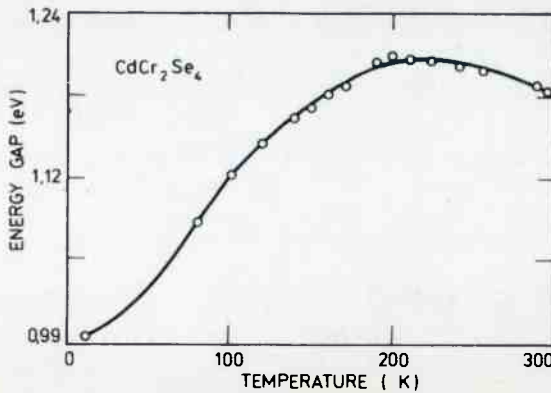

Fig.8. Temperature dependence of the optical absorption edge of $CdCr_2Se_4$ (After Shepers [42]).

15

above the Curie temperature have been made recently using single crystal $CdIn_2S_4$, and the optical constants obtained [47].

One of the most interseting ferromagnetic chalcogenide spinels is $CuCr_2Se_4$ because of its high Curie temperature (420°K) and p-metallic conductivity. The ferromagnetism is the result of an indirect exchange interaction between the localized Cr-3d spins via the antiparallel spin polarization of the free holes in the Cu- band [48]. The high concentration of the majority carriers is due to the monovalence of Cu leaving one hole per molecule, when placed on the tetrahedral site of the spinel structure [49].
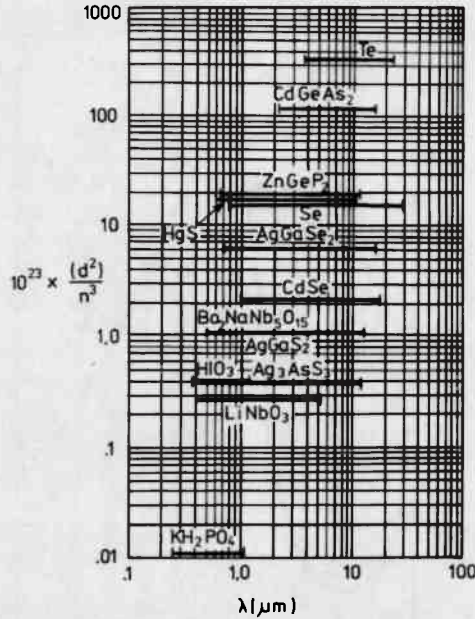

Fig.9. Comparison of the figures of merit (d^2/n^2) where d is the largest phase matchable nonlinear optical coef., and useful transparency ranges of some materials. (After Bovd [57-60]).

Lately the research on ferromagnetic chalcogenide spinel was extended to determinations of the thermoreflectance of $CdCr_2Se_4$ at temperatures above and below the Curie point [50]. Also the effect of Mn impurities was examined on the screw structure of $ZnCr_2Se_4$ [51], using neutron diffraction. By the perturbation therory, distortion of the screw spin structure due to an intersitial spin was studied. These theoretical results were compared with some experimental data and in the conclusion it was established that neither changes in Cr-Cr exchange interactions due to Mn substitution nor Mn-Cr exchange can be ignored [52].

6. NONLINEAR OPTICAL MATERIALS

The advent of the laser has stimulated many fields of study including much research on Nonlinear Optical Materials. They have become of great interest for measuring wavelengths and frequencies of infra red lasers. The interaction of the laser beam with the electronic charge distribution of atoms gives rise

16

to obervable nonlinear optical effects. By mixing the infrared laser radiation with visible light, in a nonlinear optical single crystal, two new radiation components appear, which can be measured and compared with the visible laser radiation [53,54].

A number of optical materials are available for the above use [55,56]. The best nonlinear optical materials are highly resistant semiconuctors at room temperature or even insulators. They should be highly polarizable crystals with a rarther high index of refraction. The nonlinear optical properties of several compounds have been studied during the last four years [57,58,59,60,61,62]. They have a tetrahedrally coordinated calcopyrite type of structure and are analogous to $A^{II}B^{VI}$ and $A^{III}B^{V}$ diamond cubic semidonductors. The bonding is mainly covalent although an ionic contribution is present also, because of the presence of dissimilar atoms in the lattice. It has been shown [63] that the absorption mechanisms in Ag_3AsS_3 are due to direct and indirect interband transitions. Non-linear optical materials have rather wide transparency ranges too as is shown in Fig.9. Up to now the only good quality synthetic crystal available for nonlinear interaction between CO_2 laser wavelenght and the visible. range which has appeared is Ag_3AsS_3.

7. $A^{I}B^{III}C^{VI}_2$ and $A^{II}B^{IV}C^{V}_2$ TERNARY COMPOUNDS

There is a large number of ternary tetrahedrally coordinated semiconductors. Their general formula is $A^{n-1}B^{n-1}C^{8-n}$ where n is usually 2 or 3. They are analogous of the $A^{II}B^{VI}$ and $A^{III}B^{V}$ semiconductors. The ternary compounds usually have a smaller energy gap compared with the homologous binary compounds. In this case there are two kinds of cations. The possible emission lines, which are mainly in the visible range, for these compounds, may indicate a possible application of the particular ternary compound for making a stimulated emission in the visible range [64,65,66]. The intrinsic optical properties and the band structures are not yet well understood. $CuGaS_2$ and $CuInS_2$ are the most studied $A^{I}B^{III}C^{VI}_2$ compounds [67,68], then $CuAlS_2$ and $CuAlSe_2$ [69], which are regarded as potentional candidates for non linear optical materials in the infrared region.

As a continuation of the work on copper sulfides and selenides recently the research has been extended to $AgXTe_2$ where X is Al, Ga or In compounds, which also might have a sufficiently large birefringence to be used in nonlinear optics. The alence band structure, the semiconducting properties andphase transformations of these compounds have been studied [70,71] and also far infrared resonance was obtained for $AgInTe_2$ [72] (Fig.10).

The $A^{II}B^{IV}C^{V}_2$ compounds are ternary compounds which are in some way similar to the $A^{III}B^{V}$ sphalerite compounds. There is a large list of 24 known members of this group treated in the literature [73]. From the amount of work which has been done it could be said that the main representatives of this group of semiconductors are $ZnSiP_2$, $ZnGeAs_2$. Their physical properties have been studied [74,75,76].
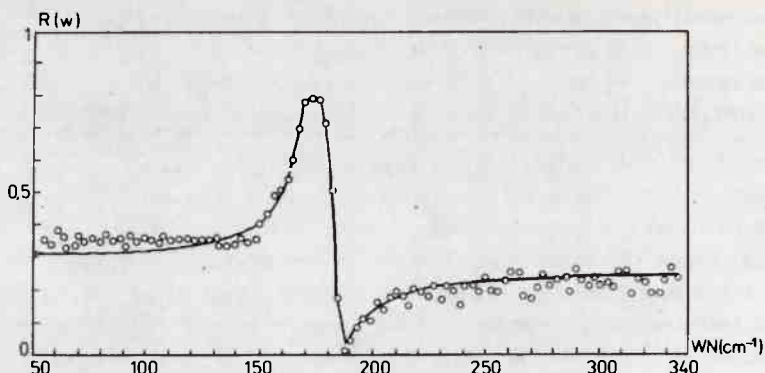
17

Fig. 10. Reflectivity of AgInTe$_2$. Experimental points and curve
obtained by a computer fitting procedure (After Nikolić [61]).

8. LAYER A^{IV}B^{VI} and A^{IV}B$^{VI}_2$ COMPOUNDS

Quite a number materials have a layer structure. For instance the transition metal dichalcogenides are about 60 in number and two thirds of them have layer structures. Crystals of these materials can be cleaved down to less than 100 A [77] ; so that some authors consider them as two dimensional materials with very weak Van der Waal's interaction between the layers.

The physical properties of these layer materials are usually characterised by extreme anisotropy. Some of their optical, magnetic and transport properties have been recently reviewed [78,79] . Most of the optical studies on layered crystals have been concentrated on those which are semiconductors. Optical transmission measurements can be made because very thin samples can be obtained in the frequency range where the absorption coefficient can be of the order of even 10^6 cm^{-1}.

Our attention will be more concentrated on tin and germanium chalcogenides (MX) and diachalcogenides (MX$_2$) e.g.: SnS, GeS, SnSe, GeSe and SnS$_2$, SnSe$_2$, GeS$_2$ and GeSe$_2$. Germanium and tin chalcogenide compounds are isomorphic semiconductors which are, in many ways, intermediate between two-dimensional (layer type) and three-dimensional crystals. They crystalize in an orthorhombic (distorted rocksalt) structure with strong intra-layer and weak interlayer forces.

SnS$_2$ and SnSe$_2$ have [88,89] the CdJ$_2$ crystal structure with symmetry of the space group D^3_{3d}, while GeS$_2$ and GeSe$_2$ crystalize in an orthorombic lattice whose symmetry corresponds to the space group D$^{13}_{2h}$. In all these cases Sn or Ge atoms (M), are sandwiched between two layers or either sulphur or selinium atoms (H), so that the unit of three layers (H-M-H) is periodically repeated along the x-axis.

All the mentioned Sn and Ge chalcogenides and dichalcogenides have been grown in our laboratory. It has been possible to obtain large single crystals from all these compounds. The crystals were of a form and size, which can be used for measurements of various optical and transport properties. Their

18

optical properties were obtained in the visible and infra red ranges extended to the far infra red range [90,91] . For all these semiconductors reflectivity measurements had been done firstly using nonpolarized [83,84] and recently polarized light beam [85,86,87,88] . For example, room temperature reflectivity of SnS in the frequency range 50-350 cm^{-1} for light polarized with E‖a; E‖b and E‖c is given [86] in Figure 11. In the case of E‖a three peaks corresponding to three oscillators are observed. Similar experimental results were also obtained [87] for GeS and are given in Figure 12. The full line corresponds to room temperature data and the broken line show the 77 K results where they differ from the former.
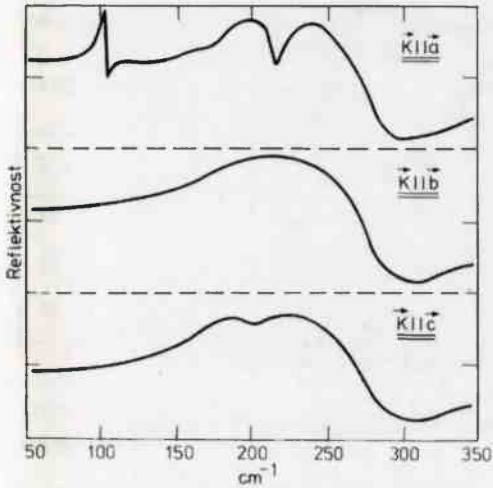

Fig. 11. Averaged reflectivity versus wavenumber curves for E‖a, E‖b and E‖c for an n-type SnS sample at 300 K. (After Chamberlain at al. [86]).
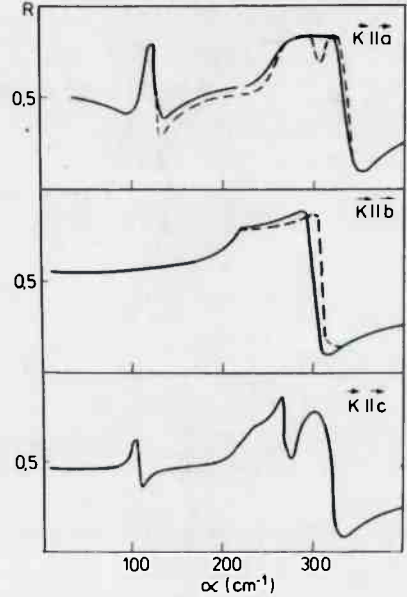
Fig. 12. Reflectivity of GeS In the far infrared for the three principal polarizations at 300 K and 77 K, which is shown with the broken line (After Mihajlović at al [87]).

The room temperature reflectivity data of GeSe$_2$ in the principal polarization [90] (E‖a and E‖b) in the range between 40 and 450 cm^{-1} are given in Figure 13. For light polarized at E‖a five "reststrahlen" peaks were observed but only one in the case E‖b. Using a fitting procedure and a four parameter semiquantum model, introduced by Gervais [91] and Piriou (1974), the frequencies and damping factors of the tranverse and longitudinal optic modes have been obtained, and are given in Table 1, together with the results of the Kramers-Krönig analysis.

These valuses for the optical mode are in good agreement with the positions of the absorption peaks obtained by Zacharov [92] et al. (1973) measuring the transmission of powdered GeSe$_2$ pressed together with polyethylene.

The ratio of intralayer and interlayer force constants can be estimated from the results of both infrared reflectivity and Ramn-scattering experiments. This ratio increases for semiconductors which are layerlike (anisotropic).

19

One should classify the infrared and Raman active phonons in terms of three dimensional symmetries for the studied crystal. For instance, GeS has a D_{2h}^{12} three-dimensional space group and the centre of inversion symmetry is for GeS between the layers. The relations between phonons for individual layers and three-dimensional crystals are given in lit. [85] , for GeS with 8 atoms per unit cell. Due to a centre of inversion between the layers, the optical active modes split into Raman (R) and infrared (IR)
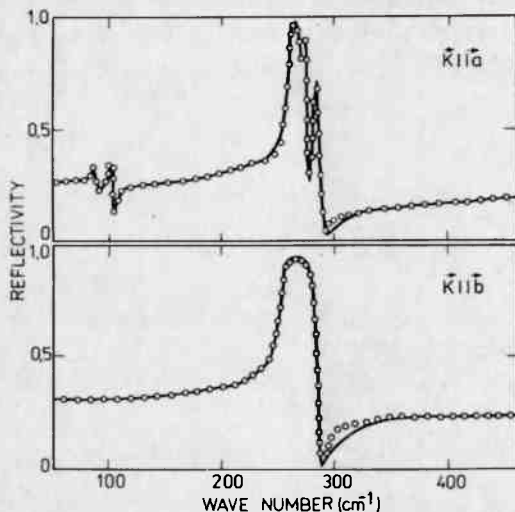

Fig. 13. Experimental infrared reflectivity (open circles) for GeSe₂ in the range between 50 and 450 cm⁻¹ at 300 K for polarized light: E ∥e and E∥b. The solid lines were calculated using the parameters given in Table 1 for the four-parameter semiquantum model. (After Popovic at al. [90]).

Table 1

PARAMETERS USED TO FIT THE INFRA RED REFLECTIVITY DATA FOR GeSe₂ WITH THE SEMIQUANTUM MODEL ARE GIVEN TOGETHER WITH RESULTS OF THE KRAMERS-KRONIG ANALYSIS. FREQUENCIES AND DAMPING CONSTANTS ARE EXPRESSED IN UNITS OF cm⁻¹. (After Popović at al [90]).

		ω_{jTO}	γ_{jTO}	ω_{jLO}	γ_{jLO}	ε_∞	ε_{jTO}	ω_{jTO}	γ_{jLO}	ω_{jLO}	ε_∞
		86.6	5	89	5		88	5	89	5	
		103.7	4	104	4		103.7	0.8	104	0.8	
E	a	260	7	272	5	3	262	1	272	15	7
		272	2	279	2		269	12	277	2	
		282	4	290	10		281	3	291	3	
E	b	256	8	288	8	5	256	2	286	2	10

active vibrations. This splitting should manage if the interlayer forces are neglectable and vice versa. Raman spectra together with the reflectivity data can be used to obtain the frequency splitting parameters besides the ratio of the force constants mentioned above. Table 2 summarizes the splittings

20

together with the frequencies ν_n of the corresponding intralayer modes, splitting factors Δ and values of $(\nu_0/\Delta)^2$ which represent a measure of the ratio of intralayer to interlayer force constants for each pair of phonons.

The obtained values indicate that GeS is a particularly interesting example of a layere compound in which the three dimentional structure, while not being dominant, has an important influence. It will be very interasting to see if the other IV-VI semiconductors isomorphs of GeS (SnS, SnSe, and GeSe)are more layer like.

Table 2

THE FREQUENCIES OF THE THREE PAIRS OF RAMAN AND INFRARED ACITVE PHONONS ARE GIVEN TOGETHER WITH THE CORRESPONDING VALUES OF THE OBSERVED PHONON FREQUENCIES (ν_+ and ν_-), ν_0 , SPLITTING FACTORS, Δ , AND THE RATIO OF INTRALAYER TO INTERLAYER FORCE CONSTANTS. (After Mihajlović at al [87]).

Splitting	ν_+	ν_-	ν_0	Δ	$(\nu_0/\Delta)^2$
B_{1u} - Ag	315	269	292	118	6
B_{1u} - Ag	258	238	248	71	12
B_{1u} - Ag	118	111	115	28	16

SnS is a compound which can be used as a detector of visible and near infrared radiation. The photoconductivity response characteristics for samples with the Sn-Sn and Sn-In contacts are shown in Figure 14. When, on the
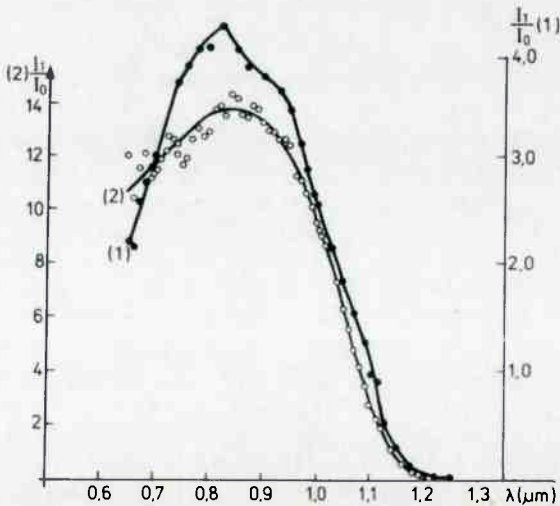

Fig. 14. The photoconductivity response characteristics for SnS samples with the Sn-Sn contact (1) and Sn-In contacts (2). (After Nikolić at al. [93]).

21

contrary, the In contact was exposed to monochromatic chopped light, the intensity of the photocurrent was much higher. The photoconductivity response for two SnS samples with Sn-In contacts are shown in Figure 15. Sample "A" had a photocurrent about 10 times lower than sample "B". For both samples two maxima were observed at the same wavelenght but the ratio of their intensity was different. The observed maxima appeared perhpas because of some lower energy band transition. This behaviour is still not quite well understood because a reliable energy band structure of SnS is still not available.
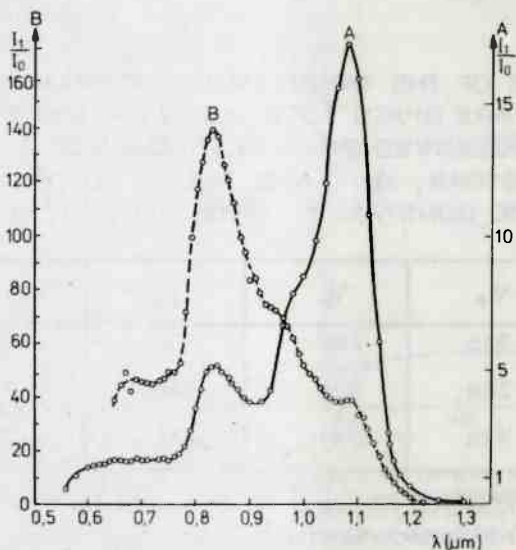

Fig. 15. The photoconductivity response characteristics of two SnS samples (A and B) with Sn-In contacts when the In contact was exposed to monochromatic chopped light. (After Nikolić et al [93]).

9. CONCLUSION

In conclusion one can say, first of all, that so much research has been done on such a great variety of new materials that it is very difficult to say which might be widely used in the near future. Our feeling is that GaAs-GaP and GaAs-InAs are alloys which might soon find the greater use.

Ferromagnetic semiconductors like $CdCr_2Se_4$ and $CdCr_2S_4$ should be mentioned too because, with the help of scientists who are familiar with sintering procedures the quality of these materials might be sufficiently improved for them to be used in practice. Moreover IV-VI compounds and alloys might become more interesting in the future for some special devices such as detectors and emitters in infrared.

Finally we believe that the technology of producing semiconductors of high melting temperature, high energy gap and high mobility will soon reach the stage that diamond, boron and some compounds (BN, SiC, AlN etc) will become of commercial interest.

22

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