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ELECTRONIC STRUCTURE AND OPTICAL PROPERTIES OF SnS<sub>2</sub> AND SnSe

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M. Y. Au-Yang and M. L. Cohen

February 1969

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Electronic Structure and Optical Properties of  $\text{SnS}_2$  and  $\text{SnSe}_2$

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February 1969

Electronic Structure and Optical Properties  
of  $\text{SnS}_2$  and  $\text{SnSe}_2$ .\*

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Abstract

The electronic band structures of  $\text{SnS}_2$  and  $\text{SnSe}_2$  are calculated by using the Empirical Pseudopotential Method. The potentials were obtained by scaling those used in other energy band calculations. The symmetry properties of the crystals are treated in detail and the optical constants are calculated. Comparison with experiment is also made.

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The concept of associating a pseudopotential with each ion core independent of its chemical state has been successfully exploited recently to yield electronic states of semiconductors both in elemental<sup>1</sup> and in compound forms<sup>2,3</sup>. In the first case, the elemental pseudopotential is extracted from semiconductors containing that element, e.g., Se from ZnSe<sup>1</sup>. In the second case, the pseudopotential form factors in a compound are derived from the known pseudopotentials of the constituent elements, e.g. Mg and Si to give Mg<sub>2</sub>Si<sup>2</sup>. In these calculations, the Empirical Pseudopotential Method<sup>4</sup> (EPM) is used. The combination of assigning a fixed pseudopotential to each ion and the use of the EPM then essentially involves the variation of pseudopotential form factors to fit optical data and the existing information on the pseudopotentials.

The success we have had on the Mg<sub>2</sub>X calculations<sup>3</sup> and the availability of good optical data has stimulated us to attempt calculations of the electronic structure of SnS<sub>2</sub> and SnSe<sub>2</sub>. The similarity of these two classes of compounds, Mg<sub>2</sub>X and SnX<sub>2</sub>, lies in the shape of the unit molecule: a dumbbell formed from the two identical atoms weighted at the center by the third, different atom. This structure preserves the inversion symmetry of the lattice. This yields a Hamiltonian with real matrix elements which is easier to diagonalize than one with complex matrix elements. In addition to calculating the electronic structure of SnS<sub>2</sub> and SnSe<sub>2</sub> we have also determined the optical constants for these materials.

The properties of SnS<sub>2</sub> and SnSe<sub>2</sub> have only been sporadically investigated experimentally and, to our knowledge, no band structure calculations exist. The direct and indirect energy gaps of SnS<sub>2</sub> have been measured optically<sup>5,6</sup>, and those of SnSe<sub>2</sub> have been measured both optically<sup>5</sup> and electrically<sup>7,8</sup>. The combined data seems to have fixed the energy gaps to

within a small range of values. Reflectivity data in the range of 1 to 12eV has also been obtained for  $\text{SnS}_2$ , for light with polarization perpendicular to the  $\underline{c}$  axis of the crystal<sup>6</sup>.

The pseudopotentials of the constituent ions in the calculation, i.e. those of Sn, Se, and S, are not available to a high degree of accuracy such as those of the group IV elements, Si and Ge. The pseudopotential for Sn, for example, differs greatly between grey tin<sup>4</sup> and for white tin<sup>9</sup>, especially for large  $|\underline{G}|$  values. We have based our calculations on the Sn, Se pseudopotentials of Animalu and Heine<sup>9</sup> and the S pseudopotential as extracted from  $\text{ZnS}$ <sup>4</sup>.

This paper is divided into three sections. In Section II the group theory of the  $\text{CdI}_2$  structure is treated in detail, and the Hamiltonian is derived. The resulting Schrodinger equation is solved and the optical constants are calculated. In Section III, we present the results and the comparison with experiment.

## II Calculations

The tin chalcogenides,  $\text{SnS}_2$  and  $\text{SnSe}_2$ , crystallize in the  $\text{CdI}_2$  structure. The lattice is simple hexagonal with one molecule per primitive cell. If one chooses the position of a Cd atom at the origin of the cell, then the positions of the I atoms can be chosen to be at the symmetric points  $\pm \underline{u}$ , where  $\underline{u} = \left(\frac{2}{3}, \frac{1}{3}, \frac{1}{4}\right)$ . (In writing the vector we have assumed the usual hexagonal coordinate system of two  $|\underline{a}|$  vectors on the x,y plane placed  $120^\circ$  apart and a  $\underline{c}$  vector in the  $\underline{z}$  direction, where a and c are the usual lattice constants.) The symmetry of the lattice is  $D_{6h}$ ; however, because of the basis, only 4 of the 24 symmetry operations in  $D_{6h}$  leave the structure invariant. These are the identity, a two-fold rotation about a corner of the hexagon in the x-y plane, inversion, and finally a reflection in the plane perpendicular to the two-fold axis. (See Fig. 1a.)

The first Brillouin zone of the hexagonal unit cell is again a hexagonal prism; however, as a consequence of the basis atoms in the  $\text{CdI}_2$  structure, the six sides of the hexagon are no longer equivalent. In fact, the direction of the two-fold axis in real space is the particular direction in the reciprocal space. It passes through the center of the zone and the midpoint of one side of the hexagon on the x-y plane (and also that of the opposite side). (See Fig. 1b.) These two points, called M points in the usual notation, are the only M points which are invariant under  $C_2$  in the  $\text{CdI}_2$  structure. The other M points are only invariant under inversion. We therefore call the M points with lower symmetry  $M'$  points.

It is easily seen from Fig. 1b that except for the  $M'$  points, all other points of symmetry are on three symmetry lines and three symmetry planes. The symmetry lines are the  $C_2$  axis ( $\Sigma$  line) and the two lines parallel to it and



displaced by  $\pm \frac{\pi}{c}$  in the  $\underline{c}$  direction (R lines). The symmetry planes are perpendicular to the  $C_2$  axis; they include two planes which contain the two previously mentioned special M points, and the third plane passes through the origin, the  $\Gamma$  point. The intersection of these planes and the x-y plane are the T and T' lines. The symmetry at  $\Sigma$  and R is the group  $C_2$ , and that at T, T' is the group  $C_s$ . The intersection of these lines and planes are the  $\Gamma$ , A, L and M points. These points therefore have the highest symmetry in the structure, the group  $C_2 \times C_s$ .

From the above discussion we see that the group theoretical treatment of the  $CdI_2$  structure is quite simple. There are only three important types of symmetry points, and the compatibility relations between them are trivial since we only have four symmetry operations. In Tables Ia, Ib, and Ic, the characters are given for the symmetry operations of the three groups and the irreducible representations are defined. The symmetry points  $M'$  are not considered because they have a low symmetry and are not joined by other symmetry points.

To calculate the optical properties of the  $CdI_2$  structure, we need to have the selection rules for optical transitions with the electric field vector both in the directions parallel and perpendicular to the  $\underline{c}$  axis. In Tables Ia, Ib, Ic, the characters for photons with both types of polarizations are given. From these characters, it is easy to compute the selection rules given in Table II.

This completes our discussion of the group theoretical aspects of the problem; next we shall derive the Hamiltonian and solve the Schrodinger equation.

We use an energy independent pseudopotential and neglect the spin-orbit interaction; the pseudo-Hamiltonian then becomes,

$$\mathcal{H} = -\frac{\hbar^2}{2m} \nabla^2 + V(\underline{r}) . \quad (1)$$

Expanding the potential in the reciprocal lattice we have

$$V(\underline{r}) = \sum_{\underline{G}} V(\underline{G}) e^{i\underline{G} \cdot \underline{r}} \quad (2)$$

and so

$$\begin{aligned} V(\underline{G}) &= \frac{1}{\Omega_{\text{cell}}} \int_{\text{cell}} V(\underline{r}) e^{-i\underline{G} \cdot \underline{r}} \\ &= \frac{1}{\Omega_{\text{cell}}} \left[ \Omega_{\text{Cd}} V^{\text{Cd}}(\underline{G}) + 2\Omega_{\text{I}} V^{\text{I}}(\underline{G}) \cos(\underline{G} \cdot \underline{u}) \right] \quad (3) \end{aligned}$$

where the  $\Omega$ 's with subscripts denote the volumes per molecule or atom in  $\text{CdI}_2$ , Cd and I and the  $V$ 's with superscripts denote the pseudopotentials of the constituent ions Cd and I. (For the actual case of  $\text{CdI}_2$ , in which  $\text{I}_2$  is a gas, we can define  $V^{\text{I}}(\underline{G})$  without the  $1/\Omega_{\text{I}}$  factor, thus getting rid of the ambiguous quantity,  $\Omega_{\text{I}}$ ). The factor,  $\cos(\underline{G} \cdot \underline{u})$ , includes all the effects of the basis atoms, and accounts for the destruction of most of the symmetries of the lattice.

The Schrodinger equation with this Hamiltonian, Eq. (1), is solved by expanding the periodic part of the Bloch state in plane waves. For  $\text{SnS}_2$  and  $\text{SnSe}_2$ , the calculations present a convergence problem if we use the full pseudo-potential curves of the constituent ions. The reason is that the hexagonal lattice contains many plane-waves with small values of  $|\underline{G}|$ , and therefore the

pseudopotential curves must be cut off for some maximum  $|G|$  value, determined by the accuracy of the computer available for the diagonalization of the Hamiltonian. The value we chose was  $G_{\max}^2 = (59/4) \frac{2\pi^2}{a}$ . (This is for  $(c/a)^2 = 8/3$ , which is approximately valid in both  $\text{SnS}_2$ ,  $\text{SnSe}_2$ ). This value of  $G_{\max}$  gives 16 non-vanishing pseudopotential form factors for Sn.

The Hamiltonian is diagonalized on a mesh of  $\underline{k}$  points one fourth the size of the Brillouin zone. The band structure thus obtained is used to calculate the optical constants of these materials<sup>10</sup>. The first optical constant to be calculated is the imaginary part of the dielectric function,  $\epsilon_2(\omega)$ . The computation uses the equation

$$\epsilon_2(\omega) = \frac{e^2 \hbar^2}{8\pi m^2 \omega^2} \sum_{c,v,i} \int \delta(E_c(\underline{k}) - E_v(\underline{k}) - \hbar\omega) |\langle u_{\underline{k},v} | \frac{d}{dx_i} | u_{\underline{k},c} \rangle|^2 d^3k \quad (4)$$

where  $c,v$  denotes the conduction and valence bands,  $u_{\underline{k},n}$  is the periodic part of the  $n^{\text{th}}$  band wavefunction and  $\delta = 2, i = 1,2$  for light with polarization perpendicular to the  $\underline{c}$  axis and  $\delta = 1, i = 3$  for light with parallel polarization. We shall approximate the wavefunctions,  $u_{\underline{k},n}$  by the eigenstates of the pseudo-Hamiltonian, Eq. (1).

Before we discuss the results of the calculation, we would like to emphasize that, because of several difficulties, the calculations should be regarded as being preliminary. In terms of accuracy they cannot be compared with the calculations performed on the  $\text{Mg}_2\text{X}$  compounds, for example. The difficulties with  $\text{SnS}_2$  and  $\text{SnSe}_2$  calculations are as follows:

- (a) The low symmetry of the  $\text{CdI}_2$  structure necessitates the computation of the band structure in one fourth the Brillouin zone which is six times as large as the amount of  $\underline{k}$  space required for the wurtzite

compounds<sup>10</sup>. The unusually large matrices to be diagonalized in the hexagonal lattice makes an accurate calculation impractical at this stage.

- (b) Although reflectivity data on  $\text{SnS}_2$  is available, an  $\epsilon_2(\omega)$  curve is not yet available. The lack of this curve makes it quite difficult to correlate the results of theory with experiment. In addition no spectrum for  $\text{SnSe}_2$  is available.
- (c) There are 8 valence bands, 6 of which are important for optical transitions under 12eV. In addition, we have to take into account 10 conduction bands for our final states in the optical transitions. The large number of bands contributing to  $\epsilon_2(\omega)$  makes the identification of the theoretical  $\epsilon_2(\omega)$  in terms of critical points quite difficult.

In view of these difficulties, little attempt has been made to adjust the potentials. The agreement between theory and experiment is therefore not expected to be better than 0.5 to 1.0eV.

### III Results and Discussion

The band structures of  $\text{SnS}_2$  and  $\text{SnSe}_2$  are shown in Figs. 2 and 3 along certain symmetry directions of the hexagonal lattice. The pseudopotential form factors used in the calculations are shown in Table III. As previously mentioned, some of the special labels (for example K) in the hexagonal lattice have no particular importance here. They are shown mainly to illustrate how the bands appear along selected general directions.

For both  $\text{SnS}_2$  and  $\text{SnSe}_2$ , the direct and indirect energy gaps are fitted to experiment. In addition, for  $\text{SnS}_2$ , the optical spectrum was used to fix some of the high energy splittings. We again remark that in both cases the potentials chosen were constrained to match those derived from analysis of other crystals.

The direct energy gaps for both  $\text{SnS}_2$  and  $\text{SnSe}_2$  are the results of transitions near the M point between the 8<sup>th</sup> and the 9<sup>th</sup> bands; the calculated values are 2.48 ( $\text{SnS}_2$ ) and 1.78eV ( $\text{SnSe}_2$ ). The calculated transition at the M point itself is forbidden by parity. Experimentally the threshold for direct transition for both compounds is forbidden as is suggested by the absorption data of Domingo et al.<sup>5</sup>. They obtained direct energy gaps (forbidden in first order) of 2.88 and 1.62eV for  $\text{SnS}_2$  and  $\text{SnSe}_2$  respectively.

The calculated fundamental energy gaps of both compounds are indirect. The maximum of the valence band is at  $\Gamma$ , and the minimum of the conduction band is at L. This fact seems to be well supported by experiment<sup>5,6</sup>. The calculated values are 2.11 and 0.81eV for  $\text{SnS}_2$  and  $\text{SnSe}_2$ , which compare fairly well with the average experimental values of 2.14 and .99eV<sup>5-8</sup> (see Table IVa).

The imaginary part of the dielectric function,  $\epsilon_2(\omega)$ , calculated from the

band structure of  $\text{SnS}_2$  is shown in Fig. 4 for light polarization both parallel and perpendicular to the  $\underline{c}$  axis. In Table IVb some prominent peaks in the reflectivity data of Greenaway and Nitsche<sup>6</sup> are compared with the results of the calculation. Because of the difficulties encountered in calculations, as mentioned in the last section, not much effort has been made to fit the theory to the experiment to better than 1.0eV, and only the most important critical points (cp) are identified for each peak. We remark also that because of the low symmetry of the structure, a great deal of contribution to the  $\epsilon_2(\omega)$  comes from "volume effects", i.e. contributions which are not related to a cp. We note that all transitions which are allowed in parallel polarization are also allowed in perpendicular polarization. If in the experiments<sup>6</sup> on  $\text{SnS}_2$  the light were not completely polarized perpendicular to the  $\underline{c}$  axis, then the peak labelled C in reference 6 would be larger than expected for purely perpendicularly polarized light. This would make the comparison between theory and experiment much better.

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Table Captions

Tables Ia, Ib and Ic:

The characters of the small representations of  $\Gamma, A, M, L, P, \Sigma$  and R where P is a general point on a symmetry plane. The bottom two rows show the characters for the photons with polarizations perpendicular and parallel to the  $c$  axis.

Table II:

Selection rules for direct optical transitions. For points of the same symmetry, only one point is shown, (i.e.  $\Gamma$  represents  $\Gamma, \Delta, M$  and  $L$ ).

Table III:

The pseudopotential form factors in Rydbergs used in the calculations of the  $\text{SnS}_2$ ,  $\text{SnSe}_2$  band structures. The Sn potentials for both compounds are identical. The differences in the table arise because of lattice constant differences.

Table IV:

- (a) Comparison between theory and experiment for the direct and indirect threshold energy gaps.
- (b) Some dominant optical transitions for perpendicular polarization in  $\text{SnS}_2$  and comparison with experiment. The subscripts in the optical transition labels indicate the band indices for the transition. The peak labels for the measured reflectivity curves are from reference 6.

Table Ia

$\Gamma, A, M, L$	E	$c_2$	I	$\sigma$
$\Gamma_1$	1	1	1	1
$\Gamma_2$	1	-1	1	-1
$\Gamma_1'$	1	-1	-1	1
$\Gamma_2'$	1	1	-1	-1
x,y	2	0	-2	0
z	1	-1	-1	1

Table Ib

P	E	$\sigma$
$P_1$	1	1
$P_2$	1	-1
x,y	2	0
z	1	1

Table Ic

$\Sigma, R$	E	$c_2$
$\Sigma_1$	1	1
$\Sigma_2$	1	-1
x,y	2	0
z	1	-1

Table IIPerpendicular Polarization

$$\Gamma_1, \Gamma_2 \leftrightarrow \Gamma'_1, \Gamma'_2$$

$$P_1, P_2 \leftrightarrow P'_1, P'_2$$

$$\Sigma_1, \Sigma_2 \leftrightarrow \Sigma'_1, \Sigma'_2$$

Parallel Polarization

$$\Gamma_i \leftrightarrow \Gamma'_i$$

$$P_i \leftrightarrow P'_i$$

$$\Sigma_i \leftrightarrow \Sigma'_j$$

where  $i \neq j$  and  $i = 1, 2$

Table III

G	Pseudopotential Form Factors			Se
	SnS <sub>2</sub> : Sn	S	SnSe <sub>2</sub> : Sn	
(001)	-.312	-----	-.36	-----
(100)	-.09	-.31	-.06	-.24
(002)	-.06	-.27	-.036	-.205
(101)	-.036	-.21	-.024	-.17
(102)	.06	-.08	.072	-.012
(003)	.074	-.042	.074	.03
(210)	.075	-.012	.073	.08
(211)	.074	.004	.071	.10
(103)	.073	.032	.069	.12
(200)	.068	.082	.057	.14
(212)	.066	.088	.054	.14
(201)	.065	.096	.05	.13
(004)	.06	.102	.048	.11
(202)	.048	.074	.035	.08
(104)	.036	.054	.029	.05
(213)	.036	.05	.029	.05

Table IVa

Threshold Energy Gaps (eV)

Direct		Indirect	
Calculated	Measured	Calculated	Measured
SnS <sub>2</sub> : 2.48 (M <sub>1</sub> ' → M <sub>1</sub> ')	2.88 (ref. 5)	SnS <sub>2</sub> : 2.11 (Γ <sub>1</sub> ' → L <sub>1</sub> ')	2.07 (ref. 5) 2.21 (ref. 6)
SnSe <sub>2</sub> : 1.78 (M <sub>1</sub> ' → M <sub>2</sub> ')	1.62 (ref. 5)	SnSe <sub>2</sub> : 0.81 (Γ <sub>1</sub> ' → L <sub>1</sub> ')	0.97 (ref. 5) 1.0 (ref. 7) 1.0 (ref. 8)

Table IVb

Energy Value of Structure		Identification of main transition in calculated $\epsilon_2(\omega)$	Type of cp
in the measured reflectivity	in the calcu- lated $\epsilon_2(\omega)$		
4.0eV (A)	4.03eV	$(M_1' \rightarrow M_1)_{8,11}$	$M_0$
4.8eV (B)	5.35eV	$(M_1 \rightarrow M_1')_{6,9}$	$M_1$
5.7eV (C)	5.83eV	$(L_1 \rightarrow L_1')_{5,9}$	$M_1$
7.0eV (D)	7.72eV	$(\Gamma_1' \rightarrow \Gamma_1)_{8,12}$	$M_0$
7.8eV (E)	8.67eV	$(M_1' \rightarrow M_1)_{8,12}$	$M_1$

Figure Captions

Figures 1a, 1b:

- (a): The projection on the x-y plane of the  $\text{CdI}_2$  structure in real space. The triangles denote the positions of the Cd atoms, and the circles denote those of the I atoms. The triangles are on the x-y plane; the open and the closed circles are on the planes  $z = \pm c/4$  respectively, where  $c$  is the usual lattice constant.
- (b): The section of the first Brillouin zone of the simple hexagonal lattice on the x-y plane. The symmetry lines for the  $\text{CdI}_2$  structure are labelled and they are drawn in heavy lines. The two-fold axis and the reflection planes are indicated by  $C_2$  and  $\sigma$  in both figures.

Figure 2:

The calculated band structure of  $\text{SnS}_2$ . The first two bands are not shown; they are lower than the third band by at least 4.5eV.

Figure 3:

The calculated band structure of  $\text{SnSe}_2$ . As in Figure 2, the first two bands are not shown.

Figure 4:

The calculated imaginary part of the dielectric function for light polarizations perpendicular and parallel to the  $z$  axis, for  $\text{SnS}_2$ .

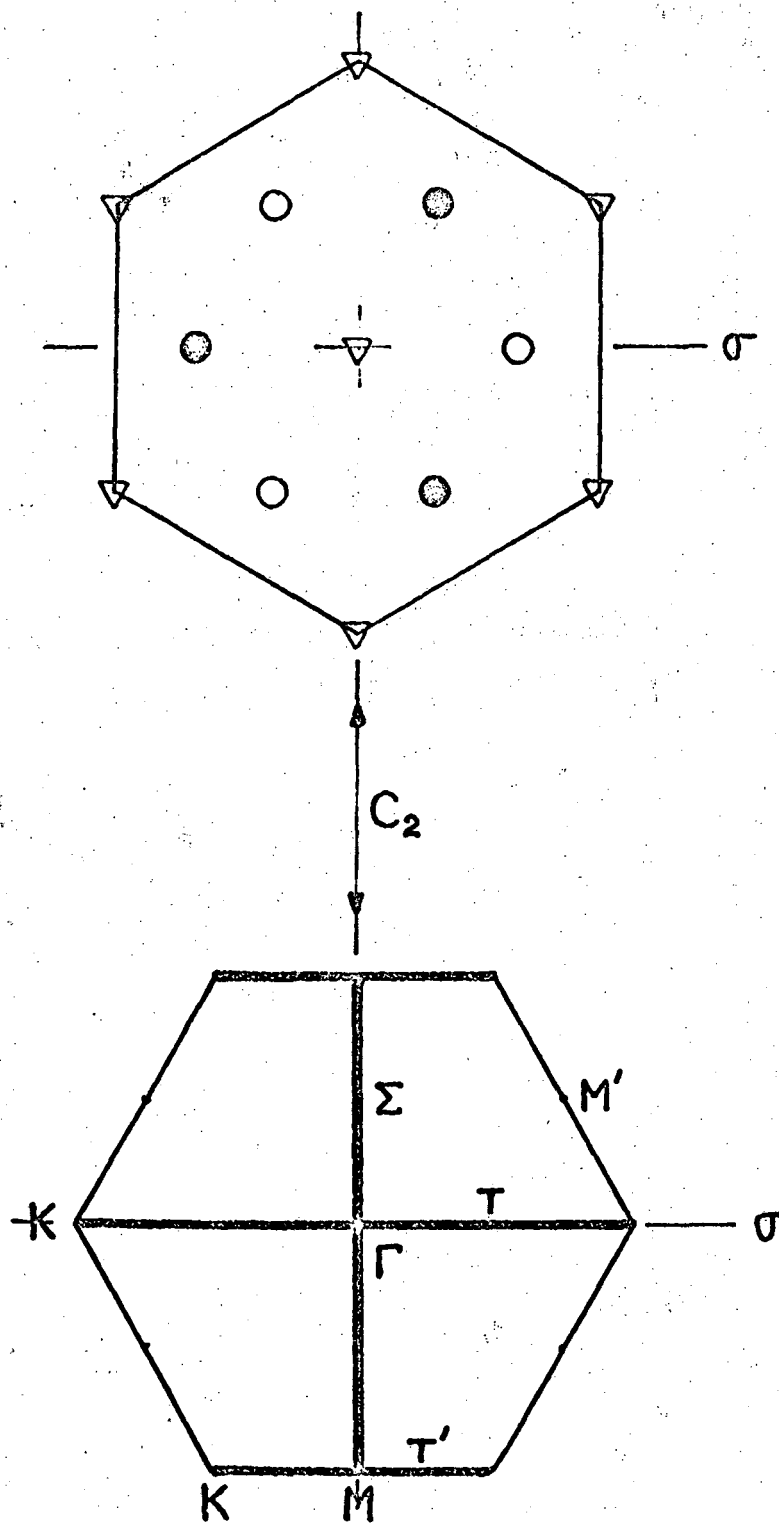
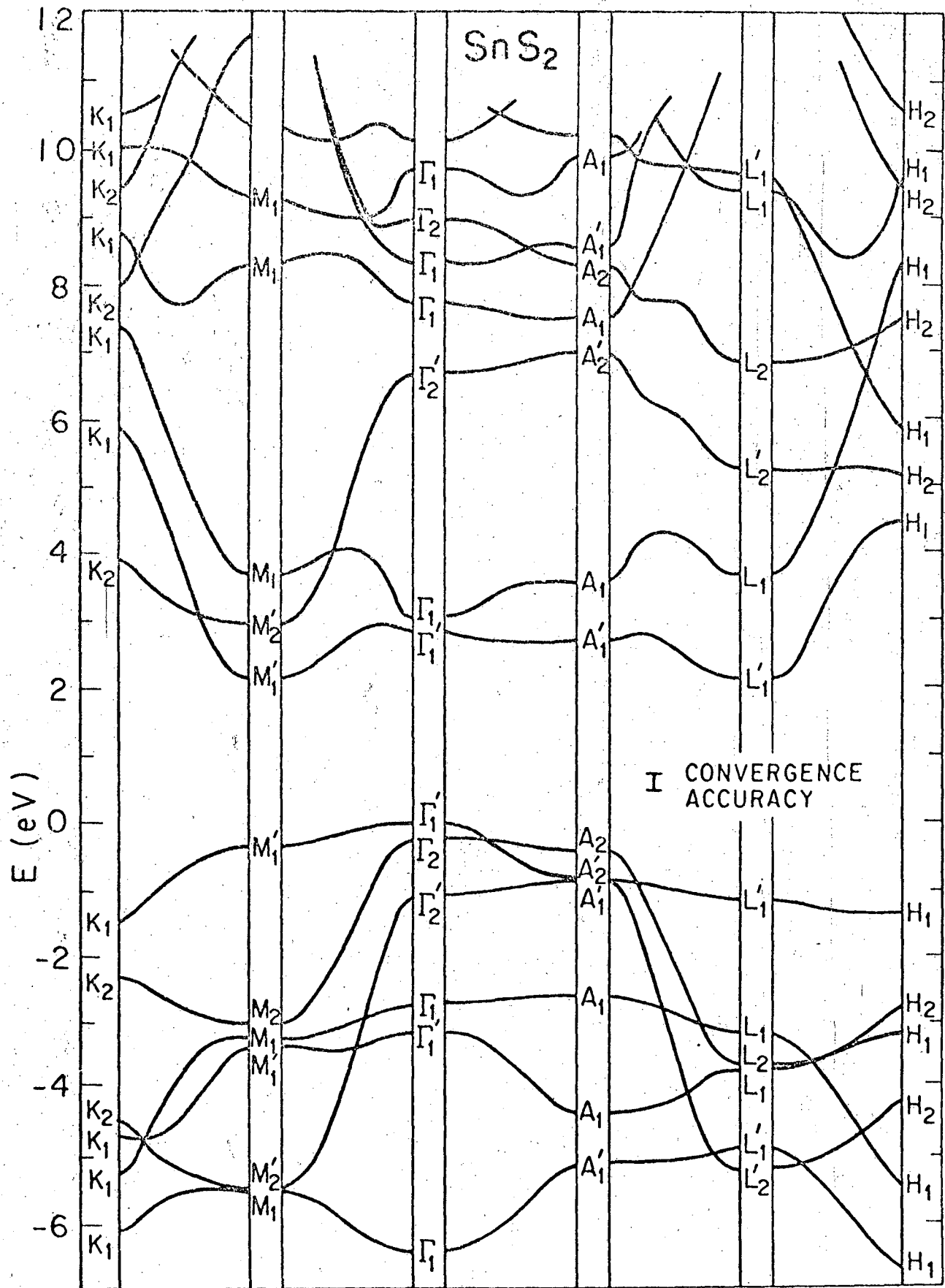


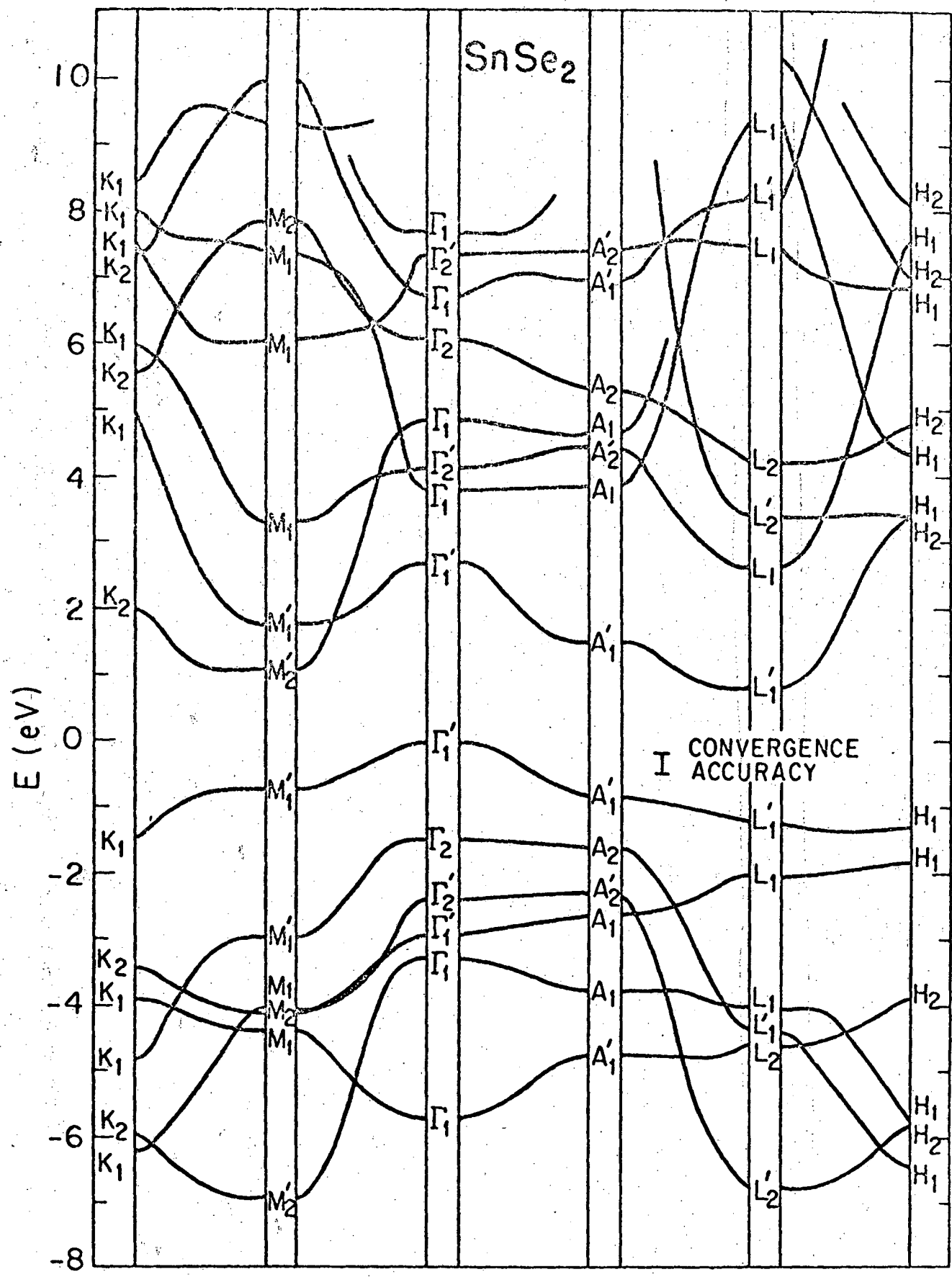
Fig. 1





k

Fig. 2



k

Fig. 3

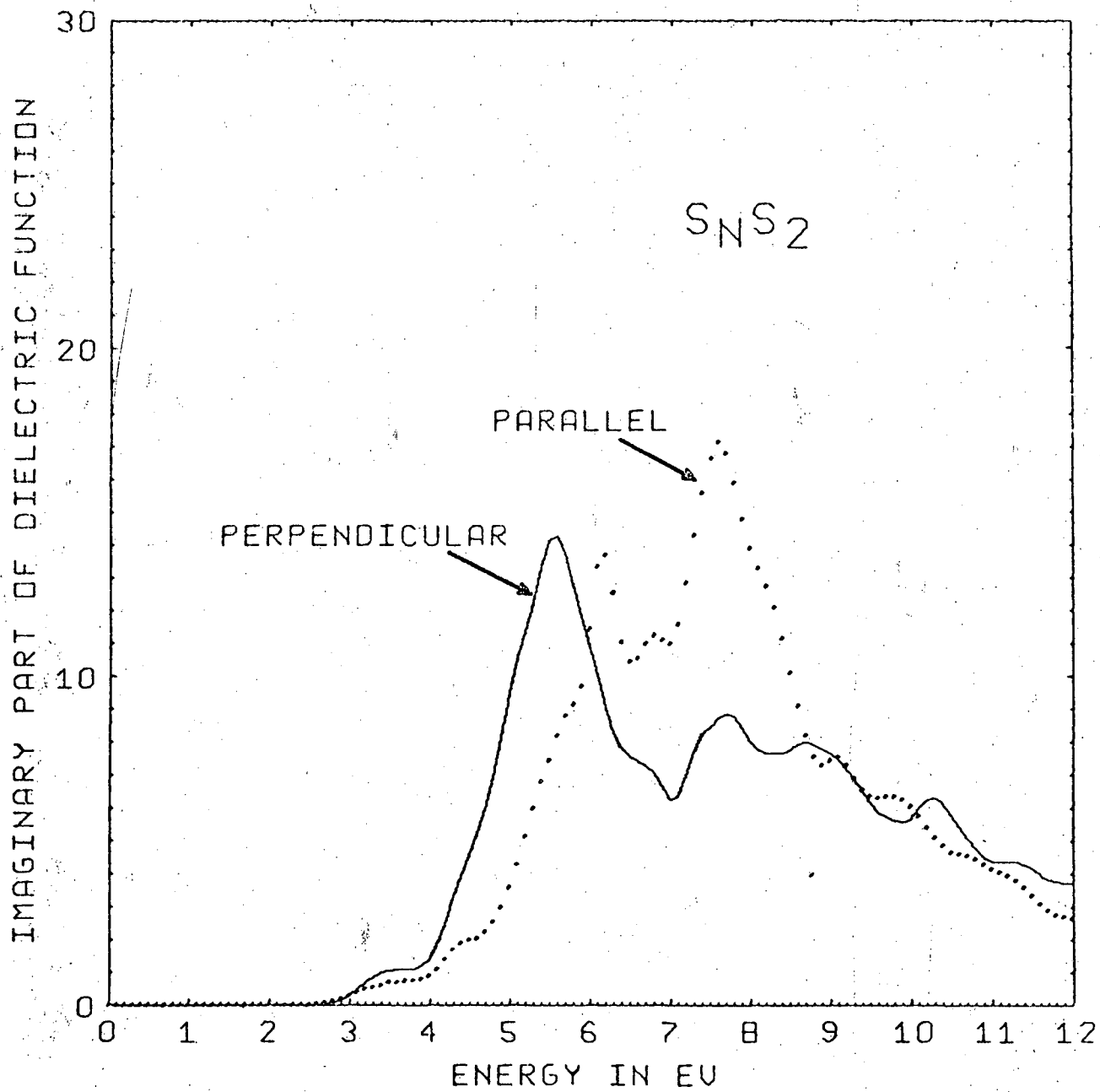


Fig. 4

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