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OPTICAL PROPERTIES, BAND STRUCTURE, AND SUPERCONDUCTING PROPERTIES OF SnTe AND GeTe (*)

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Abstract. — We present calculations of the electronic band structure (including spin orbit effects), the imaginary part of the frequency dependent dielectric function, and the superconducting transition temperature as a function of carrier concentration for SnTe and GeTe.

Résumé. — Nous présentons des calculs de la structure de bandes électronique (en tenant compte de l'interaction spin-orbite) de la partie imaginaire de la fonction diélectrique dépendant de la fréquence, et de la température de transition supraconductrice en fonction de la concentration de porteurs pour SnTe et GeTe.

Introduction. — SnTe and GeTe are extremely interesting materials. Despite the fact that in recent years some very able researchers have greatly increased our knowledge of these materials and other IV-VI compounds, several mysteries and controversies still remain. In particular, SnTe has recently been accused of having new, previously unsuspected, properties. Questions [1] have been raised about the crystal structure and phonon spectrum at low temperatures. Is SnTe fcc or fct like GeTe at low temperatures? Is it ferroelectric or a pseudoferroelectric, i.e. does the lowest TO phonon frequency ever get to zero frequency at very low temperatures?

Questions have also been raised about the electronic structure near the band edge and away from the band edge. It has been suggested [2] that the ordering of the levels at the band edge (L point of the Brillouin zone) reverses as one goes from PbTe to SnTe. It is also known that SnTe is a many valley semiconductor, but despite the beautiful experimental work using the de Haas-Shubnikov effect, optical and tunneling spectroscopy and other techniques, a complete model of the Fermi surface is still not available. Work has also been done on the visible and ultraviolet optical properties [3] of these materials to explore the electronic structure away from the fundamental gap, and a band structure and dielectric function calculation [4] for SnTe has been done to analyze this data. Because spin-orbit effects were not included in this calculation the ordering of the levels near the gap was not determined accurately. (We will discuss a new calculation with spin orbit effects in this paper.)

Finally, both GeTe [5, 6] and SnTe [7] are superconductors, and these crystals offer the possibility of doing detailed studies of the mechanisms which cause superconductivity.

In this paper, we will only focus on a few of these questions. In particular, the electronic band structure including spin-orbit effects will be discussed for both SnTe and GeTe; the results of the calculation of the imaginary part of the frequency dependent dielectric function will be described and compared with the ultraviolet optical spectrum; and a calculation of the dependence of the superconducting transition temperature on carrier concentration will be presented and compared with experiment.

Pseudopotential Calculation. — We have used the Empirical Pseudopotential Method [8] (EPM) to calculate the electronic band structure of SnTe and GeTe. We begin by describing the calculation without

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(†) Alfred P. Sloan Fellow.
spin-orbit interactions, we'll then go on to include these interactions and finally we will compare the results with experiment.

The pseudopotential Hamiltonian without spin-orbit interactions has the form
\[ H = -\left( \frac{\hbar^2}{2m} \right) V^2 + V(r) \]  
where \( V(r) \) is a weak effective potential. This potential is expanded in the reciprocal lattice
\[ V(r) = \sum_{\mathbf{G}} V(G) e^{-i\mathbf{G} \cdot \mathbf{r}} \]  
where \( \mathbf{G} \) is a reciprocal lattice vector and
\[ V(G) = \sum_{\alpha} V_{\alpha}(G) S_{\alpha}(G) \]  
\[ S_{\alpha}(G) = e^{-i\mathbf{G} \cdot \tau_{\alpha}}, \]  
\[ V_{\alpha}(G) = \frac{1}{Q} \int V_{\alpha}(r) e^{-i\mathbf{G} \cdot \mathbf{r}} d^3r \]  
where \( \alpha \) labels the element in a given compound and \( \tau_{\alpha} \) is the vector locating the elements in a unit cell.

For the rocksalt structure, the form factors \( V(G) \) can be divided into symmetric and antisymmetric components given by
\[ V(G) = V_4(G) + V_8(G) = 2 S^S(G) \]  
\[ V(G) = V_4(G) - V_8(G) = 2 S^A(G) \]  
where subscripts 1 and 2 refer to the two atoms in the unit cell. The above calculation does not include spin-orbit effects and it is essentially the formulation used by Lin et al. [4]. The form factors derived in this calculation are given in Table I.

To include spin-orbit effects, we use the model introduced by Weisz [9] for white tin. The total pseudopotential matrix element in the plane wave representation then becomes
\[ \mathcal{H}_{k',k} = \frac{\hbar^2}{2m} \delta_{kk'} \delta_{ss'} + \]  
\[ + S^S(k - k') [V^S(|k' - k|) \times \]  
\[ \times \delta_{ss'} - i\lambda_1(k' \times k) \sigma_{ss'}] \times \]  
\[ + i S^A(k - k') [V^A(|k' - k|) \times \]  
\[ \times \delta_{ss'} - i\lambda^A(k' \times k) \sigma_{ss'}]. \]  

where \( S^S \) and \( S^A \) are the symmetric and antisymmetric structure factors. Using the method of Herman et al. [11] for estimating s-o splittings in crystal from atomic splittings, we assume the splitting at \( \Gamma \) to be 0.98 eV and 0.77 eV for SnTe and GeTe respectively. The parameters \( \lambda^S \) and \( \lambda^A \) are chosen to give the estimated splitting at \( \Gamma \) and their ratio is taken to be consistent with the ratio of atomic s-o splitting of the two types of atoms. With this modification, we apply the EPM to SnTe and GeTe. Both compounds are assumed to have the rocksalt structure; the lattice constant is 6.313 Å for SnTe and 5.996 Å for GeTe.

The potentials are chosen in the following way. For SnTe, we start with the same set of symmetric form factors used in the previous EPM calculation [4] and vary the two antisymmetric form factors to give a few of the principal gaps. The symmetric form factors are then varied slightly in an attempt to get even closer to experiment. The form factors are given in Table I along with those of Lin et al. [4]. The largest variation is 0.004 Ryd in the symmetric potential and 0.02 Ryd in the antisymmetric potential.

<p>| Table I |</p>
<table>
<thead>
<tr>
<th>---</th>
<th>SnTe (Lin et al.)</th>
<th>SnTe (present calculation)</th>
<th>GeTe</th>
</tr>
</thead>
<tbody>
<tr>
<td>( V_4^S )</td>
<td>0.233 Ryd</td>
<td>0.232</td>
<td>0.245</td>
</tr>
<tr>
<td>( V_8^S )</td>
<td>0.028</td>
<td>0.024</td>
<td>0.022</td>
</tr>
<tr>
<td>( V_4^S )</td>
<td>0.016</td>
<td>0.018</td>
<td>0.032</td>
</tr>
<tr>
<td>( V_4^A )</td>
<td>0.041</td>
<td>0.555</td>
<td>0.060</td>
</tr>
<tr>
<td>( V_{11}^A )</td>
<td>0.004</td>
<td>0.023</td>
<td>0.017</td>
</tr>
</tbody>
</table>

Pseudopotential form factors for SnTe and GeTe.

For GeTe, the starting set of form factors was taken as an average of the Sb and As form factors.
FIG. 1. — Electronic energy band structure of SnTe.

FIG. 2. — Electronic energy band structure of GeTe.
extracted from the symmetric and antisymmetric form factors of InSb [8] and GaAs [8] (with appropriate scaling factors to account for the lattice constant changes). After this set was chosen, the form factors were varied slightly to give some of the observed gaps. The GeTe form factors are given in Table I.

The resulting band structures are given in figure 1 and figure 2. The valence band maximum and the conduction band minimum are both at the L point of the Brillouin zone as in the previous EPM calculation. However, the ordering of the levels near the gap is opposite to that found before for SnTe [4] and opposite to the PbTe [12] ordering. There is a second valence band maximum and conduction band minimum along ∏ and there is experimental evidence [13] for a second valence band maximum in SnTe. The ∏ maximum is a possible choice.

To analyze the visible and ultraviolet optical properties of these crystals, we have computed the imaginary part of the frequency dependent dielectric function, \( \varepsilon_2(\omega) \), for both crystals. This function has the form

\[
\varepsilon_2(\omega) = \frac{e^2 \hbar^2}{3 \pi m^2 \omega^2} \sum_k \left| \langle \delta(E_c(k) - E_v(k) - \omega) \right| < U_{k,v} | \nabla | U_{k,c} > \right|^2 d^3k,
\]

where \( U_{k,v} \) and \( U_{k,c} \) are the periodic parts of the valence and conduction band wave functions. The interband energies and the dipole matrix elements are obtained from the EPM eigenvalues and the eigenvectors.

\[
\begin{array}{cccc}
\text{Energy (in eV)} & \text{SnTe} & \text{Band Transition} & \text{GeTe} \\
0.33 & M_0 & L (5 \rightarrow 6) & 0.23 & M_0 & L (5 \rightarrow 6) \\
0.85 & M_0 & \Sigma (5 \rightarrow 6) & 1.06 & M_1 & \Sigma (5 \rightarrow 6) \\
1.75 & M_0 & \Sigma (5 \rightarrow 7) & 1.81 & M_1 & \Sigma (5 \rightarrow 7) \\
1.99 & M_2 & \Delta (5 \rightarrow 6) & 2.25 & M_2 & \Delta (5 \rightarrow 6) \\
3.0 & M_1 & \Sigma (4 \rightarrow 7) & 3.24 & M_1 & \Sigma (4 \rightarrow 7) \\
3.02 & M_2 & \Delta (4 \rightarrow 6) & 3.57 & M_2 & \Delta (4 \rightarrow 6)
\end{array}
\]

Energies and symmetries of critical points and band transitions associated with prominent optical structure are given in Table II.

The procedure used for evaluating Eq. (10) is the same as that used in reference [4]. In figure 3 we give the plots of \( \varepsilon_2(\omega) \); the energies and symmetries of the critical points responsible for the prominent optical structure are given in Table II.

For SnTe the onset of our \( \varepsilon_2(\omega) \) spectrum occurs near 0.3 eV due to the fundamental band gap at \( L \) and this is in good agreement with tunneling data [14]. The drop of intensity at 0.7 eV is not attributed to any critical point, but rather to a lack of bands with direct energy differences in this energy range. The spectrum rises again above 0.85 eV to give the first peak at 1.1 eV as compared to the experimental transmission peak at 0.97 eV [3]. This peak comes from a \( M_0 \) critical point arising from transitions from
band 5 to 6 at the second valence band maximum along $\Sigma$. Transitions from this same valence band maximum to band 7 give rise to the main peak at 2.0 eV; the decrease above this peak is due to a strong $M_2$ critical point form transitions along $\delta$. The 3.2 eV shoulder observed in the optical reflectivity spectrum is identified as arising from a $4 \rightarrow 7$ band transition along $\Sigma$ (critical point symmetry $M_1$ at 3.0 eV) and a $4 \rightarrow 6$ band transition along $\delta$ (critical point symmetry $M_2$ at 3.02 eV). The relative intensity of this shoulder is too low compared with optical reflectivity data. We attribute this to the fact that our pseudowavefunction dipole matrix elements for the $\delta$ transition here are about two orders smaller than that of the $\delta$ transition responsible for the main peak at 2.0 eV. The three high energy bumps 6.9 eV, 7.5 eV, 8.7 eV in our $e_2(\omega)$ are shifted considerably from their experimental [3] values 6.1 eV, 7.4 eV, and 9.5 eV respectively; they appear to arise mainly from clusters of critical points, i.e. there are a large number of transitions between bands with interband energy in the above range.

The analysis of GeTe $e_2(\omega)$ spectrum is very similar to that of SnTe. The onset of the spectrum comes from the fundamental band gap [15] at $L$ at 0.32 eV. The $e_2$ function then drops slightly because of lack of volume, but it rises quickly after 0.63 eV, as predicted by the onset of the experimental [16] transition spectrum. The main peak occurs at 2.1 eV. The shoulder at 3.3 eV is again low in intensity because of small matrix elements as well as lack of volume. The two high energy bumps 7.5 eV, 8.25 eV are shifted from the experimental [3] values 6.2 and 7.8 eV. Again these arise from clusters of critical points.

The agreement between the calculated optical spectrum and experiment are better at low energies than at higher energies. This is what one usually expects for the EPM, however, the shifts from experiment at high energies are relatively larger than observed for several other crystals [1, 17, 18]. We also note that we are comparing the calculated $e_2(\omega)$ with reflectivity since an experimental $e_2(\omega)$ is not available. A more detailed analysis for SnTe and GeTe will be published [19] elsewhere.

Superconductivity. — We now turn to the superconducting properties of GeTe and SnTe. In early investigations into the possibility of superconductivity in degenerate semiconductors our calculations [20, 21] indicated that superconductivity was possible in highly doped, many valley semiconductors with large static dielectric constants. It was expected that these materials would be superconducting around 0.1 °K, that the superconducting transition temperature, $T_c$, should be a function of carrier density, $n$, and that these materials would be type II superconductors.

Both GeTe and SnTe have the desired normal state properties, and the superconducting properties [6, 7] predicted above. At first there was some question about the superconductivity of these crystals particularly for GeTe which was investigated first. To show that superconductivity was a bulk property, the superconducting GeTe samples were investigated spectroscopically for superconducting impurities and none were found in any substantial amount. Experiments were done to test the original Ge and Te as well as the GeTe samples down to 0.04 °K and a superconducting transition was observed only for the compound. This ruled out the possible effects of any foreign impurity. To rule out the possibility that some other phase (formed in the fabrication of the compound) was responsible for the observed superconducting properties, the samples were powdered and tested magnetically. Resistive measurements would not be conclusive since, in this case (depending on the current) a transition could be observed when a single filament of superconductor was present across the sample. Magnetic susceptibility measurements of bulk samples could also be called into question as it was argued that a collection of superconducting filaments shielding the interior could give the appearance of bulk superconductivity. This was the reason for measurements on powdered samples. Although the positive results for powdered crystals should be conclusive, heat capacity measurements were done [22]. Such measurements show whether superconductivity is a bulk property of a sample or only a property of a small part of a sample. For the case of GeTe, the measurements showed that the major part of the GeTe sample was superconducting. This experiment firmly established GeTe as a superconductor. SnTe was subjected to similar investigations. Powdered samples were superconducting and heat capacity measurements [23] showed that superconductivity was a bulk effect.

The first superconducting property to be explored in detail was the dependence of the superconducting transition temperature on carrier concentration, $T_c(n)$; and for both GeTe and SnTe superconductivity was found to exist over a wide range of carrier concentration [6, 7, 24]. We present next a calculation of $T_c(n)$ for SnTe and GeTe. The calculation involves one adjustable parameter; this parameter, $\xi$, measures the strength of the coupling of electrons to short wavelength intervalley phonons.
The calculation follows the theory previously developed [25-27] for low carrier density superconductors. The theory is based on the BCS [28] theory of superconductivity, but retardation and renormalization effects are included [3].

The gap equation solved was

\[ D(\epsilon_k) = \frac{1}{Z(\epsilon_k)} \int_{\epsilon_F}^{\epsilon_{\text{max}}} D(\epsilon_{k'}) \frac{E(\epsilon_{k'})}{2 k_B T} \text{d} \epsilon_{k'} \times \]

\[ \times \frac{D(\epsilon_k)}{E(\epsilon_k)} K(c, \delta) \tanh \left( \frac{E(\epsilon_k)}{2 k_B T} \right) \text{d} \epsilon_k, \]  

(11)

where \( Z(\epsilon_k) \) is the renormalization,
\[ E(\epsilon_k) = (\epsilon_k^2 + A_k^2)^{1/2} \]

is the quasi-particle energy,
\[ D(\epsilon_k) = (k/k_F) A_k, \quad D(\epsilon_{k'}) = (k'/k_F) A_{k'}, \quad c = k/k_F, \]

\[ \delta = \hbar \omega_k \epsilon_k = (\epsilon_k - \epsilon_0) / E_k, \]

and \( A_k \) is the energy gap. \( \epsilon_{\text{max}} \) is a maximum energy determined from the band structure of the material and is of the order of the energy width of the band.

The kernel \( K(c, \delta) \) is given by

\[ K(c, \delta) = K^c(c, \delta) + K^e(c, \delta) + K^e(c, \delta), \]  

(12)

where \( K^c(c, \delta) \) arises from intravalley interactions (both Coulomb and phonon), \( K^e(c, \delta) \) arises from intervalley Coulomb interactions and \( K^e(c, \delta) \) arises from the intervalley interaction between electrons through phonons. The kernel essentially measures \( \frac{1}{2} N(O) V \) where \( N(O) \) is the density of states and \( V \) is the effective electron-electron interaction.

Since a description [25-27] of the procedure for obtaining the kernels from the intravalley and intervalley interactions is available we describe here only the interactions themselves.

The intravalley interaction is given by a bare Coulomb interaction divided by a frequency and wavevector dependent dielectric function containing both the lattice and free carrier polarizabilities

\[ V_{\text{in}} = \frac{4 \pi e^2}{q^2 \epsilon_0 + 4 \pi (\alpha_0^2 + \alpha_{\text{ph}}^2)} \]  

\[ 4 \pi \alpha_{\text{ph}} = \frac{\epsilon_0 - \epsilon_\infty}{\epsilon_0^2 - \omega^2} \]  

(13)

(14)

where \( \omega \) and \( q \) are the frequency and wavevector, \( \epsilon_0 \) and \( \epsilon_\infty \) are the low and high frequency dielectric constants and \( \alpha_0 \) is the free carrier polarizability which can be evaluated using the Lindhard model [25-27]. The interaction of Eq. (13) is equivalent [29] to a Coulomb interaction screened by a free carrier dielectric function plus an interaction of the Bardeen-Pines form between electrons.

For the intervalley interaction, it is convenient to separate the total interaction directly into a Coulomb and phonon part. The Coulomb part can be evaluated using a dielectric function arising from core polarizability (e. g. using Penn's [30] model) plus a free carrier polarization [25-27]. The intervalley phonon interaction is chosen to be of the Bardeen-Pines form with a parameter, \( \xi \), to measure the electron-phonon coupling.

The renormalization function \( Z(\epsilon_k) \) is taken to be

\[ Z = 1 + \lambda = 1 + 2 K_{\text{ph}} \]  

(15)

where \( K_{\text{ph}} \) is the phonon contribution to the kernel; the Coulomb renormalization is small.

The kernels are evaluated using essentially the band structure model described before. The Fermi surface is assumed to consist of hole pockets near the \( L \) point of the Brillouin zone. The intervalley phonon frequency is chosen to be an average of the \( LO \) and \( LA \) frequencies [31] at \( X \). Since the electron-phonon interaction is treated with an adjustable parameter, some inaccuracies of the above model are hidden in this parameter.

The results of the calculation using the above model are given in figure 4 along with the experimental results.
We find that screening effects become important at carrier concentrations, $n_c$, at which the free carrier contribution to the dielectric function becomes comparable to core polarizability. At this point the intervalley phonons become screened and the transition temperature is lowered. Such effects are seen in SrTiO$_3$ [27] at low carriers. For GeTe, we estimate that screening becomes important around $n_c = 5 \times 10^{21}/\text{cm}^3$; for SnTe we find $n_c$ to be one order of magnitude larger. The SnTe case is then particularly simple; it involves the coupling of relatively unscreened intervalley phonons to the electrons. Because only intervalley phonons dominate, the kernel of the gap equation can be fit by a two square-well model as a function of energy. This is similar to the model used by McMillan [32] (with Eliashberg kernels) for studying metallic superconductors. This model yields

$$T_c = \omega_0 \exp\left\{ \frac{-(1 + \lambda)}{\lambda - \mu^* - \omega_0 \omega_0^* - \lambda \mu^*} \right\}$$

(16)

$$\mu^* = \mu/1 + \mu \ln\left( \frac{\omega_{\text{max}}}{\omega_0} \right)$$

(17)

where $\mu$ is a Coulomb pseudopotential equal to twice the Coulomb kernel, $\omega_0$ is taken as the intervalley phonon energy, $\omega_0$ is an average phonon energy and $\omega_{\text{max}}$ is the cutoff for the Coulomb kernel ($\sim 1.0 \text{ eV}$). The BCS kernels discussed above, including retardation and renormalization effects, become identical to the Eliashberg kernels used by McMillan when evaluated at the Fermi surface. The intervalley deformation potential, $\xi$, is determined separately for the McMillan model. The values of $\xi$ along with the normal state parameters used are given in Table III. The dependence of $\lambda$ on carrier density is given in figure 5.

Hulm et al. [24] have also used the two square-well model to evaluate $T_c(n)$. However, these authors take a jellium model for the electron-phonon and Coulomb interactions and Fermi-Thomas screening. The results of their calculations for $T_c$ are smaller than the observed values. We believe that the jellium model is not appropriate for SnTe and GeTe.

![Figure 5. The phonon renormalization factor $\lambda$, as a function of carrier density for SnTe and GeTe.](image)

### Table III

<table>
<thead>
<tr>
<th></th>
<th>SnTe</th>
<th>GeTe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice parameter, a</td>
<td>6.319 Å</td>
<td>5.996 Å</td>
</tr>
<tr>
<td>Heat capacity $\gamma$ (mJ mole$^{-1}$K$^{-2}$)</td>
<td>1.38 (a)</td>
<td>1.32 (b)</td>
</tr>
<tr>
<td>Static dielectric const. $\varepsilon_0$</td>
<td>1770</td>
<td>~ 40. (c)</td>
</tr>
<tr>
<td>Infrared dielectric const. $\varepsilon_{\infty}$</td>
<td>45.0 (d)</td>
<td>~ 20. (c)</td>
</tr>
<tr>
<td>Intervervalley dielectric const. $\varepsilon_{\infty}^{\text{int}}$</td>
<td>~ 5.5</td>
<td>~ 4.0</td>
</tr>
<tr>
<td>LO phonon frequency at $q = 0$, $\omega_l$</td>
<td>0.017 eV (e)</td>
<td>~ 0.020 eV (c)</td>
</tr>
<tr>
<td>Intervalley phonon frequency $\omega_{\text{int}}$</td>
<td>0.007 eV (e)</td>
<td>~ 0.008 4 eV</td>
</tr>
<tr>
<td>Interval deformation potential $\xi$</td>
<td>2.35 eV</td>
<td>2.25 eV</td>
</tr>
<tr>
<td>McMillan</td>
<td>2.63 eV</td>
<td>2.75 eV</td>
</tr>
</tbody>
</table>


*c) Barker (S. A.), private communication.*

*d) Riedl (H. R.), Dixon (J. R.) and Schoolar (R. B.), Solid State Comm., 1965, 3, 323.*


Normal state properties of SnTe and GeTe.
In conclusion, we find that we are able to fit the observed $T_c(n)$ curve for both GeTe and SnTe with only one adjustable parameter for each crystal. Further details of this calculation will be presented elsewhere [33].

Note added in proof: In figures 1 and 2 there should be a small splitting between $\Delta_7$ and $\Delta_6$ bands when they cross bands of the same symmetry. This is not shown in the figures.

References

[10] Bloom (S.) and Bergstresser (T. K.), to be published.
[15] Chang (L. L.), Stiles (P. J.) and Esaki (L.), IBM Research and Development Journal, 1966, 10, 484.
[19] Tung (Y.) and Cohen (M. L.), to be published.
[25] See, for example, references 5 and 21.
[33] Allen (P. B.) and Cohen (M. L.), to be published.

DISCUSSION

Birman, J. — Can you give more detail on the calculation of pairing interaction due to intervalley scattering (electron-ion) and the screening of intervalley Coulomb interaction? Did you consider dispersion in the pairing interaction, or in the intervalley and intravalley Coulomb interaction? Did you use RPA type approximation for the «dielectric function»?

Cohen, M. — The intervalley phonon-electron interaction was evaluated using the deformation potential which I described. Dispersion was not included since $k_v \ll$ intervalley wave vector. For the intervalley Coulomb interaction, we estimated $\varepsilon_0$ using Penn's model. The RPA was used for the intravalley Coulomb interaction. The inaccuracies in this model and perhaps even some effects of other valence bands are all «hidden in» the intervalley deformation potential parameter.

Stiles. — If you plotted the transition temperature versus $1/Re$, what would a changing R factor do?

Cohen, M. — I don't know. We have compared our theoretical results with the published experimental $T_c(n)$ curve. We have assumed that the experimental curve contained the best available estimate of the carrier concentration.