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RECENT STUDIES OF LEAD POLONIDE (PbPo)

Richard Dalven

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RECENT STUDIES OF LEAD POLONIDE (PbPo)

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ABSTRACT

Lead polonide (PbPo) is the fourth member of the lead salt group of semiconductors, and has the same NaCl crystal structure as do PbS, PbSe, and PbTe. The empirical relation stating that the energy gap E_o of a semiconductor is directly proportional to $(1/a_o^2)$, where a_o is the lattice constant, has been found to be valid for a number of semiconductors with the NaCl crystal structure. Using this empirical relation, a plot of E_o as a function of $(1/a_o^2)$ for PbS, PbSe, PbTe, and PbPo has been used to calculate a value of $E_o \equiv E(L_6^-) - E(L_6^+) \cong -0.1$ eV for PbPo. Based on this result, it has been suggested that the band structure of PbPo may be inverted from that of PbS, PbSe, and PbTe. This inversion would result in PbPo having an L_6^+ conduction-band edge and an L_6^- valence-band edge. It would therefore imply a negative value of the temperature coefficient (dE_o/dT) of the energy gap in PbPo. The f-sum rule has been applied to this proposed inverted band structure and used to calculate values of the longitudinal and transverse effective masses as follows: for electrons, $(m_\ell^*/m) = 0.040$, $(m_t^*/m) = 0.024$; for holes, $(m_\ell^*/m) = 0.045$, $(m_t^*/m) = 0.028$. All of these results predicted for PbPo await the test of experimental study. It is suggested that experimental measurements on lead polonide of the energy gap, of the sign of its temperature coefficient, and of carrier effective masses would provide an interesting test of the correctness of the hypotheses on which the conclusions above are based.

ÉTUDES RÉCENTES DE POLONIDE DE PLOMB (PbPo)

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ABSTRAIT

Le polonide de plomb est le quatrième membre du groupe "PbS" des semi-conducteurs, et a le même structure crystallographique "NaCl" comme PbS, PbSe, et PbTe. La relation empirique énoncant que la zone interdite E_o d'un semi-conducteur est directement proportionnelle à $(1/a_o^2)$, où a_o est la constant du treillis, a été trouvée d'être valide pour un nombre des semiconducteurs avec le structure de NaCl. Faisant usage de cette relation empirique, un graphique d'E_o comme function de $(1/a_o^2)$ pour PbS, PbSe, PbTe, et PbPo a été employé pour calculer un valeur d'$E_o \equiv E(L_6^-) - E(L_6^+) \cong -0.1$ eV pour PbPo. Fondé sur ce résultat, on a suggéré que le structure des bandes de PbPo est peut-être renversé de ceux de PbS, PbSe, et PbTe. Ce renversement résulterait en le PbPo ayant un bord L_6^+ du bande de conduction, et un bord L_6^- du bande de valence. Cela impliquerait un valeur négatif du coefficient de la température (dE_o/dT) de la zone interdite de PbPo. Le regle "f-sum" a été appliqué à ce structure renversé des bandes et a été employé de calculer des valeurs des masses effectives longitudinales et transversales comme suivant: pour les électrons, $(m_\ell^*/m) = 0.040$, $(m_t^*/m) = 0.024$; pour les trous, $(m_\ell^*/m) = 0.045$, $(m_t^*/m) = 0.028$. Tous ces résultats prédit pour le PbPo attend l'épreuve d'étude expérimentale. C'est suggéré que des mesurages expérimentals, sur le PbPo, du valeur de la zone interdite, du signe du coefficient de la température, et des valeurs des masses effectives des porteurs, fourniraient des épreuves intéressantes du correction des hypothèses sur lesquels les conclusions en haut sont fondés.

I. Introduction

The compound semiconductors PbS, PbSe, and PbTe, collectively known as the lead salts, have been the subject of intensive research for many years. However, PbPo (lead polonide), the fourth member of this group, has been the object of only one experimental study[1], and the only data available are its crystal structure and lattice constant at room temperature. The work in this paper, published earlier in preliminary form[2,3], proposes a possible band structure for lead polonide. Using this band structure, values of the energy gap and of effective masses are calculated. While these proposals regarding PbPo are based on empirical evidence, the proposed band structure is amenable to an unequivocal experimental test.

The band edge structure of PbS, PbSe, and PbTe is comprised of a direct minimum energy gap E_o at the L point. The conduction band edge is an L_6^- state and the valence band edge is an L_6^+ state. Lead sulfide, selenide, and telluride all have the rock-salt crystal structure. Lead polonide also has the NaCl crystal structure, with a lattice constant[1] $a_o = 6.590$ Å at room temperature. This fact suggests that PbPo very likely has the same band structure as do PbS, PbSe, and PbTe, including a direct energy gap at the L point. Given this assumption, it is of considerable interest to calculate the energy gap $E_o \equiv E(L_6^-)-E_o(L_6^+)$ of PbPo. As discussed in detail below, knowledge of E_o for lead polonide would also be most useful in the study[4] of the anomaly in the energy gap values $[E_o(\text{PbTe}) > E_o(\text{PbSe})]$ in the lead salt group.

II. The Empirical Relation between Energy Gap and Lattice Constant

In order to calculate E_o for PbPo, the well-known[5] empirical relation between the energy gap E_o of a semiconductor and its lattice constant a_o was

considered. This relation states that E_o is directly proportional to $(1/a_o^2)$. It is known[5] to hold quite well for the Group IV elemental semiconductors diamond, silicon, germanium, and α-tin, thus covering a wide range of energy gap values.

In order to apply this relation to the lead salts, its validity for semiconductors with the rock-salt crystal structure was investigated. The following groups of semiconductors were considered, and values of E_o were plotted as a function of $(1/a_o^2)$ for: (1) solid solutions of SnSe in PbSe; (2) solid solutions of SnTe in PbTe; (3) BaS, BaSe, BaTe; (4) CaS, CaSe, CaTe; (5) SrS, SrSe, SrTe.

The values of E_o (at 12 K) for solid solutions $Pb_{(1-x)}Sn_xTe$, where $0.15 \leq x \leq 0.27$, were determined by laser emission studies.[6] The energy gap range corresponding to these values of x is $0.106\ eV \geq E_o \geq 0.044\ eV$. All solid solutions of SnTe in PbTe have the rock-salt crystal structure, and the linear experimental relation[7] between a_o and x was used to determine values of a_o (at ∿ 300 K) corresponding to the values of x for which E_o had been measured[6]. These values of E_o are plotted as a function of $(1/a_o^2)$ in Figure 1. The horizontal error bars shown were calculated by the author using an estimated uncertainty in x of ± 3%, a value typical of the results of electron beam microprobe analysis[8]. Also shown in Figure 1 are values of E_o for PbTe of 0.190 eV[9] (at 4 K) and 0.186 eV[10] (at 12 K); the horizontal error bars were also estimated by the author. It is clearly seen from Figure 1 that the empirical relation $E_o \propto (1/a_o^2)$ holds very well for the cubic solid solutions $Pb_{(1-x)}Sn_xTe$ in the range $0 \leq x \leq 0.27$.

A similar plot is shown in Figure 2 for solid solutions $Pb_{(1-y)}Sn_ySe$, which have the cubic rock-salt crystal structure for $y \lesssim 0.4$. Values of E_o

at 12 K for $Pb_{(1-y)}Sn_ySe$, with $0 \leq y \leq 0.276$, were also determined by laser emission[11,12]. The range of values of E_o corresponding to this range of composition is $0.149\ eV \geq E_o \geq -0.102\ eV$. The error bars shown correspond to uncertainties estimated by the author. It should be noted that these data cover both positive and negative values of the energy gap $E_o \equiv E(L_6^-) - E(L_6^+)$, where negative values of E_o corresponed to a band structure[10,11] in which the conduction band edge is an L_6^+ state and the valence band edge is an L_6^- state. This band structure is often referred to as inverted relative to the band structures of PbS, PbSe, and PbTe. The values of the lattice constant a_o of solid solutions $Pb_{(1-y)}Sn_ySe$ (at $\sim$300 K) were calculated using the linear relation between a_o and y observed[13] experimentally. From Figure 2 it is again seen that the relation $E_o \propto (1/a_o^2)$ is followed very well by these solid solutions of SnSe in PbSe. It is especially interesting that this empirical relation is valid for both positive and negative values of the energy gap. (It should be noted that this relation, shown in Figure 3, holds also for CdTe[14], HgTe[15], and solid solutions[16-19] $Hg_{(1-x)}Cd_xTe$ ($0.86 \geq x \geq 0.63$) with the zinc-blende structure[20], for both positive and negative values of the energy gap.)

The II-VI semiconductors BaS, BaSe, BaTe; CaS, CaSe, CaTe; and SrS, SrSe, SrTe, all have the rock-salt crystal structure[21]. They have large energy gaps[22], with $E_o > 3$ eV at $\sim$ 80 K for all nine compounds. These values[22] of E_o, however, appear to have been estimated from optical spectra[23] at $\sim$ 80 K. Since these spectra show strong exciton absorption, the energy gap values for these semiconductors must be regarded as approximate. Figure 4 shows E_o plotted as a function of $(1/a_o^2)$, using values[21] of a_o at $\sim$ 300 K for these nine semiconductors. While the fit of these points to straight lines is not as good as that of the data in Figures 1 and 2, it seems reasonable to conclude

that the relation $E_o \propto (1/a_o^2)$ is followed, at least approximately, by these semiconductors.

Based on the results presented in Figures 1,2, and 4, it is concluded that this empirical relation holds at least as well for compound semiconductors with the NaCl crystal structure as it does for the Group IV elemental semiconductors with the diamond structure. Given this conclusion, it is of interest to apply the relation $E_o \propto (1/a_o^2)$ to the sequence PbS, PbSe, PbTe, PbPo in order to calculate a value of the energy gap of lead polonide.

III. Calculated Value of the Energy Gap of PbPo

To calculate E_o for lead polonide using the known value[1] of a_o for PbPo, it is necessary to extrapolate a plot of E_o as a function of $(1/a_o^2)$ for PbS, PbSe, and PbTe. Using values of E_o at 77 K[9] and at 300 K[24], and values[25] of a_o at 300 K, for PbS, PbSe, and PbTe, the plots of E_o as a function of $(1/a_o^2)$ shown in Figure 5 were made. From the points in Figure 5, it is quite clear that the sequence Pbs, PbSe, PbTe does <u>not</u> follow the $E_o \alpha (1/a_o^2)$ relation. However, the straight lines connecting the points for PbS and PbSe have been extrapolated; the points for PbTe have <u>not</u> been used. This choice may be justified in the following manner. In recent papers[4], it has been proposed that PbTe is the irregular member of the sequence PbS, PbSe, PbTe, and this irregular nature of PbTe is the reason that $E_o(\text{PbTe}) > E_o(\text{PbSe})$. In making this proposal, it was pointed out that the outer s-electron energies[26] of the S, Se, and Po atoms are a linear function of their atomic number, whereas the Te atom 5s electron energy does <u>not</u> lie on this S-Se-Po line. This observation suggests the existence of an empirical regularity in the series of semiconductors PbS-PbSe-PbPo, a regularity <u>not</u> shared by PbTe.

For this reason, and because of their common crystal structure, it is suggested that PbS, PbSe, and PbPo form a regular sequence of related semiconductors with common properties. Specifically, it is assumed that (1) PbS, PbSe, and PbPo have similar band structures, including a direct gap E_o at L; (2) E_o for PbPo is smaller that E_o for PbSe; (3) the empirical relation $E_o \propto (1/a_o^2)$ holds for the sequence PbS, PbSe, PbPo; (4) E_o for PbPo may be determined by an extrapolation of the PbS-PbSe lines on a plot of E_o vs. $(1/a_o^2)$. Figure 5 shows this extrapolation of the PbS-PbSe lines for both 77 K and 300 K. It is seen that both lines intersect the value of $(1/a_o^2)$ for PbPo at <u>negative</u> values of E_o. Since the assumption has been explicitly made that the energy gap for PbPo is at the L point, this result implies that, for PbPo, $E_o \equiv [E(L_6^-) - E(L_6^+)] < 0$, leading to the conclusion that $E(L_6^+) > E(L_6^-)$ for PbPo. This result is interpreted as indicating that the band structure of PbPo may be <u>inverted</u> from those of PbS, PbSe, and PbTe, leading to an L_6^+ conduction band edge and an L_6^- valence band edge in PbPo.

A second conclusion drawn from Figure 5 concerns the temperature coefficient of the energy gap of PbPo. Since PbS, PbSe, and PbTe all have unusual <u>positive</u> values of (dE_o/dT), it is of particular interest that an inverted band structure implies a negative value of the temperature coefficient of the energy gap of PbPo between 300 K and 77 K. If we let $E_o' \equiv E(L_6^+) - E(L_6^-)$ be the energy gap of PbPo, then from Figure 5, the values of E_o' for PbPo are approximately 0.03 eV and 0.10 eV at 300 K and 77 K, respectively. The temperature coefficient (dE_o'/dT) for PbPo would then have (assuming the usual linear variation of energy gap with temperature) a value of approximately -3×10^{-4} eV/K. This value is typical of the magnitude and sign of the temperature coefficient of the energy gap for most semiconductors. It may also be noted

that this change in the sign of the temperature coefficient from positive to negative, as the energy gap changes sign from positive to negative, has been observed[8,11] experimentally for solid solutions of SnSe in PbSe.

IV. Calculated Carrier Effective Masses in PbPo

It has been shown[4] that experimental values of longitudinal and transverse effective masses and of the energy gap for PbTe do not follow the same f-sum rule[27] as do the corresponding data for PbS and PbSe. This result implies that the f-sum matrix element (considering for simplicity a two-band f-sum with only one term) is the same for PbS and PbSe. If the assumption (equivalent to assumption (1) in Section III) is now made that PbPo follows the same f-sum rule as do PbS and PbSe, effective mass values for PbPo may be calculated using values[4] of the f-sum matrix elements previously determined for PbS and PbSe. For a band model involving only the L_6^+ conduction band and L_6^- valence band of PbPo, the effective mass m* of electrons in the L_6^+ band is given[4] by

$$m_j^*/(m - m_j^*) = (m/2|M_j|^2)E_o', \qquad (1)$$

where $E_o' \equiv E(L_6^+) - E(L_6^-)$, M_j is the f-sum matrix element, j = longitudinal (ℓ) or transverse (t), and m is the free electron mass. For holes in the L_6^- band, the negative sign on the left hand side of equation (1) is replaced by a plus sign. Using values[4] of $|M_\ell|$ and $|M_t|$ obtained for electrons and holes in PbS and PbSe, the longitudinal and transverse effective masses m_ℓ^* and m_t^* were calculated using $E_o' = 0.1$ eV and assuming the (inverted) band structure proposed for PbPo. For electrons (L_6^+ band), $|M_\ell| = 3.32$, $|M_t| = 4.32$; for holes (L_6^- band), $|M_\ell| = 3.26$, $|M_t| = 4.08$; all in units of 10^{-14} $g^{1/2}$- $(eV)^{1/2}$. The calculated effective mass values are shown in Table I, and are

appropriate for 80 K.

Table I. Calculated Effective Masses in PbPo

Carrier	(m_t^*/m)	(m_ℓ^*/m)	(m_c^*/m)
Electrons	0.024	0.040	0.029
Holes	0.028	0.045	0.032

Since PbS and PbSe have ellipsoidal surfaces of constant energy it is reasonable to assume the same for PbPo, and the values of the conductivity effective mass $m_c^* = (3m_\ell^* m_t^*)/(2m_\ell^* + m_t^*)$ for PbPo shown in Table I were so calculated. It should be noted that the calculated values of m_ℓ^* and m_t^* for PbPo are smaller than the corresponding values for PbS and PbSe. This is a direct consequence of the assumed appropriateness of the f-sum rule, since the effective mass so calculated will vary directly with the energy gap of the semiconductor.

V. Relation to the Energy Gap Anomaly in the PbS Group

As mentioned earlier[4], the fact that E_o(PbTe) is larger than E_o(PbSe) is an anomaly in that E_o does not exhibit the usual monotonic decrease with anion atomic number. The proposal[4] that PbTe is the irregular member of the sequence was based on an examination of the f-sum rule at the conduction and valence band edges in PbS, PbSe, and PbTe. The fact that PbTe does <u>not</u> follow the same f-sum rule as do PbS and PbSe is interpreted as indicating that the f-sum matrix element for PbTe is different from that for PbS and PbSe. This effect is ascribed to a qualitative difference in the L_6^- conduction band edge state for PbTe, reflecting the irregular value of the Te atom

5s electron energy, which is responsible also for the anomalously large value of E_o(PbTe).

An experimental study[28] of E_o' and (dE_o'/dT) for PbPo would provide interresting evidence concerning the validity of the premises and conclusions mentioned above. The properties herein proposed for the band structure of PbPo $[E(L_6^+) > E(L_6^-), (dE_o'/dT) < 0]$ stem directly from the proposal above concerning the irregular nature of PbTe. This proposal was the justification for deriving the properties of PbPo by extrapolation of the properties of PbS and PbSe alone. Should experiment confirm the properties suggested for PbPo, it would provide corroborative evidence regarding the irregular nature of PbTe. Further, an experimental determination of carrier effective masses in PbPo in agreement with the values calculated in this paper would provide indirect confirmation of the proposed value of the energy gap of PbPo. Such agreement would then also confirm the applicability of the PbS-PbSe f-sum rule to PbPo and thus would also confirm the appropriateness of the extrapolation shown in Figure 5. If observed, then, experimental values of effective masses in PbPo in agreement with the calculations of this paper would provide further information concerning the interesting energy gap anomaly in the lead salt group. For these reasons, it is suggested that experimental determinations of the energy gap, its temperature coefficient, and effective masses in PbPo would be very worthwhile studies.

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PbSe is E_o = 0.149 eV, differing from the value (E_o = 0.145 eV) determined by interband magneto-absorption studies[9].

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28. The preliminary report (Phys. Rev. Letters 28, 91 (1972)) of this work makes suggestions concerning a possible optical method of measuring the sign of (dE_o'/dT) in PbPo.

Figure Captions

Figure 1. E_o plotted as a function of $(1/a_o^2)$ for solid solutions of SnTe in PbTe.

Figure 2. E_o plotted as a function of $(1/a_o^2)$ for solid solutions of SnSe in PbSe.

Figure 3. $E_G = E(\Gamma_6) - E(\Gamma_8)$ plotted as a function of $(1/a_o^2)$ for solid solutions of HgTe in CdTe.

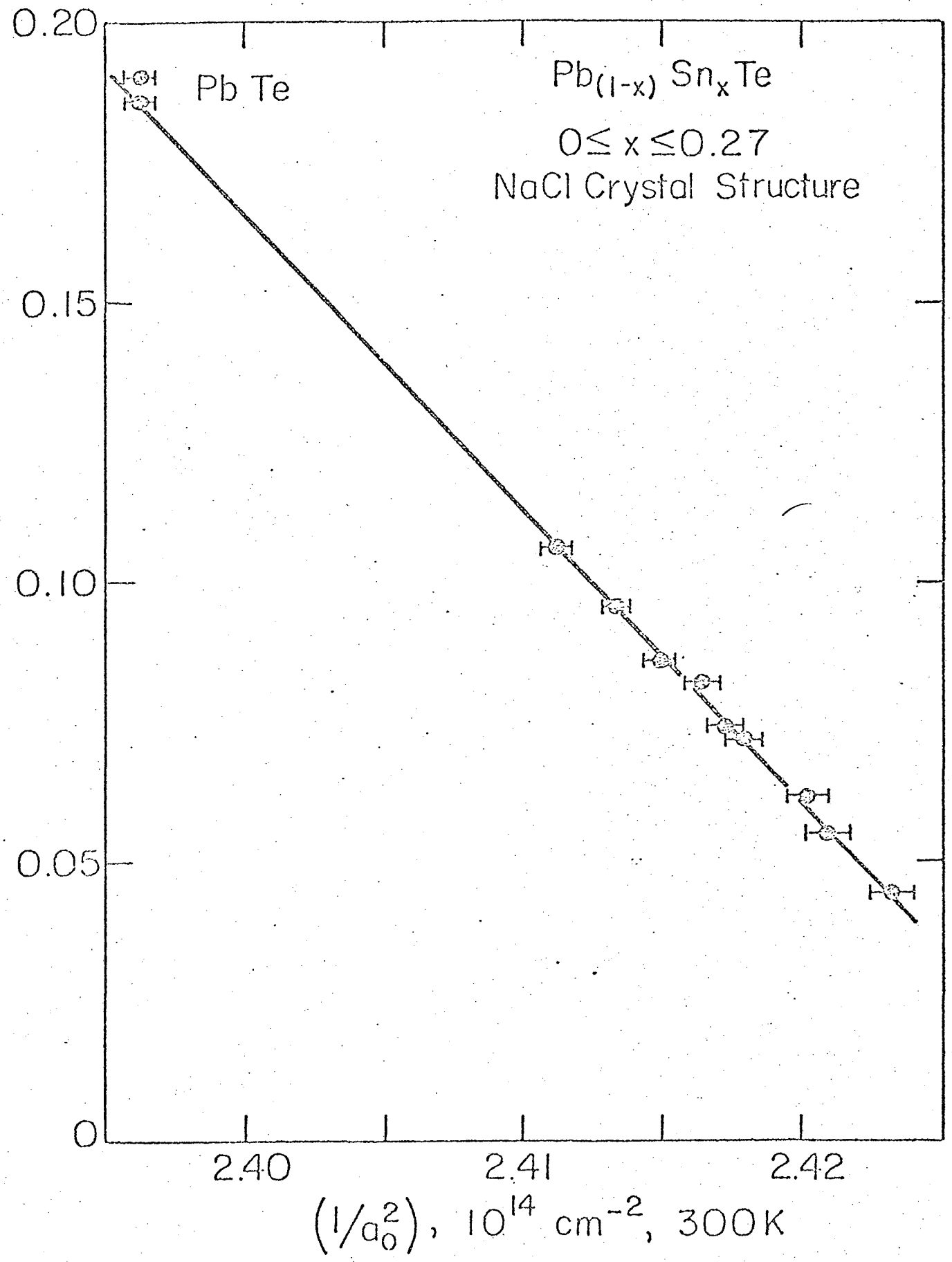
Figure 4. Energy gap plotted as a function of $(1/a_o^2)$ for CaS, CaSe, CaTe; SrS, SrSe, SrTe; BaS, BaSe, BaTe.

Figure 5. E_o plotted as a function of $(1/a_o^2)$ for PbS, PbSe, PbTe, and PbPo.

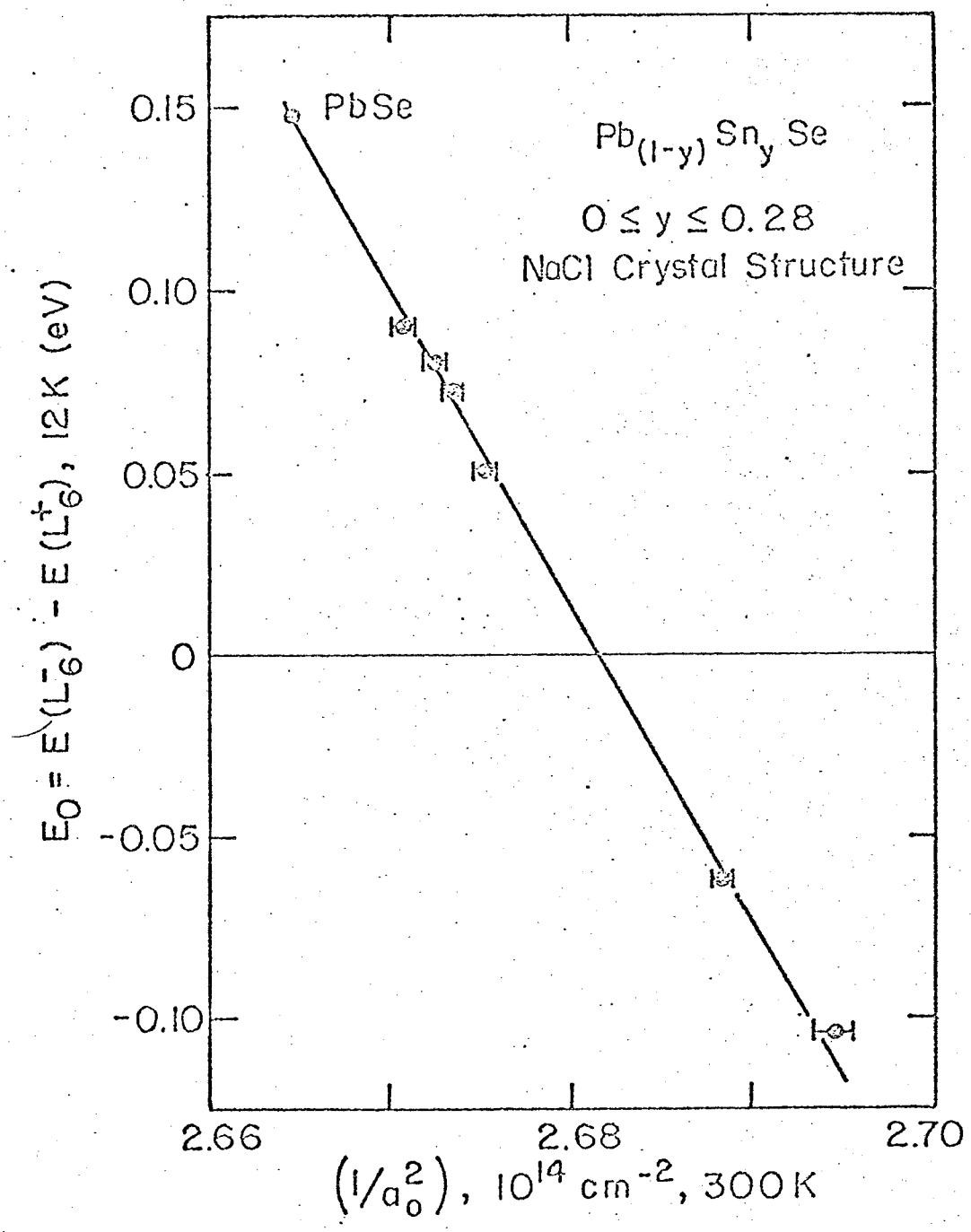
Tables

Table I. Calculated Effective Masses in PbPo.

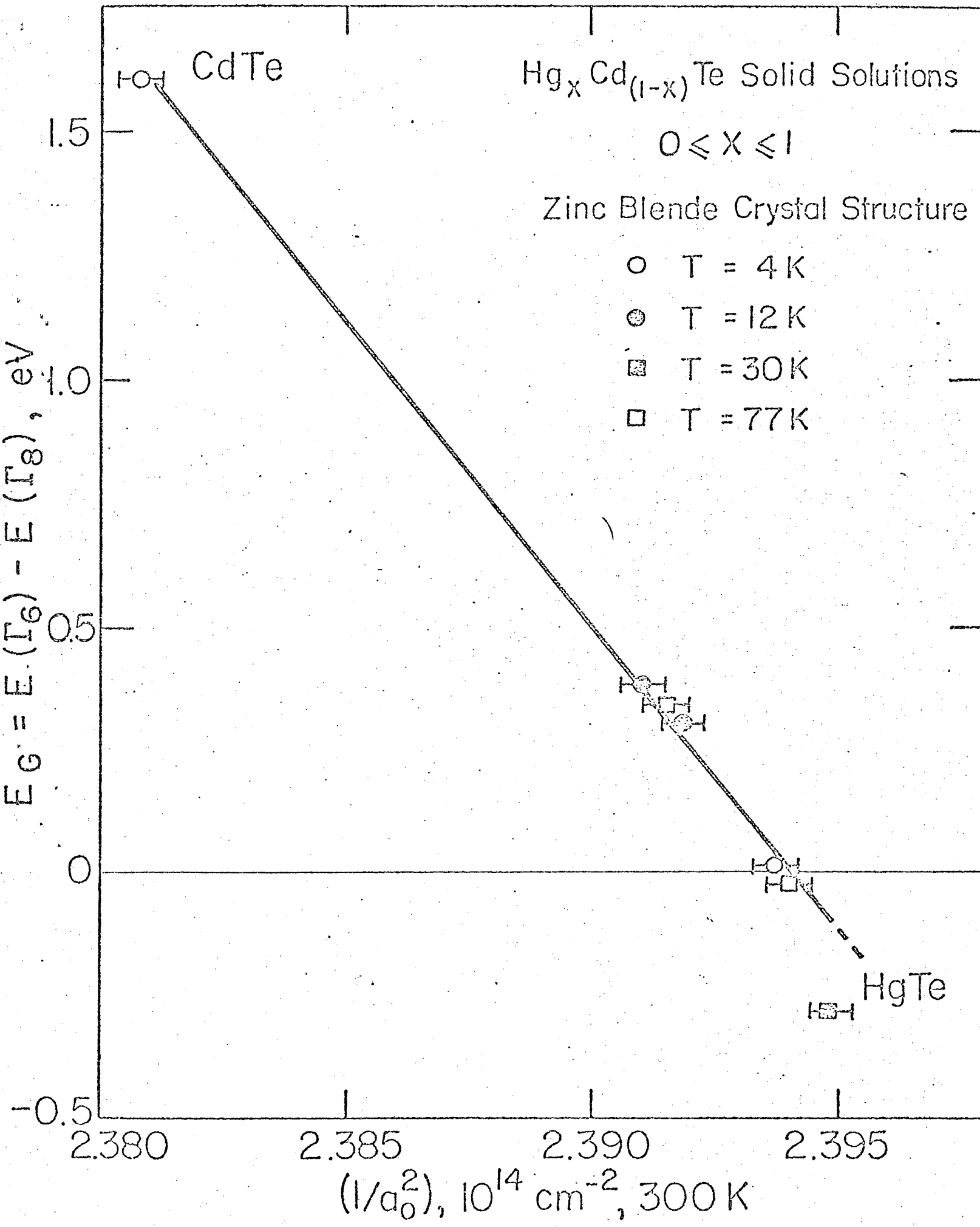
1.

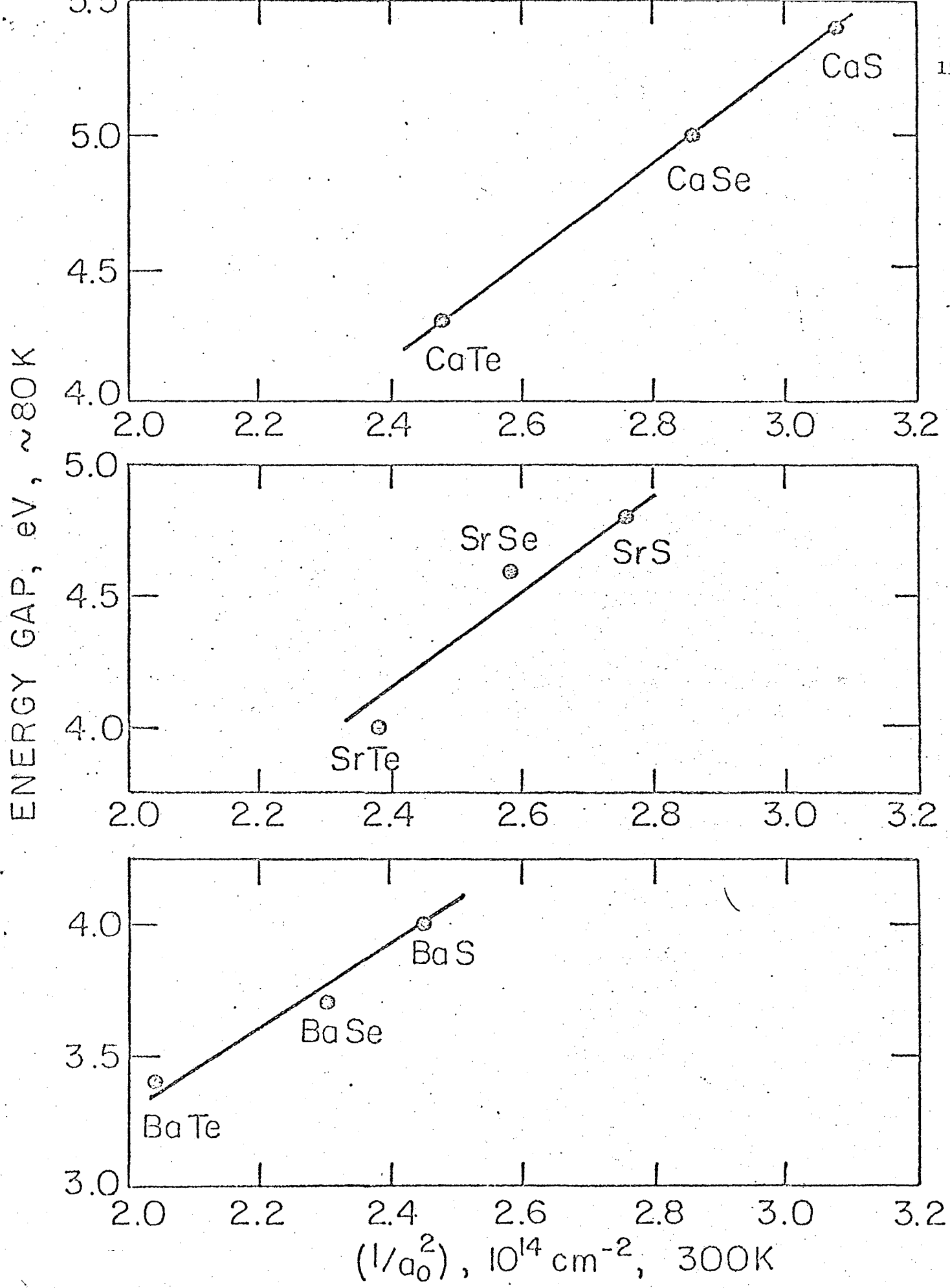


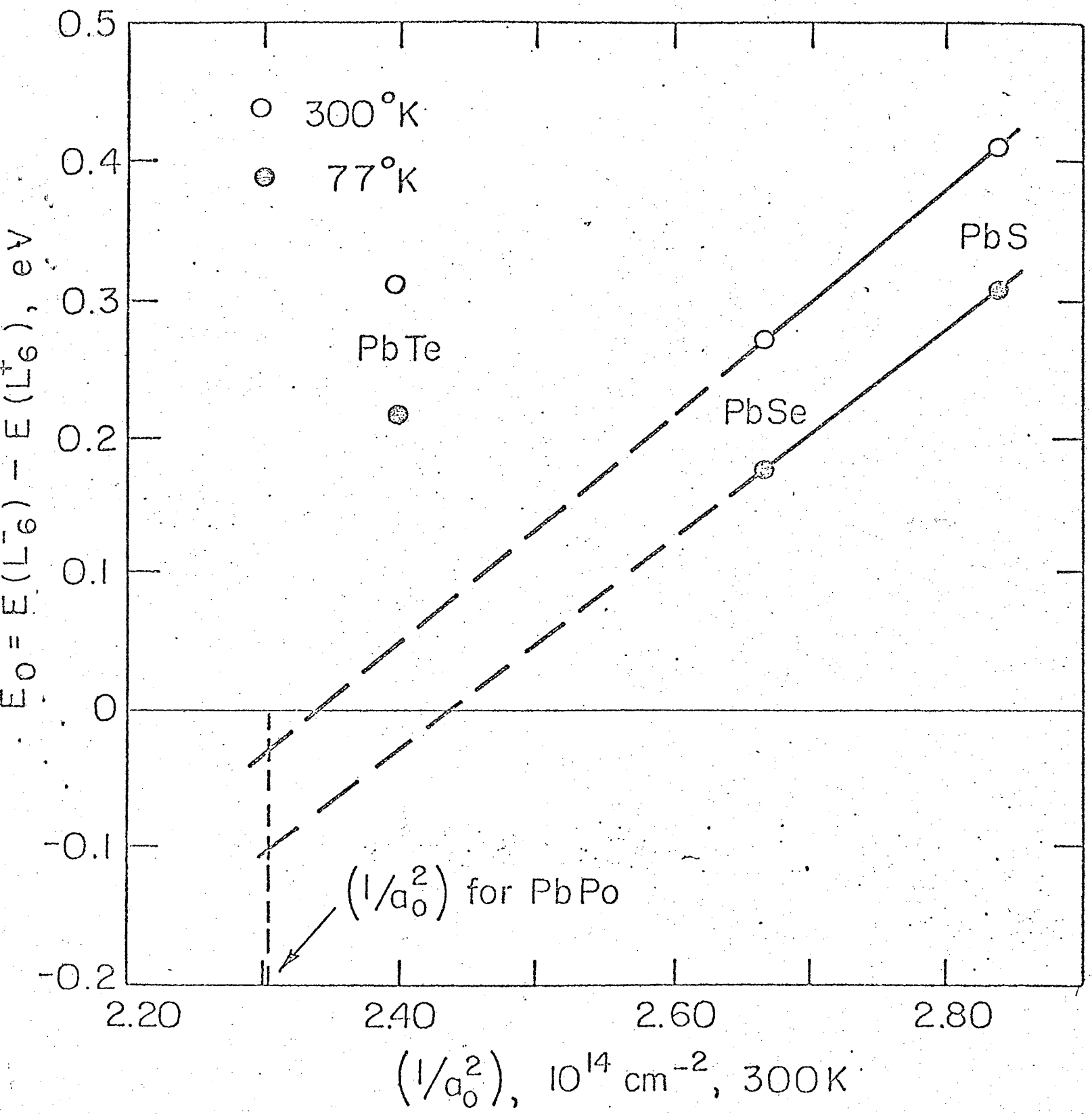
XBL 728-6823


PbSe
$Pb_{(1-y)}Sn_ySe$
$0 \leq y \leq 0.28$
NaCl Crystal Structure
0.15
0.10
0.05
0
-0.05
-0.10
$E_0 = E(L_6^-) - E(L_6^+)$, 12 K (eV)
2.66
2.68
2.70
$(1/a_0^2)$, 10^{14} cm^{-2}, 300 K

XBL728-6822


CdTe
$Hg_xCd_{(1-x)}Te$ Solid Solutions
$0 \leqslant X \leqslant 1$
Zinc Blende Crystal Structure
T = 4 K
T = 12 K
T = 30 K
T = 77 K
HgTe
$E_G = E(\Gamma_6) - E(\Gamma_8)$, eV
1.5
1.0
0.5
0
−0.5
2.380
2.385
2.390
2.395
$(1/a_0^2)$, 10^{14} cm^{-2}, 300 K


ENERGY GAP, eV, ~80K
5.0
4.5
4.0
CaS
CaSe
CaTe
2.0
2.2
2.4
2.6
2.8
3.0
3.2
5.0
4.5
4.0
Sr Se
SrS
SrTe
4.0
3.5
3.0
Ba S
Ba Se
Ba Te
$(1/a_0^2)$, 10^{14} cm^{-2}, 300K


0.5
0.4
0.3
0.2
0.1
0
-0.1
-0.2
$E_0 = E(L_6^-) - E(L_6^+)$, eV
300°K
77°K
PbTe
PbSe
PbS
$(1/a_0^2)$ for PbPo
2.20
2.40
2.60
2.80
$(1/a_0^2)$, 10^{14} cm^{-2}, 300K

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