

Point Defects in $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ Crystals

J. Horák,* S. Karamazov,† P. Nesládek,† and P. Lošťák†

**Institute of Inorganic Chemistry, Academy of Sciences of the Czech Republic, Pelléova 24, 16000 Prague, Czech Republic; and*

†*Faculty of Chemical Technology, University of Pardubice, Čs. legii sq. 565, 53210 Pardubice, Czech Republic*

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Improved ideas on point defects in the $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ crystals are formulated in this paper. Measurements of the Hall coefficient for a series of $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ ($x=0-1$) crystals served to determine the concentration of holes as a function of the selenium content in the lattice of the mixed crystals. The obtained variations of the hole concentration, linked hitherto solely with the decrease in the concentration of antisite (AS) defects of the Sb'_{Te} type, are explained using a model describing the interaction of the Se atoms entering the lattice not only with AS defects but also with the vacancies in the tellurium sublattice. The original concentration of AS defects in undoped Sb_2Te_3 crystals, approximately 11.3×10^{19} , drops with increasing x , reaching for the $\text{Sb}_2\text{Te}_2\text{Se}$ crystal a value of $3 \times 10^{19} \text{ cm}^{-3}$; the concentration of vacancies in the anion sublattice drops slightly: from a value of $2.5 \times 10^{19} \text{ cm}^{-3}$ down to $0.7 \times 10^{19} \text{ cm}^{-3}$. The calculated variations of the concentration of holes, AS defects, and $\text{V}_{\text{Te}}^{\bullet\bullet}$ vacancies are in agreement with the idea that the increase in the polarity of bonds gives rise to a decrease in the concentration of AS defects and in the concentration of anion vacancies. © 1997

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1. INTRODUCTION

The mixed $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ ($x = 0-1$) crystals rank among the family of layered semiconductors with the space group $D_{3d}^5 - R\bar{3}m$. The structure of $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ and the variations of the structure parameters connected with the gradual incorporation of Se atoms into the Sb_2Te_3 lattice were determined in Refs. (1, 2). Increasing selenium content in the crystal lattice gives rise to variations in the physical properties of the mixed crystals, namely those of the transport coefficients and the optical properties in the infrared region; the reflectivity and transmittance of the crystals with various values of x were studied in Refs. (3, 4); the transport coefficients and reflectivity in the plasma resonance region were studied also in our earlier papers (5, 6). It follows unambiguously from these studies that gradual substitution of selenium atoms for tellurium in the crystal lattice of Sb_2Te_3 gives rise to an increase in the Hall constant, to

a drop of the electrical conductivity, and to a shift of the reflectivity minimum in the IR region toward longer wavelengths, which means that the concentration of holes decreases with increasing x . This effect is explained in Ref. (6) by the idea that the introduction of Se atoms into the tellurium sublattice is connected with the interaction of antisite (henceforth AS) defects with Se atoms, which results in a decrease in the concentration of AS defects.

The idea formulated in Ref. (6) is based on the assumption that the dominant defects in the Sb_2Te_3 lattice are AS defects of the Sb'_{Te} type, carrying one negative charge. This, however, does not respect the finding (7) that the native defects in the Sb_2Te_3 lattice are represented not only by AS defects but also by a nonnegligible concentration of positively charged vacancies in the tellurium sublattice.

In this article we present an improved view of the interaction between Se atoms entering the lattice and the native defects in Sb_2Te_3 ; we consider not only the variations in the concentration of AS defects but also the variations in the concentration of vacancies in the tellurium sublattice connected with the introduction of Se atoms into the crystal. Also, we focus our attention on the changes in the bonding parameters due to the incorporation of Se atoms into the crystal lattice of $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$.

2. EXPERIMENTAL

2.1. Sample Preparation

$\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ single crystals were prepared from 5N purity elements using a modified Bridgman method; the technique and conditions of growing perfect single crystals were described in our earlier paper (5).

The grown single crystals, 50–60 mm long and 8–10 mm in diameter, were well cleavable along (0001) planes—the orientation of cleavage faces was performed by means of the Laue back reflection technique. The content of selenium in the crystals was analyzed by means of a Link Systems 860, series 2, energy dispersion analyzer and a Tesla BS 300 scanning electron microscope. The samples for the measure-

ments of Hall constant were cut from central parts of the single crystals.

2.2. Measurements of Hall Constant and Determination of the Concentration of Free Current Carriers

The room temperature values of the Hall constant were determined using samples of dimensions $10 \times 3 \times 0.1\text{--}0.3 \text{ mm}^3$. The experimental geometry corresponded to $R_H(\mathbf{B} \parallel \mathbf{c})$. The samples were connected to an AC current supply of 170 Hz frequency, the constant magnetic field induction \mathbf{B} was equal to 1.1 T. The results are shown in Fig. 1 in the form of the R_H plot. Using the relation $R_H = \gamma A_1 / Pe$, where A_1 is the Hall factor, γ the anisotropy factor, and e the elementary charge, we determined the values of the concentration of the free carriers P from the experimental values of R_H . In view of the fact that the band structure of $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ mixed crystals is not known, the calculations were performed with the value $\gamma = 0.72$, published for Sb_2Te_3 (8); i.e., a simplification was adopted that—in the first approximation—the value of γ in $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ crystals is independent of x . This assumption is not in disagreement with the results published in Ref. (9), from which it follows that the value of the density-of-states effective mass and hence also the value of γ only very slightly varies with increasing Se content. Further it was assumed that, given the relatively high concentration of the free charge carriers, the value of A_1 is approximately equal to unity. The results are shown in Fig. 1. It is evident that with increasing Se content in the $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ crystals, the obtained dependence $p = f(x)$ is monotonously decreasing.

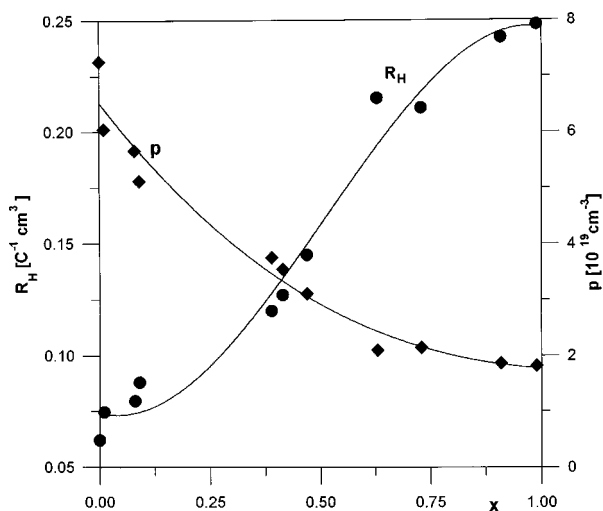


FIG. 1. Dependence of the Hall constant R_H and of the concentration of the free carriers of the content of selenium at room temperature.

3. DISCUSSION

Sb_2Te_3 crystals consist of layers, each layer comprising 5 atomic planes oriented perpendicular to the trigonal axis c and alternating according to the scheme



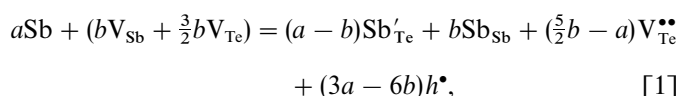
Characteristic coordination of the crystals is octahedral; the octahedra with Te^2 in the center are surrounded by 6 Sb atoms from the two neighboring atomic planes and are regular; the remaining octahedra centered about Sb or Te^1 are irregular. On formation of $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ mixed crystals, Se atoms always preferentially occupy the Te^2 sites (2, 10).

The Sb_2Te_3 crystals prepared from the melt of stoichiometric composition $2\text{Sb}/3\text{Te}$ exhibit always excess Sb, because tellurium partially segregates during growth. Superstoichiometric Sb atoms occupy prevalingly the Te^2 sites in the tellurium sublattice and thus give rise to AS defects of the Sb'_{Te} type, which are ionized at room temperature and whose charge is compensated by holes.

From the analysis of the relations among stoichiometry, concentration of free charge carriers and density of the crystals, the energy of formation AS defects in undoped Sb_2Te_3 crystals was determined and found to be $0.32 \pm 0.01 \text{ eV}$ (7). It was also stated that not all sites in the Te sublattice are occupied by superstoichiometric Sb atoms, but that there is also a nonnegligible concentration of vacancies in the Te sublattice of these crystals.

In (6) we formulated an idea on the explanation of the decrease in the concentration of holes observed on incorporation of Se atoms into the lattice: Se atoms substitute the sites in the tellurium sublattice, forming uncharged substitution defects $\text{Sb}^{\times}_{\text{Te}}$. Incorporation of an element more electronegative than Te into the tetradymite lattice leads to an increase in the polarity not only of the $\text{Sb--Se}_{\text{Te}^2}$ and Sb--Te^2 bonds, but also of the Sb--Te^1 bonds. The increase of the bond polarity results in an increase of the energy required to move Sb atoms into Te^2 sites, thereby decreasing the probability of formation of AS defects of the Sb'_{Te} type, the concentration of AS defects drops down and, consequently, the concentration of holes decreases. We improve this qualitative idea by taking into consideration the variations of not only of the concentration of AS defects but also those of the vacancies in the tellurium sublattice on incorporation of Se atoms into the crystal lattice. We proceed from the model of point defects presented in our earlier paper (7).

The incorporation of superstoichiometric Sb atoms, which always arises during the growth of Sb_2Te_3 crystals, can be described by the equation



where coefficients a and b stand for the stoichiometric ratios of interacting particles, V_{Sb} and V_{Te} are vacancies in the antimony and tellurium sublattice, Sb_{Sb} and Te_{Te} stand for Sb and Te atoms in the respective sublattice, and h^\bullet is a hole. This general formulation of the incorporation of superstoichiometric Sb into the lattice implies that in addition to the AS defects a certain concentration of $V_{\text{Te}}^{\bullet\bullet}$ defects in the crystal must be taken into account.

In our model of the point defects in Sb_2Te_3 crystals we adopt the following principles:

(a) The defects in question arise during the crystal growth, i.e., on transition from the melt into the solid phase; on further cooling their concentration can further vary, remaining constant below a certain temperature T_a . This assumption is acceptable in view of the fact that the formation energy of the defects is relatively high.

(b) As the crystal prepared from the melt of stoichiometric composition ($2\text{Sb} + 3\text{Te}$) shows high content of AS defects (more than 10^{20} cm^{-3}) (7), we suppose that

$$[\text{AS}] \gg [V_{\text{Sb}}^{-\Delta Z}].$$

(c) On the basis of the analysis given in Ref. (7) we also adopt the assumption that

$$[V_{\text{Te}}^{\bullet\bullet}] \gg [V_{\text{Sb}}^{-\Delta Z}].$$

(d) In this model we ascribe to a tellurium vacancy at room temperature 2 negative charges. The possibility of formation of defects of this type is considered also in the isostructure Bi_2Te_3 crystal (11).

(e) Under given conditions we can express the concentration of free carriers by the relation

$$[h^\bullet] = [\text{AS}] - 2[V_{\text{Te}}^{\bullet\bullet}]. \quad [2]$$

In undoped Sb_2Te_3 crystal the concentration of holes is given by

$$[h^\bullet] = k_1 \frac{[\text{Sb}_{\text{Sb}}]}{2} \exp\left(-\frac{E_{a,\text{AS}}(0)}{k_B T_a}\right) - 2[\text{Te}_{\text{Te}}] \exp\left(-\frac{E_{a,\text{V}}(0)}{k_B T_a}\right), \quad [3]$$

where k_1 is a statistical factor referring to the formation of a single AS defect in the tetradymite sublattice; irrespective of energy considerations it takes on a value of 1.2 (12). $[\text{Sb}_{\text{Sb}}]$ and $[\text{Te}_{\text{Te}}]$ are the numbers per 1 cm^3 of Sb and Te sites, k_B is the Boltzmann constant, $E_{a,\text{AS}}$ and $E_{a,\text{V}}$ are energies of formation of AS defects and $V_{\text{Te}}^{\bullet\bullet}$ vacancies corresponding to the model according to which the concentration of AS defects and $V_{\text{Te}}^{\bullet\bullet}$ vacancies does not change in the

temperature region $T < T_a$; knowing neither $E_{a,\text{AS}}$ and $E_{a,\text{V}}$ nor T_a , we introduce the substitution

$$\frac{E_{a,\text{AS}}(0)}{k_B T_a} = A(0), \quad \frac{E_{a,\text{V}}(0)}{k_B T_a} = B(0). \quad [4]$$

The values of $A(0)$ and $B(0)$ are constant for undoped material, whereas for the $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ mixed crystal the A and B parameters depend on the concentration c_{Se} of selenium atoms built into the lattice.

The value of $A(0)$ was determined in the same way as mentioned in our earlier paper (7), i.e., on the basis of chemical analysis, accurate values of lattice constants, and density of the crystal. Using these experimentally determined values the concentration of AS defects was calculated and found to be $1.13 \times 10^{20} \text{ cm}^{-3}$; this value was used to determine the parameter $A(0)$. We note that the values of the free-carrier concentration and crystal density are well reproducible if growing the Sb_2Te_3 or $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ crystals from stoichiometric melt. Therefore, one can assume that also the concentration of native defects will not vary.

Selenium atoms entering the Sb_2Te_3 crystals give rise to substitution defects $\text{Se}_{\text{Te}}^{\times}$; the polarity of $\text{Sb}-\text{Se}_{\text{Te}}^{\times}$ bonds increases, thus also increasing the energy of formation of AS defects, i.e., the energy required to move an Sb atom into a Te^2 site. This increase of the formation energy of AS defects depends on the concentration of Se atoms, c_{Se} ; the energy of formation of vacancies changes, too. In a mixed crystal, we can express the concentration of holes at room temperature by means of a relation

$$[h^\bullet] = k_1 \frac{[\text{Sb}_{\text{Sb}}]}{2} \exp(-A(c_{\text{Se}})) - 2[\text{Te}_{\text{Te}}] \exp(-B(c_{\text{Se}})). \quad [5]$$

Experimentally determined $[h^\bullet] = f(c_{\text{Se}})$ dependence was fitted using Eq. [5] and the obtained values of $A(c_{\text{Se}})$ and $B(c_{\text{Se}})$ are given in Table 1.

It is evident from these results that the formation energy of AS defects increases with increasing content of Se atoms in the lattice. Increasing formation energy of AS defects with increasing c_{Se} corresponds with the decrease of the concentration of AS defects, as documented in Table 1. The concentration of $V_{\text{Te}}^{\bullet\bullet}$ vacancies in the anion sublattice, i.e., the sum of concentration of vacancies $[V_{\text{Te}}^{\bullet\bullet}] + [V_{\text{Se}}^{\bullet\bullet}]$, drops somewhat, and that from a value of approximately $2.5 \times 10^{19} \text{ cm}^{-3}$ for undoped crystals to $0.7 \times 10^{19} \text{ cm}^{-3}$ for $\text{Sb}_2\text{Te}_2\text{Se}$. The mentioned relations are plotted in Fig. 2. Curve 1 shows the decrease in the concentration of AS defects, curve 2 shows the decrease of the concentration of electrons compensating the charge of the $V_{\text{Te}}^{\bullet\bullet}$ vacancies, and curve 3 corresponds to the change of the concentration of holes caused by the gradual substitution of Te^2 sites by Se atoms.

TABLE 1
Concentration of Holes, Calculated Concentrations of AS Defects, and $V_{\text{Te}}^{\bullet\bullet}$ Vacancies, and Formation Energies of AS Defects and $V_{\text{Te}}^{\bullet\bullet}$ Vacancies Using the Approximation $T_a = T_m$

x [-]	p [10^{19} cm^{-3}]	[AS] [10^{19} cm^{-3}]	[$V_{\text{Te}}^{\bullet\bullet}$] [10^{19} cm^{-3}]	p_{fit} [10^{19} cm^{-3}]	$A(x)$ [-]	$B(x)$ [-]
0.00	6.710	11.333	2.508	6.317	4.194	6.619
0.01	6.048	11.185	2.453	6.235	4.207	6.632
0.08	5.661	10.205	2.258	5.689	4.299	6.723
0.09	5.120	10.072	2.228	5.615	4.312	6.737
0.38	3.125	6.887	1.524	3.839	4.692	7.117
0.39	3.753	6.798	1.504	3.789	4.705	7.130
0.42	3.541	6.535	1.446	3.643	4.744	7.169
0.47	3.103	6.121	1.354	3.412	4.810	7.235
0.63	2.694	4.963	1.098	2.767	5.020	7.444
0.73	2.137	4.353	0.963	2.427	5.151	7.575
0.91	2.026	3.439	0.761	1.917	5.387	7.811
0.99	1.815	3.096	0.685	1.726	5.491	7.916

These results confirm the ideas on the relations between the bond polarity and the change of the concentration of AS defects, as qualitatively formulated above. The increase of the bond polarity and the resulting increase of the crystal ionicity with increasing Se content is corroborated also by the observed shift of the short-wavelength absorption edge toward shorter wavelengths, as shown in Refs. (2, 3).

Also, the decrease of the concentration of $V_{\text{A}}^{\bullet\bullet}$ vacancies during the process of substitution of Te sublattice sites by Se atoms is understandable from the following considerations.

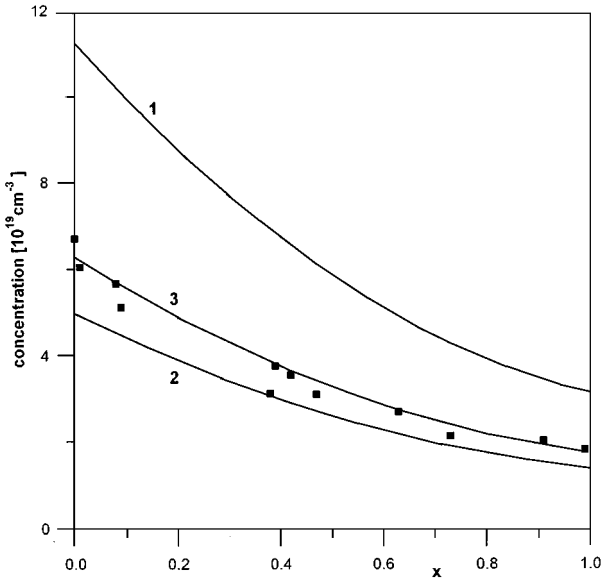
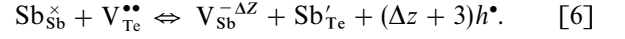
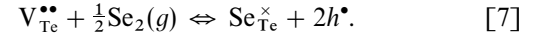


FIG. 2. Calculated concentration of AS defects (curve 1), concentration of free carriers compensating the charge of doubly charged vacancies $V_{\text{Te}}^{\bullet\bullet}$ (curve 2), and concentration of the free-charge carriers (curve 3) as functions of the content of Se.

In the lattice of the tetradymite crystals, where AS defects arise, we can formulate the interaction of superstoichiometric Sb atoms with vacancies in the Te sublattice by means of the equation¹



If we provoke by certain action, e.g., by incorporation of Se atoms into the Te sublattice, a suppression of the concentration of anion vacancies $V_{\text{Te}}^{\bullet\bullet}$, then in the sense of the equilibrium characterized by Eq. [6] the concentration of AS defects will decrease as well. This process is characterized by a decrease of the concentration of holes and connected with the transition of Sb atom from the AS defect site into a cation vacancy $V_{\text{Sb}}^{-\Delta Z}$, or, as the case may be, an Sb atom from the AS defect site will occupy the Sb sublattice site, which results in an increase of the crystal volume. Both these considered processes result in suppression of the concentration of holes. We note, however, that besides the suppression of the concentration of AS defects due to Se atoms entering the Te sublattice, one must also take into account the formation of substitution defects of the $\text{Se}_{\text{Te}}^{\times}$ type, which depends on the selenium vapor pressure over the crystal during growth which influences the concentration of holes according to the equation



As follows from Table 1, the concentration of vacancies $[V_{\text{A}}^{\bullet\bullet}]$ decreases on incorporating Se into Te sublattice. This fact confirms the correctness of the idea on the equilibrium between the point defects, as embodied in Eq. [6]. From the chemical point of view the result can be interpreted as follows: on increasing the selenium content the system $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ approaches the composition corresponding to the stoichiometry.

These results, however, do not make it possible to obtain data on the formation energies $E_{a,\text{AS}}$ and $E_{a,\text{V}}$. Admitting, however, that the system at the melting point approaches the equilibrium state and that the concentration of the native defects does not vary any more, we can substitute the melting point T_m for the temperature T_a . Under these conditions it holds that

$$\frac{E_{a,\text{AS}}}{T_a} = \frac{E_{\text{AS}}}{T_m}, \quad \frac{E_{a,\text{V}}}{T_a} = \frac{E_{\text{V}}}{T_m} \quad [8]$$

and, consequently, would give for the formation energy of the AS defects in the undoped crystal and in $\text{Sb}_2\text{Te}_2\text{Se}$ the values 0.32 and 0.42 eV, respectively.

¹ Equation [6] describes the interaction of defects as an equilibrium process, even if the formation of defects during crystal growth cannot be taken for an equilibrium. Nevertheless, we admit that the interaction of defects described by Eq. [6] approaches the equilibrium state.

In order to support the above-mentioned model based on qualitative ideas on the change in the bond polarity connected with the incorporation of Se atoms into the crystal, we performed a calculation of the charges on individual atoms in Sb_2Te_3 and $\text{Sb}_2\text{Te}_2\text{Se}$ and of the bond order using the quantum chemical program MOPAC 93 (13). Input data, definitions of bonding parameters, and the calculation procedure are given in the Appendix.

The results on calculated charges on individual atoms in Sb_2Te_3 and $\text{Sb}_2\text{Te}_2\text{Se}$ crystals are given in Table 2. The calculations also indicate that, in the first approximation, there is nearly linear dependence between the magnitude of the charge and the concentration of Se atoms built into the crystal lattice (Fig. 3). The magnitude of the charges of cation and anion sublattices essentially confirms the original bonding model proposed by Drabble and Goodman (14) for the tetradymite structure crystals; in this model it is assumed that the $6s^2$ electrons of the Te^2 atom are bonding and form together with $6p^4$ electrons of Te and 5 electrons of the Vb group element and one electron of the Te^1 atom a strong bond by the sp^3d^2 hybridization, whereas smaller bond ionicity is ascribed to the Te^1 atom (14); this conception is somewhat different from the presented calculation of the electron density given in Table 2. The calculated value of the charge is higher for the Te^2 atom than that for the Te^1 atom. For this reason the Te^2 -Sb bond is more polar than the Sb - Te^1 bond; this is in agreement with the crystallographic analysis, which shows that the Te^2 -Sb distance corresponding to the more polar bond is somewhat larger than the Sb - Te^1 distance (see Table 2). Substitution of Se atoms for Te^2 atoms gives rise to a pronounced increase of the charge of the Se atoms on Te^2 sites, the positive charge on the cation sublattice atoms increases. Simultaneously, the charge on Te^1 sites increases too. This results in a change of the distance Te^1 - Te^1 between the neighboring atomic planes of two layers; the increase of the charge on Te^1 implies an increase of the repulsive forces and thus an

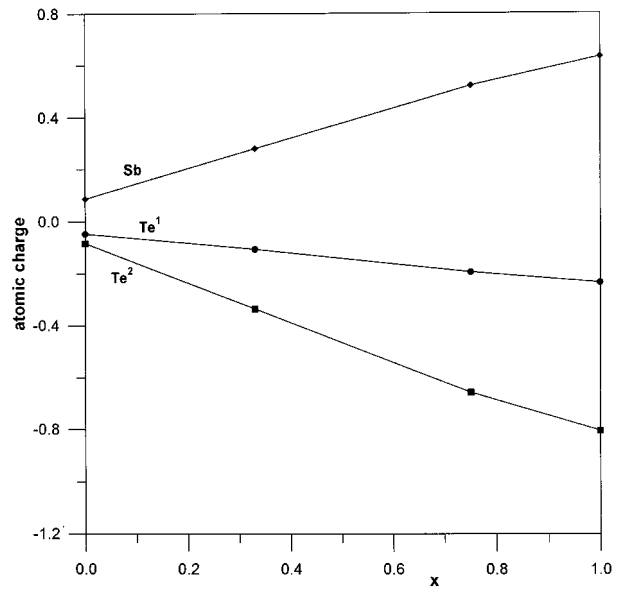


FIG. 3. Dependence of the atomic charge on the content of Se calculated using quantum-chemical program MOPAC 93.

increase of the Te^1 - Te^1 interatomic distances. This effect of higher negative charge of Te^1 atoms is more pronounced in the increase of the distance of the Te^1 - Te^1 atomic planes of two neighboring layers, i.e., of the van der Waals gap, which amounts to 2.87 Å for $\text{Sb}_2\text{Te}_2\text{Se}$ and 2.81 Å for Sb_2Te_3 (15). The increase of the polarity of Sb - Te^1 bonds in $\text{Sb}_2\text{Te}_2\text{Se}$ crystals in consequence of the increase of the positive charge of Sb atoms and the increase of the negative charge of the Te^1 sublattice atoms leads to a slight change in chemical bonds. The distance of the center of the regular octahedron from the nearest Sb atoms remains unchanged, although the covalent radius of the Se atom ($r_{\text{Se}} = 1.16$ Å) is noticeably smaller than that of the Te atom ($r_{\text{Te}} = 1.36$ Å); this is due to the fact that the Se charge on Te^2 site is, according to calculations, almost ten times larger than the charge on Te^2 atom in the undoped Sb_2Te_3 crystal. This calculated increase of the charge $\text{Se}_{\text{Te}^2}^{\times(-\delta)}$ is responsible for the lowering of the bonding order of the Sb - $\text{Se}_{\text{Te}^2}^{\times(-\delta)}$ bond.

The discussion of the properties of the $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ crystals implies that the substitution of selenium for the Te^2 atom results in a pronounced increase of the crystal ionicity, an increase of the Sb - Te^2 bond polarity, a decrease of the concentration of AS defects, and a decrease of the concentration of $\text{V}_{\text{Te}}^{\bullet\bullet}$, following from the discussion of the equilibrium described by Eq. [6]. In our opinion, the relations between the point defects expressed by the equilibrium in Eq. [6] are of general validity for the crystals of tetradymite structure.

TABLE 2
Charge on Individual Atoms and Structural Parameters of Sb_2Te_3 and $\text{Sb}_2\text{Te}_2\text{Se}$ Compounds

		Sb_2Te_3	$\text{Sb}_2\text{Te}_2\text{Se}$
Interatomic distances in Å (15)	Sb- Te^2 (Se^2)	3.17	3.03
	Sb- Te^1	2.98	2.98
	Te^1 - Te^1	3.74	3.75
Atomic charge	Te^1	-0.050	-0.23
	Sb	+0.093	+0.63
	Te^2	-0.085	-0.80
Bond order	$\text{BO}_{\text{Sb}-\text{Te}^1}$	0.538	0.430
	$\text{BO}_{\text{Sb}-\text{Te}^2}$	0.240	0.202

APPENDIX

With the objective to confirm the idea that the incorporation of Se atoms into the Te^2 sites leads to an increase in the bond polarity in the $\text{Sb}_2\text{Te}_{3-x}\text{Se}_x$ mixed crystals and hence increasing their ionicity, we used a quantum-chemical program MOPAC 93 to calculate the charges on individual atoms of the investigated crystal lattice.

Since the studied mixed crystals exhibit a relatively narrow gap and a considerable concentration of the free-charge carriers, we respected the requirement given in the manual (13), namely that the crystal section used to calculate the chemical bond parameters should have sufficiently large dimensions and suitable shape. Therefore, the calculations were carried out for a section of a crystal layer between the van der Waals gap, which contained 120 atoms and had prismatic shape, approximately 20 Å in the x - y plane. At the same time, we fulfilled the required conditions to close the chosen spatial form in such a way that conserved bonds between the initial and final atomic plane.

Definition of the Notions Used

Electron density refers to one atom and is given in units of elementary charge. It is given by the main diagonal members of the density matrix (13).

Atomic charge is calculated on the basis of the determined electron density, respecting the number of electrons on an atom.

Bond order between A and B atoms is an average order of the bonds of the same kind. It describes the multiplicity of a bond (i.e., corresponds to the electron density in the space occupied by a bond); it is a dimensionless quantity denoted

by BO . It is given by equation

$$\text{BO}_{AB} = \sum_{\lambda \in A} \sum_{\sigma \in B} P_{\lambda\sigma}^2, \quad [9]$$

where $P_{\lambda\sigma}^2$ is an element of the density matrix.

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