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Radiation-induced point defect in epitaxial layers of SnTe

B.K.Ostafiychuk, Ya.P.Saliy, V.M.Chobanuk, G.D.Mateik, M.V.Pyts
Departament of Solid State Physics, Precarpathian University, Shevchenko Str. 57, Ivano-Frankivsk, 76000, Ukraine

Dependence of lattice parameter and holes concentration in epitaxial layers p-SnTe on integral α-particles flux are investigated. Comparing rated and experimental data the conclusion on acceptor character of tin vacancies vjn and tellurium vacancies V_{Te}^{2-} have been made.

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I. Introduction

Type and concentration of free charge carriers in semiconductors $A^{IV}B^{VI}$ in the area of impurity conduction when the content of impurity does not exceed the background value are conditioned by crystal lattice intrinsic defects. Among the possible elementary defects types the vacancies of metal and chalcogen have been investigated thoroughly [1-8], the interstitial atom [9,10] and antistructural defects [11] have been reseached to a less extent. At present there is no single view point concerning the prevailed type of defects in $A^{IV}B^{VI}$ and its charge state. In addition, one and the same conduction type, i.e. for SnTe, in the material may be enriched with metal as well as with chalcogen.

In energetic spectrum SnTe chalcogen vacancy level is set in valence band, that's why V_{Te} must be not the donor, but the acceptor, as it is in chalcogenids of plumbum. What is quite a disputable question at present. The withdrawal of double valenced electrons with the atom of metal causes the two holes, then the vacancies in the metallic sublattice SnTe the most probably are the double-charged acceptors V_{Sn}. Some authors consider them to be the double-charged ones [12].

The treatment of the semiconductors by the high energy particles is a long-term method of modification of its properties [9, 14]. Last time some data about the influence of the irradiation by high-speed electrons [15-18], protons [l9], *a*-particles [20-22], ions of different atoms [23,24], γ-quantums [25] and laser [26] have been recieved on the compound $A^{IV}B^{VI}$. However the nature of radiation-induced point defect and its charge state have not been finally determined. It has not even been mentioned about more complicated defect types because of their small concentrations.

To study the nature and charge state of the radiation-induced point defect, created in SnTe by the α-particles irradiation is the main purpose of this article.

II. Samples preparation and experiment technique

The subject of this research is p-SnTe layers ~8 mkm thickness resulted from vapour phase by the hot-wall method on the substrates (111) BaF_2 [27]. According to the data of the X-ray double-crystal spectrometry and topography the films were like monocrystalline blocks with the dimension ~10 mkm and patchiness up to 1', orientated by the plane (111) paralelly to surface of the substrate. The lattice parameter has been determined by the Bond method with the accuracy

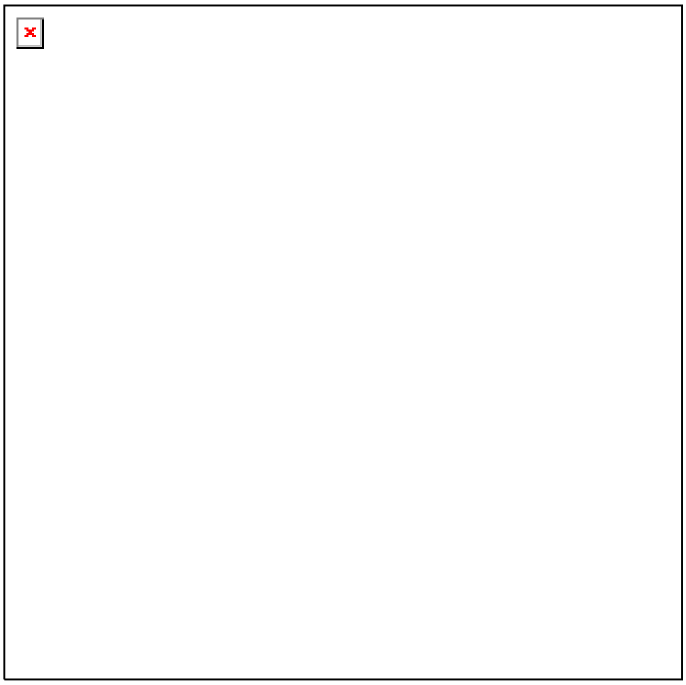

Fig.l. Dependence of lattice parameter $\Delta a/a_0$ on defects N_D accordingly: 1,2,3 - calculation for crystal structure like NaCI with interstitial atoms (1) ($N_D = N_i$, vacancies (2) ($N_D=N_v$) and Frenkel pairs (3) ($N_D=N_i+N_v$); 4,5 - experiment for crystals (4) according [13] (N_D=p/2) and irradiated epitaxial layers (5) (N_D=GΦ) SnTe.

$\Delta a = \pm 0{,}0001\ \overset{0}{A}$, The samples have been irradiated by the α-particles nonfocusing flux with the energy ~5 MeV from source ^{238}Pu on the irradiation facility in the vacuum under the room temperature. The α-particles flux density was p= $5\text{-}10^7 s^{-1} cm^{-2}$.

The electric parameters of the layers have been measured in the stationary electric and magnetic fields by the compensation method before and after a defined irradiant dose.

III. Model and identification of the point defect

Purpousing to identificate the point defect in crystal SnTe the structure like NaCI with defect has been modelled by the Vinyard method [28]. For model construction the potential Lenard-Jonson

$$\varphi(r) = 4c\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right], \qquad (1)$$

has been chosen, used earlier for the defect calculation in PbTe [29]. Where r - the distance to atom of the first coordination sphere; *c*, φ- the empiric parameters (*c* = 0,002 eV, σ = =3,98 $\overset{0}{A}$ [30]). The distribution of the interplanar distances (x = $\Delta a/a_0$) the chosen "cluster" strained by the intrinsic atom defects has been aproximated by Gauss dependence

$$f(x) = \frac{1}{\sqrt{2\pi\varepsilon}} e^{\frac{(x-x_0)^2}{2\varepsilon^2}}, \qquad (2)$$

where ε = $5\text{-}10^{-3}$ - the standart deviation of the normal distribution. The increase of vacancies concentration causes the decrease of lattice parameter (fig. 1-2), and the increase of the interstitial atoms causes the increase of the lattice parameter (fig. 1-1). For Frenkel pairs formation we have the dependence, analogous to the dependence for the vacancies, but more steep (fig. 1-3).

The comparison of the model calculation with the experiment results provides the opportunities to determine the type of intrinsic defect. It is known that the lattice parameter SnTe within the homogenous region is determined by the relation

$$a\left(\overset{0}{A}\right)=6{,}327-1{,}7\cdot 10^{-23}\,p, \qquad (3)$$

where a_0 = 6,327 $\overset{0}{A}$ - lattice parameter SnTe, saturated by the tin; p(sm^{-3}) - holes concentration. The increase of the defect concentration N_D (N_D = 1/2 p - for double-charged defect) leads to the lattice parameter decrease according to the regularity, which is douse to vacancy mechanism of the defect formation (fig. 1-4). It is connected with the tin vacancy formation in the metal sublattice by the increasing of the tellurium content.

The α-particles irradiation of the epitaxial layers SnTe leads to small change of the lattice constant. Besides the increase of the irradiation dose causes the decrease of the interplanar distance to some degree (fig. 1-5). The latter can be explained by the two competing processes, namely: by the generation of Frenkel pairs in the two sublattice and by the thermal recombination of the defects determined by their diffusion.

IV. Generation-recombination mechanism of radiation defect formation

Discriping the mechanism of Frenkel pairs radiation formation the model with the next charged state of the. defects has been accepted $Sn_i^{+}-V_{Sn}^{2-}$, $Te_i^{0}-V_{Te}^{2-}$. It was established, that the tin vacancies were the intrinsic prevailed defects in SnTe films caused by the full displacement of the gomogenous region to tellurium side [12,13]. Thus, in a noncompensation sample p-SnTe with primary holes concentration the next primary concentration of the defects

$$\left[V_{Sn}^{2-}\right]_0=p_0/2,\ \left[Sn_i^{+}\right]_0=0,\ \left[Te_i^{0}\right]_0=0,\ \left[V_{Te}^{2-}\right]_0=0$$

will be natural ones.

The Frenkel pairs generation caused by the irradiation and their next recombinations can be described by the relations

$$\begin{gathered}\frac{d}{dt}\left[V_{Sn}^{2-}\right]=G\cdot p-\mu\cdot D_{Sn}\left[V_{Sn}^{2-}\right]\cdot\left[Sn_i^{+}\right],\\ \left[Sn_i^{+}\right]=\left[V_{Sn}^{2-}\right]-\left[V_{Sn}^{2-}\right]_0\end{gathered} \qquad (4)$$

for the vacancy cocentrations and interstitial tin atoms correspondently [31]. Here G - coefficient of the point defects generations, p - a-particles flux density, μ - coefficient of the tin recombination, D_{Sn} - coefficient of the tin diffusion. The solution of the differential equation (4) considering the initial conditions determines the dependence of vacancy concentrations on the time

$$\left[V_{Sn}^{2-}\right]=\frac{1}{2}\left[V_{Sn}^{2-}\right]_0+R\frac{1-A}{1+A}, \qquad (5)$$

where

$$R=\left[\frac{G\cdot p}{\mu\cdot D_{Sn}}+\frac{1}{4}\left\{\left[V_{Sn}^{2-}\right]_0\right\}^2\right]^{1/2},\quad A=A_1\cdot exp(A_2 t),$$

$$A_1=\frac{R-\frac{1}{2}\left[V_{Sn}^{2-}\right]_0}{R+\frac{1}{2}\left[V_{Sn}^{2-}\right]_0},\quad A_2=-2R\mu D_{Sn}.$$

The analogous dependence will be determine the temporal changes of the vacancies concentration and interstitial tellurium atoms.

The closeness of the atomic mass and atomic number Te and Sn causes the equal

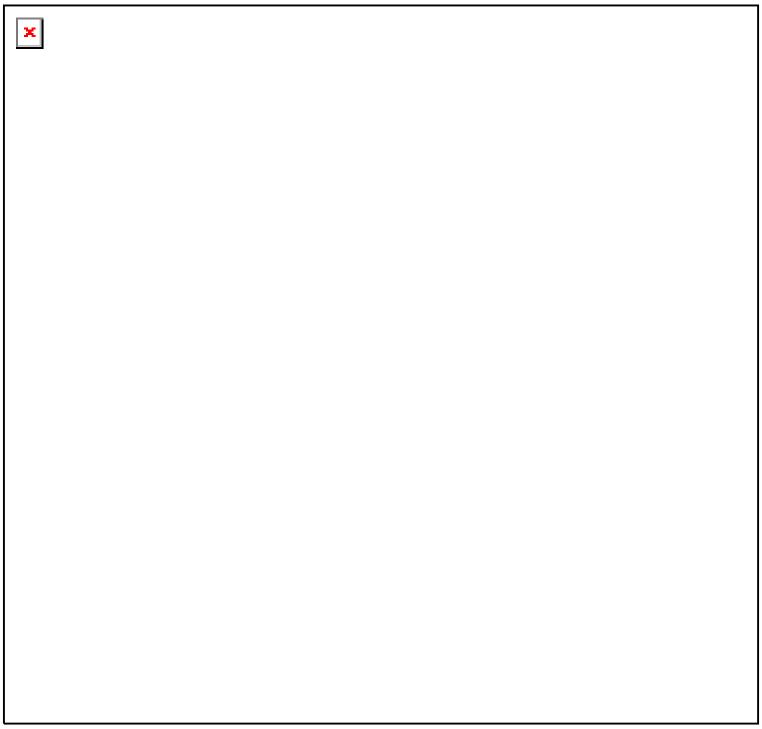

Fig.2. Dependence of charge carriers concentration in epitaxial layers p-SnTe on *a*-particle irradiated dose Φ (o- experiment, -calculation for Frenkel pairs)

$$[Sn_i^+]-[V_{Sn}^{2-}] \text{ and } [Te_i^0]-[V_{Te}^{2-}] - (1)$$
$$[Sn_i^+]-[V_{Sn}^{2-}] \text{ and } [Te_i^0]-[V_{Te}^{2+}] - (2)$$
$$[Sn_i^+]-[V_{Sn}^{-}] \text{ and } [Te_i^0]-[V_{Te}^{2+}] - (3)$$
$$[Sn_i^+]-[V_{Sn}^{-}] \text{ and } [Te_i^0]-[V_{Te}^{2-}] - (4)$$

value of the interaction cross-section in the process of the cascade forming, and also causes the equal value of generation coefficient *G* for the two sublattice. The experimental value of the generation. coefficient has been determined from the initial field of dose dependence of the cocentration charge carriers.

The concentration change of the charge carriers expresses itself through the change of the defects concentration considering their full ionization and charge state

$$p = 2\cdot[V_{Sn}^{2-}] + 2\cdot[V_{Te}^{2-}] - 1\cdot[Sn_i^+] - 0\cdot[Te_i^0]. \quad (6$$

At the figure 2. the data of the calculations for the different states of the tellurium vacancy $(V_{Te}^{2-}, V_{Te}^{2+})$ and the tin vacancy (V_{Sn}^{2-}, V_{Sn}), and the experimental results are displayed too. The parameters of the generation process and recombination of the radiation defects, used in the calculations, are displayed on the table. The analysis of the dose dependence of the charge carriers concentrations in the a-particles irradiated films p-SnTe (fig.2) indicates the sufficiently good concordance of the experimental results and the calculations for case of Frenkel pairs formation with double-charged tellurium vacancies of the acceptor type V_{Te}^{2-} and double-charged tin vacancies V_{Sn}^{2-} (fig. 2-1). The results of the radiational defect formation model of the single-charged tin vacancies V_{Sn} and double-charged tellurium vacancies of the

Table

Electrical properties (p_0 – initial concentration of charge carriers) and parameters of generation-recombination mechanism of defect formation (*G* – generation coefficient, μ - recombination coefficient, D_0 – diffusion coefficient, K=μ*D* coefficien by recombination addend, E_0 – energy of defect diffusion) of epitaxial layers p-SnTe by *a*-particles irradiation.

Model of defects	P_0, cm^{-3}	G, cm^{-3}	(μD), cm^{-3}s^{-1}	D_0, cm^{-3}s^{-1} [31]	E_0, eV [31]	μ, cm
$Sn_i^+ - V_{Sn}^{2-}$ $Te_i^0 - V_{Te}^{2-}$	$8{,}5\cdot10^{19}$ $8{,}5\cdot10^{19}$	$8{,}3\cdot10^{8}$ $8{,}3\cdot10^{8}$	$8{,}9\cdot10^{-25}$ $1{,}4\cdot10^{-25}$	$6{,}64\cdot10^{-2}(Sn_i)$ $3{,}25\cdot10^{-2}(V_{Te})$	1,02 1,04	$1{,}86\cdot10^{-6}$ $1{,}26\cdot10^{-6}$
$Sn_i^+ - V_{Sn}^{2-}$ $Te_i^0 - V_{Te}^{2+}$	$8{,}5\cdot10^{19}$ $8{,}5\cdot10^{19}$	$8{,}3\cdot10^{8}$ $8{,}3\cdot10^{8}$	$8{,}0\cdot10^{-26}$ $4{,}4\cdot10^{-25}$	$6{,}64\cdot10^{-2}(Sn_i)$ $3{,}25\cdot10^{-2}(V_{Te})$	1,02 1,04	$4{,}8\cdot10^{-6}$ $4{,}0\cdot10^{-6}$

donor type V_{Te}^{2-} have given the great differencies between calculation and experiment (fig. 2-2, 3, 4). Besides the increasing of *α*-particles integral flux irradiation of the layers p-SnTe leads to the growth of the hole concentration. The contribution of two types defects (interstitial atoms and vacancies) to the mechanism of recombination is equivalent. It is obvious from the fact that the approximation parameter *K* = *μD* value and table value of the diffusion coefficient determine in reasonable limits the radius recombinations *r (r = μ/4π)* values, which are comparable with (a_0= 6,327 $\overset{0}{A}$ - lattice parameter SnTe (table).

V. Conclusion

The change of charge carriers concentration in the epitaxial layers p-SnTe by a-particles irradiation has been explained through the formation model of the double charged acceptor type vacancies of the tin and tellurium $Sn_i^+ - V_{Sn}^{2-}$, $Te_i^0 - V_{Te}^{2-}$.

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Радіаційні точкові дефекти в ерітаксійних плівках SnTe

Б.К.Остафійчук, Я.П.Салій, В.М.Чобанюк, Г.Д.Матеїк, М.В.Пиц
Прикарпатський університет,
76000, Україна, Івано-Франківськ, вул.Шевченка, 57

Розглянуто залежність сталої кристалічної гратки і концентрації дірок в епітаксійних шарах p-SnTe від інтегрального потоку α–частинок. Зроблене порівняння оцінок та експериментальних даних приводить до висновку про акцепторний характер вакансій олова та телуриду V_{Te}^{2-}.