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Thermodynamics of defect subsystem in zinc telluride crystals

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Using method on the base of minimizing of thermodynamic potential in "crystal-vapor" system as a function of defect concentration the equilibrium concentration of point defects and free charge carriers in zinc telluride (ZnTe) crystals have been calculated depending on the technological factors of two-temperature annealing (annealing temperature T and vapor pressure P_{Zn} of zinc or P_{Te} of tellurium). It is shown that the dominant defects are zinc vacancies the charge state of which depends on the technological conditions of annealing.

Keywords: Zinc telluride; point defects; two-temperature annealing.

1. Introduction

Actuality of defective subsystem research of A^2B^6 semiconductor crystals is caused by the use of these materials for quantum-sized structures and semiconductor lasers.[1–4] Zinc telluride (ZnTe) is a relatively little studied direct band gap semiconductor and it is interesting in terms of the expansion of element basis of modern electronics.[1] Also, as a direct band gap semiconductor, ZnTe has high photosensitivity and can be successfully used for efficient solar energy conversion.[1]

Point defects of crystal lattice effect on almost all range of semiconductor materials properties. Even in Ref. 6 it was suggested and confirmed in later studies

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that the dominant defects in ZnTe are doubly ionized zinc vacancies. Based on this hypothesis, Vinogradov *et al.*[2] explained the experimental dependence of Hall carrier concentration on the zinc vapor pressure. However, annealing in tellurium vapor leads to slope of Hall dependencies that may indicate the charge state change of the dominant type of defects, or even to increase the impact of other point defects. In addition to interstitial Tellurium atom, studies[7–9] indicate the prevalence of such types of defect as Zn_i, V_{Te}. It should be noted that changing of the defect's charge state may change the effectiveness of self-compensation of impurities, including donor–acceptor complexes that are typical for ZnTe.[10,11] The binding energy of these complexes is the sum of the deformation and Coulomb components. Changing the defect charge, such as vacancy zinc, from double to single, will twice reduce Coulomb component of binding energy, which will reduce the effectiveness of impurity compensation by vacancy complexes.

In most studies, including Refs. 10 and 11, conclusions about the possible types of point defects in ZnTe crystals with different structural perfection are based on the analysis of optical spectra. However, due to the significant amount of reflexes in spectrum and the substantial dependence of their properties on the history of the samples the dominant type it is not always possible to establish the dominant type of point defects clearly. Thus, the problem of determining the dominant types of defects and their concentration in the crystals obtained at different technological conditions is relevant, and its solution is important for the practical use of the materials.

The simulation of defect subsystem relying on the experimental data of Hall measurements and using quasi-chemical formalism[12] is one of the various methods for establishing of the dominant type of defects in crystals. In particular, the method of thermodynamic potentials is very promising in this task[13] and is the reason why used in this paper. In this case, the possibility of formation of the neutral, and singly and doubly ionized vacancies in the cation and anion sublattices is considered. The main objective of this paper was to determine the dominant type of defects in ZnTe at different conditions of two-temperature annealing and to establish the differences in the defective subsystem of crystals annealed in zinc and tellurium vapors.

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2. The Equilibrium Equation of the "Crystal-Vapor" System

The equilibrium concentration of point defects in the crystal during two-temperature annealing is directly determined from the system of equations describing the equilibrium of heterogeneous multicomponent systems at a given pressure P and temperature T[14]:

$$\sum \mu_i^{s(g)} = 0\,, \tag{1}$$

where $\mu_i^{s(g)}$ is chemical potential of the ith component ($i =$ Zn, Te) in a vapor g or crystal s.

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The chemical potential of the gas[15]:

$$\mu^g = kT \ln P + \mu_0 \,. \tag{2}$$

For the monatomic gas Zn

$$\mu_0 = kT(-\ln(kT) + \ln(h^3/(2\pi mkT)^{\frac{3}{2}}))\,. \tag{3}$$

For the diatomic gas Te_2:

$$\mu_0 = kT(-\ln(kT) + \ln(h^3/(2\pi mkT)^{\frac{3}{2}}) + \ln(h^2/8\pi^2 IkT) + \ln(h\nu/kT))\,, \tag{4}$$

where m is the mass of the atom or molecule, $I = ml^2$ is the moment of inertia of the molecule, l is the distance between the nuclei of the molecule, ν is the internal frequency of vibrations of the molecule.

The chemical potential of a defect equals the chemical potential of the crystal component taken with the "+" or "−" sign. We used the method of differentiation of Gibbs energy G with respect to defect concentration to determine these potentials. The Gibbs energy is represented as[13]:

$$G = G_0 + \sum (E + F_{\text{vib}})[D] + nE_C - pE_V - T(S_n + S_p + S_k)\,, \tag{5}$$

where G_0 is part of Gibbs energy that is independent of the presence of defects, E is energy of defect formation, F_{vib} is free vibrational energy of defect, $[D]$ is concentration of defect D, n and p are concentrations of electrons and holes, E_C, E_V are energies of the bottom of conduction band and of the top of valence band, S_k is configuration entropy, S_n, S_p are entropies of electrons in the conduction band and of holes in the valence band. The summation is over all sublattices and all defects in the sublattice.

Entropy is determined by the Boltzmann law:

$$S_k = k \ln \left(\prod W_j \right), \tag{6}$$

where W_j is the thermodynamic probability of the jth sublattice. For a sublattice with several distinct defect types:

$$W_j = \frac{N_J!}{(N_J - \sum [D])! \cdot \prod [D]!}\,, \tag{7}$$

where N_J is the concentration of units, in which a defect can be formed. For electrons and holes thermodynamic probabilities equal to

$$W_n = \frac{N_C!}{(N_C - n)!n!}, \quad W_p = \frac{N_V!}{(N_V - p)!p!}\,, \tag{8}$$

where

$$n = N_C e^{\frac{\mu}{kT}}, \quad p = N_V e^{-\frac{\mu + E_g}{kT}}\,. \tag{9}$$

N_C and N_V are densities of states in the conduction band and valence band, respectively; E_g is band gap; μ is the chemical potential of electrons, which is determined from the electroneutrality equation

$$\sum Z[D] = N_C e^{\frac{\mu}{kT}} - N_V e^{-\frac{E_g + \mu}{kT}}\,. \tag{10}$$

Then, the chemical potential of the defect is

$$\mu^s_{D_i} = E_i + F_{\mathrm{vib},i} - kT\ln\left(\frac{N_J - \sum[D]}{[D_i]}\right) + \left[n\left(\frac{E_C}{kT} - \ln\left(\frac{N_C - n}{n}\right)\right) + p\left(\frac{E_V}{kT} + \ln\left(\frac{N_V - p}{p}\right)\right)\right] \times \frac{kT \cdot Z_i}{\sqrt{(\sum Z[D])^2 + 4N_C N_V \exp(-E_g/kT)}}\,. \quad (11)$$

The main point defects considered are the zinc and tellurium vacancies: V_{Zn}, V_{Te}, each of which can be in three charge states: neutral, singly or doubly charged. The formation energies of neutral defects E_0 are determined by the method proposed in Ref. 16.

$$E_0 = E' - E_Z + E_K + \Delta E_1 + \Delta E_2\,. \quad (12)$$

Value E' is determined by multiplying of energy of one bond $D = 1.14$ eV (Ref. 17) on the coordination number $k_n = 4$ ($E' = 4.56$ eV).

E_Z is the formation energy of new bonds, and according to Ref. 16, it is equal to

$$E_Z = \frac{x}{a} \cdot \delta_1\,. \quad (13)$$

Here, $x = 6$ is the number of new bonds A–A (for V_B) or B–B (for V_A), $a = 4$,[16] δ_1 is the energy of one bond, which is equal to the melting energy of pure Zn (0.077 eV)[18] or Te (0.18 eV).[18]

E_K in (12) defines the Coulomb interaction energy of atoms around vacancies

$$E_K = \frac{1}{4\pi\varepsilon_0} \cdot \frac{z^*_{A,B} \cdot z^*_{A,B}}{\varepsilon \cdot r}\,. \quad (14)$$

Here, z^* is the effective charge of atoms, ε_0 is the electric constant, ε is the static dielectric constant (10.1),[19] r is the distance between atoms of the same sort. According to Ref. 16 during the vacancy formation the charge excess, which was formed as a result of the removing of the cation or anion, equally distributed between the atoms of the first coordination sphere. Thus, in Eq. (14), value z^* should be multiplied by the values, which take into account the type of atoms that form compound and their number in the first coordination sphere. For compounds A^2B^6, it is equal to: $z^*_A = \frac{6}{4}z^*$, $z^*_B = \frac{2}{4}z^*$. Effective charges were found using data about the level of ionicity in a crystal[20] $z^* = n\sqrt{f_i}/k_n$. Here, n is the refractive index (3.56),[21] f_i is the ionicity (0.55),[22] $k_n = 4$ is the coordination number. Thus, $z^* = 0.66\,e_0$.

The fourth and fifth terms in (12) are responsible for the relaxation of the atoms around the vacancy. However, we ignored them paying attention to the proximity of our calculation.

As a result of our calculation, we have obtained the formation energies of neutral defects $E_0(V_{\mathrm{Zn}}) = 4.51$ eV, $E(V_{\mathrm{Te}}) = 6.38$ eV.

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The energies of formation of singly and doubly ionized defects are determined by the formulas

$$E_1 = E_0 - \frac{Z}{|Z|}\varepsilon_1; \quad E_2 = E_2 - \frac{Z}{|Z|}(\varepsilon_1 + \varepsilon_2)\,, \tag{15}$$

where E_0 is the formation energy of the neutral defect, Z is the charge state of the defect and ε_1, ε_2 are the first and second levels of ionization of the formed defect.

Information about localized energy levels in the energy spectrum of ZnTe that is presented in the literature, including Refs. 20–29, cannot uniquely identify the level of intrinsic point defects. Almost always the studied samples in addition to their own defects have included uncontrolled impurity atoms and their systems. Situation with donor levels is rather difficult because ZnTe has predominantly p-type of conductivity and their observation is difficult. Therefore, based on the information presented in the literature, one can build several models of the energy spectrum of defective crystals. In Ref. 23, based on the analysis of common patterns in change of photo-stimulated electron paramagnetic resonance and photoluminescence spectra for a number of A^2B^6 compounds, the following model of energy levels placement of vacancies in ZnTe has been offered: $V_{\mathrm{Te}}^{+} - E_v + 0.729\,\mathrm{eB}$, $V_{\mathrm{Te}}^{2+} - E_v + 0.212\,\mathrm{eB}$, $V_{\mathrm{Zn}}^{-}E_v - 0.106\,\mathrm{eB}$, $V_{\mathrm{Zn}}^{2-} - E_v + 0.400\,\mathrm{eB}$. Considering that the method used by the authors is one of the most reliable for studying the energy levels of point defects, for calculation of the concentration of point defects we used the same energetic scheme.

The change of free vibrational energy of the crystal during the formation of a defect is[30,31]:

$$E_{\mathrm{vib}} = \pm\left\{3kT\ln\left(\frac{T_\theta}{T}\right) - kT\right\} + x\cdot 3kT\ln\left(\frac{\omega}{\omega_0}\right), \tag{16}$$

where x is the number of atoms that have changed their vibrations frequency from ω_0 to ω. The change of the oscillation frequency of atoms ω/ω_0 is considered to be the variation parameter of the model and has been chosen so that curves $n(T)$ and $n(P)$ should be maximum close to experimental curves. Defined optimal values for them are $\omega/\omega_0 = 0.4$ for V_{Zn} and $\omega/\omega_0 = 3.0$ for V_{Te}.

Thus, to determine the equilibrium concentration of defects, we obtained a system of six equations (1). The problem of finding their solutions is equivalent to that of minimizing any increasing function $L_{\min}$ of absolute values of residuals $L_{\min} = \sum_1^6 |\sum \mu_i^{s(g)}|^t$.

The numerical convergence rate of the iterative process to the minimum is the main criterion for choosing the parameter t. The coordinates of the minimum of $L_{\min}$ are equilibrium values of the six types of defects (V_{Zn}^0, V_{Zn}^-, V_{Zn}^{2-}, V_{Te}^0, V_{Te}^{1+}, V_{Te}^{2+}) for a given vapor pressure of the additional component P and annealing temperature T. The whole procedure is implemented numerically in MAPLE. The finding of the minimum is performed by the method of random perturbations, and the initial values of the coordinates are randomly simulated. Parameters used in the calculation are presented in Table 1.

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Table 1. Calculation parameters.

Parameter	Value	Reference
Internal oscillation frequency of the molecule Te_2	$0.625102 \cdot 10^{13}\,s^{-1}$	32
The distance between the nuclei of molecule Te_2	$2.59 \cdot 10^{-10}$ m	32
Constant $K = P_{Te_2} \cdot P_{Zn}^2$	$K = 10^{-33440/T+20,33} \cdot (101325)^3$, Pa^3	33
Effective mass of electrons	$0.2\,m_0$	34
Effective mass of holes	$0.1\,m_0$	34
Band gap	$E_g = 2.394 - 8.1 \cdot 10^{-4} \cdot T^2/(T + 210)$, eV	35
Debye temperature	$T_0 = 300$ K	34

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3. Results and Discussion

The calculation results of the concentration dependences of free charge carriers in ZnTe crystals under conditions of two-temperature annealing on zinc vapor pressure for different annealing temperatures are shown in Fig. 1. It is seen that in the temperature range 966–1226 K theoretical curves satisfactory describe the experimental data that indicates on adequacy of accepted model of defect subsystem and its parameters. It should be noted that in energy calculation the values of neutral vacancies have been changed for determining the more optimal values in comparison with the result of approximate calculation by Eq. (12). However, it was found that the optimal value of formation energy of zinc neutral vacancy is $E_0 = 4.59$ eV, which differs from just 0.08 eV from the previous calculation. This fact is another proof of the adequacy of the vacancy defect subsystem models and the objectivity results received on their base and of the conclusions made.

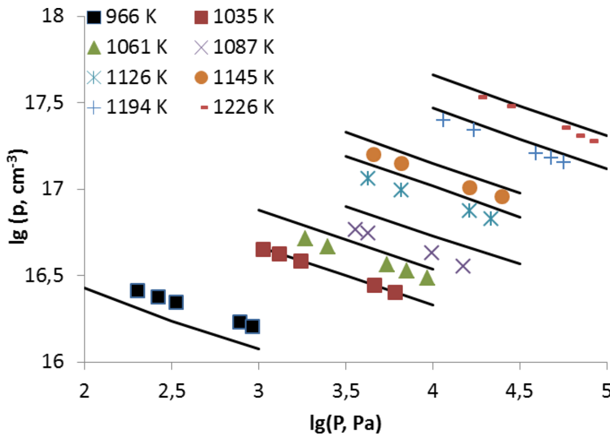

Fig. 1. (Color online) Dependence of the concentration of free charge carriers p on the vapor pressure of zinc P_{Zn} under two-temperature annealing at different temperatures T. The solid curves are calculations; the points are experimental data.[6]

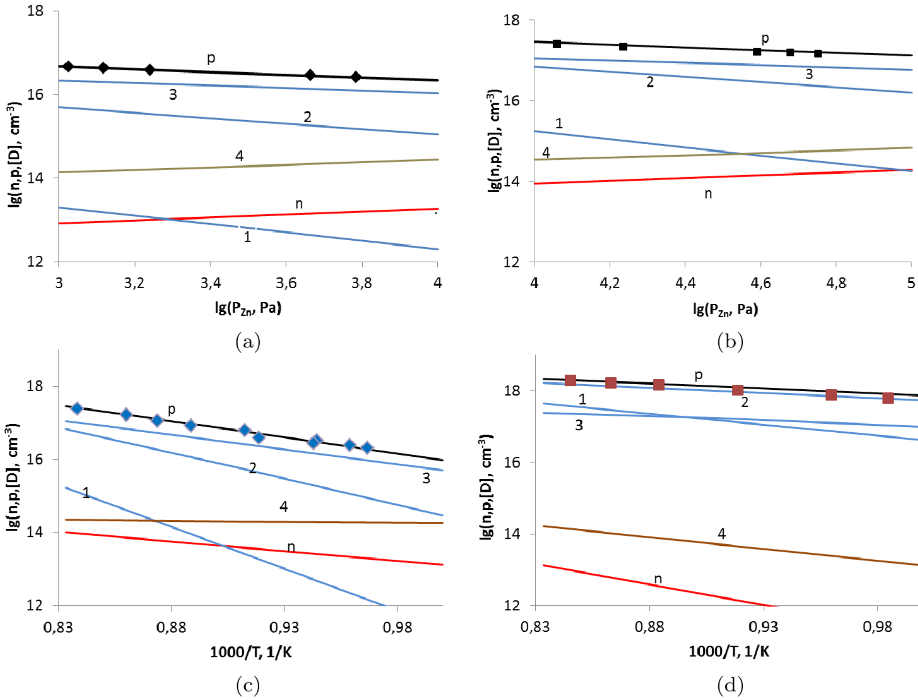

Fig. 2. (Color online) The dependence of concentrations of electrons (n), holes (p), point defects ($[D]$) ($1 - [V_{\mathrm{Zn}}^{0}], 2 - [V_{\mathrm{Zn}}^{-}], 3 - [V_{\mathrm{Zn}}^{2-}], 4 - [V_{\mathrm{Te}}^{2+}]$) under two-temperature annealing: (a) on the vapor pressure of zinc P_{Zn} at temperature $T = 1035$ K, (b) on the vapor pressure of zinc P_{Zn} at temperature $T = 1194$ K, (c) on the annealing temperature at vapor pressure of zinc ($P_{\mathrm{Zn}} = 13,300$ Pa) and (d) on the annealing temperature at vapor pressure of tellurium ($P_{\mathrm{Te}} = 13,300$ Pa). The solid curves are calculation; the points are experiment.[6]

The calculated dependences of the point defects concentrations in ZnTe crystals under different conditions of two-temperature annealing are shown in Fig. 2. Figures 2(a) and 2(b) show that in the range of vapor pressure of zinc 10^3–10^5 Pa and of annealing temperature ≈ 1000–1200 K the dominant type of defect is doubly ionized zinc vacancies, the concentration of which increases with both increasing of T and decreasing of P_{Zn}. At the same time, under these conditions, the concentration of singly ionized zinc vacancies V_{Zn}^{1-} increased and its increasing is sharper than the increasing of V_{Zn}^{2-}.

During annealing in tellurium vapor the dominance defect is also a zinc vacancy, but with lower charge state $-V_{\mathrm{Zn}}^{-}$ (Fig. 2(c)). One should also pay attention to concentration of neutral zinc vacancies, which is sufficiently high for such annealing conditions, which at temperatures above ≈ 1100 K even exceeds the concentration of singly ionized defects of this type. This change in charge state of the dominant defect is caused by a shift of the Fermi level toward the valence band, which in turn is caused by an increase of free hole concentration in the band with increasing total concentration of acceptor defects.

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No significant impact of donor tellurium vacancies in the studied range of technological parameters was observed. The greatest number of chalcogen vacancies are in charge state 2+ (Fig. 2, curve 4), and numbers of neutral and singly ionized vacancies are much smaller. Obviously, this is the result of high formation energy of the defect, which almost exceeds the formation energy of cation vacancies by 2 eV.

We should note that the used calculation model is relatively simple, but at the same time, very productive, and we could explain the proposed experimental data not only qualitatively but also quantitatively. We also have to pay attention for almost complete linearity of experimental and calculated concentration dependences of point defects and free carriers from the technological parameters of two-temperature annealing. Proposed annealing temperature range of crystals ≈ 900–1300 K, and vapor pressure of components (10^2–10^5 Pa) is wide enough and covers almost all region of parameters in which the conducting of annealing is effective. In this range, we can see a satisfactory correlation between the calculated and experimental dependences. So, in our opinion there is no reason to doubt the adequacy of the adopted model. Linearity dependencies apparently caused by the fact that Hall carrier concentration is determined mostly by one type of defect (zinc vacancies). Minor changes of slope of the dependencies are observed in the area where the dominance of doubly ionized defects is changed by once ionized defects. More significant changes in the functional dependence of concentration of point defects and free charge carriers can occur in the areas where the concentrations of acceptor defects are commensurate with concentrations of donors. However, at the investigated range of annealing temperatures and vapor pressures of components such areas were not found.

The obtained results are important in assessing the level of self-compensating impurities introduced into the crystal, because, as follows from the results of our calculations, the changing of technological conditions of high-temperature treatment of samples leads to change of charge state of the dominant defect which, in turn, changes the compensation effectiveness of the doping elements.

4. Conclusion

The defect subsystem of ZnTe crystal has been investigated under the process of two-temperature annealing in a pair of components using the method of thermodynamic potentials. The concentrations of point defects and of free charge carriers as functions of the annealing temperature T and partial vapor pressure of components $P_{\mathrm{Zn,Te}}$ have been calculated.

It has been established that under annealing in zinc vapor $P_{\mathrm{Zn}} = 13,300$ Pa at temperatures $T = 1000$–1200 K, the dominant defects are doubly ionized zinc vacancies, and under annealing in tellurium vapor $P_{\mathrm{Te}} = 13,300$ Pa in the same temperature range the dominant defects are once ionized zinc vacancies.

A close agreement between the formation energy of neutral zinc vacancy that has been calculated in our work and theoretically known values, and a satisfactory correlation of our values of concentration of free charge carriers with experimental data indicate the adequacy of the presented defect subsystem model and of conclusions that have been made on its basis.

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