Particularities of the interstitial atoms and vacancies clusters formation in a thin cadmium telluride foil during in situ electron irradiation in a TEM

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Abstract. The formation of interstitial atoms and vacancies, as well as their clusters in the form of dislocation loops and voids in CdTe is simulated. The sizes and features of the growth of dislocation loops and voids were determined depending on the irradiation time, taking into account the decrease in the number of nodes of the semiconductor crystal lattice with the irradiation time, since in experiments we studied a thin CdTe foil in a transmission electron microscope (TEM). The calculated and experimental data are compared.

1. Introduction

Cadmium telluride (CdTe) relating to compounds $A_2B_6$ is one of the main materials of semiconductor microelectronics. This material is promising for the production of gamma and x-ray detector devices, new hard x-ray detection systems for space science, small pixel imaging detectors, ultra-high-resolution SPECT with CdTe detectors [1-5]. However, upon irradiation of CdTe, in particular by electrons, the structural defects are formed in it [6, 7], which affect the electrophysical properties of this material and devices made on its basis.

Computer simulation of the interstitial atoms and vacancies clusters formation in cadmium telluride upon irradiation with electrons was carried out in [7, 8]. In real experiments on the analysis of defect formation upon electron irradiation in a TEM, a thin foil is studied and the formation of clusters of point defects in this foil over time tends to decompose the material [9]. This means that in a thin foil, when modeling the processes of defect formation in TEM, it is also necessary to take into account the change in the number of crystal lattice sites of the material under study, which decreases with the duration of irradiation.

2. Results and Discussion

In previous studies, it was found that the types of defects, their concentration and the rate of defect formation in semiconductors depend on the stacking fault energy (SFE) [6, 10]. One of the options for the formation of dislocation loops is the presence of partial dislocation loops. TEM studies have shown that Frank and Shockley dislocations are involved in these processes [10]. The results made it possible to establish the “critical” radii of dislocation loops $r_{crit}$, which determine the boundaries that separate processes with different qualitative and quantitative changes in the evolution of a defective
network of cadmium telluride. It was assumed that the value of $r_{\text{crit}}$ is a measure of the material's resistance to defects.

To determine this parameter, the classical model describing the energy of the formation of Frank loops was upgraded, taking into account that the formation of this loop occurs from partial Shockley dislocations, which is more energy-efficient.

$$E_{(l,\text{perfect})} = E_{(SF)} + E_{(l,\text{Frank})} + 2E_{(\text{Shockley})},$$

$$E_{SF} = \pi r^2 \gamma,$$

$$E_{l,\text{Frank}} = 2\pi r \frac{\mu b_{\text{Frank}}^2}{4\pi (1-\nu)} \left[ \ln \left( \frac{8\alpha r}{b_{\text{Frank}}} \right) - 1 \right],$$

$$E_{\text{Shockley}} = 2\pi r \frac{\mu b_{\text{Shockley}}^2}{4\pi} \left[ \cos(\beta)^2 - \frac{\sin(\beta)^2}{1-\nu} \right] \ln \left( \frac{\alpha r}{b_{\text{Shockley}}} \right).$$

As a result, we get:

$$\gamma = \frac{\mu a^2}{4800\pi r_{\text{crit}}^2 (1-\nu)} \left[ 400\nu \ln \left( \frac{\alpha r_{\text{crit}}}{a} \right) + 32 + 34\nu \right],$$

where $E_{(l,\text{perfect})}$ is the total energy of dislocation loop formation, $E_{SF}$ is the energy of the stacking fault, $E_{l,\text{Frank}}$ is the elastic energy of the dislocation line, $E_{\text{Shockley}}$ is the energy of the partial Shockley dislocation, $\mu$ is the shear modulus, $\nu$ is the Poisson coefficient, $\alpha$ is the dislocation core factor, $a$ is the lattice parameter, $\gamma$ is the total energy of the stacking fault, $b$ is the module of the Burgers vector, $r$ is the radius of the dislocation loop.

The values of some parameters and $r_{\text{crit}}$ for CdTe “generalized model” are shown in table 1.

**Table 1. The values of some parameters and $r_{\text{crit}}$ for CdTe “generalized model”.

| Sample | lattice parameter, $nm$ | Poisson coefficient [11] | Shear modulus, $Pa$ [11] | SFE, $J/m^2$ [6,10] | Critical radius, $nm$ |
|--------|-------------------------|--------------------------|--------------------------|---------------------|----------------------|
|        |                         |                          |                          |                     | Classic model        |
|        |                         |                          |                          |                     | Classical model taking into account Shockley dislocations |
| CdTe   | 0.648                   | 0.41                     | 9.2⋅10^9                 | (11 ± 1.9)⋅10^{-3}  | 72.4 – 113           | 26.3 – 41            |

The results show a good correlation with the experimental data obtained previously [6, 10]. However, the mechanisms of development and evolution of a defective network in various conditions require detailed study.

In this paper, as part of a theoretical study, the formation of interstitial atoms and vacancies, as well as their clusters in the form of dislocation loops and voids in CdTe under electron irradiation is modeled.

Using effective activation energy values and effective concentrations of interstitial $c_I$ atoms and $c_V$ vacancies, we can write the equations simulating changes in the concentrations $c_I$ and $c_V$, as well as the growth of interstitial loops with radii $r_I$ and concentrations $c_{bI}$ and voids with radii $r_V$ and concentrations $c_{bV}$ in the following form:
\[
\frac{dc_I}{dt} = c_0 G - K_2 c_I c_V - 2 K_I c_I^2 - K_J \frac{2\pi I}{b} c_I c_{bl} - K_S J c_I c_S,
\]
\[
\frac{dc_V}{dt} = c_0 G - K_2 c_I c_V - 2 K_V c_V^2 - K_V \frac{4\pi V}{b^2} c_V c_{bV} - K_S V c_V c_S,
\]
\[
\frac{dc_{bl}}{dt} = c_0 K_J c_I^2,
\]
\[
\frac{dc_{bV}}{dt} = c_0 K_V c_V^2,
\]
\[
\frac{dr_I}{dt} = c_0 K_I c_I b,
\]
\[
\frac{dr_V}{dt} = c_0 K_V c_V b.
\]

Here \(c_0 = 1.5 \cdot 10^{22} \text{cm}^{-3}\) is the concentration of the nodes of the unirradiated crystal, \(c_S\) is the concentration of "sinks" on the surface. The values \(2\pi I / b\) and \(4\pi V^2 / b^2\) are the number of places where point defects attach along the perimeter of the loop and on the void surface, respectively, and \(b\) is a parameter of the order of interatomic distance magnitude (Burgers vector). The product of the electron scattering cross section and the reaction cross section \(j = 10^{19} \text{cm}^{-2} \text{c}^{-1}\) is denoted by \(G = \sigma \cdot j = 3 \cdot 10^{-3} \text{s}^{-1}\), then the value \(G \cdot c_0\) is the rate of generation of point defects (Frenkel pairs). If \(\nu = 10^{13} \text{s}^{-1}\) is the frequency of atomic oscillations in the lattice, \(E_2\) is the recombination energy of point defects, \(E_I\) is the energy of interstitials formation, \(E_V\) is the energy of vacancies formation, then the coefficients of the corresponding recombinations and generations can be written as \(K_{2,I,V} = b^3 \nu \cdot \exp(-E_{2,I,V} / kT)\), respectively, where \(k\) is the Boltzmann constant, and \(T\) is the temperature.

As a result of the diffusion of point defects to the surface of a thin sample, they recombine with the surface. The concentration of point defects that change as a result of their migration to the surface can be represented as
\[
\frac{\partial c_{I,V}}{\partial t} = K_S c_{I,V} c_S = -D_{I,V} \frac{\partial^2 c_{I,V}}{\partial z^2},
\]
where \(z\) is the coordinate perpendicular to the surface of the sample, the values of \(D_{I,V} = b^2 \nu \cdot \exp(-E_{mI,mV} / kT)\) are the diffusion coefficients of interstitial atoms and vacancies, and the migration energies of these defects are denoted by \(E_{mI}\) and \(E_{mV}\), respectively.

Taking into account the formula (2), it is possible to convert expressions (1) into a system of partial differential equations taking into account the decrease in the number of lattice nodes under irradiation as
\[
\frac{du_0}{dt} = -Gu_0 + c_0 K_2 u_1 u_2,\]
\[
\frac{du_1}{dt} = Gu_0 + D_I u_1 - c_0 K_2 u_1 u_2 - 2 c_0 K_I u_1^2 - 2 \pi I c_0 K_I u_1 u_3,
\]
\[
\frac{du_2}{dt} = Gu_0 + D_I u_2 - c_0 K_2 u_1 u_2 - 2 c_0 K_V u_2^2 - 4 \pi I c_0 K_V u_2 u_4 u_6,
\]
\[
\frac{du_3}{dt} = c_0 K_I u_1^2,
\]
\[ \dot{u}_4 = c_0 K_V u_2, \]
\[ \dot{u}_5 = c_0 K_I u_1, \]
\[ \dot{u}_6 = c_0 K_I u_2, \]

where \( u_0 = c(t) / c_0 \), and \( c(t) \) is the concentration of the lattice nodes during irradiation, \( u_1 = c_I / c_0 \), \( u_2 = c_V / c_0 \), \( u_3 = c_{bI} / c_0 \), \( u_4 = c_{bV} / c_0 \), \( u_5 = r_I / b \), \( u_6 = r_b / b \). The dot and dash represent the partial derivatives of the variables \( t \) and \( z \), respectively.

The numerical solution of the obtained system of equations gives the dependences \( u_0, \ldots, u_6 \) on \( z \) and the irradiation time \( t \) at the temperature \( T = 300 \, \text{K} \) (the parameter values are taken from [6]) (figures 1-4).

The results presented in figure 1 show that in the center of sample \( z = 100 \, \text{nm} \) the mobility of interstitial atoms is higher than that of vacancies. Interstitial atoms quickly move to the periphery, their concentration decreases faster than the concentration of vacancies. In the time interval from 0.01 to 0.1 seconds, the concentration of vacancies reaches a maximum that almost coincides with the minimum concentration of interstitial atoms. This behavior is explained by the dynamics of the generation and recombination of Frenkel pairs, as well as the agglomeration of Frenkel pairs in the di-interstitials and divacancies, which are supposed to be the nuclei for the formation of dislocation loops and vacancy voids.

![Figure 1](image1.png)

**Figure 1.** Dependence of \( u_1 \) and \( u_2 \) on the irradiation time at 300 K. (left, \( 10^{-5} \, s \leq t \leq 10^3 \, s \), right, \( 10s \leq t \leq 10^3 \, s \)).
Figure 2. Dependence of $U_3$ and $U_4$ on the irradiation time at 300 K. (left, $10^{-5} \leq t \leq 10^3$ s, right, $10 s \leq t \leq 10^3$ s).

Figure 3. Dependence of the radii of loops and voids on the irradiation time at 300 K.

Figure 4. Dependence of the number of lattice nodes on the irradiation time at 300 K.

When the sample is irradiated for more than 50 seconds, both concentrations $C_I$ and $C_V$ decrease. Figures 2 and 3 show that the increase in the concentration of dislocation loops $C_{bI}$ and voids $C_{bV}$, as well as their radii $r_I$ and $r_V$, significantly slows down. It should be noted that the real radii of dislocation loops and voids are greater than the lattice parameter. In the figures, these radii begin to exceed the parameter $b$ only a few seconds after the start of irradiation, and their values at long times are almost independent of the initial conditions.

Figure 4 shows the change in the number of lattice nodes during irradiation. A model that takes this change into account is displayed in solid lines in the figures. A model with a constant number of lattice nodes is indicated by dashed lines in the figures. The difference between these models is noticeable in figures 1-3 only at long times.
3. Conclusion
The obtained data were compared with experimental data both for determining the "critical" radii of dislocation loops and for numerical simulation of the formation of point defects clusters of interstitial and vacancy types in cadmium telluride [6, 9]. There is a good agreement between theory and experiment. It is particularly important that we were able to significantly improve the results by taking into account two important features in the process of defect formation in CdTe. First, a contribution from partial Shockley dislocations was added to the model describing the formation of Frank loops. Second, the simulation takes into account the decrease in the number of nodes of the semiconductor crystal lattice with the time of irradiation, which is an important physical point, since the crystal (foil) has a limited size when irradiated in situ in TEM.

Taking into account the foregoing, the obtained results quite adequately reflect the mechanisms of defect formation in cadmium telluride and can be applied to similar semiconductors taking into account their physicochemical properties and the value of the SFE.

References
[1] Elsharkawy M, Kanda G, Abdel-Hady E and Keeble D 2016 Appl. Phys. Lett. 108 242102
[2] Veale M, Bell S, Seller P, Wilson M and Kachkanov V 2012 Journ. of Instrumentation 7 P07017
[3] Ogawa K and Muraishi M 2010 IEEE Transactions on Nuclear Science 57 17
[4] Limousin O, et al. 2011 Nuclear Instruments and Methods in Physics Research A 647 46
[5] Iniewski K and Awadalla S 2015 Solid-State radiation detectors: technology and applications (CRC Press LLC)
[6] Loginov Y, Brown P and Durose K 2003 The Regularities of Structural Defect Formation in Semiconductors II–VI (Logos, Moscow)
[7] Gué A, Djafari-Rouhani M, Estève D and IdrissiSaba H 1991 J. Phys. I 1 97
[8] Loginov Y, Mozzherin A and Paklin N 2019 IOP Conf. Series: Materials Science and Engineering 467 012007
[9] Loginov Yu and Brown P 1992 Phys.Stat.Sol.(a) 132 323
[10] Loginov Y, Mozzherin A and Brilkov A 2014 Phys. Solid State 56 720
[11] Gorichok I 2012 Phys. Solid State 54 1373