

CHARACTERISTICS OF THE EFFECT OF RADIATION DEFECTS ON THE CURRENT-CARRYING MECHANISM IN THE p-CuTlS SINGLE CRYSTAL

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Abstract: The electrical conductivity and volt-ampere characteristics of the p-CuTlS single crystal with specific resistance $\rho = 40 \text{ } \Omega \cdot \text{cm}$ and irradiated by γ -quantum were studied in the range of 100-300 K temperature and $10 \cdot 10^4 \text{ V/cm}$. It was determined that the cause of the conduction disorder observed in the CuTlS single crystal at low electric fields and high radiation doses is the formation of defect clusters dominated by cation vacancies. A sharp increase in current at high electric fields and temperatures occurs as a result of thermo-field ionization of the acceptor level with activation energy $\Delta E_a = 0,08 \text{ eV}$ and the ionization voltage decreases with increasing radiation dose. Based on the determination of the parameters ($\lambda, \Gamma_m, n_0, \epsilon$) that determine the mechanism of current flow, the dependence of the shape of the potential hole on the radiation dose was determined.

Keywords: Radiation, semiconductors, electrical conductivity, current, defects

1. Introduction

One of the issues facing of radiational material science is to increase the sustainability of the parameters of materials to ionizing radiation and particles [1-3]. [4-6] - studies show that cation and anion vacancies and interstitial ions are formed as a result of the displacement of atoms from the nodes of the crystal lattice during irradiation. The emergence of new impurity levels of acceptor and donor types as a result of the interaction of radiation and structural defects changes the free charge carriers and electrical conductivity in the crystal. Dependence of the relationship between the interaction between defects and the concentration of free charge carriers on the parameter (E_g) of the semiconductor material updates the traditional conductivity model in narrow-band semiconductor materials [7, 8]. Existing models [9-12] are mainly applied to broadband (Si, GaAs, etc.) semiconductors, while in narrowband materials, the high concentration of charge carriers creates certain difficulties in the application of the model. For this reason, the research of the influence of defect regulation process on conduction regulation process in narrowband semiconductors has practical and scientific significance. The study of the effect of γ -quantum on the electrophysical properties of the layered p-CuTlS single crystal as an object of research, which is representative of chalconides and has thermal conducting properties, is scientifically and practically important. CuTlS belongs to the group of single-crystalline copper-thallium chalcogenides and belongs to the group of compounds with the formula $\text{TlCu}_{2n}\text{X}_{n+1}$ ($X = \text{S, Se}$) [1-5]. The compound has a defective structure and the structure of it has been studied in [4, 5]. It is shown that their structure can be described as a tetrahedral set of Tl atoms located between separate layer planes. Depending on the position and amount of the Cu atom in the lattice, a transition from metallic conductivity to semiconductor conductivity is observed [7, 8]. This feature can be caused by a violation of stoichiometry with the substitution of cation atoms, which changes the

electrophysical properties of the compound. To clarify the mechanism of the anomaly, the study of the mechanism of defect formation in narrow-band semiconductors under the influence of ionizing radiation allows clarifying the characteristics of the current.

The electrical conductivity, thermoelectric and optical properties of the CuTlS crystal are partially described in the scientific literature [8]. Although there is some information about their electronic structure and conductivity anomalies at low temperatures, they do not allow us to propose a model of the effect of radiation defects on the conductivity of narrow-band semiconductors. To complete the theoretical and experimental results and obtain new data, the effect of γ -quantum on the current transfer mechanism in the CuTlS single crystal was studied in the temperature range of 100-300 K in low and high electric fields.

2. Experimental Techniques

The studied p-CuTlS single crystal growth by the Bridgman-Stockbarger technique at high-temperature gradient. Using materials with a high degree of purity (Tl 99.99%; Cu 99.999%; S 99.99%) in the crucible is completely melted in the hot zone at 775 K of the two-zone Bridgman furnace and is transferred to the cold zone (580 K) at speed of 1.2 mm/h. The obtained single crystal had a diameter of 1 cm, a length of 8 cm, and a specific resistance of $\sim 40 \text{ Ohm} \cdot \text{cm}$. Crystal structure and lattice parameters of the sample obtained by X-ray analysis method $a= 4.08$; $c= 8.16 \text{ \AA}$; $z = 2$ was calculated and it was determined that the compound crystallizes in a tetragonal crystal system and the results were given in [8]. The silver paste was used for the conductive ohmic contact and the distance between the contacts is $L = 6 \text{ mm}$. The size of the studied sample is $2 \times 0.5 \times 6 \text{ mm}$. Measurements were made on a B7-30 universal ampere-voltmeter at a voltage of 0-20V ($E \sim 10 \cdot 10^5 \text{ V/cm}$) and a temperature range of 100-300 K. Radiation with γ -quantum was carried out at Co^{60} device and the radiant flux was $\sim 40 \text{ W/sec}$.

3. Results and Discussion

In Fig.1 the effect of γ -quantum on volt-ampere characteristics (VAC) of p-CuTlS single crystal at $T=300 \text{ K}$ is given. In a non-irradiated CuTlS crystal, the voltage-current dependence obeys the law $I \sim U^n$ ($n = 1-3$) (Fig.1- curve 1). Variation of the voltage applied to the sample in the range of 0.05 - 0.5 V has a linear character and corresponds to the ohmic part ($n=1$). The dependence of $I \sim U^n$ in the voltage range of 0.5-5 V ($n = 2$) is quadratic and obeys the theory of current [Lam] is limited by the volume charge. In the region corresponding to a sharp increase in current at values of voltage $U > 5 \text{ V}$, the mode of full filling of traps is observed, and in the subsequent increase of voltage, again the quadratic region is observed. The $I \sim U^n$ ($n = 1-3$) dependence observed in the non-irradiated CuTlS crystal corresponds to the injection and ionization state of the traps. During radiation with $\Phi = 500 \text{ krad}$ γ quantum, a decrease in the current (curve 2) is observed, and the transition voltage from the ohmic part to the quadratic part increases. At 1 Mrad of dose, the value of the current increases concerning to the value before radiation and in the $I \sim U^n$ dependence, ohmic, quadratic, and sharply increasing of current are observed (curve 2). At higher values of the radiation dose (curves 4 and 5) the regularity $I \sim U^n$ is maintained and in this case the value of the current increases depending on the radiation dose. Fig.1 shows that with increasing radiation dose, the transition voltage from the ohmic region to the quadratic region shifts to smaller values of the voltage axis. [9-10] - based on the information provided in Fig. 1, the coefficient of retention, the concentration of free charge carriers and traps were calculated from the volume charge area and the extrapolation of the current corresponding to the full charge of the trap before and after irradiation: $\theta_0 = 2 \cdot 10^{-5}$ and $\theta_\Phi = 1.7 \cdot 10^{-6}$; $n_0 = 7 \cdot 10^{16} \text{ cm}^{-3}$ and $n_\Phi = 2 \cdot 10^{15} \text{ cm}^{-3}$; $N_{t0} = 3.2 \cdot 10^{13} \text{ cm}^{-3}$ and $N_{t\Phi} = 1,2 \cdot 10^{15} \text{ cm}^{-3}$.

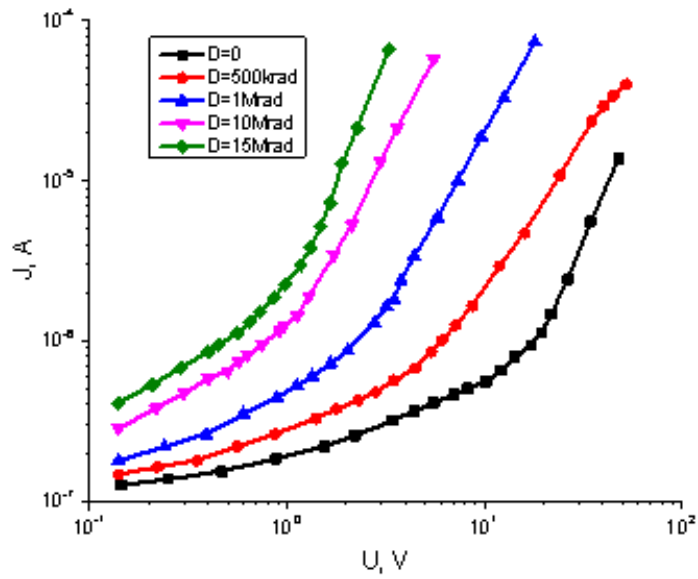


Fig. 1. The effect of γ -quantum on volt-ampere characteristics (VAC) of p -CuTlS single crystal at $T=300$ K

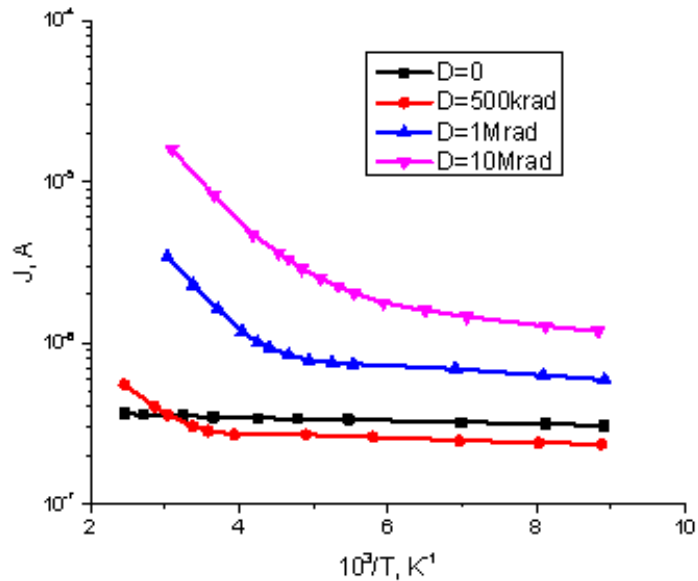


Fig. 2. The temperature dependence of the dark current in a CuTlS crystal

In Fig.2 the temperature dependence of the dark current in a CuTlS crystal irradiated at 500 krad and 1Mrad radiation doses are given. As can be seen from Fig.2, the current change is very weak in the temperature range of 100-250 K and the activation energy of the trap which calculated from the 1st curve is ~ 0.025 eV. At $T > 250$ K, a reincrease of current is observed. At 500 krad of radiation dose, an increase in current at a lower temperature (curve 2) is observed relative to the previous amount of radiation. Current begins to increase at lower temperatures at higher values of radiation dose ($\Phi > 1$ Mrad) (curve 3), which indicates the formation of a deeper energy level due to the effects of γ -quantum.

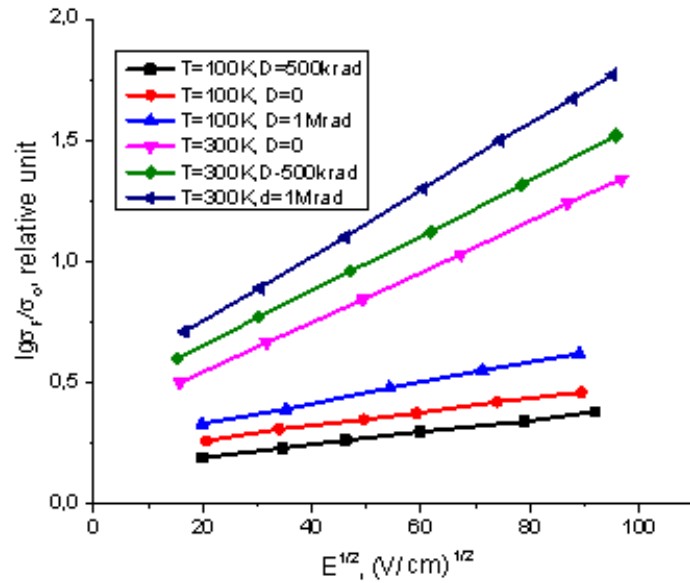


Fig. 3. The dependence of $\lg\sigma_f/\sigma_0 \sim F^{1/2}$ at different temperatures on CuTIS crystals irradiated at different doses.

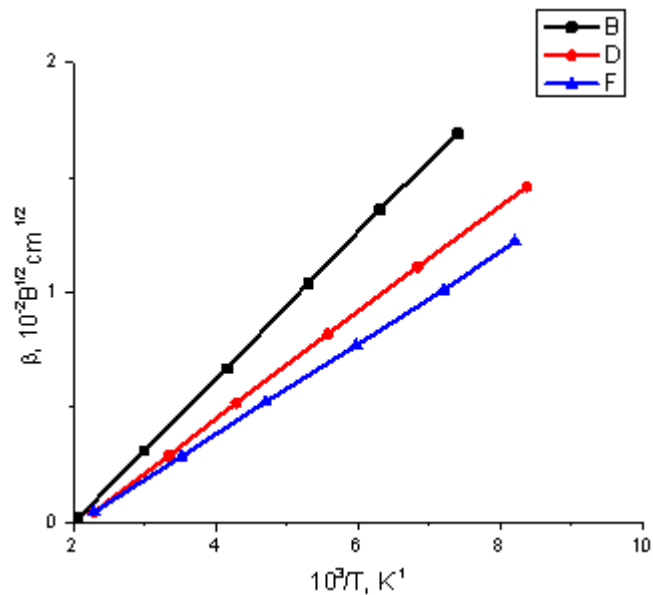


Fig. 4. The dependence of B-Frenkel coefficient on temperature

In Fig.3 the dependence of $\lg\sigma_f/\sigma_0 \sim F^{1/2}$ at different temperatures on CuTIS crystals irradiated at different doses is shown. As can be seen from Fig. 3, At $T = 100$ K, the gradient of $\lg\sigma_f/\sigma_0 \sim F^{1/2}$ increases weakly with increasing radiation dose and this may be because the resulting defects create a deep energy level. At $T = 300$ K, an increase in current and an increase in gradient are observed and this indicates an increase in the concentration of cation-type defects. Based on the obtained experimental results, the β -Frenkel coefficient was calculated and shown in Fig. 4.

It can be seen from the figure that the decrease in the β - coefficient after irradiation is related to the increase in the permittivity of the medium- ϵ_0 , as in [11]. This is due to the redistribution of defects in the crystal lattice by the interaction of defects in CuTIS crystals under

the influence of γ -quantum with primary defects. Defective changes were examined under an electron microscope (Fig. 5) and it has been established that the regulation of defects (b) occurs during low-dose radiation (500 krad), a large set of local defects occurs at high doses (5 Mrad). This fact leads to a decrease in the concentration of free charge carriers in the crystal and an increase in the transition voltage from the ohmic region to the quadratic region at low temperatures ($T < 300$ K) in VAC.

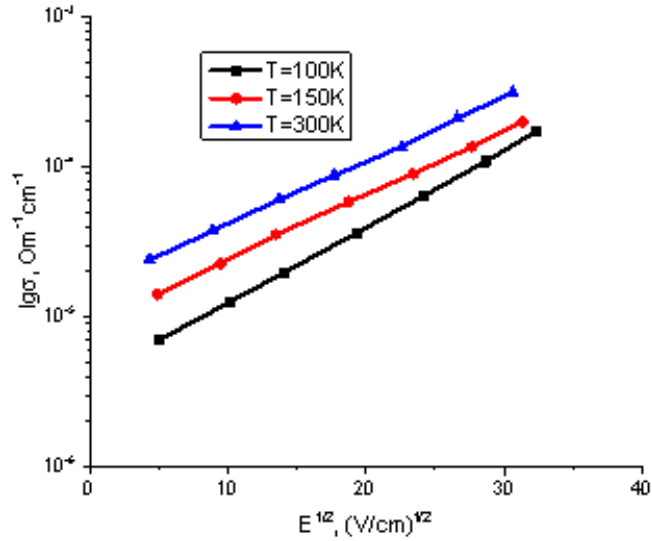


Fig. 5. The dependence of electric field on the electrical conductivity in CuTlS single crystal at different temperatures.

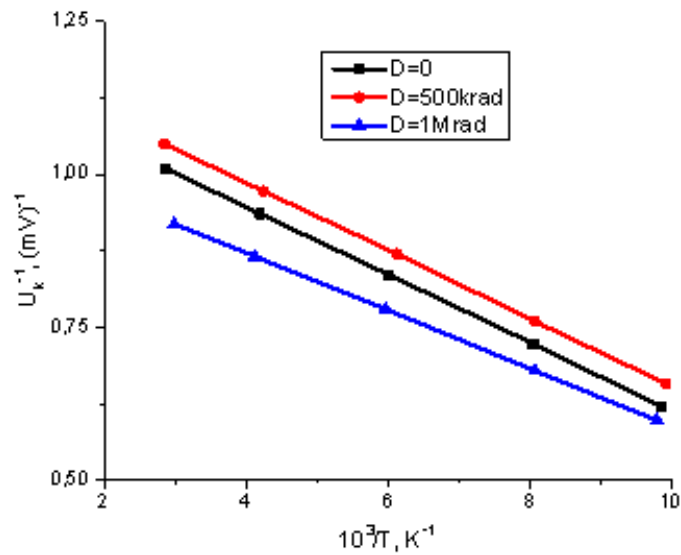


Fig. 6. The temperature dependence of the transition voltage of CuTlS single crystal in the VAX quadratic region

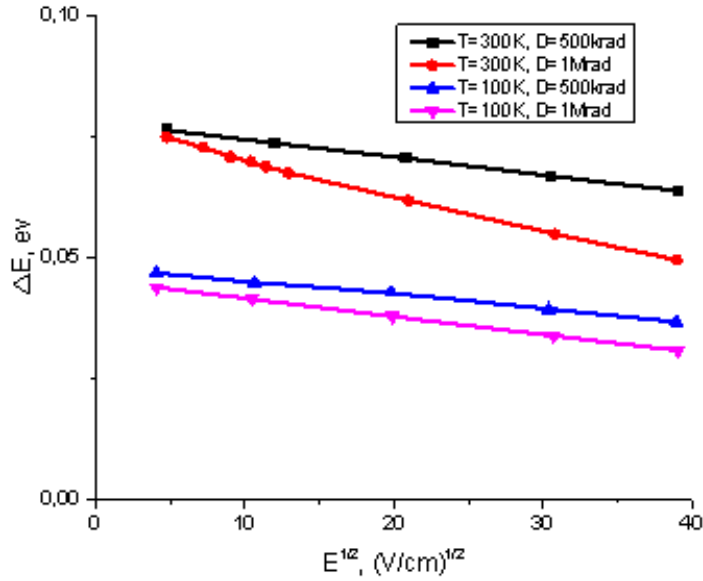


Fig. 7. The dependence of the activation energy of the trap on $E^{1/2}$ at different temperatures and doses

The Full filling of traps slides into a higher voltage range and its temperature dependence at different radiation doses is given in Fig. 6. Comparison of curves 1; 2 and 3 in Fig. 6 show that at high radiation doses in the $U_{ad} \sim 1/T$ dependence, the gradient of the curve (3) increases relative to the state before radiation and the activation energy determined by the retention level $T \rightarrow 0$ is equal to ~ 0.07 eV. At low radiation dose values ($\Phi < 500$ krad; 2-curve), the gradient of $U_{ad} \sim 1/T$ dependence remains the same as before radiation (1). According to the results, donor-type defects, which arising at radiation doses $\Phi < 500$ krad in the narrow-band CuTIS single crystal, partially reduce conductivity, and at high doses the conductivity increases due to the higher concentration of acceptor-type defects. For this reason, in Figures 3 and 6, the gradient of the curve varies depending on the radiation dose at current-temperature dependence. To determine the relationship of this change with the mechanism of current, let's analyze the result obtained in Figure 3, taking into account Frenkel's theory [11]:

$$\begin{aligned} \sigma &= \sigma_0 \exp \beta \sqrt{E}, \\ \sigma_0 &= A \exp(-\Delta E_0 / 2kT) \end{aligned}$$

here,

E - electric field intensity, ΔE_0 - activation energy, β -Frenkel - coefficient, k -Bolsman constant, T - absolute temperature.

Taking into account the expression σ_0 in the Frenkel formula, the dependence $\Delta E \sim f(E^{1/2})$ can be written as follows [12]:

$$\Delta E_t(E) = E_t(0) - (e^3 E / \pi \epsilon \epsilon_0)^{1/2}$$

and its graphical description at different radiation doses is given in Fig. 7. It can be seen from the figure that after irradiation $\Delta E_t \sim E^{1/2}$ - dependence also retains its linear character, but its gradient changes concerning the original sample. It is known from the study that the ionization process in metallic materials with a high concentration of free charge carriers does not affect the process of defect formation and distribution of structural defects. As shown in [14] work, this can be related to the change in the permittivity of the medium- ϵ_0 which is caused by the defects that were created during the emission. The activation energy of the trap from the extrapolarization of the linear dependence $\Delta E - f(E^{1/2})$ under the condition $E = 0$ was determined and was 0.05 eV for low

temperature and 0.08 eV for high temperature. Experimental results show that the activation energy of the retention center during radiation depends on the radiation dose, and the gradient of the curve does not change at $\Phi < 500$ krad, increases at $\Phi > 500$ krad, and the resulting defects have a deeper energy level. In Fig. 1, curve 3, it can be seen that the radiation defects caused by the increase in current after irradiation have led to an increase in the concentration of charge carriers involved in the conduction. [13-17] - studies show that the permittivity of the medium increases to a certain value of the dose due to the parabolic dependence on the radiation dose, which decreases with the subsequent dose increase. According to this result, it can be assumed that the dependence of the gradient of $\Delta E_r \sim E^{1/2}$ and dependence of the β - coefficient on the radiation dose is due to changes in the permittivity of the medium.

Based on the experimental results and literature data [11-15], the length of the free path of charge carriers (λ) in the p-CuTIS single crystal and the distance from the potential fence to the bottom of the conductive zone (r_m) are calculated based on $T = 250$ K, $E_c = 40$ V/cm, $\beta = 2,7 \cdot 10^{-2} \text{ V}^{-1/2} \text{ cm}^{1/2}$: Before radiation $\lambda = 6 \cdot 10^{-6}$ cm, $r_m = 2,3 \cdot 10^{-6}$ cm and $\epsilon = 15$, at $\Phi < 500$ krad doses $\lambda = 6,2 \cdot 10^{-6}$ cm, $r_m = 2,5 \cdot 10^{-6}$ cm and $\epsilon \approx 15$ and at $\Phi > 1$ Mrad doses $\lambda = 2,3 \cdot 10^{-6}$ cm, $r_m = 7,3 \cdot 10^{-7}$ cm and $\epsilon = 200$. The satisfaction of the condition $\lambda > r_m$ shows that in the p-CuTIS single crystal a thermoemission occurs as a result of the interaction of anion and cation-type defects with structural defects caused by γ -quantum and satisfies the [15, 16, 17]-condition. The increase in the concentration of free charge carriers at doses $\Phi > 1$ Mrad and in the areas $E > 40$ V / cm occurs as a result of ionization of the local level with the activation energy of 0.08 eV.

The mechanism of defect formation in the p-CuTIS single crystal under the influence of gamma quantum: Under the influence of gamma quantum, an electron-hole pair is created in the crystal lattice. These pairs migrate in the crystal and are captured by lattice defects, as a result, charged vacancies and interstitial ions arise. The mobility defects interact with the primary defects, are captured by the appropriate centers, and annihilate or associate. The speed of these processes, according to experimental results, depends on the radiation dose. Taking into account that, the fact that the probability of creation of anion vacancy is more than the probability of creation of cation vacancy in the CuTIS single crystal, donor-type defects occur at initial doses of radiation. For this reason, the electrical conductivity decreases during the initial radiation (figure 3). At high radiation doses, the concentration of charge carriers increases due to the predominance of acceptor-type defects, resulting in increasing the electrical conductivity of the crystal. The dependence of p-CuTIS VAC and electrical conductivity on the radiation dose proves the correctness of the above mechanism and corresponds to the results obtained in [14-16] - studies.

Thus, the effect of γ -quantum on the current transfer mechanism in the p-CuTIS single crystal was studied in different ranges of electric field and temperature and compared with the corresponding theoretical and experimental results [9, 11, 12]:

- In the p-CuTIS single crystal at $T < 250$ K, the value of current limited by the volume charge at the electric field intensities $E < 40$ V/cm does not depend on the radiation dose, and in the $E > 40$ V/cm fields, under the influence of γ -quantum a sharp increase in current occurs as a result of ionization of the acceptor level with energy $\Delta E_i = 0,08$ eV;
- The reason for the conductivity anomaly observed in crystals irradiated at high doses is the formation of defect clusters dominated by cation vacancies as a result of the interaction of radiation defects with structural defects. Based on the determination of the parameters (λ , r_m , n_0 , ϵ) that determine the mechanism of current flow, the dependence of the shape of the potential hole on the radiation dose was determined.

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ВЛИЯНИЕ γ -ИЗЛУЧЕНИЯ НА ТОКОПРОВОДЯЩИЙ МЕХАНИЗМ В МОНОКРИСТАЛЛЕ p-CuTlS

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Резюме: Электропроводность и вольт-амперные характеристики монокристалла p-CuTlS с удельным сопротивлением $\rho = 40 \text{ } \Omega \cdot \text{см}$, облученного γ -квантом, исследованы в диапазоне температур 100–300K и $10\text{--}10^4 \text{ В/см}$. Установлено, что причиной беспорядка проводимости, наблюдаемого в монокристалле CuTlS при малых электрических полях и высоких дозах облучения, является образование кластеров дефектов с преобладанием катионных вакансий. Резкое увеличение тока при высоких электрических полях и температурах происходит в результате термополевой ионизации акцепторного уровня с энергией активации $\Delta E_a = 0,08 \text{ эВ}$, а напряжение ионизации уменьшается с увеличением дозы облучения. На основе определения параметров (λ , γ_m , n_0 , ϵ), определяющих механизм протекания тока, была определена зависимость формы потенциальной ямы от дозы облучения.

Ключевые слова: Излучение, полупроводники, электропроводность, ток, дефекты

p-CuTlS MONOKRİSTALINDA γ -ŞÜANIN CƏRƏYANIN KEÇMƏ MEKANİZMİNƏ TƏSİRİ

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Xülasə: Xüsusi müqaviməti $\rho=40\Omega\cdot\text{sm}$ və γ -kvantlarla şüalandırılmış p- CuTlS monokristalının elektrik keçiriciliyi və volt-ampere xarakteristikaları 100-300 K temperatur və $10\text{-}10^4 \text{ V/sm}$ intervallarında tədqiq edilmişdir. Müəyyən edilmişdir ki, zəif elektrik sahələlərində və yüksək şüalanma dozalarında CuTlS monokristalında müşahidə olunan keçiriciliyin anomaliyasının səbəbi, kation vakansiyaları üstünlük təşkil edən defekt klasterlərinin yaranmasıdır. Yüksək elektrik sahələrində və temperaturlarda cərəyanın kəskin artması $\Delta E_a=0,08\text{eV}$ aktivləşmə enerjili akseptor səviyyəsinin termo-sahə ionlaşması nəticəsində baş verir və şüalanma dozasının artması ilə ionlaşma gərginliyi azalır. Cərəyanın keçmə mexanizmini müəyyən edən parametrlərin ($\lambda, \gamma_m, n_0, \epsilon$) təyin edilməsi əsasında potensial çuxurun formasının şüalanma dozasından asılılığı müəyyən edilmişdir.

Açar sözlər: Radiasiya, yarımkəçirici, elektrik keçiriciliyi, cərəyan, defektlər