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CHARGE TRANSPORT MECHANISM IN IMPLANTED p-GaSe:H$^+$ SINGLE CRYSTAL

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The study analysed the impact of radiation defects on p-GaSe single crystal implanted with H$^+$ ions (70 keV) on its charge transport mechanism. The research was conducted at 100 K and 300 K in an electric field of 10^2-10^4 V/cm. The study found that the activation energy of charge carriers injected at low temperatures and electric fields $E < 10^3$ V/cm ranged from 0.23-0.39 eV. This was observed due to the trapping of charge carriers in concentration traps of approximately $9\cdot10^{13}$ cm^{-3}, leading to monopolar injection. In the fields, $E > 10^3$ V/cm, a sharp increase in current was observed, which was explained by the thermal ionisation of local levels following the Frenkel mechanism. The study determined that the charge transport mechanism in GaSe:H+ crystals at low temperatures has a non-activated character.

Keywords: *Implantation; GaSe; Single crystal; Frenkel mechanism*
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1. INTRODUCTION

Studying the effects of materials under external influences helps us determine their potential applications. Recently, there has been a significant interest in studying functional materials under external influences like pressure, temperature, and radiation. With the development of computing technologies, some processes can now be studied by modelling them. As a result, complex studies are being conducted in this direction [1-5].

Among functional materials, chalcogenide conductors hold a special place. Recently, there has been a wide exploration of these materials' structure, thermal, and electrical properties. Researchers are studying the effects of ionising radiation to expand their application possibilities [6-10]. Radiation defects formed in the crystal lattice under the influence of radiation rays cause changes in the status of the Fermi level in the forbidden zone. Thus, the electrophysical properties of the crystal are modified [11,12].

The nature and properties of radiation defects formed in the crystal, especially in complex semiconductors, depend on the structure of the material and the distribution of primary defects [13,14]. Complex defects formed due to the interaction of radiation defects with structural and additive atoms lead to changes in the electronic characteristics of the crystal. Studying these mechanisms for the first time in layered crystals may allow us to propose a mechanism for purposeful control of the radiation resistance and properties of crystals.

The defects that arise from the impact of charged particles, such as H$^+$ and Sb$^+$, in complex semiconductors are complex and dynamic. Various factors, including the structure of the crystal lattice, radiation, temperature, and more can cause these defects. To understand these factors and determine the mechanism of charge transport, researchers have chosen to study a GaSe single crystal with a layered structure. To control the distribution of defects in the crystal lattice, they have used H$^+$ implantation and thermal infusion methods.

The GaSe single crystal D_6H_4 crystallizes in a hexagonal lattice with spatial plane symmetry. The elementary lattice has two layers, with each layer consisting of two molecules. Weak Van der Waals forces connect the atoms of neighbouring layers. As a result, many physical properties of these crystals are two-dimensional. These crystals' concentration of structural defects is estimated to be around 10^{17} cm^{-3}.

The effect of γ-quanta on the electrical and photoelectric properties of GaSe single crystals has been partially studied. It has been shown that resulting defects can be located within the layer and in the interlayer region. As a result of their influence, the conductivity increases due to the decrease of the specific reluctance of the crystals. However, information about the nature of radiation defects and the mechanism of their formation was not provided.

To clarify the mechanism of formation of these defects and the effect of external factors on them, researchers studied the effect of H$^+$ ions (70 keV) and subsequent thermal infusion on the mechanism of current passage in GaSe single crystal.

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2. EXPERIMENTS

The GaSe single crystal was studied, which was obtained using the Bridgman-Stockbarger method. The crystal had p-type conductivity, and its specific resistance was $2\cdot10^9$ Ohm·cm and $1\cdot10^8$ Ohm·cm in the directions parallel and perpendicular to the layers, respectively, at room temperature.

To investigate the effect of H^+ ions on the physical properties of the GaSe single crystal, samples were studied before implantation, and after being implanted with $2\cdot10^{15}$ ions/cm^2 of H^+ ions with an energy of 70 keV, followed by thermal annealing at 100-200°C for 1 hour. A sample thickness of ~50 μm was selected for the study. The average penetration depth of a proton with an energy of 70 keV in a GaSe crystal is Rp ~ 1.40 μm [12]. The GaSe crystal was implanted with H^+ ions using an ESU-2 accelerator, with the entire surface of the sample being implanted with H^+ ions in the indicated doses. The energy of H^+ ions was 70 keV, and the current density was 0.15 μA/cm^2.

The samples' volt-ampere characteristics and specific conductance were studied before and after irradiation in the ranges of electric field 10-10^4 V/cm and temperature 100-300 K. The distance between the contacts was ~50 μm. After the study, the characteristics of the implanted samples were studied again after being annealed (1 hour) in the temperature range of 100-200°C. The obtained results were analysed based on relevant theories.

3. RESULTS AND DISCUSSIONS

Figure 1 displays the volt-ampere characteristics of a GaSe single crystal before implantation (1), after implantation (2) and after thermal annealing (3) at different temperatures. The I-u curves consist of three regions: ohmic, quadratic and sharp growth. It can be observed that after implantation, the transition voltage from the ohmic region to the quadratic region decreases, as shown by curve 2. After thermal annealing of the implanted samples, the transition voltage value increases, as shown by curve 3. These results indicate that the defects caused by H+ ion implantation are mainly of an acceptor nature and that the specific resistance of the samples decreases as a result of implantation. This suggests that the injected charge carriers from the electrode cannot fill the traps, and conduction occurs due to thermal ionization of shallow levels. As the voltage applied to the samples increases, the concentration of the injected charge carriers also increases, partially filling the traps. After the traps are filled, there is a sharp increase in the current, up to the quadratic region, as shown by curve 2.

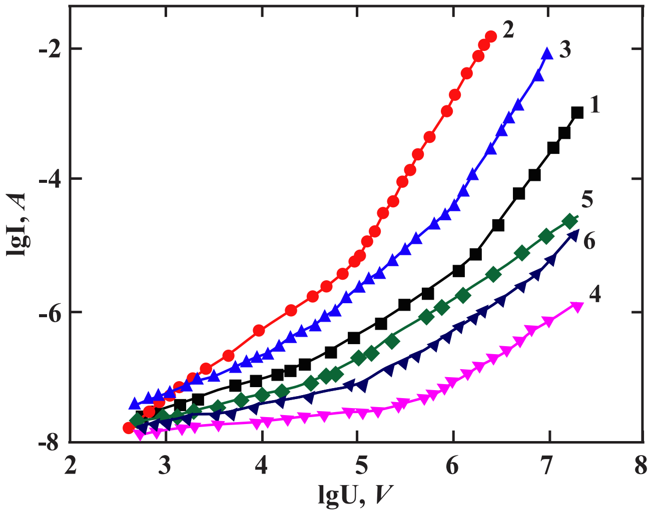

Figure 1. Volt-ampere characteristic of GaSe single crystal at room and nitrogen temperatures. 1. GaSe-initial, 2. GaSe(H^+), 3. GaSe(H^+) T = 300 K, 4. GaSe-initial, 5.GaSe(H^+), 6.GaSe(H^+) T = 100 K.

Based on [15], the concentrations of charge carriers in equilibrium in GaSe crystals were calculated using Figure 1. The concentration values were determined based on the transition voltage from the ohmic region to the quadratic region for the crystals. The values were found to be n_0 ~ $2.8\cdot10^{10}$ cm^{-3}, $6.8\cdot10^{11}$ cm^{-3} and $8.2\cdot10^{11}$ cm^{-3}, respectively. The concentration of traps was also calculated based on the electric capacitance of the sample at an input voltage of $U = 0$ ($C = 5\cdot10^{-10}$ F) and the transition voltage of the traps to a fully charged state (U_{fft}). For GaSe crystals from curves 1-3, $N_{t01} = CU_{\text{fft}}/qv = 5\cdot10^{14}$cm^{-3}, $N_{t02} = 9\cdot10^{13}$ and N_{to3}=$4\cdot10^{13}$ cm^{-3}.

The comparison of the calculated parameters shows that the transition voltage decreases from the ohmic region to the quadratic region due to the formation of acceptor-type defects after H^+ ion implantation. After thermal annealing, the transition voltage at both temperatures increases again (curves 3 and 6). The results of the $I \sim U^n$ dependences show that the defects (Frenkel pair) formed during H^+ implantation in GaSe crystals interact with structural defects to form acceptor-donor complexes and affect the charge transport mechanism.

A comparison of curves 1-3 in Figure 1 shows that the increase in the current value at the temperature $T = 300$ K after implantation with H^+ ions is due to the rise in the concentration of acceptor-type levels. The decrease in current after thermal annealing occurs due to the recombination of defects created during implantation. The obtained results correspond to the theory of the current limited by the area of charges, and the parameters of the energy levels are presented in Table 1.

Table 1. Physical parameters of implanted and non-implanted GaSe samples.

Sample	ΔE_t (eV)	ΔE_{1_2}(eV)	λ (cm)	r_m (cm)	N_t(cm^{-3})	p_0 (cm^{-3})
GaSe (pure)	0.23	0.35	$8.9\cdot10^{-6}$	$5\cdot10^{-6}$	$5\cdot10^{14}$	$1.4\cdot10^{13}$
GaSe ($5\cdot10^5$ ion impl)	0.26	0.32	$7\cdot10^{-6}$	$14\cdot10^{-6}$	$9\cdot10^{13}$	$3.44\cdot10^{13}$
GaSe (H^+) + TT = 100°	0.28	0.39	$7.9\cdot10^{-6}$	$4.5\cdot10^{-6}$	$4\cdot10^{13}$	$3.55\cdot10^{13}$

Figure 2 depicts the temperature dependence of the electrical conductivity of a GaSe crystal before and after implantation at varying electric field intensities. Comparing curves 1-3 reveals that two energy levels (0.19 and 0.34 eV) were present in the forbidden zone before implantation (curve 1). After implantation, the conductivity of the crystal increased partially (curve 2), and new energy levels with ionization energy of 0.18 eV and 0.45 eV were added. However, after thermal infusion at 370 K, the conductivity of the sample decreased, returning to its previous character. By comparing the electrical conductivity values in curves 1-3, it can be concluded that the defects created in the Ga and Se lattices during implantation are dominated by acceptor-type defects and V_{Ga} - vacancy.

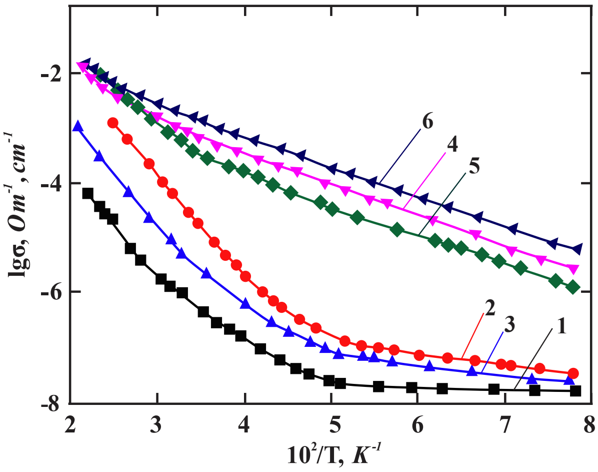

Figure 2. Temperature dependence of the conductivity of GaSe single crystal in the dark at a voltage of 10 V:
1. GaSe-initial, 2. GaSe(H^+), 3. GaSe(H^+)+T; 4. GaSe-primary, 5. GaSe(H^+), 6. GaSe(H^+)+T, U = 10 V under the influence of light

Thermal annealing shows that the defects created during implantation undergo partial recombination at low temperatures due to their excitation, and hence the conductivity of the crystal (curve 2) decreases. The nature of the levels formed during implantation is determined by curves 4-6. It has been revealed that since the trapping-filling process takes place at local levels during illumination, the conductivity in the temperature range of 110-300 K increases linearly with increasing temperature. The ionisation energy of the level was calculated for all three cases and is approximately 0.32 eV from the slope of the curves. By comparing curves 1-3, it can be inferred that the defects created in layered crystals during implantation are of the same nature as primary structural defects. After thermal annealing, only the concentration of defects changes, and this fact is compensated during illumination (curves 4-6).

In Figure 3, we can see the temperature dependence of the specific electrical conductivity ($\lg \sigma_F/\sigma_0$) in a GaSe crystal at different voltages (10-40 V). Curves 1-6 in Figure 3 show that the curves' inclination and the current value increase with the electric field intensity. The dependence of $\ln\sigma \sim f(E^{1/2})$ in a GaSe crystal follows Frenkel's theory.

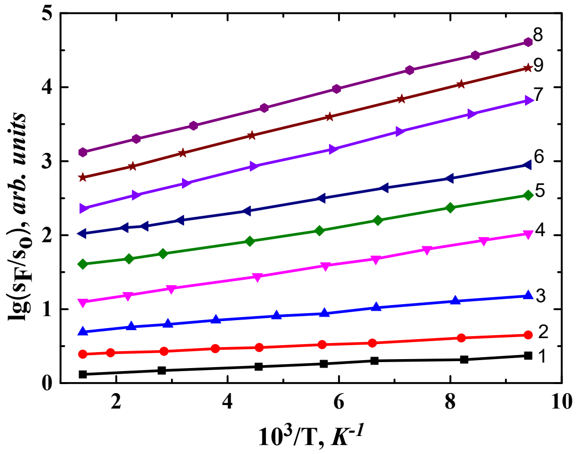

Figure 3. Temperature dependence of the conductivity of GaSe single crystal at different (10-40 V) voltage:
1. GaSe-initial, 2. GaSe(H^+), 3. GaSe(H^+)+T, 4. GaSe-initial, 5. GaSe(H^+), 6. GaSe(H^+).

Based on the experimental results, it can be said that conductivity $\sigma_0 = A exp(-\Delta E_0/2kT)$ in weak electric fields (ohmic region), and in high fields $\sigma_0 = A exp(-\Delta E_r - 2e(eE/\varepsilon)^{1/2}/2\varepsilon kT)$ follows. The slope of the curve increases as the temperature increases in the dependence of $ln\sigma \sim f(E^{1/2})$. This result is confirmed in Figure 4, which shows that the dependence $ln\sigma \sim f(E^{1/2})$ consists of two parts. At weak values of the field intensity ($E < 10^2$ V/cm), the conductivity does not change, and at high values, a linear exponential dependence is observed.

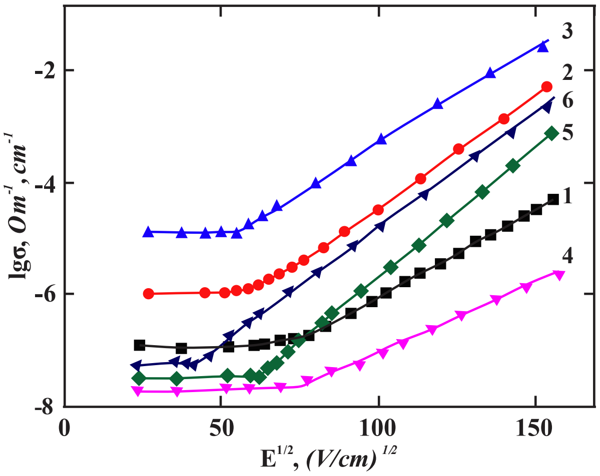

Figure 4. Dependence of conductivity on electric field intensity at different temperatures:
1. GaSe-initial, 2. GaSe(H^+), 3. GaSe(H^+)+T T = 100 K; 4. GaSe-initial, 5. GaSe(H^+), 6. GaSe(H^+)+T T = 300 K

When temperature decreases, the transition voltage value increases (curves 4-6). By comparing curves 1-6 at different temperatures, we observe that samples implanted with high field intensities exhibit a less steep curve in low and high-temperature ranges (curve 5). This implies that the activation energy associated with the defect level depends on the field intensity. This effect is more noticeable in implanted samples, and the concentration of electroactive centres influences the activation energy at the local level (Et). The increase in curve steepness due to the electric field indicates a change in the energy of the local levels. Considering the Paul-Frenkel effect, the reduction of Et under the field's influence is determined by the following expression:

$$\Delta E_t(E) = E_t(0) - (e^3 E / \pi \varepsilon \varepsilon_0)$$

where $\Delta E_t(0)$ is the activation energy of the level corresponding to the ohmic region ($E = 0$), $\Delta E_t(E)$ is the activation energy of the energy level in high fields.

The dependence of $\Delta E_t(E) \sim f(E^{1/2})$ is shown in Fig. 5 for thermally brewed GaSe crystals before and after implantation and after implantation. It can be seen from the figure that the energy of local levels decreases linearly with the increase of $\Delta E_t \sim E^{1/2}$. The activation energy of the trap was determined from the extrapolarization of the linear dependence of $\Delta E \sim f(E^{1/2})$ under the condition $E = 0$ and was 0.23, 0.26 and 0.28 eV for low temperatures and 0.35, 0.32 and 0.31 eV for high temperatures, respectively.

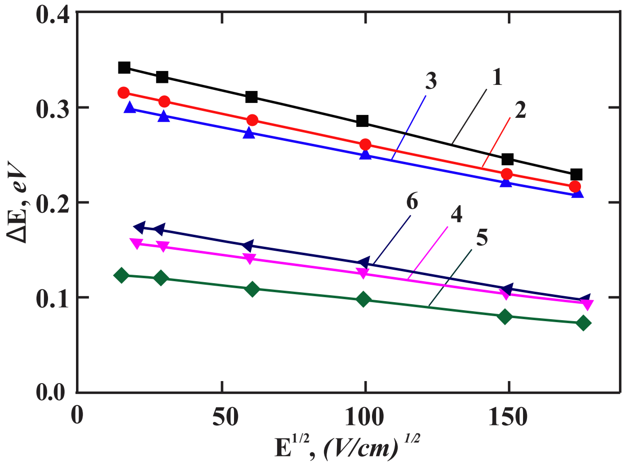

Figure 5. Dependence of trap activation energy ($\Delta E_t(E)$) on $E^{1/2}$ at different temperatures

At values of critical field intensity $E_k > 10^2$ V/cm, the field dependence of electrical conductivity obeys Frenkel's law. It can be seen from Figure 5 that the range of 5-40 V/cm of the field corresponds to the ohmic part, and the dependence of the conductivity on the field intensity is not observed in the dependence of $ln\sigma \sim E^{1/2}$. After the Ek value of the field intensity, the conductivity line increases according to the law, and with increasing temperature, the value of E_k and the slope of the dependence decrease. This indicates the thermal ionization of local levels of conductivity and corresponds to Frenkel's theory. Based on the experimental results, the value of the β-Frenkel coefficient was calculated according to the following expression: $\beta = 1/kT\,(e^3/\pi\varepsilon\varepsilon_0)^{1/2}$ and the dependence of $\beta \sim 1/\mathrm{T}$ is given in Figure 6. It can be seen from the figure that the regularity $\beta \sim 1/\mathrm{T}$ is fulfilled in GaSe crystals before implantation and thermally affected. It is proven that the conductivity depends on the intensity of the electric field and the temperature. From the comparison of curves 1-3 in Figure 6, it can be seen that the variation of the Frenkel coefficient β at the same temperature may be related to the possibility of the dielectric permittivity ε to accumulate radiation defects in the intralayer and interlayer regions.

The ionization of the local level depends on the field intensity, the height of the potential barrier, and the free escape path of the charge carrier. For this process to occur, the condition $\lambda > r_m$ must be met. To determine the length of the escape path of free carriers (λ) and the distance from the potential barrier to the bottom of the conduction zone (r_m) before and after implantation in a p-GaSe single crystal, as well as after thermal annealing, various conditions were taken into account such as $T = 300$ K, $E_k > 10^3$ V/cm, $\beta = 2.7\cdot10^{-2}$ V$^{-1/2}$ cm$^{1/2}$. The results were obtained and presented in a Table 1.

After comparing the values of λ and r_m given in the table, it can be concluded that the condition $\lambda > r_m$ is fulfilled for pure and thermally brewed GaSe crystals after implantation. This means that the electro-thermal ionization of local levels occurs, leading to an increase in current at values of $E_k > 10^3$ V/cm. In the case of samples implanted with H^+ ion, the condition $\lambda < r_m$ is met. This indicates that GaSe: H^+ local levels are filled with injected charge carriers at high field values, and the conductivity is determined by monopolar injection.

Based on these experimental results, the parameters of local energetic levels in GaSe: H+ crystals were calculated and presented in Table 1.

A model using proton stimulation was used to explain how H^+ ion defects are formed in layered crystals. When a proton interacts with an atom, the electrons break away and the atom becomes excited. This causes the electron layer to become ionized, and the nucleus interacts with the crystal lattice, resulting in defects. These defects are known as Frenkel pairs. In semiconductors, the defects caused by protons in the forbidden zone are energy levels that act as recombination centres for charge carriers of acceptor and donor type or non-equilibrium. During irradiation, Ga-vacancies increase the rate of stimulated diffusion of defects. Because these levels are unstable, their mobility and nature depend on temperature, making them important in modifying physical properties. Based on experimental results analyzed in [16-18], a mechanism for defect formation in layered p-GaSe crystals under low-energy H^+ (70 keV) ions was determined. According to this mechanism, defects are primarily formed at the end of the escape path (radiation loss ~ 1-2 keV, escape path ~1.4 µm). The generation of defects (Frenkel pairs) along the trajectory is unequal in terms of volume, and the resulting vacancies and interstitial atoms migrate. The acceptor nature of the defects created during initial irradiation ($2\cdot10^{15}$ ions/cm^2) leads to an increase in conductivity in the GaSe crystal. As a result of thermal annihilation, the migration rate of interstitial atoms increases, and defects are annihilated in the runway distance (radiation annihilation), resulting in a decrease in the conductivity of the crystal.

CONCLUSIONS

It investigated how electric field and temperature affect the mechanism of charge transport in GaSe crystal implantation with H+ ions and thermally soaked crystals. The study was conducted in the temperature range of 100-200 K, and the following results were determined:

1. During the implantation of 70 keV H^+ ions into p-GaSe layered crystal, dynamic local point defects are formed at a running distance of ~1.5 µm. When the surface layer undergoes thermal annihilation (100-200 °C) of defects formed, partial annihilation of defects occurs in the lattice due to ion-stimulated diffusion of metastable defects, and defects become stable. This increases the crystal's resistance to radiation.

2. The study determined that local point defects (0.23-0.39) eV and ~ 9·1013 cm^{-3}) formed in the p-GaSe crystal as a result of implantation of H^+ ions determine the charge transport mechanism in the temperature range of 100-300 K. At electric field values Ek < 103 V/cm, primary and implanted crystals exhibit monopolar injection. At values of E > 103 V/cm, an increase in current due to the Frenkel mechanism is observed as a result of thermal ionization of local levels in all crystals. At low temperatures in thermally annealed GaSe:H+ crystals, the charge transport mechanism has a non-activated character.

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МЕХАНІЗМ ТРАНСПОРТУ ЗАРЯДУ В ІМПЛАНТОВАНОМУ МОНОКРИСТАЛІ p-GaSe:H+

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У дослідженні проаналізовано вплив радіаційних дефектів монокристала p-GaSe, імплантованого іонами H+ (70 кеВ), на механізм переносу заряду. Дослідження проводили при 100K і 300K в електричному полі 10^2-10^4 В/см. Дослідження показало, що енергія активації носіїв заряду, інжектованих при низьких температурах і електричних полях $E < 10^3$ В/см, коливалася в межах 0,23-0,39 еВ. Це спостерігалося через захоплення носіїв заряду в концентраційні пастки приблизно $9\cdot10^{13}$ см$^{-3}$, що призвело до монополярної інжекції. У полях $E > 10^3$ В/см спостерігалося різке зростання струму, що пояснювалося термоіонізацією локальних рівнів за механізмом Френкеля. Дослідженням встановлено, що механізм переносу заряду в кристалах GaSe:H+ при низьких температурах має неактивований характер.

Ключові слова: *імплантація; GaSe; монокристал; механізм Френкеля*