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EFFECT OF GAMMA-IRRADIATION ON THE ELECTROPHYSICAL PROPERTIES OF LAYERED GaS:Er SINGLE CRYSTALS

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The electrical and photoelectrical properties of layered GaS:Er single crystals, gamma-irradiated at room temperature at absorbed dose range D = 20…100 krad have been investigated. It has been established, that both donor- and acceptor-type defects are formed during doping and irradiation, leading to a decrease in electrical conductivity. At low temperatures (T = 125…180 K), the activation energy of the levels was determined to be 0.036 eV for Er-doped GaS single crystals, while for irradiated samples it was found to be 0.068 and 0.120 eV for doses of D = 20 and 50 krad, respectively. After irradiation, the band gap of the sample arises with increasing dose from 2.48 to 2.91 eV and the photocurrent spectrum maximum shifts toward higher energy regions. The obtained results demonstrate that GaS:Er single crystals can be considered promising functional materials for optoelectronic applications and radiation-resistant semiconductor devices through controlled defect engineering.

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INTRODUCTION

The role and significance of impurity and defect interaction processes in the science of controlling the properties of semiconductor materials are appeared in complicated semiconductor compounds, including, in particular, compounds of the $A^{III}B^{VI}$ group [1–8]. Due to inadequate research and the challenge of controlling the interactions between impurities and defects in them, the compounds in this group are currently underutilised in modern optoelectronic devices despite having distinct photoelectric, radiation, and electro-optical capabilities. Investigating the physico-chemical regularities of the structural defect remodelling and impurity-defect interactions in single crystals of $A^{III}B^{VI}$ compounds, as well as the relationships between these processes, is a fundamental work [9–12].

GaS monocrystal, which corresponds to the $A^{III}B^{VI}$ group of compounds, has a broad band gap, high sensitivity in the UV region of the spectrum, radiation resistance, and anisotropy of chemical bonds, which make it easier to manipulate and utilise its fundamental features. [13–23]. The two primary characteristics of layered crystals are weak van der Waals connections between the layers and covalent bonds within the layer. The previously mentioned bonds' controlled properties through doping and ionizing radiation allow for intentional modification of the physical properties of multilayer crystals. The electrical [18], optical, and photoelectric [24] properties of the layered GaS monocrystal have been well studied, but little is known about how defects in the structure impact the flux mechanism of the crystals. Consequently, in the study [25], the effects of metal and dopant atoms of rare earth elements (REE) on the conductivity of layered crystals were analysed using the partially-disordered system model. It has been shown that the properties of multilayer crystals can be altered by manipulating the height of the potential barrier between regions through the impact of dopant atoms. Additional research [26–28] provides information on how the conductivity of GaS monocrystals is impacted by the gamma-radiation exposure. These results explain the development mechanism of radiation defects in pure crystals. Determining the influence mechanism of defect-dopant interaction in layer and interlayered areas is one of the main obstacles in providing a control mechanism for the physical properties of defective (~10^{17} cm^{-3}) layered crystals, according to an analysis by [29]. To solve this issue, it is essential to investigate the structures and features that govern how the physical properties of multilayer crystals change with the type and concentration of dopant and defect atoms. Ionising exposure to radiation can cause radiation defects in semiconductor materials, which can lead to vacancies, dislocations, and interstitials in the crystal lattice [30–34]. By altering the material's optical characteristics, electronic structure, and transport properties, these flaws can influence the material's anisotropic behaviour across multiple crystallographic orientations.

The aim of this work is to investigate the effect of gamma irradiation on electrophysical (electrical and photoelectric) properties of layered GaS:Er single crystals with ~ 0.1 at.% concentration of erbium and to elucidate the interaction of radiation-induced defects with structural defects. Radiation-induced defects are primarily defined as vacancy centers (sulfur and gallium vacancies), Frenkel pairs, and Er-vacancy complexes formed during γ irradiation. These defects arise as a result of the synthesis of vacancies and interstitials caused by high-energy secondary electrons generated during irradiation, which break Ga-S bonds.

1. EXPERIMENTAL PART

The GaS single crystal investigated in this study was synthesised using the Bridgman–Stockbarger method at elevated temperatures. During crystal growth, Erbium (Er) was introduced as a dopant into the layered GaS monocrystal. Due to the volatility of sulfur, its

concentration tends to decrease during crystal growth; therefore, to compensate for sulfur deficiency, the GaS monocrystal was additionally annealed in sulfur vapour.

Doping was performed during the crystal growth process. Erbium (Er) was selected as the dopant element with a concentration of approximately 0.1 at.%. The obtained GaS:Er monocrystals exhibited p-type conductivity with a resistivity of about $\rho \approx 10^9$ Ω·cm, similar to that of undoped crystals. High-purity source materials were used: Ga (99.99%), S (99.999%), and Er (0.1%). The charge was completely melted in the hot zone of a two-zone Bridgman furnace at 1360 K and then slowly transferred to the cold zone (823 K) at a rate of 1.2 mm/h.

For measurements, samples with a thickness of d ~ 200 μm were prepared, oriented along the (001) direction, and polished to obtain mirror-like surfaces.

GaS belongs to the class of wide-bandgap semiconductors (Eg ≈ 2.5 eV), crystallizing in the hexagonal system with lattice parameters a = b = 0.378 nm and c = 1.547 nm, and belonging to the $P6_3/mm$ space group [3]. Strong covalent bonds exist within the layers, while weak Van der Waals interactions occur between the layers.

The samples were irradiated with γ-rays (E = 1.25 MeV) at a dose rate 106 rad/s using a ^{60}Co-based irradiation facility at room temperature. Ohmic contacts were formed by applying silver paste perpendicular to the crystal layers (parallel to the c-axis). Electrical and photoelectrical measurements were carried out using standard experimental techniques and measurement circuits [4].

2. RESULTS AND DISCUSSION

The aim of the work is to dope layered GaS single crystals with the rare earth element Er, to investigate the energy level spectra before and after irradiation with γ-quanta, and to determine a number of parameters of these levels.

The temperature dependence of the electrical conductivity of the investigated samples is presented in Fig. 1. As can be seen from the figure (curve 1), the electrical conductivity of the undoped GaS single crystal remains unchanged in the temperature range T = 125…250 K. With a further increase in temperature, the conductivity rises sharply, and the activation energy associated with this level is determined to be 1.461 eV.

The nature of the temperature dependence of the electrical conductivity of the Er-doped GaS single crystal differs from that of the undoped sample (curve 2). Thus, at low temperatures (T = 125…190 K), the electrical conductivity decreases sharply, and from the slope of the curve, the activation energy of conductivity in the range T = 125…175 K is determined to be 0.036 eV. In the temperature range T = 175…260 K, thermal quenching of conductivity is observed. In the range T = 260…300 K, the electrical conductivity increases sharply, and the activation energy in this region is determined to be 0.833 eV.

When the GaS:Er single crystal is irradiated with a dose of D = 20 krad, the character of the curve does not change, but the conductivity value decreases (curve 3). In the temperature range T = 125…175 K, the conductivity increases with temperature, and the activation energy of conductivity is calculated as 0.068 eV. In the temperature range T = 175…230 K, thermal quenching of electrical conductivity is observed, and the depth of thermal quenching increases and shifts toward the lower temperature region. In the temperature range T = 230…300 K, the electrical conductivity increases, and in this region, the activation energy is determined to be 0.684 eV.

When GaS:Er single crystal is irradiated with a dose of D = 50 krad (curve 4), the value of electrical conductivity decreases. In the temperature range T = 125….175 K, the conductivity increases with increasing temperature, and from the slope of the curve, the activation energy of electrical conductivity is calculated to be 0.120 eV.

In the temperature range T = 175…230 K, thermal quenching of electrical conductivity is observed. The value of electrical conductivity decreases and shifts toward the lower temperature region. With further increase in temperature, the electrical conductivity increases sharply, and the activation energy of the level is calculated to be 0.867 eV.

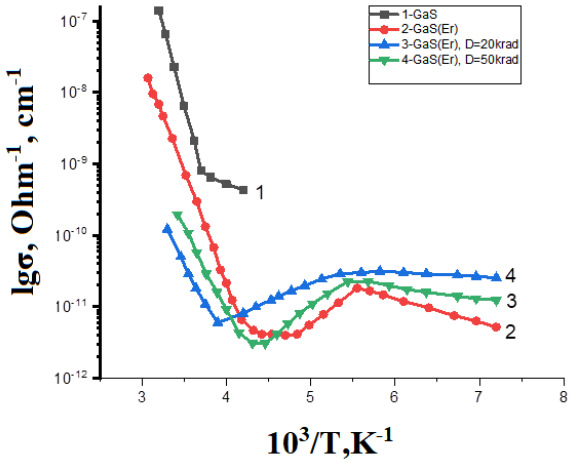


Fig. 1. Temperature dependence of electrical conductivity of initial (curves 1, 2) and gamma-irradiated (curves 3,4) GaS and GaS:Yb single crystals

When GaS single crystals are doped with Er, due to the similarity between the ionic radius of erbium atoms and that of the host component atoms, erbium atoms can substitute gallium atoms within the layers or occupy interstitial positions both within and between the layers.

As a result, the concentration of vacancy-type structural defects decreases, since sulfur atoms interact with erbium atoms, leading to a reduction in hole concentration.

Furthermore, erbium atoms fill vacancies and, being distributed in different layers, form stronger covalent bonds between adjacent layers compared to the weak Van der Waals interactions, which leads to a decrease in electrical conductivity.

Fig. 2 shows the spectral distribution of photocurrent for pure GaS (curve 1), Er-doped GaS (curve 2), and γ-irradiated GaS:Er single crystals (curves 3 and 4) at T = 300 K. As can be seen from the figure, the photocurrent maximum shifts within the range of 2.48, 2.6, 2.7, and 2.91 eV, respectively. The shift of the photocurrent maximum is associated with changes in the band gap width.

In addition, the magnitude of the photocurrent increases both after doping and irradiation. The increase in photosensitivity of both doped and γ-irradiated GaS single crystals, as well as the change in the band gap width, is attributed to the redistribution of hole recombination flux from fast s-recombination centres to slower r-recombination centres.

Various structural defects act as s-centres, while cation vacancies (V_{Ga}) serve as r-centres. An increase in the band gap width of the GaS single crystal leads to an increase in the distance between the Fermi level and the s- and r-centres, which in turn affects the filling of these levels with charge carriers and the recombination flux through these centres.

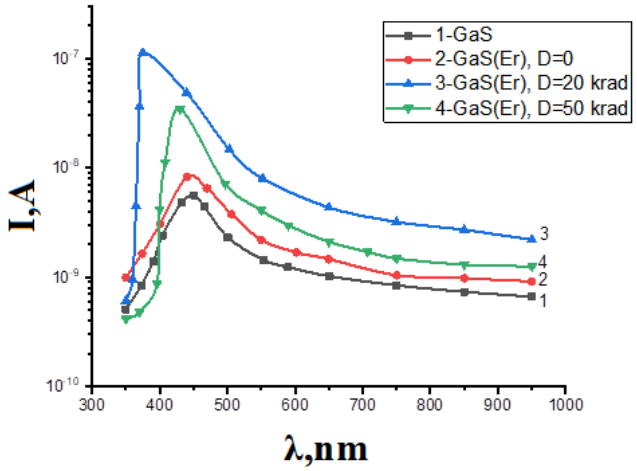


Fig. 2. Spectral distribution of photocurrent (U = 50 V) of initial (curves 1, 2) and gamma-irradiated (curves 3, 4) GaS and GaS:Er single crystals

Let us assume that the t-centre is associated with the valence band, such that its position does not change relative to the valence band. In this case, as the band gap increases, the energetic position of the t-centre shifts upward relative to the conduction band. Under these conditions, the t-centre acts both as a trapping centre and as a recombination centre for electrons.

For compounds with a small t-centre energy (Et), electrons are not effectively excited into the conduction band. The probability of electron capture by trapping centres is very high, and almost all electrons recombine with nonequilibrium holes at r- and s-centres. In this case, the occupation of r- and s-recombination centres by electrons reaches a maximum, and the lifetime of holes in the valence band becomes minimal.

$$\tau_p = 1/\upsilon s_p n_e. \quad (1)$$

Here n_e is concentration of electrons in recombination centres (r and s) depends on the capture cross-section of holes by these centres in the valence band. Photosensitivity is proportional to the lifetime and mobility of holes. As a result of both irradiation and doping, the increase in the band gap width and the t-centre energy enhances the efficiency of trapping centres (t-centres), leading to an increased filling of these centres with electrons and a decrease in their concentration in the conduction band. This, in turn, increases the lifetime of holes and consequently enhances the photosensitivity of the sample.

CONCLUSIONS

The conducted comprehensive experimental studies have shown that structural changes occurring in GaS single crystals doped with the rare earth element Er and irradiated with γ-quanta at various doses significantly affect their electrophysical and photoelectrical properties. As a result of doping and irradiation processes, both donor- and acceptor-type localised energy levels are formed in the crystal, leading to changes in the concentration of charge carriers and their transport mechanisms.

Temperature-dependent electrical conductivity measurements indicate that, in the low-temperature region (T ≈ 125…180 K), conductivity is mainly governed by an activation-type mechanism, and the corresponding activation energies increase significantly for doped and irradiated samples. This behaviour is explained by the deepening of defect levels and their redistribution within the energy spectrum.

In addition, with increasing γ-irradiation dose, an expansion of the band gap (from 2.48 to 2.91 eV) and a shift of the photocurrent spectrum maximum toward higher energy regions are observed. This effect is associated with the redistribution of recombination centres (from fast s-centres to relatively slower r-centres), as well as changes in the position of the Fermi level.

As a result, defect complexes formed due to doping and radiation influence the recombination kinetics and

lifetime of charge carriers, leading to an increase in the photosensitivity of the material. The obtained results demonstrate that GaS:Er single crystals can be considered promising functional materials for optoelectronic applications and radiation-resistant semiconductor devices through controlled defect engineering.

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ВПЛИВ ГАММА-ОПРОМІНЕННЯ НА ЕЛЕКТРОФІЗИЧНІ ВЛАСТИВОСТІ ШАРУВАТИХ МОНОКРИСТАЛІВ GaS:Er

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Було досліджено електричні та фотоелектричні властивості шаруватих монокристалів GaS:Er, опромінених гамма-випромінюванням за кімнатної температури в діапазоні поглинутої дози D = 20…100 крад. Встановлено, що під час легування та опромінення формуються дефекти як донорного, так і акцепторного типів, що призводить до зниження електропровідності. За низьких температур (T = 125…180 K) енергія активації рівнів для легованих ербієм монокристалів GaS становила 0,036 еВ, тоді як для опромінених зразків вона дорівнювала 0,068 та 0,120 еВ за доз D = 20 і 50 крад відповідно. Після опромінення ширина забороненої зони зразка збільшується зі зростанням дози від 2,48 до 2,91 еВ, а максимум спектра фотоструму зміщується в бік вищих енергетичних областей. Отримані результати демонструють, що монокристали GaS:Er можуть розглядатися як перспективні функціональні матеріали для оптоелектронних застосувань і радіаційно стійких напівпровідникових пристроїв завдяки керованому формуванню дефектів.