

EFFECT OF STOICHIOMETRIC DISTORTIONS ON ELECTRICAL AND PHOTOELECTRIC PROPERTIES OF LAYERED GeS CRYSTAL

R.S. Madatov¹, A.S. Alekperov^{2,3}, V.A. Abdurahmanova², R.K. Huseynov⁴,
R.J. Bashirov⁵,  Y.I. Aliyev^{2,6*}

¹*Institute of Radiation Problems, Ministry of Science and Education Republic of Azerbaijan, Baku, AZ-1143, Azerbaijan*

²*Azerbaijan State Pedagogical University, Baku, AZ-1000, Azerbaijan*

³*Baku Engineering University, Khirdalan, AZ-0101, Azerbaijan*

⁴*Ganja State University, Ganja, AZ-2000, Azerbaijan*

⁵*Azerbaijan Technical University, Baku, AZ-1073, Azerbaijan*

⁶*Western Caspian University, Baku, AZ-1001, Azerbaijan*

*Corresponding Author email: yusifafshar@gmail.com

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The current-voltage characteristic, electrical conductivity, thermally stimulated current, and photoelectric properties of a layered GeS crystal with excess sulfur were investigated under an external electric field of 10-104 V/cm and at temperatures of 100-300 K. It was found that donor-type defects formed as a result of stoichiometric distortion due to excess sulfur in a GeS crystal obtained by the Bridgman method, leading to impurity conductivity. Charge transport occurs by a monopolar injection current limited by the volume of the charge region. It was found that thermal activation of photocurrent and thermal quenching of photocurrent in doped GeS crystals are associated with electron exchange between capture traps and recombination centers.

Keywords: Layered crystal; Electrical conductivity; Monopolar injection; Thermally stimulated current; Photoconductivity

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1. INTRODUCTION

In low-dimensional layered crystals (GeS, GeSe, SnS, GaS, GaSe, InSe), the quantum effects observed during structure formation depend on the nature of the chemical bonds of the components and the shape of the crystal lattice. In such systems, the relationship between physical properties and structure was studied in binary systems $A^{III}B^{VI}$ and $A^{IV}B^{VI}$ [1-4]. It was found that the electrical, photoelectric, and radiation-resistance properties of layered crystals depend on the nature of the interactions between layers and within layers. Within the layer, this interaction is powerful and ionic-covalent in nature [5-7]. The formation of weak van der Waals bonds between layers leads to the emergence of anisotropic properties characteristic of layered crystals. The effects observed in low-dimensional structures form new areas of research in layered crystals and expand the possibilities of their practical application. Therefore, by selecting different components, it is possible to control the type of chemical bond intentionally.

A comparative analysis of the results obtained in the study of binary systems $A^{III}B^{VI}$ and $A^{IV}B^{VI}$ shows that the electrical, especially photoelectric properties of layered crystals depend on the electron configuration of the components and the nature and properties of the electron transitions of cation vacancies, which occur depending on external influences (temperature, illumination, radiation rays, etc.). On the other hand, elements with a large ionic radius and chemical activity, electrons in the p-layer of cations during the formation of chemical bonds have a strong effect on the electrical, photoelectric, and optical properties of the crystal [8]. In the GeS crystal, which is one of the binary compounds $A^{IV}B^{VI}$ with the above properties, information on the formation of chemical bonds, the structure of the crystal lattice and the influence of impurity atoms on the physical properties has been studied [9]. The studies show that the GeS crystal is a semiconductor with a complex band structure. The elementary lattice consists of two layers, each layer of two molecules. Since the atoms of adjacent layers are linked to each other by weak van der Waals bonds, many of the physical properties of the crystal are two-dimensional [10].

Since layered semiconductors crystallize in defect structures, the study of the electronic structure of localized defects is particularly important. Dopant atoms are distributed non-periodically in different spaces (inside a layer or in the interlayer region). Although the above facts were investigated in a doped GeS crystal, the results obtained do not support the development of a universal model explaining the mechanism of dopant-atom distribution. One of the important properties of a GeS crystal is that it has high concentrations (10^{17} - 10^{18} cm⁻³) of specific defects (cation and anion vacancies, Frenkel defects), especially cation vacancies. This property causes low mobility of charge carriers in the direction of the c axis of the crystal and a short diffusion path. A high concentration of specific defects, coupled with their compensation by dopant atoms, enables targeted control of properties. The above is also supported by the results of the study of the electrical and photoelectric properties of the GeS crystal [11].

Despite the existence of a number of research works on the study of electrical, photoelectric and optical properties of semiconductor compounds of the $A^{IV}B^{VI}$ type, including the GeS crystal, there is no information on the mechanism of

the effect of defects on the current [12-14]. The obtained results do not allow us to study the mechanism of the effect of specific defects in the GeS crystal, including uncontrolled impurity atoms, on conductivity. The study of defects present in semiconductor crystals provides extensive information on the formation of their structural features and physical properties. Therefore, research has been carried out in this direction recently [15,16]. Therefore, in order to obtain additional information on the distribution, concentration and energy states of defects in layered crystals, it is planned to study the volt-ampere characteristics, electrical conductivity and photoconductivity of the GaS crystal with excess sulfur at temperatures of 100-300 K and external electric field values of 10^{-4} V/cm.

2. EXPERIMENTS

The GeS single crystal was grown by the Bridgman method, a standard method for chalcogenide semiconductors. High purity Ge and S (99.999%) were used in the synthesis process. Ge with a specific resistance of 50 Ohm·cm and B5 grade sulfur were used in the synthesis. To maintain the stoichiometric ratio and minimize sulfur vacancies, the quartz ampoule filled with components taken with excess sulfur was welded after reducing the pressure to 1.33×10^{-2} Pa. During synthesis, the ampoule heating rate at the first stage was set to 1.5 degrees/minute, and after melting the germanium, the heating rate was set to 2.5 degrees/minute. To ensure the safety of the synthesis process, the germanium crystal was ground into powder, and the evaporation and condensation zone temperatures were 1173 K and 1073 K, respectively. The homogeneity of the grown crystals and their single-phase nature were tested using differential thermal analysis (DTA), X-ray diffraction (XRD) and electron microscopy (SEM). The obtained crystals had *p*-type conductivity and a specific resistance of 10^5 - 10^6 Ohm·cm. The sample size during measurements was $2.0 \times 5.0 \times 0.2$ mm³. The conductive contacts were applied to a natural flat surface using aquadag and had an ohmic character. The study of electrical and photoelectric properties was carried out at 100-300 K and in an electric field of 10^4 V/cm.

RESULTS AND DISCUSSIONS

During the research, the structure of the GeS crystal obtained with excess sulfur was studied by X-ray diffraction. The X-ray diffraction spectrum obtained at room temperature is shown in Figure 1. The spectrum shows six main maxima over the diffraction angle range $2\theta = 20$ - 70° . It was determined that these maxima correspond to the atomic planes (111), (020), (022), (131), (222) and (040). The obtained single crystal is single-phase and orthorhombic. The lattice parameters correspond to the values: $a = 4.291$ Å, $b = 3.641$ Å and $c = 10.471$ Å.

The volt-ampere characteristics of a GeS single crystal with excess sulfur at different temperatures were studied. The obtained dependences are shown in Fig. 2. From the nature of curves 1-3 it is evident that the $I \sim U^n$ dependences consist of ohmic, quadratic and sharply increasing regions. From the dependences it is evident that the GeS crystal has a semiconductor property, i.e. with decreasing temperature the resistance of the sample increases, due to which the ohmic region observed at room temperature shifts towards the region of high voltages.

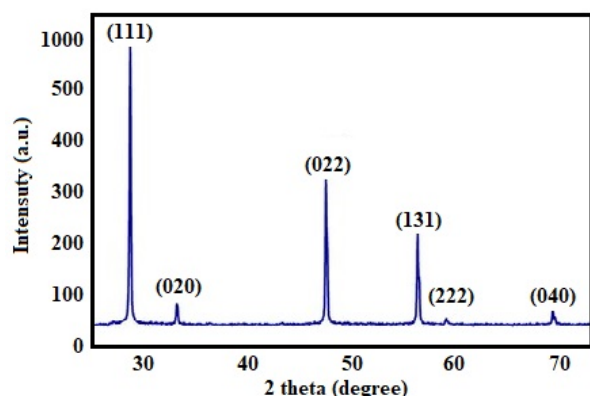


Figure 1. X-ray diffraction spectrum of a GeS crystal

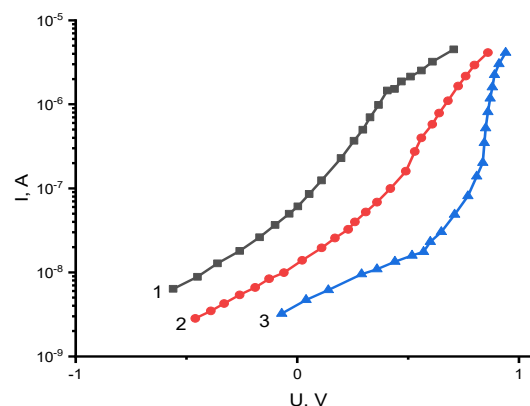


Figure 2. Current-voltage characteristics of a GeS crystal at different temperatures: 1) $T = 100$ K, 2) $T = 160$ K, 3) $T = 300$ K

According to Lambert's theory, the charge carriers injected from the electrode cannot completely fill the traps, and conductivity occurs due to thermal ionization of shallow levels. With a further increase in the voltage applied to the sample, the concentration of injected charge carriers increases and the traps are partially filled. After the traps are completely filled, with increasing voltage, the current increases sharply to the quadratic region without jumps (curves 1-3). Based on the value of the transition voltage from the ohmic part to the quadratic part for the crystals from Figure 2, the concentration and mobility of charge carriers in the equilibrium state in GeS crystals were calculated according to Lampert's theory ($n_0 \sim 4 \cdot 10^{13}$ cm⁻³, $\mu = 20$ -30 cm²/V·s) [12].

Based on the measured value of the electrical capacitance of the GeS sample at the input voltage $U = 0$ and the transition of the traps to a completely filled state ($C = 5 \cdot 10^{-10}$ F, E7-20 is an impedance meter), the trap concentration was calculated to be $Nt = CUtd/q \cdot V = 5 \cdot 10^{14}$ cm⁻³ (where V is the sample volume). The shift of the Fermi level from the equilibrium state and $\Delta E_f \approx 0.02$ eV was. This indicates that at low voltages (in the ohmic region) and low temperatures,

the charge carriers injected from the electrode cannot completely fill the traps and conductivity occurs due to the thermal ionization of shallow levels.

To study the electrical properties of semiconductor materials, it is necessary to analyze the temperature dependences of electrical conductivity. Previous studies have shown that a number of electronic processes occurring in these materials can be explained based on the temperature dependence of electrical conductivity [17-22]. Therefore, for the GeS crystal, a mechanism for changing electrical conductivity depending on temperature was determined. Figure 3 shows the temperature dependence of the electrical conductivity of the GeS crystal at different electric field strengths.

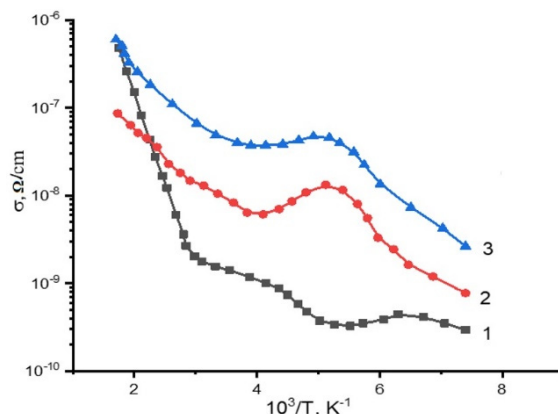


Figure 3. Electrical conductivity of doped GeS crystal at different field strengths: 1) 10^2 V/cm, 2) 10^3 V/cm, 3) 10^4 V/cm

The dependences show that in an undoped GeS crystal, a specific and additive region is observed, and with increasing temperature, the dark conductivity increases, passes through a minimum and increases again (curve 1). At high electric fields, an increase in temperature leads to the dark conductivity passing through a maximum and increasing again (curves 2 and 3). From a comparison of the experimental curves, it is clear that at low and high field strengths, the dark conductivity is explained by two opposing processes: thermal ionization and electroionization. Thus, in the GeS crystal, the dark conductivity in the layer direction obeys the law $\sigma_0 = A \cdot \exp(-\Delta E_0/2kT)$, and in strong fields – the law $\sigma_0 = A \cdot \exp(-\Delta E_r - 2e(eE/\epsilon)^{1/2}/2\epsilon kT)$. The calculated values of the activation energy of the energy levels from the slope of the curve from the dependence $\ln \sigma_0 \sim f(1/T)$ were 0.22 eV and 0.45 eV. From a comparison of curves 1 and 3 in Figure 3, it is evident that the slope of the curve increases with increasing field strength in the range of 120-250 K. At low field strengths ($E < 10^2$ V/cm), the conductivity occurs due to thermal ionization of the defect level, and in strong fields it increases exponentially as a result of field ionization.

To determine the energy depth, concentration and cross-section of filling centers located in the forbidden zone in the GeS crystal, the thermally stimulated current was studied, the results are presented in Figure 4.

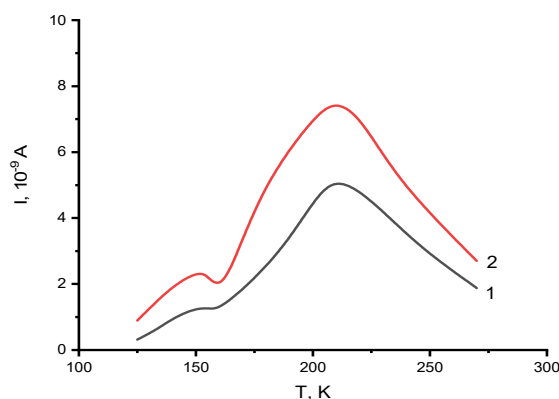


Figure 4. Thermally stimulating current in a doped GeS crystal at different field strengths: 1) 10^2 V/cm, 2) 10^4 V/cm

According to the research methodology, the GeS sample was cooled to a temperature of 100 K in vacuum, then illuminated with natural light for 5 minutes and heated at a rate of 0.40 degrees/second at different field strengths (10^2 V/cm, 10^4 V/cm), the resulting signal was recorded on a special recorder. From Figure 4 it is evident that the thermal signal increases with increasing temperature, and the maxima of low and high intensity are observed at temperatures of 150 K and 210 K, respectively. Based on the conditions given in the research [12], according to the values of T_{max} and $T_{1/2}$ from Figure 4, the activation energy of the filling center ΔE_i was $\Delta E_1 \approx 0.19$ eV and $\Delta E_2 \approx 0.27$ eV, the concentration $N_{t1} \approx 2 \cdot 10^{13}$ cm $^{-3}$ and $N_{t2} \approx 1 \cdot 10^{17}$ cm $^{-3}$, and the filling cross-section $S_{t1} \approx 10^{-20}$ cm $^{-3}$ and $S_{t2} \approx 10^{-19}$ cm $^{-3}$.

Figure 5 shows the spectral characteristics of photoconductivity in the layer direction in a GeS crystal at temperatures of 300 K and 100 K. It is evident from the dependences that two maxima ($\lambda = 0.75$ μ m and $\lambda = 0.90$ μ m) are observed on

the curve of the spectral distribution of photoconductivity in an undoped GeS crystal. The first maximum falls on the region of specific absorption, and the second on the doped region; therefore, it corresponds to the defect-zone transition. The observation of a doped absorption region in an undoped GeS crystal can be explained by the formation of partially irregular regions (low-resistance regions) in layered compounds.

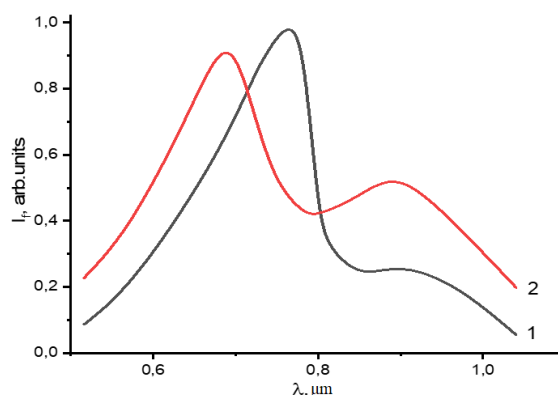


Figure 5. Spectral distribution of photoconductivity in undoped GeS crystal. 1) $T = 300$ K, 2) $T = 100$ K

During the synthesis of the GeS crystal, the excess of sulfur causes a violation of stoichiometry, which leads to the formation of a local impurity band in the crystal [23]. Figure 6 shows that at a temperature of $T \sim 100$ K (curve 2), both maxima shift to the short-wave region. This proves that the absorption outside the fundamental absorption band in the long-wavelength region is due to impurities. The temperature dependence of the photoconductivity in the GeS crystal upon excitation by natural light of varying intensity in the direction perpendicular to the layer surface and at an electric field strength of 10^4 V/cm is shown in Figure 6.

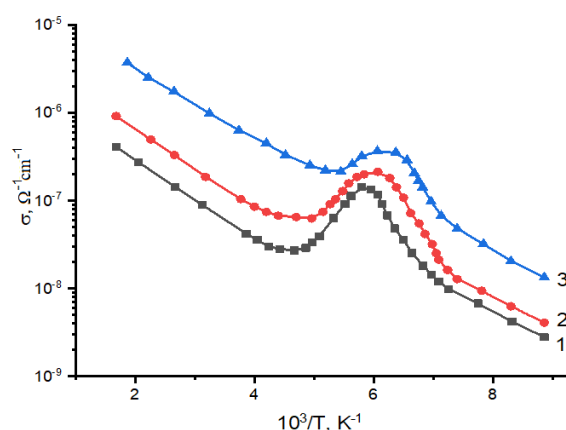


Figure 6. Temperature dependence of photocurrent in undoped GeS crystal. 1) 10 lx, 2) 10^2 lx, 3) 10^4 lx

It is evident from the figure that in the range of 100–160 K, as a result of thermal activation, the photocurrent increases (thermal activation), and in the region of 160–200 K, thermal quenching of photoconductivity is observed. With a further increase in temperature, the photocurrent increases again. It should be noted that with an increase in the illumination intensity the depth of the maximum decreases. It is evident from the experimental results that the dependence $\sigma_f \sim f(1/T)$ obeys the law $\sigma_f = \sigma_0 \exp(\Delta E_f/kT)$ and two processes are observed - thermal activation and thermal quenching of photoconductivity. The activation energy determined from the slope of the curve was ~ 0.27 eV. It should be noted that there is no information on thermal quenching of photoconductivity in the doped GeS crystallite, but it is noted that there is specific conductivity. The activation of the photocurrent observed by us in the GeS crystal with excess sulfur occurs as a result of the nonequilibrium exchange of the majority charge carriers between the photoactivation trap and the recombination center, as shown in [24].

The results of the study of electrical conductivity and photoelectric properties of alloyed GeS crystals (with an excess of sulfur), obtained by the Bridgman method at different temperatures and external electric fields, show that the capture and recombination processes in alloyed GeS crystals largely depend on the chemical nature of the components, including alloying atoms, and less on the degree of disorder in the crystal. In contrast to previous studies, we found that complex defects (complexes, dislocations, point defects) formed in alloyed GeS crystals with an excess of sulfur, as well as in alloyed GeS, have a strong influence on the electronic processes occurring in the crystal under external excitation (light, temperature, radiation). Therefore, as a result of exposure to an electric field and illumination in the alloyed region of conductivity of the GeS crystal, thermal quenching is observed (Fig. 3, curve 1). The reason for this is that excess sulfur causes the formation of defect regions with activation energies of 0.17 eV and 0.27 eV. This fact is confirmed by the

dependence presented in Figure 4. As you can see, excess sulfur causes partial disorder of the crystal, both due to stoichiometric distortion and due to the formation of new structural defects.

At the same time, the temperature dependence of the charge-carrier mobility partially confirms the correctness of the observed effect (Fig. 7).

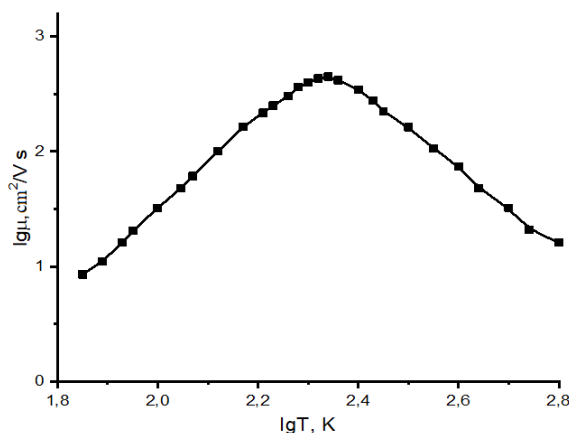


Figure 7. Temperature dependence of charge-carrier mobility in an undoped GeS crystal

According to the theory, the centers formed as a result of the introduction of an impurity atom into a crystal, especially a layered crystal, can cause thermal quenching of photoconductivity, thermal activation and quenching of the photoluminescence process. As shown in Fig. 6, the effects of thermal activation and thermal quenching of photoconductivity were observed in the dependence $\sigma_f \sim f(1/T)$.

CONCLUSIONS

The obtained results show that the activation energy resulting from the stoichiometric disorder in layered crystals, depending on the ratio of components, plays the role of the center of generation and recombination of the majority charge carriers, the defect level of which is 0.27 eV in the low-temperature region and 0.41 eV in the high-temperature region, depending on the ratio of components. The concentration of these centers varies with temperature and illumination intensity in undoped GeS crystals. The results obtained in a comprehensive study of the electric, photoelectric, and thermally stimulating current in an undoped layered GeS crystal show that the defects resulting from the stoichiometric disorder caused by excess sulfur in the crystal lattice of the orthorhombic GeS structure create the possibility of targeted control of electronic processes, i.e. electrical and photoelectric properties. This, as shown in the scientific literature, enables the practical application of the layered GeS crystal, particularly for the creation of highly efficient photo- and thermal converters.

ORCID

©Y.I. Aliyev, <https://orcid.org/0000-0001-8896-2180>

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ВПЛИВ СТЕХІОМЕТРИЧНИХ СПОТВОРЕНЬ НА ЕЛЕКТРИЧНІ ТА ФОТОЕЛЕКТРИЧНІ ВЛАСТИВОСТІ ШАРУВАТИХ КРИСТАЛІВ GeS

Р.С. Мадатов¹, А.С. Алекперов^{2,3}, В.А. Абдурахманова², Р.К. Гусейнов⁴, Р.Дж. Баширов⁵, Ю.І. Алієв^{2,6*}

¹Інститут радіаційних проблем, Міністерство науки та освіти Азербайджанської Республіки, Баку, AZ-1143, Азербайджан

²Азербайджанський державний педагогічний університет, Баку, AZ-1000, Азербайджан

³Бакинський інженерний університет, Хирдалан, AZ-0101, Азербайджан

⁴Гянджінський державний університет, Гянджа, AZ-2000, Азербайджан

⁵Азербайджанський технічний університет, Баку, AZ-1073, Азербайджан

⁶Західно-Каспійський університет, Баку, AZ-1001, Азербайджан

Вальт-амперна характеристика, електропровідність, термостимульований струм та фотоелектричні властивості шаруватого кристала GeS з надлишком сірки досліджувалися в умовах зовнішнього електричного поля 10-104 В/см та при температурах 100-300 К. Було виявлено, що дефекти донорного типу утворюються в результаті стехіометричного спотворення через надлишок сірки в кристалі GeS, отриманому методом Бріджмена, що призводить до домішкової провідності. Перенесення заряду відбувається за допомогою монополярного струму інжекції, обмеженого об'ємом області заряду. Було виявлено, що термічна активація фотоструму та термічне гасіння фотоструму в легованих кристалах GeS пов'язані з електронним обміном між пастками захоплення та центрами рекомбінації.

Ключові слова: шаруватий кристал; електропровідність; монополярна інжекція; термостимульований струм; фотопровідність