TRANSPORT PHENOMENA AND DIELECTRIC PROPERTIES OF MULTICOMPONENT $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ (x = 0 – 1.0)

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The dielectric properties of the prepared single-crystal samples of solid solutions $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ were studied in alternating electric fields with a frequency of $f = 5 \times 10^4$–3.5×10^7 Hz. The relaxation character of the complex permittivity, the nature of dielectric losses, and the hopping mechanism of charge transfer in the $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ samples were established. It was shown that in $(TlGaSe_2)_{1-x}(TlGaS_2)_x$, with increasing x, the conductivity, average distance, and hopping time of charge carriers over the forbidden band decrease, while the energy spread of states localized near the Fermi level and their concentration increase.

Keywords: transport phenomena, $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions, alternating electric fields, frequency, complex permittivity, dielectric losses, charge transfer, localized states.
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Compounds of the $TlGaX_2$ (X = S, Se, Te) type [1-5] and solid solutions based on them $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ [6,7] are promising for use as active elements of semiconductor and optoelectronic devices in the visible range. The direct and indirect band gaps (E_g) of $TlGaSe_2$ are 2.08 and 1.93 eV, respectively [8]. For $TlGaSe_2$, the following data for E_g are given in [9]: 1.95 eV for the indirect band gap and 2.11 eV for the direct band gap. E_g of the $TlGaS_2$ compound at 10 K is 2.62 eV [10], and the optical indirect and direct band gaps are 2.45 and 2.63 eV. $TlGaSe_2$ and $TlGaS_2$ crystals have a layered structure and are characterized by anisotropy of properties. $TlGaSe_2$ and $TlGaS_2$ crystals can be fabricated as two-dimensional (2D) layered structures due to weak van der Waals forces between their adjacent monolayers. $TlGaSe_2$ and $TlGaS_2$ are p-type semiconductors. Studies of crystals of solid solutions $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ have shown that they are characterized by a noticeable change in physical properties depending on the composition [6,7] and an intense photoluminescence spectrum in the visible range for all compositions.

The purpose of this work is to study the influence of the composition of the obtained solid solutions $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ (x = 0 – 1.0) on their dielectric characteristics and to establish the mechanism of charge transfer in them in alternating electric fields of the radio frequency range.

Samples $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ were synthesized from elements taken in stoichiometric ratios by directly alloying them in quartz ampoules evacuated to 10^{-3} Pa. High-quality single crystals were obtained from synthesized $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ by the Bridgman method.

The dielectric coefficients of the $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ samples were measured by the resonance method. The frequency range of the alternating electric field was 5×10^4–3.5×10^7 Hz. Samples of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ for electrical measurements were made in the form of flat capacitors. Silver paste was used as electrodes. Dielectric properties were measured in the direction perpendicular to the layers of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals. All dielectric measurements were carried out at 298 K.

The frequency dispersion of the real and imaginary components of the complex dielectric constant of samples $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ in the frequency range $f = 5 \times 10^4$–3.5×10^7 Hz was revealed (Figures 1 and 2).

An increase in x in $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals led to a significant decrease in the real and imaginary components of the complex dielectric permittivity.

The frequency dependences of the dielectric loss tangent (tanδ) in $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions were characterized by a hyperbolic drop, indicating losses in through conductivity. An increase in the concentration of x in the $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals led to a decrease in tanδ.

We also studied the frequency-dependent conductivity (σ_{ac}) of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions (Figure 3). The value of σ_{ac} for $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions decreased by approximately two orders of magnitude as x increased from 0 to 1.0. In all the solid solutions studied, the frequency dependence $\sigma_{ac}(f)$ exhibited power-law regions $\sigma_{ac} \sim f^n$. In $TlGaSe_2$, initially there was a dependence $\sigma_{ac} \sim f^{0.8}$, which then became flatter. In $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals at $x > 0$, the sublinear sections $\sigma_{ac} \sim f^{0.8}$ were replaced by superlinear sections ($n > 1$).

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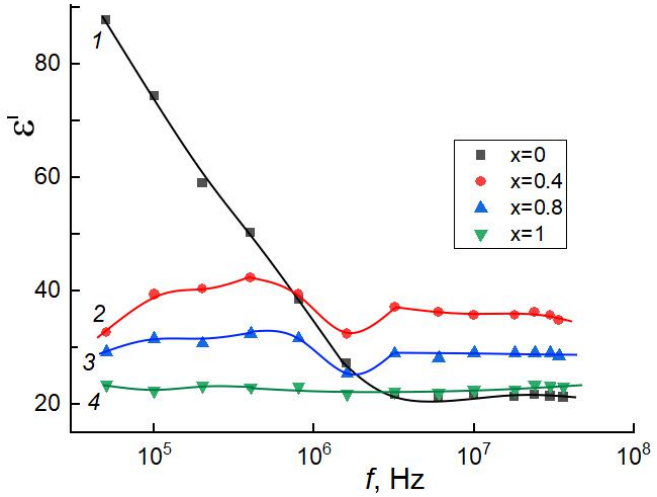

Fig. 1. Frequency dependences of the real part of complex dielectric permittivity of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals at various x : 0 (*1*); 0.4 (*2*); 0.8 (3) and 1.0 (*4*). T = 298 K.

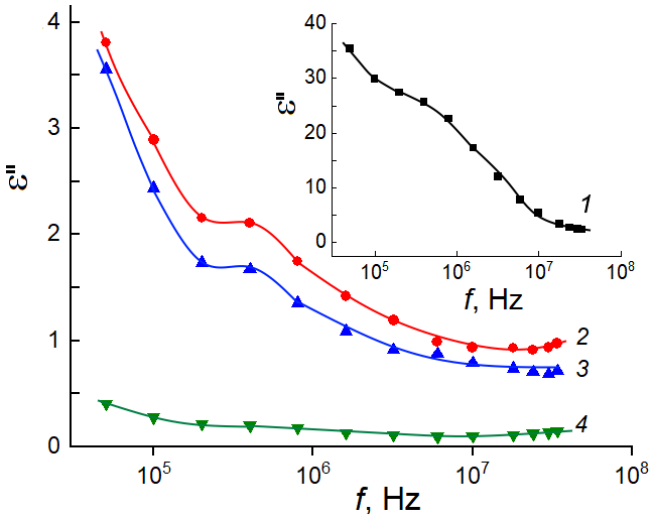

Fig. 2. Frequency dependences of the imaginary part of complex dielectric permittivity of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ crystals at various x : 0 (*1*); 0.4 (*2*); 0.8 (3) and 1.0 (*4*). T = 298 K.

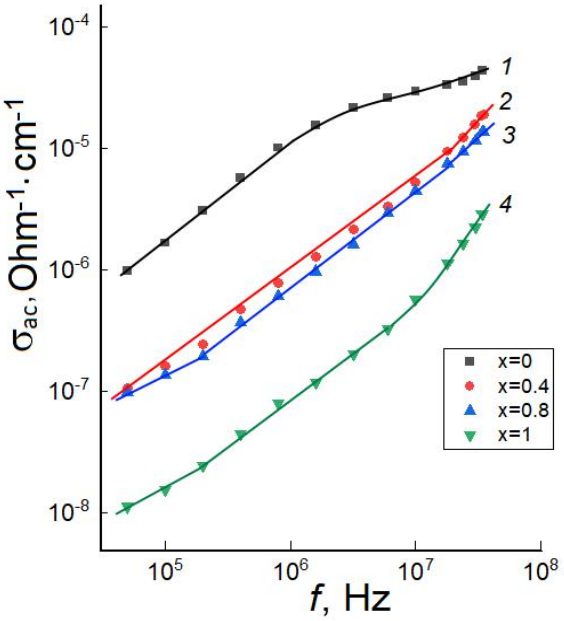

Fig. 3. Frequency-dependent conductivity of $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions at various x : 0 (*1*); 0.4 (*2*); 0.8 (3) and 1.0 (*4*). T = 298 K.

The sublinear (power-law) frequency dependence of conductivity $\sigma_{ac} \sim f^{0.8}$ indicates the hopping nature of transport. In this case, such a dependence is usually associated with jumps of charge carriers across localized states with the participation of phonons. This is the so-called relaxation hopping conduction.

The real part of conductivity at relatively low frequencies is determined by the phonon mechanism, and with increasing frequency, phononless hopping conductivity becomes predominant. This conductivity is characterized by a superlinear dependence $\sigma_{ac} \sim f^{n}$, where $n > 1$. In view of the above, the law we obtained $\sigma_{ac} \sim f^{0.8}$ indicates a hopping mechanism of charge transfer across states localized in the vicinity of the Fermi level [11].

The parameters of localized states in $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions have been calculated in terms of Mott theory for hopping conductivity: the density of states near the Fermi level $N_F = 2.2 \cdot 10^{18}$–$1.7 \cdot 10^{19}$ $eV^{-1}cm^{-3}$ and their energy spread $\Delta E = 0.005$–0.15 eV, average time $\tau = 9.9 \cdot 10^{-8}$–$1.2 \cdot 10^{-6}$ s and distance $R = 81$–240 Å of jumps.

An increase in x from 0 to 1.0 in $(TlGaSe_2)_{1-x}(TlGaS_2)_x$ solid solutions led to a decrease in the average values of the time and length of jumps, as well as to an increase in the energy spread of states localized near the Fermi level and the concentration of deep traps responsible for the transfer of charge carriers in alternating electric fields of the radio frequency range.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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