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CHARGE TRANSFER AND EXTERNAL FACTOR-DEPENDENT CHARACTERISTICS OF 2D MULTICOMPONENT CHALCOGENIDE MATERIALS

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Abstract- There have been developed synthesis regimes and growth technology of layer-chain single crystals of $A^{III}B^{III}C_2^{VI}$ type and solid solutions on their base. Physical properties of obtained low-dimensional semiconductors and influence of different external factors on them have been investigated. Effective detectors of visible and IR-radiation, neutron detectors, tenzo-and piezo-photo-resistors are proposed on the base of obtained semiconductors.

Keywords: Single Crystal, Solid Solution, Semiconductor, Physical Properties, Roentgendetector.

I. INTRODUCTION

Search of new semiconductor crystal materials using in electron technique is one of the main problems of crystal physics. In the course of many years we carry out the works on the development of synthesis regime and growth technology of layered-chain single crystals of $A^{III}B^{III}C_2^{VI}$ (A–Tl, B-In, Ga, Ag, Cu, Co, Cr, Mn, Ni, Fe, Sb, REE; C–S, Se, Te) type. There have been developed processing and crystallochemical aspects of synthesis and growth of low-dimensional chalcogenides of III B subgroup elements, REE and transition metals, and also solid solitions on their base [1-10]. There also have been developed optimum regimes of Li ions intercalation of layered-chain semiconductors of $A^{III}B^{III}C_2^{VI}$ type by pulling electric field from especially developed electrolyte [11, 12].

The aim of this work was investigation of influence of particular substitution of metal (Tl, Ga) atoms by Ag, Cu, Co, Cr, Mn, Ni, Fe, Sb, REE in grown $A^{III}B^{III}C_2^{VI}$ single crystals on electric, dielectric, photoelectric, optical and thermal properties of studied specimens.

II. EXPERIMENTAL RESULTS

We carry out the study of physical properties (electric, photoelectric, optical, thermal, roentgen dosimetry) of obtained low-dimensional semiconductors under the effect of different external factors (ionizing radiation, DC- and AC-electric and magnetic fields, temperature and pressure). The following results have been obtained.

By investigation of intercalation, temperature and deformation influense on exciton absorption spectra of single crystals of $A^{III}B^{III}C_2^{VI}$ type it is established that temperature dependence of exsiton absorbtion band maximum has a complex character. In particular, for $TlGaS_2$ single crystal the temperature coefficient of energetic position of the ground state of direct exciton has a positive sign as for $TlGaSe_2$ single crystal which has a negative one.

In this case temperature and baric shift coefficients are undergone to abrupt changes at structural phase reconstructions, characteristic of crystals of this type. Li intercalation of single crystals $TlGaSe_2$ leads to shift of exciton peak E_{EX}^{II} in the direction of small energies in all temperature range under the investigation; coefficient of temperature shift is also decreased in absolute value, besides at temperatures $T \leq 20$ K and $90K \leq T \leq 110$ K this change takes place abruptly. The second low-energy band of exciton absorption E_{EX}^{II} observed at $T \leq 120$ K shifts in the direction of high energies, and values $\partial E_{EX}^{II} / \partial T$ are undergone to abrupt changes at temperatures about 90, 50 and 20 K.

It was shown that intercalation of $TlGaSe_2$ single crystals with Li allow to control their optical properties. The structural and magnetic characteristics of the $(TlInS_2)_{1-x}(TlFeSe_2)_x$ crystals $(0 \leq x \leq 0.015)$ are investigated. It's shown that a continuous series of solid solutions are formed in this system. It is found that the unit cell parameters of these compounds increase with increasing $FeSe_2$ concentration. The abrupt change of the specific magnetization value is discovered on the temperature dependences of the $(TlInS_2)_{0.995}(TlFeSe_2)_{0.005}$ solid solution magnetization in the fields with strength of less than 1T, that is probably associated with the canted magnetic structure formation bellow 115 K. The growth of the $FeSe_2$ concentration in the studied solid solitions leads to the formation of a superparamagnetic state in the diamagnetic matrix.

Analysis of investigation results of influence of solid solutions composition $(TlInS_2)_{1-x}(TlFeSe_2)_x$ $(0 \leq x \leq 0.015)$

on their physical properties showed that as increasing x the widh of forbidden gap and energy of exciton peak are linearly increased, roentgencondutivity rises 2–3 times, and the temperatures of phase transitions shift in the direction of low temperatures.

Absorption spectra of exciton luminescence and combination scaterring of light in $TlGaS_2$ single crystals at 1.8 K are investigated. Multiband photoluminescence in 2.48–2.54 eV due to irradiation recombination of non-direct excition with phonon emission is revealed. Energy positions of direct (2.606 eV) and non-direct (2.540 eV) exsitions and their energies are found.

As a result of investigation of λ-modulation photoelectric spectra of $TlGa_{1-x}Fe_xS_2$ single crystals in the region of exsiton absorption it was established that Ga→Fe (1–2 at%) substitution leads to significant shift of exsiton band; at increasing consentration of Fe exciton peak at the absorption edge is disappeare. The latters is connected with strong change of interlayer bounds in single crystals.

Analysis of study results of influence of partial substitution of Ga atoms by Fe in $TlGa_{1-x}Fe_xS_2$ single crystals (x = 0–0.01) on their physical properties showed that as increasing x long-wave edge of photoconductivity is displaced to longer waves from 1.5 to 1.1 ev (from 826 nm to 1127 nm); phonon spectra in freguency range 250–350 cm^{-1} are recontructed and temperature of phase transition is shift to the side of low temperature (from 200 to 185 K).

The influence of particular substitution of metal (Tl, Ga) atoms by Ni in grown $TlGaS_2$ single crystals on dielectric properties and conductivity of specimens measured at alternate electric fields have been studied. This study presents the results of studying the frequency dependence of real (ε') and imaginary (ε'') components of the complex dielectric permittivity, loss tangent ($\tan\delta$), ac-conductivity across the layers of obtained $TlGaS_2$<Ni> (1 mol.% Ni) samples at frequencies $f = 5\times10^4$–3.5×10^7 Hz. The results demonstrate that the dielectric dispersion in the studied $TlGaS_2$<Ni> single crystals has a relaxation nature (Figure 1).

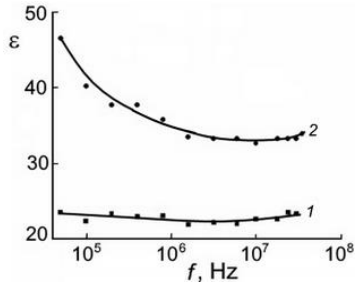

Figure 1. Dispersion curves of dielectric permittivity for $TlGaS_2$ (1) and $TlGaS_2$<1 mol.% Ni > (2).

The particular substitution of metal (Tl, Ga) atoms by Ni in $TlGaS_2$ single crystal leads to modification of $\varepsilon'(f)$ and $\varepsilon''(f)$ dispersion curves. At $f = 5\times10^4$–3×10^7 Hz $\tan\delta$ descends hyperbolically with an increase in frequency, which indicates conductivity losses in $TlGaS_2$<Ni>. The experimental dependence $\sigma_{ac} \sim f^{0.8}$ that we observed at $f = 2\times10^5$–10^7 Hz indicates that it is conditioned by hops of charge carriers between the states localized in the forbidden band of $TlGaS_2$<Ni>. We calculated the density of states at the Fermi level for $TlGaS_2$<Ni> single crystals: $N_F = 1.1\times10^{19}$ $eV^{-1}\cdot cm^{-3}$, the scattering of trap states near the Fermi level $\Delta E = 6.6\times10^{-2}$ eV, the average time of charge carrier hopping from one localized state to another τ = 0.2 microseconds, the average hopping distance R = 86 Å.

The results of studying the frequency dependence of real and imaginary components of the complex dielectric permittivity, loss tangent, ac-conductivity across the layers of $(TlGaSe_2)_{1-x}(TlInS_2)_x$ (x = 0; 0.2; 0.4 and 1.0) solid solutions at frequencies from 50 kHz up to 35 MHz have been presented. Samples from $(TlGaSe_2)_{1-x}(TlInS_2)_x$ solid solutions were made in sandwich form with electrodes of silver paste. The thickness of the crystal samples was 0.02–0.3 cm. All measurements were performed at 300 K by the resonance method.

The results demonstrate that the dielectric dispersion in the studied crystals has a relaxation nature. It was shown that increasing the $TlInS_2$ content of the $(TlGaSe_2)_{1-x}(TlInS_2)_x$ solid solutions decreases real and imaginary parts of their complex dielectric permittivity, dielectric loss tangent and conductivity. At x = 0.2; 0.4 and 1.0 $\tan\delta$ descends hyperbolically with an increase in frequency, which indicates conductivity losses.

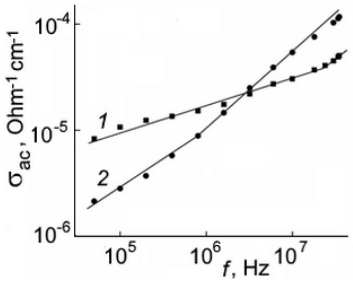

Figure 2. Frequency dependence of ac-conductivity for $TlInSe_2$ (1) and $TlIn_{0.995}Sb_{0.005}Se_2$ (2) at T = 300 K

We measured also the frequency dependence of the *ac*- conductivity of $(TlGaSe_2)_{1-x}(TlInS_2)_x$ (x = 0; 0.2; 0.4 and 1.0) solid solutions. Calculated values of N_F for these crystals were equal to 1.5×10^{18}–7.5×10^{18} $eV^{-1}\cdot cm^{-3}$. Evaluated values for the average time τ of charge carrier hopping from one localized state to another were equal to 2×10^{-7}–1.2×10^{-6} s. Calculated values for the average hopping distance R for studied crystals were equal to 86–240 Å.

From the experimental dependence $\sigma_{ac} \sim f^{0.8}$ for $TlIn_{0.995}Sb_{0.005}Se_2$ crystals (Figure 2) calculated value of N_F was equal to 1.8×10^{18} $eV^{-1}\cdot cm^{-3}$. Evaluated value for the average time τ of charge carrier hopping from one localized state to another was equal to 5.6×10^{-8} s. Calculated value for the average hopping distance R for studied single crystals was equal to 319 Å. We estimated scattering of trap states near the Fermi level: ΔE = 8 meV for $TlIn_{0.995}Sb_{0.005}Se_2$ crystals and determined the concentration of deep traps in these crystals: $N_t = 1.4\times10^{16}$ cm^{-3}.

Doping the $TlInSe_2$ single crystals with antimony (x = 0.005 Sb) was found to modify the dispersion curves $\tan\delta(f)$, $\varepsilon'(f)$ and $\varepsilon''(f)$ (Figures 3 and 4).

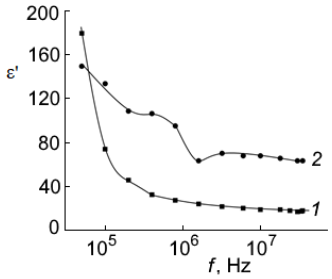

Figure 3. Frequency dependences of real part of complex dielectric permittivity for $TlInSe_2$ (1) and $TlIn_{0.995}Sb_{0.005}Se_2$ (2)

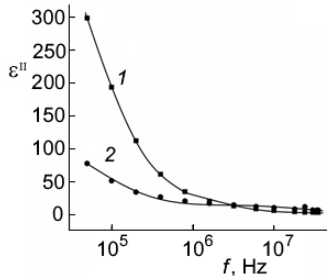

Figure 4. Frequency dependences of imaginary part of complex dielectric permittivity for $TlInSe_2$ (1) and $TlIn_{0.995}Sb_{0.005}Se_2$ (2)

The frequency dependence of the complex dielectric permittivity, loss tangent and ac-conductivity across the layers of $TlGa_{0.999}Sb_{0.001}S_2$ single crystals have been studied at frequencies from 50 kHz up to 35 MHz. The results demonstrate that the dielectric dispersion in the studied crystals has a relaxation nature. It is seen from measurements that $\tan\delta$ descends hyperbolically with an increase in frequency, which indicates conductivity losses (Figure 5).

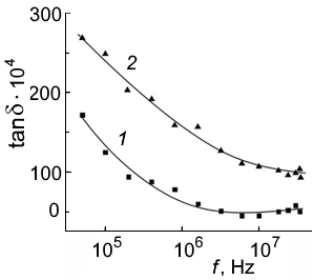

Figure 5. Loss tangent in $TlGaS_2$ (1) and $TlGa_{0.999}Sb_{0.001}S_2$ (2) vs frequency

For $Tl_{0.98}Ag_{0.02}GaS_2$ crystals calculated value of N_F was equal to 1.3×10^{19} $eV^{-1}\cdot cm^{-3}$. Evaluated value for the average time τ of charge carrier hopping from one localized state to another was equal to 4.4×10^{-8} s. Calculated value for the average hopping distance R for studied single crystals was equal to 76 Å. We estimated scattering of trap states near the Fermi level: ΔE = 84 meV for $Tl_{0.98}Ag_{0.02}GaS_2$ crystals and determined the concentration of deep traps in these crystals: $N_t = 1.1\times10^{18}$ cm^{-3}.

There have been investigated roentgenconductivity and roentgendosimetric characteristics of single crystals A^3B^6, $A^3B^3C^6_2$-type and their solid solitions $[(TlGaSe_2)_{1-x}(TlInSe_2)_x$; $(TlInS_2)_{1-x}(TlFeSe_2)_x$; $TlGa_{1-x}Fe_xS_2]$.

Prospectives of their practial use as active elements for roentgen radiation detectors are revealed [13]. On the base of experimental measurements it is revealed that these single crystals showed high roentgensensitivity at all fixed values of accelerating potential in 25–50 keV and dose rates from 0.78 to 78 R/min. Dependence of roentgencurrent on intensity of dose (E) has follow character:

$$\Delta I_{E,0} = I_E - I_0 \sim E^{\alpha} \quad (1)$$

where, α varies from 0.5 to 2.0.

We measured the spectral dependences of photoconductivity and photosensitivity R_d/R_{ph} (R_d is the dark resistance, and R_{ph} is the resistance of the sample under above-gap illumination) at a steady illumination, as well as the roentgen sensitivity and other photoelectric parameters. Table gives the photoelectric and roentgen dosimetry characteristics of the obtained $(TlGaS_2)_{1-x}(TlInSe_2)_x$ solid solutions.

From Table 1 we can see that the photosensitivity maximum (λ_{max}) linearly shifts from 0.50 to 0.73 μm as x increases from 0 to 0.5. This shift is associated with a decrease in the band gap with increasing x. Increasing x leads to a redshift of the sensitivity range $\Delta\lambda$ and a considerable rise in R_d/R_{ph} at 200 lx. For example, the R_d/R_{ph} of $(TlGaS_2)_{0.5}(TlInSe_2)_{0.5}$ is 5 to 6 times greater than that of pure $TlGaS_2$ (Table 1). The rise in R_d/R_{ph} with increasing x is apparently related to an increase in both the lifetime and mobility of the photogenerated carriers. Roentgen sensitivity K_σ of $(TlGaS_2)_{1-x}(TlInSe_2)_x$ was characterized by relative change in conductivity per unit dose rate,

$$K_\sigma = \frac{\Delta\sigma_{E,0}}{\sigma_0 \cdot E} \quad (2)$$

where, σ_0 is the conductivity of the nonirradiated crystal and $\Delta\sigma_{E,0} = \sigma_E - \sigma_0$ is the change in the conductivity under X-ray irradiation with dose rate E (R/min). Table 1 lists K_σ values at accelerating voltages from 25 to 30 keV and dose rates from 0.75 to 10 R/min. One can see that the K_σ of $(TlGaS_2)_{1-x}(TlInSe_2)_x$ solid solutions exceeds that of pure $TlGaS_2$. As the $TlInSe_2$ content increases, K_σ rises to 0.142–0.252 min/R at x = 0.5.

There have been carried out experimental investigations of hard radiation influence on photoelectric properties of three-component semiconductors $A^{III}B^{III}C_2^{VI}$ ($TlGaSe_2$, $TlInSe_2$) and photodiodes on their base. In particular we investigated influence of preliminary gamma (sourse is cobalt-60) (10^4–10^8 R), impulse neutron with energy $E \geq 0.1$ Mev ($F = 10^{12}$–10^{14} n/cm^2) and electron with energies 6 and 25 MeV ($\Phi=10^{13}$–10^{16} e/cm^2) irradiations on spectral characteristics and integral photosensitivity structures on their base.

It is revealed that as a result of neutron irradiation up to 5×10^{13} n/cm^2 elements on $TlGaSe_2$ -based photosensivity at a maximum of spectral characteristics (0.56–0.58 mkm) does not change practically, slight increase of photosensitivity is simulalataneously observed in impurity range up to 1.2 mkm. Over the range of shorter waves (0.34–0.55 mkm) the change of spectral photosensitivity due to mentioned neutron irradiation is resiprocal in its behaviour. The value of integral photosensitivity of single crystals $TlGaSe_2$ under the effect of neutron (up to 5×10^{13} n/cm^2) and qamma radiation (up to 10^8 R) at other equal conditions depends on value of applied voltage.

This study presents also results of studying the frequency dependence of complex dielectric permittivity and AC-conductivity of high-resistive $TlInS_2$ single crystals at frequencies from 50 kHz up to 35 MHz and the effect on them electron-irradiation.

Samples from $TlInS_2$ were made in sandwich form with electrodes of silver paste. The thickness of the single-crystal samples of $TlInS_2$ was ~200 microns. All measurements were performed at 300 K by the resonance method.

Electron-irradiation of the samples was carried out on the linear accelerator. The energy of electron-irradiation was 4 MeV. Measurements of samples were carried out after each irradiation.

It is shown that electron-irradiation of $TlInS_2$ single crystal with doses of 2×10^{12}–2.4×10^{13} e/cm^2 resulted in a decrease in the real component of the complex dielectric permittivity and tangible increase in its imaginary part, loss tangent and ac-conductivity across the layers at relatively low frequencies. At high frequencies, however, some noticeable changes of ε'', $\tan\delta$ and σ_{ac} due to electron-irradiation were not observed.

The experimental frequency dependence of the dissipation factor $\tan\delta$ for $TlInS_2$ single crystal at f = 50 kHz – 35 MHz is characterized with a monotonic descending before and after electron-irradiation. The hyperbolic decrease of $\tan\delta$ with frequency is evidence of the fact, that conductivity loss becomes the main dielectric loss mechanism in the $TlInS_2$ single crystal at studied frequency range. The investigation of the frequency dependences of ac-conductivity of the electron-irradiated $TlInS_2$ single crystal made it possible to elucidate the hopping charge-transfer mechanism.

It is established that due to Li intercalation of $A^{III}B^{III}C_2^{VI}$ single crystals [11, 12] one can control electric conductivity along C-direction of crystal by increasing anisotropy degree. Introduction of Li ions in interlayer and interchain spaces of these crystals favours increasing photosensitivity, roentgenosensitivity, and also extends the range of sensibility of mentioned crystals. Intecalation by Li leads to delay of photocurrent kinetics in crystals, and also to the formation in samples of enternal electric field controlled by different wave lengths of visible range.

Table 1. Photoelectric and roentgen dosimetry characteristics of the $(TlGaS_2)_{1-x}(TlInSe_2)_x$ solid solutions

Solid solution composition	$\Delta\lambda_{max}$, μm	R_d, Ohm	R_d/R_{ph} at 200 lx	K_σ, min/R
$TlGaS_2$	0.46-0.57	$(3\text{-}5)\times10^{10}$	5-8	0.063-0.159
$(TlGaS_2)_{0.9}(TlInSe_2)_{0.1}$	0.50-0.62	$(1\text{-}2)\times10^{10}$	10-25	0.075-0.178
$(TlGaS_2)_{0.8}(TlInSe_2)_{0.2}$	0.55-0.66	$(3\text{-}4)\times10^{9}$	15-30	0.089-0.198
$(TlGaS_2)_{0.7}(TlInSe_2)_{0.3}$	0.59-0.71	$(2\text{-}3)\times10^{8}$	21-37	0.098-0.213
$(TlGaS_2)_{0.6}(TlInSe_2)_{0.4}$	0.64-0.76	$(1\text{-}2)\times10^{7}$	23-42	0.107-0.219
$(TlGaS_2)_{0.5}(TlInSe_2)_{0.5}$	0.69-0.81	$(3\text{-}5)\times10^{6}$	25-46	0.142-0.252

III. CONCLUCIONS

On the base of investigation of electrical, dielectric, photoelectric, optical and roentgendosimetric properties of obtained low-dimensional semiconductors of $A^{III}B^{III}C_2^{VI}$ and solid solutions on their base have been suggested in effective detectors of visible and IR-radiation, detectors of roentgen-and qamma-radiation, electron detectors, tenso-and piezo-photoresistors.

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BIOGRAPHIES

Solmaz Nariman Mustafaeva was born in Dalian, China, 1950. She graduated from Azerbaijan State University (Baku, Azerbaijan) in 1972 and received scientific degree of Candidate of Physical-Mathematical Sciences in 1978, and degree of Doctor of Physical-Mathematical Sciences, in 1990. She is the main scientific worker in Institute of Physics, Azerbaijan National Academy of Sciences, Baku, Azerbaijan. She has published 700 papers and 35 patents.

Salim M. Asadov was born in Baku, Azerbaijan, 1982. He graduated from Azerbaijan State Oil Academy (Baku, Azerbaijan) in 2003, Scientific degree of Doctor of Technical Sciences in 2008. Currently, he is working as Senior Researcher at Institute of Catalysis and Inorganic Chemistry, Azerbaijan National Academy of Sciences (Baku, Azerbaijan). He has 77 published papers and 3 patents.