

MONOPOLAR INJECTION CURRENTS IN THE LAYERS OF $\text{Ge}_x\text{As}_y\text{Te}_{100-x-y}$ SYSTEM

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The features of current passage mechanism through sandwich of Te- $\text{Ge}_x\text{As}_y\text{Te}_{100-x-y}$ -Al structure type are investigated. It is established that the currents limited by space charges which are formed by hole monopolar injection, form from tellurium at positive potential on electrode. Using the Lambert's classic theory of injection currents, it is established that the electricity transfer is controlled by small trap centers situated between valence band and Fermi level. The parameters of the given centers are defined: the depth of occurrence and concentration. It is shown that the current in third VAC region increases in the region of trap filling limit not by vertically according to Lambert theory for monoenergetic levels of hole traps, but by power law. The given feature is explained by Pool-Frenkel effect taking under consideration the transition of the distribution function from Boltzmann approximation up to Fermi-Dirac one caused by the change of Fermi level position. The critical values of electric field strength at which the thermo-field decrease of Coulomb barrier up to zero blocking to thermo-field free current carriers are evaluated and the electric strength value at transition from trap quadratic law to non-trap one is defined.

Keywords: volt-ampere characteristic (VAC), Pool-Frenkel effect, Poisson equation.

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1. INTRODUCTION

Last years the interest in chalcogenide glass-like semiconductors (ChGS) on the base of S, Se and Te elements in combination with appropriate elements from Mendeleev's table (Ge, As and etc.) has increased. This is caused by one hand by their unique properties, in particular, high transparency in IR spectrum, high value of refraction index, high optical nonlinearity and by other hand by technological process similarity necessary for applied tasks and also by possibility of unlimited doping and chemical composition variation causing to material obtaining with optimal parameters that allows us to widen the regions of their applications [1-4]. However, the achievements of applied goals require the material obtaining with predicted properties and optimal parameters necessary for concrete goals.

The many-component ChGS materials containing the elements differing by the valence electron number are the more suitable for the given goals. According to the rule $8-N$ [atom with N valence electrons ($N \geq 4$) forms $8-N$ bonds, i.e. has $8-N$ close neighbors], coordination numbers of Ge, As and chalcogene (S, Se and Te) atoms are equal to 4, 3 and 2 correspondingly. Such difference in coordination number values promotes to change of glass matrix structure changing the chemical composition, i.e. allows us to obtain the glass with both one-dimensional and two- and three-dimensional structures. Such change in amorphous matrix should be accompanied by the change of electron properties. The investigation results of physical property peculiarities of ChGS system Ge-As-Se and their correlation with local structure parameters in scales of local and average atomic orders (correlation length, quazi-period in the region of average order, packing coefficient, compactness, average value of atomic volume, average coordination number, quantity of

limits and cohesion energy and also the chemical bonds and their percent parts in the investigated materials) are presented in our works [5-6]. ChGS system marks out by parameters required for practical goals. In particular, it has the wide transparency window covering the wavelength interval 3-20 μm and high value of refraction index ($n \sim 3,5$ at 1,55 μm wavelength) [7-8]. The investigation of crystallization kinetics [9] is predicted by perspective of the given material with low Ge concentration (<10 at%) for the use in optical record with reversible phase change. The investigation of reversible switching of memory [10] show that the given material can be mainly used for the readout of memory applications.

Taking under consideration the above mentioned we can conclude that the complex investigations of both atomic and energy structure and also physical properties of ChGS system Ge-As-Te are the actual ones. The investigation of charge transport mechanism through structures of Te-- $\text{Ge}_x\text{As}_y\text{Te}_{100-x-y}$ --Al sandwich type is the aim of the present work.

2. EXPERIMENT TECHNIQUE AND SAMPLE PREPARATION

ChGS synthesis of $\text{Ge}_x\text{As}_y\text{Te}_{100-x-y}$ system is carried out in following sequence: the purest elementary substances in required atomic percent are filled in quartz ampoules and after air exhaust they are heated up to temperature ~ 900 °C during 3 hours up to pressure 10^{-4} millimeter of mercury, further they are endured for about 12 hours at this temperature. The synthesis is carried out in rotating stove and the cooling is carried out in the off mode stove with aim of the promotion of sample homogeneity. The films of the different width used in the investigations are obtained by thermal evaporation with velocity $0,2 \div 0,4$ $\mu\text{m}/\text{sec}$ on glass substrates in vacuum at pressure 10^{-4} millimeter of mercury. The following

ChGS compositions Ge_{8,33}As_{16,67}Te₇₅, Ge₁₀As₂₀Te₇₀, Ge_{8,33}As_{16,67}Te_{62,5}, Ge_{18,2}As_{18,2}Te_{63,6} and Ge₁₈As₁₇Te₅₅ are synthesized.

The density of obtained samples ρ is measured by Archimed principle with the use of liquid (water) and it is calculated using the following formula:

$$\rho = \left[\frac{w_0}{(w_0 - w_L)} \right] \rho_L$$

where w_0 and w_L are material height in air and liquid correspondingly. ρ_L is liquid density (water) at room temperature is 1gr/cm³. The measurement accuracy isn't less than ± 0.02 gr/cm³.

VAC of Te-Ge_xAs_yTe_{100-x-y}-Al structure is investigated on constant current at application of electric strength of both polarities. The plane- parallel electrodes from aluminum and tellurium are also marked by vacuum evaporation. This square is 3,2 m². The space charge limited current (SCLC) mode is observed at the application of positive potential to Te, i.e. the holes are the injected charge carriers. The current values are registered by device 64876 Picoammeter voltage source (Keithley).

3. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

VACs for ChGS of compositions Ge_{8,33}As_{16,67}Te₇₅, Ge₁₀As₂₀Te₇₀ and Ge_{8,33}As_{16,67}Te_{62,5} at room temperature are shown in fig.1. The other compositions have the similar VACs.

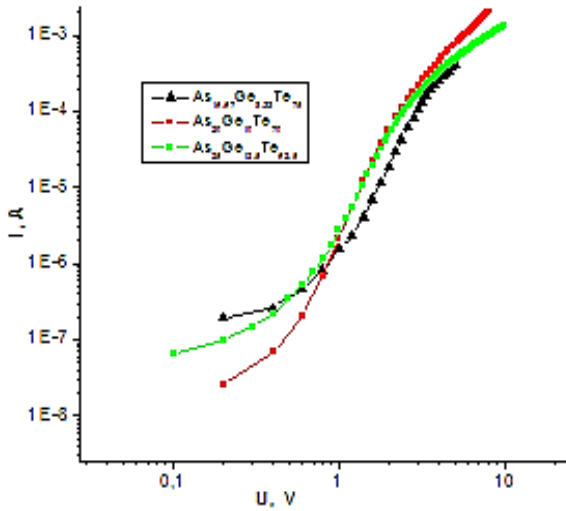


Fig. 1. VACs of ChGS compositions Ge_{8,33}As_{16,67}Te₇₅, Ge₁₀As₂₀Te₇₀ and Ge_{8,33}As_{16,67}Te_{62,5}.

As it is seen from the figure VACs of all compositions beginning from ohmic law with increasing voltage transit to the region corresponding to power law, i.e. $I \sim V^n$, where $n \approx 2$. Further, the section where $n > 2$ which transits into the region where $n \approx 2$ with increasing voltage. According to injection current theory [11], such VAC behavior proves the fact that the electric charge transfer in the given materials is carried by the monopolar injection

currents controlled by small trap capture centers for the holes situated below Fermi level. Using the classic theory of injection currents [11], some parameters controlling the electric charge transfer in wide interval of applied voltage are defined.

The values of specific resistance are calculated from VAC ohmic section and using them the concentrations of equilibrium free holes (table 1) are estimated according to formula $\rho = (ep_0\mu)^{-1}$, where e is elementary charge and μ is drift mobility of charge carriers. The mobility values are estimated using the component element mobility [12 -14] taking under the consideration their percent part in investigated ChGS compositions. Using these data Fermi level position in forbidden band ($F_0 - E_V$) is defined by following formula:

$$p = N_V \exp\left(-\frac{F_0 - E_V}{kT}\right) \quad (1)$$

where N_V is effective state density in valence band and for many chalcogenide glasses $N_V = 10^{20}$ cm⁻³ [15], kT is thermal energy.

In table 1 p_{01} is charge carrier concentration taking part in current passing in VAC section corresponding to Ohm law, $F_{01} - E_V$ corresponds to Fermi level position; p_{02} are the given concentration values in the beginning region of non-trap quadratic law and $F_{02} - E_V$ corresponds to Fermi quazi-level position.

Using the voltage values at which the current excursion begins (the region of Trap Filling Limit V_{TFL}) the concentration of attachment level which doesn't filled in thermodynamic equilibrium is defined by following formula [11]

$$p_{t,0} = 1,1 \cdot 10^6 \frac{\epsilon V_{TFL}}{L^2} \text{ cm}^{-3} \quad (2)$$

where ϵ is relative dielectric constant which is also estimated using ϵ of component elements [16] taking into consideration their percent part in investigated ChGS compositions. As in our investigations the quadratic section precedes to almost vertical section of the characteristics (fig.1), so we can accept that $p_{t,0} = N_t$ according to [11]. Energy position of trap centers is estimated by following formula [11].

$$E_t - E_V = kT \ln \frac{N_V}{\theta g N_t} \quad (3)$$

where θ is part of charge free carriers (holes) on all injected ones. Its values are defined as relation of current value corresponding to trap quadratic law to extrapolated current value from the section of non-trap quadratic law. The obtained values of concentration and energy position of trap centers are presented in table 1. The treatment of experimental results mentioned above, are carried out within the framework of Lambert theory [11] for injection currents in the solid states. According to theory began by Rous and continued by Lambert [11], the monopolar injection currents in the solid states (when one from the contacts is injecting one) have the following peculiarities: at low voltages of applied

field the charge transfer is carried out by thermal equilibrium carriers and obeys to Ohm law. The injected carrier concentration exceeds their equilibrium values with increasing voltage and Ohm law is exchanged by Mott-Gurni law [18], i.e. VAC obeys to law $I \sim V^2 L^{-3}$. Here V is applied voltage, L is sample thickness (the distance between plane-parallel electrodes). In the case of the samples the part of injected carriers is captured by traps and don't take part in current passing. That's why the transition from ohmic law into quadratic one takes place at relative high voltages. At further increase of applied voltage the total filling of trap centers should be observed and the vertical increase of current value obeys to Mott-Gurni law without traps. However, in many cases in particular, in our experiments, the current excursion takes place not vertically and by power law ($I \sim V^n$). This fact is explained by Lambert and Rouse by the fact that trap centers aren't monoenergetic ones and their densities are exponentially distributed on energies [11,17]. The alternative explanations of such type VAC behavior in TFL region are shown by authors of [18] with accepting of Gaussian distribution of defect states.

However, note that the experiments on investigation of injected currents are carried out in wide interval of applied field, moreover, VAC section at which the current excursion is observed, corresponds to electric field strength $10^3 \div 10^4$ V/cm. At such high values of applied electric field strength, Pool-Frenkel effect (PF) influences on mechanism of current passing [19]. The mechanism of the given effect is in thermo-field release of charge carriers from Coulomb traps existing in material volume amplified by electric field [20]. The author of [20] taking under consideration that only monoenergetic defect states exist in materials and $(E_F - E_V) \gg kT$ condition carries out, proposes the formulae expressing the dependences of conductivity and current density on applied electric field strength caused by PF effect.

$$\sigma = C \exp\left(-\frac{e\varphi - \beta\sqrt{\varepsilon}}{\zeta kT}\right) \quad (4)$$

$$J = C \varepsilon \exp\left(-\frac{e\varphi - \beta\sqrt{\varepsilon}}{\zeta kT}\right) \quad (5)$$

where σ is conductivity, J is current density, ε is electric field strength, β and C numerical parameters depending on material, k is Boltzman constant, ζ is inclination parameter of PF effect which varies between 1 and 2 in the dependence on concentration of acceptor traps. In work [19] this parameter is equal to 2.

The last equation can be written in the form:

$$\ln \frac{J}{\varepsilon} = \frac{\beta}{\zeta kT} \sqrt{\varepsilon} + \left[\ln C - \frac{e\varphi}{\zeta kT} \right] \quad (6)$$

The dependence $\ln \frac{J}{\varepsilon} \sim \sqrt{\varepsilon}$ in VAC region corresponding to current excursion is presented in fig. 2. Parameter β , the values of which are also presented in table 1, is defined from inclination of the given graph $\left(\frac{\beta}{\zeta kT}\right)$.

For the first time the authors of works [21, 22] developed the theory of monopolar injection currents using the classic model of PF effect. In classic PF model [23] it is proposed that firstly, the existing of the monoenergetic levels; secondly, $E_F - E_V \gg kT$, i.e. Boltzmann approximation is correct for energetic distribution of charge carriers in allowed bands.

VACs constructed according to model proposed in [21, 22] at low voltages well correspond to Lambert theory but at high electric field strengths the results strongly differ with both SCLC theory and experimental results. This is explained by the fact that Boltzmann approximation is accepted for both free and captured carriers. In modern PF models Boltzmann approximation is applied for free charge carriers and Fermi-Dirac distribution is used for captured carriers. Indeed, the current passing is carried out by thermal-equilibrium holes at low values of applied voltage and obey to Ohm law. Moreover, Fermi levels locate in the middle of forbidden band, i.e. far from trap center level. The use of Boltzmann distribution is reasonable one. The concentration of injected holes with increasing voltage strongly increases and position of Fermi quasi-level approximates to energetic position of trap centers (table 1). That's why it is more real to use Fermi-Dirac distribution. The observable VAC peculiarities at the use of classic and modern model, mainly reveal at high values of applied voltage. In first case the current increases as quadratic one with participation of small traps. And in second case the current strongly increases similar to TFL low as in SCLC. However, the non-trap quadratic law at this isn't observed that it should take place according to Lambert theory. According to [20] such disagreement is connected with inaccuracy at Poisson equation formation which has the following general form:

$$\frac{d\varepsilon}{dx} = \frac{\rho + \rho_t}{\varepsilon} \approx \frac{\rho_t}{\varepsilon} \quad (7)$$

Here \mathcal{E} is applied electric field strength, ρ and ρ_t are densities of free and captured charge carriers (holes), ε is dielectric constant of investigated ChGS compositions. In works [21, 22] at solving of Poisson equation ρ doesn't take under consideration, so $\rho \ll \rho_t$. Such approximation is correct at small voltages, i.e. when VAC region isn't reached, where TFL law is satisfied. That's why VAC in the given region agrees with Lambert theory. Upon reaching voltage V_{TFL} the current density values strongly increase, i.e. the transition in the region of non-trap quadratic law doesn't take place. The author of work [23] giving the refinements in Poisson equation taking under consideration both captured and free charges, obtains VAC consisting in three sections predicted by Lambert theory with the difference that the transition from trap quadratic law in non-trap one takes place not vertically, but by power law. The values of electric field strength, at which the height of Coulomb barrier decreases up to zero (saturation of electric field strength of PF effect is ε_s) and the transition from current excursion with into non-trap quadratic law

(ϵ_{TFL}) with increasing voltage takes place, are evaluated (table 1) by following formulae proposed by author of work [23]:

$$\epsilon_S = \left(\frac{e\varphi}{\beta}\right)^2 \quad (8)$$

$$\epsilon_{\text{TFL}} = \frac{eN_t L}{\epsilon} \quad (9)$$

where $e\varphi$ is height of a Coulomb barrier for the carriers captured by trap centers that is equal to their energy depth of occurrence, N_t is trap center concentration, ϵ is dielectric constant, L is film thickness. As it is seen from the table, ϵ_S values are almost an order greater than ϵ_{TFL} . Indeed, at ϵ_{TFL} all trap centers are filled by electrons according to classic theory of injected currents. And at ϵ_S the depletion of the given centers which still aren't fully captured by carriers takes place. That's why its value are less than required values of electric field strength (ϵ_{TFL}) for carrying out of the transition from the trap quadratic law to non-trap one. It is known that the defect states in chalcogenide glass-like semiconductors is connected with broken chemical bonds, nano-voids, with existence of many structural elements, relative quantities of homeopolar and heteropolar bonds and etc. By other words, they are connected with peculiarities of short-range order in atom disposition. For the establishment of the given position the densities of all investigated compositions are measured and such parameters of short-range order as packing coefficient [24] and R parameter defined by ratio of number of possible chalcogene atom covalent bonds to the number of non-chalcogenide one [25].

$$\delta = \frac{\sum_i (x_i A_i) / \rho_i - \sum_i (x_i A_i) / \rho}{\sum_i (x_i A_i) / \rho} \quad (10)$$

where ρ and ρ_i are densities of chemical compositions and elements including in ChGS composition.

$R = 1$ presents itself the case of stoichiometric composition consisting only from energy profitable heteropolar bonds. $R > 1$ values correspond to chemical compositions rich by tellurium (samples with numbers 1 and 2 in the table) in which homeopolar bonds Te-Te also take place along with existence of heteropolar bonds As-Te and Ge-Te. At $R < 1$ values the chemical compositions differ by deficiency of chalcogene atoms for total satisfaction of valence requirements of non-chalcogene atoms (samples 4 and 5, table). As it is seen from the table the least values of trap center concentrations correspond to ChGS composition $\text{As}_{25}\text{Ge}_{12.5}\text{Te}_{62.5}$ where $R=1$. According to principles of chemical bond methods (Chemical bond approaches (CBA) model) [7] the probability of heteropolar bond formation exceeds the probability of homeopolar bond one, moreover, firstly the strongest bonds are formed (bonds with high energies).

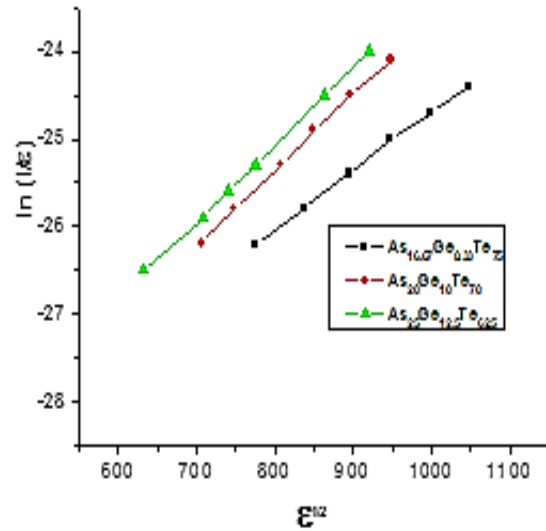


Fig. 2. $\ln \frac{j}{\epsilon} \sim \sqrt{\epsilon}$ dependence for ChGS compositions $\text{Ge}_{8.33}\text{As}_{16.67}\text{Te}_{75}$, $\text{Ge}_{10}\text{As}_{20}\text{Te}_{70}$ and $\text{Ge}_{8.33}\text{As}_{16.67}\text{Te}_{62.5}$ in VAC region corresponding to current excursion.

Table 1

The parameters of local states controlling the current passing through $\text{Al--As}_x\text{Ge}_y\text{Te}_{100-x-y}\text{--Te}$ structure.

	$\text{As}_{16.67}\text{Ge}_{8.33}\text{Te}_{75}$	$\text{As}_{20}\text{Ge}_{10}\text{Te}_{70}$	$\text{As}_{25}\text{Ge}_{12.5}\text{Te}_{62.5}$	$\text{As}_{18.2}\text{Ge}_{18.2}\text{Te}_{63.6}$	$\text{As}_{17}\text{Ge}_{28}\text{Te}_{55}$
R	1.8	1.4	1	0.998	0.675
δ	0.016	-0.152	-0.015	-0.003	-0.002
θ	$9 \cdot 10^{-3}$	$4 \cdot 10^{-3}$	$6 \cdot 10^{-3}$	$8 \cdot 10^{-3}$	10^{-2}
ϵ	23.18	22.32	21.03	21.44	20.21
μ , (cm ² /V·s)	29.78	32.34	36.18	38.51	46.22
p_{01} , cm ⁻³	$8.07 \cdot 10^8$	$2.93 \cdot 10^8$	$4.31 \cdot 10^8$	$2.14 \cdot 10^8$	$1.54 \cdot 10^8$
$F_{01}\text{-E}_v$, eV	0.661	0.687	0.677	0.695	0.704
p_0 , cm ⁻³	$2.9 \cdot 10^{15}$	$2.8 \cdot 10^{15}$	$2.6 \cdot 10^{15}$	$3.2 \cdot 10^{15}$	$3.9 \cdot 10^{15}$
$E_{t1}\text{-E}_v$, eV	0.374	0.397	0.388	0.375	0.364
p_{02} , cm ⁻³	$9 \cdot 10^{12}$	$13 \cdot 10^{12}$	$8.5 \cdot 10^{12}$	$4.72 \cdot 10^{12}$	$1.44 \cdot 10^{12}$
$F_{02}\text{-E}_v$, eV	0.420	0.410	0.421	0.436	0.467
β , C(m/V) ^{1/2}	$6 \cdot 10^{-23}$	$9 \cdot 10^{-23}$	$7 \cdot 10^{-23}$	$8 \cdot 10^{-23}$	$6.2 \cdot 10^{-23}$
ϵ_S , V/cm	$9.3 \cdot 10^3$	$5 \cdot 10^3$	$7.8 \cdot 10^3$	$5.6 \cdot 10^3$	$8.8 \cdot 10^3$
ϵ_{TFL} , V/cm	$4.52 \cdot 10^4$	$4.54 \cdot 10^4$	$4.47 \cdot 10^4$	$5.39 \cdot 10^4$	$6.97 \cdot 10^4$

Thus, amorphous matrix of the given composition is formed mainly from heteropolar bonds. From above mentioned we conclude that the matrix of the given composition is the more perfect.

The main principles of chemical bond method (Chemical bond approaches (CBA) model) [26] are used in this goal. According to CBA the probability of heteropolar bond formation exceeds the homeopolar one, moreover, firstly the strongest bonds form (bonds with high energies). The consistency of given bond formation corresponds to consistency of energy decrease until the achievable atom valence won't be satisfied. Finally, according to CBA the bond energy

is additive one. The supposed chemical formulae of the investigated compositions taking under consideration of the given principles are presented in table 1. In third composition the glass structure consists in totally matched tetrahedral and pyramidal structural units which have energy profitable heteropolar bonds. In 1st, 2nd and 4rd compositions along with the given element the homeopolar bonds between chalcogene atoms are also take part, i.e. the excess selenium atoms take part in bounding of given elements and also join between each other in chain and ring formation.

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