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Low-temperature anomalies and nanosized levels formation of self-organized structures in the non-crystalline solids of As(Ge)-S(Se) systems

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The relationship between low-temperature anomalies of physical and chemical properties and the formation of self-organized structures in non-crystalline solids of the As(Ge)-S(Se) systems is discussed. Obtained temperature dependences for the specific heat capacity and coefficient of linear expansion in the temperature domain $T < 100$ K, detailing the linear and nonlinear contributions. The influence of the obtaining conditions of non-crystalline solids on the low-temperature behavior of physico-chemical properties and the change in the ratios of various contributions and temperature intervals is considered. The correlation and common features of the formation of self-organized structures of non-crystalline materials in the region of low temperatures and softening temperatures, manifested in the presence of nanolevels of structuring, are analyzed.

Keywords: low-temperature anomalies, nanosized effects, non-crystalline materials, self-organized structures, synergetics, soft atomic configurations, two-level states.

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Introduction

Non-crystalline solids of systems As(Ge)-S(Se) are very interesting objects with wide practical and fundamental applications [1,2]. The fundamental researches using ideas of synergetics for the self-organizing processes and formation of self-organized structures in them, also is extremely also important and unique [2]. In the region of low temperatures ($T < 50$ K), the behavior of a number of thermal properties (the heat capacity, the coefficient of thermal expansion, the velocity of sound propagation) for non-crystalline solids is different from the temperature characteristics of the crystalline compounds. The low-temperature anomalies of physical properties for non-crystalline solids are described at present on the basis of representations of two-level tunnel states and soft atomic configurations without a single interpretation of their microscopic nature [3-5]. An interesting application of such phenomena is also the Bose & Einstein condensation, which is being studied intensively over the past decades [6]. Processes of

self-organization and a synergetic approach to low-temperature anomalies can cardinaly change the views and methods of implementation without dissipative dissemination of information through the realization of fractal structures [7].

In order to explain the nature of the low-temperature phenomena of non-crystalline materials of As(Ge)-S(Se,Te) systems in a wide temperature range, it is necessary to consider the behavior of physical-mechanical properties in conjunction with the features of the structure and factors that determine it. These factors include the conditions for obtaining non-crystalline materials and the effect of external fields (electromagnetic radiation, external noise). Non-crystalline materials are characterized by the presence of self-organized and fractal structures [2, 7-10], whose parameters, due to influence on the near and medium order, are largely determined by the conditions of obtaining. For example, such factors include the synthesis temperature, cooling rate, annealing regimes, which determine the spatial fluctuations of structural parameters. In addition, the self-consistent consideration

of the dynamics of atomic oscillations, the reorganization of local atomic potentials, and the spectrum of low-frequency excitations of non-crystalline materials in terms of conditions for their obtaining is of fundamental interest. The nature of the transition to a non-crystalline state as a self-organized process and low-temperature anomalies also have much in common [7, 11]. From this perspective, the study of the formation processes of orderly self-organized structures in non-crystalline solids continues to arouse constant interest in the synergetic approach and their practical application in the creation of non-volatile memory and technologies, the development of intelligent non-crystalline materials of artificial intelligence [7]. The above-mentioned approaches are discussed in this article.

I. Model of system

Let's consider the relationship between the behavior of physical properties (mean-square displacements and frequency of oscillations of atoms, heat capacity and thermal expansion) of non-crystalline solids in the region of low temperatures $T < 100$ K with the conditions for their obtaining. The fluctuations of the parameters of the non-crystalline structure, which are described at the near-order level by the dispersion of the interatomic distances and angles between the bonds, cause the local heterogeneity of the matrix and lead to the formation of regions with different levels of ordering. The Hamiltonian H of such a system can be given as follows

$$H = \sum_f \sum_l \left(\frac{(\bar{p}_l^f)^2}{2m} + U_f(\vec{r}_l) \right) \sigma_l^f + \frac{1}{2} \sum_{f,f'} \sum_{l,l'} \Phi_{f,f'}(\vec{r}_l - \vec{r}_{l'}) \sigma_l^f \sigma_{l'}^{f'}. \quad (1)$$

Here $U_f(\vec{r}_l) = \sum_{f,f'} \Phi_{f,f'}(\vec{r}_l - \vec{r}_{l'})$ is the single-particle potential, $\Phi_{f,f'}(\vec{r}_l - \vec{r}_{l'})$ is the paired interaction potential, σ_l^f are the local characteristic functions. Effective Hamiltonian \tilde{H} of two-level states ($f = 1, 2$) the system has the look:

$$\tilde{H} = \sum_l \tilde{H}_f, \tilde{H}_f = \sigma_f \sum_l \left(\frac{(\bar{p}_l^f)^2}{2m} + U_f(\vec{r}_l) - \mu \right) + \frac{A_f}{2} \sum_{l,l'} \Phi_f(\vec{r}_l - \vec{r}_{l'}). \quad (2)$$

Where $A_1 = 1 - \sigma_2^2$, $A_2 = \sigma_2^2$, $\sigma_1 + \sigma_2 = 1$. In the future, we will introduce the notation $\sigma \equiv \sigma_2$. Configuration entropy $dS_i = -k_B \ln \left[\prod_f \left\{ \frac{g_f^{j_f}}{N_f^{j_f} (g_f - N_f)^{j_f}} \right\} \right]$, associated with fractal regrouping the regions of soft-atomic configurations inside the system, is determined for this case by the number of distribution methods N_f atoms in f possible states with degrees of degeneracy g_f . Degree of degeneracy g_f takes into account the splitting of the corresponding energy level due to the influence of neighbors, and $g_f > N_f$. So, $g_2 > N_{2max}$ ($N_{2max} = (10^{-4} \div 10^{-2}) \cdot N$) (is the density of possible soft atomic configurations in non-crystalline material, which is determined by its structure and conditions of obtaining [11]) and $g_1 \geq N_{1max} \cong N$, which gives an estimation $g_2/g_1 = 10^{-4} \div 10^{-2}$. Since in the non-crystalline material the proportion of atoms contained in soft atomic configurations is significant ($\sigma \cong 10^{-4} \div 10^{-2}$), and the displacement of the atoms for the system near the order (10 ÷ 30)% the interatomic distances, then for the study of the temperature behavior

of the non-crystalline system, it is necessary to involve methods that take into account strongly non-harmonic effects and structural rearrangements. We use the self-consistent pseudo-harmonic approximation, which enables us to determine the renormalization of the force constants and local potentials as a result of the anharmonicity of atomic oscillations [11-14].

Let's consider the solution of the variational problem [11, 12] for the functional of energy

$$\begin{aligned} \partial F / \partial \sigma &= a_e \cdot (\sigma - \sigma_e), \\ F &= \sum_f F_f - T \cdot dS, \quad dS = dS_i + dS_e, \\ a_e &= (\partial^2 F / \partial \sigma_e)_e. \end{aligned} \quad (3)$$

Where $dS_i \geq 0$ is a flow of entropy within a non-crystalline system, $dS_e < 0$ is a flow of syentropy from the external environment [7], σ_e is an equilibrium value σ . The variational problem leads to self-consistent equations:

$$\sigma = \frac{\tilde{\lambda}_1 - \tilde{\lambda}_2 - \vartheta \ln \frac{g_2/N_2 - 1}{g_1/N_1 - 1} - a_e \cdot (\sigma - \sigma_e)}{2(\tilde{\Phi}_2 - \tilde{\Phi}_1)}, \tilde{\lambda}_f = \langle \sum_l \frac{(\bar{p}_l^f)^2}{2m} \rangle, \tilde{\Phi}_f = \langle \sum_{l,l'} \Phi_f(\vec{r}_l - \vec{r}_{l'}) \rangle. \quad (4)$$

The system of equations with respect to renormalized power constants $\tilde{v}_f(l)$ and mean-square displacements $\tilde{D}(l) = \langle \frac{[u(l) - u(0)]^2}{l^2} \rangle$ in the approximation of the pair interaction of near neighbors Z , it is possible to investigate the temperature behavior of the non-crystalline system and has the form

$$z \tilde{D}_f(l) = \frac{\hbar}{N} \sum_k \frac{\omega(k)}{\tilde{v}_f(l)} \text{cth} \frac{\hbar \omega(k)}{2\vartheta}, \quad \tilde{v}_f(l) = A_f \frac{\partial^2}{\partial r_l^2} \left[\exp \left\{ \frac{1}{2} \tilde{D}_f \frac{\partial^2}{\partial r_l^2} \right\} \Phi_f(r_l - r_{l'}) \right] \quad (5)$$

Describing the effective interaction of atoms with the Morse potential, we find for a self-consistent interaction

potential:

$$\tilde{\Psi}_f(l) = A_f V_{0f} \exp\{-Q^*\} \left[\exp\{-2\alpha(l - \alpha_0)\} \exp\{2y_f\} - 2 \exp\{-\alpha(l - \alpha_0)\} \exp\{y_f/2\} \right], \quad (6)$$

where $\alpha = \alpha_0 + 3Q^*/2\alpha$ is an averaged interatomic distance taking into account the static disordering of equilibrium positions characterized by a parameter $Q^* = \alpha^2 Q^2$; $y_f = \alpha^2 \tilde{D}_f(l)$ are given reduced mean-square displacements of atoms; α and V_{0f} are the parameters of the potential. Thus, according to the principle of the local quasi-equilibrium of non-crystalline material in the presence of external applied pressure P , the average inter atomic distance is equal

$$l_f = \alpha_0 + \frac{3y_f}{2\alpha} + \delta_f, \quad \delta_f = -\frac{P^* \exp(y_f)}{(3\alpha(1-\xi\sigma_f^2))},$$

$$\xi = 1 - \frac{V_{02} \exp\{y_1 - y_2\}}{V_{01}} \ll 1, \quad P^* = \frac{6P\alpha\alpha_0^2}{z_f}$$

is the reduced pressure, $f = 2\alpha V_{0f} \exp\{-Q^*\}$ is the power constant in a harmonic approximation, then the potential energy and force constants in the region of low temperatures ($\theta \ll k_B T_D$, where T_D is a Debye's temperature) are defined by expressions

$$\begin{aligned} \tilde{\Psi}_f(l_f) &= -A_f V_{0f} \exp\{-Q^*\} \exp\{-y_f\}, \\ \tilde{v}_f(l_f) &= A_f f \left(\exp\{-y_f\} + \frac{P^*}{1-\xi\sigma_f^2} \right) \\ y_f &= \frac{\beta_f}{A_f^2 \lambda_f} \vartheta^4 \left(1 + \frac{5\beta_f}{2A_f^2 \lambda_f} \vartheta^4 + \dots \right) \end{aligned} \quad (7)$$

where $\lambda_f = \frac{V_{0f} \alpha_0 (1+P^*) \cdot V \cdot \exp\{-Q^*\}}{\pi^2 \hbar v_{0f} S_{0f}}$, $\beta_f = \frac{\alpha^4}{60(\hbar S_{0f})^4}$, $S_{0f}^2 = z_f \alpha_0^2 (1 + \frac{P^*}{1-\xi\sigma_f^2}) / 2m$ is a sound speed in harmonic approximation, v_{0f} is a geometric structural factor ($0 < v_{0f} < 1$). The system of equations (4) - (7) allows us to investigate the behavior of the mean-square displacements, the atomic fraction in soft atomic configurations and a number of other physical properties that are determined through $y_f(T)$ and $\sigma_f(T)$ in the temperature range $T < 100^\circ K$. Because $A_f \tilde{\Phi}_f = zN \tilde{\Psi}_f(l)/2$, $\sigma_f \tilde{\Lambda}_f = zN \tilde{v}_f(l)/\alpha^2$, Then equation (4) using the relation (7) is rewritten in this way:

$$\sigma = \frac{V_{02} y_2 \sigma \left(\exp\{-y_2\} + \frac{P^*}{1-\xi\sigma^2} \right) - V_{01} y_1 (1+\sigma) \left(\exp\{-y_1\} + \frac{P^*}{1-\xi\sigma^2} \right) + \vartheta \ln \frac{\frac{\vartheta^2 - N}{\frac{\vartheta^2}{1-\sigma}} \exp\{Q^*\} + \alpha e^{-(\sigma - \sigma_e)}}{\frac{\vartheta^2}{1-\sigma}}}{V_{01} \exp\{-y_1\} - V_{02} \exp\{-y_2\}}. \quad (8)$$

In particular, taking into account (7), for $T < 10^\circ K$ we get

$$\sigma = \frac{(1+P^*)\vartheta^4}{2\varepsilon} \left(\frac{\beta_2 V_{02}}{\lambda_2 \sigma^3} - \frac{\beta_1 V_{01}}{\lambda_1 (1-\sigma^2)(1+\sigma^2)} \right). \quad (9)$$

From here we have $\sigma \cong \gamma\theta$, $\gamma^4 = \frac{\beta_2(1+P^*)V_{02}}{2\lambda_2\varepsilon} \ll 1$, $\varepsilon = V_{01} - V_{02}$.

Internal energy and volume

$$\tilde{E} = (1 - \sigma^2) \tilde{\Phi}_1 + \sigma^2 \tilde{\Phi}_2 + \sigma \tilde{\Lambda}_2 + (1 - \sigma) \tilde{\Lambda}_1, \quad V = N((1 - \sigma) \cdot v_1 + \sigma \cdot v_2), \quad v_f = v_{0f} l_f^3, \quad (10)$$

there are functions σ , $y_{1,2}$ and given this way:

$$\tilde{E} = \frac{z}{2} \exp\{-Q^*\} \times \left[-V_{01} e^{-y_1} + \sigma^2 (V_{01} e^{-y_1} - V_{02} e^{-y_1}) + V_{01} y_1 (1 - \sigma^2) \left(e^{-y_1} + \frac{P^*}{1-\xi\sigma^2} \right) + V_{02} y_2 \sigma^2 \left(e^{-y_2} + \frac{P^*}{1-\xi\sigma^2} \right) \right], \quad (11)$$

$$\frac{V}{N} = (1 - \sigma) v_{01} \left(\alpha + \frac{3y_1}{2\alpha} - \frac{P^* e^{y_1}}{3\alpha(1-\sigma^2)} \right) + \sigma v_{02} \left(\alpha + \frac{3y_2}{2\alpha} - \frac{P^* e^{y_2}}{3\alpha(1-\xi\sigma^2)} \right). \quad (12)$$

Thus, the low-temperature thermal capacity and the coefficient of thermal expansion of the system are described by the relations

$$\begin{aligned} C_p &= \frac{M\alpha}{m} \cdot \frac{\partial \left(\frac{\tilde{E} + PV}{N} \right)}{\partial T} \Bigg|_2 = \frac{z M \alpha}{2 m} \cdot e^{-Q^*} \cdot \\ &\left([V_{01} e^{-y_1} (1 - y_1) - V_{02} e^{-y_2} (1 - y_2)] - (V_{01} y_1 - (V_{02} y_2)) \frac{P^*}{1-\xi\sigma^2} \frac{\partial(\sigma^2)}{\partial T} + V_{01} e^{-y_1} (1 - \sigma^2) (2 - y_1) \frac{\partial y_1}{\partial T} + V_{02} e^{-y_2} \cdot \sigma^2 \cdot \right. \\ &\left. (2 - y_2) \frac{\partial y_2}{\partial T} + [V_{01} (1 - \sigma^2) \frac{\partial y_1}{\partial T} + V_{02} \sigma^2 \frac{\partial y_2}{\partial T}] \frac{P^*}{1-\xi\sigma^2} + P^* (v_2 - v_1) \frac{\partial \sigma}{\partial T} + P^* \frac{\partial v_1}{\partial T} + P^* \sigma \frac{\partial}{\partial T} (v_2 - v_1) \right), \end{aligned} \quad (13)$$

$$\alpha_T = \frac{1}{V} \left. \frac{\partial V}{\partial T} \right|_P = \frac{1}{V} \left[(v_2 - v_1) \frac{\partial \sigma}{\partial T} + \frac{\partial v_1}{\partial T} + \sigma \frac{\partial}{\partial T} (v_2 - v_1) \right].$$

Here M_α is molar mass. Using (11), (12) for E and V in the temperature range $T < 10^\circ K$ we will get:

$$\tilde{E} = \frac{z}{2} e^{-Q^*} \left[-V_{01} + \gamma^2 \vartheta^2 \left(\varepsilon + \frac{\beta_2 V_{02} (2+P^*)}{\lambda_2 \gamma^4} \right) + \frac{\beta_1 V_{01} \vartheta^4}{\lambda_1 (1-\gamma^2 \vartheta^2)^2} (2+P^* - \gamma \vartheta (1+P^* \gamma^2 \vartheta^2)) \right], \quad (14)$$

$$\frac{V}{N} = \alpha_0^3 \left(1 - \frac{P^*}{3\alpha\alpha_0(1-\xi\gamma^2\vartheta^2)} \right)^3 \times \left\{ v_{01}(1-\gamma\vartheta) \left[1 + \frac{\beta_1 \vartheta^4}{6\alpha\alpha_0\lambda_1(1-\gamma^2\vartheta^2)^2} \left(9 - \frac{2P^*}{(1-\xi\gamma^2\vartheta^2)} \right) \right]^3 + v_{02}\gamma\vartheta \left[1 + \frac{3\beta_2}{2\alpha\alpha_0\gamma^4\lambda_2} - \frac{\beta_1 P^* \vartheta^4}{6\alpha\alpha_0\lambda_1(1-\gamma^2\vartheta^2)^2(1-\xi\gamma^2\vartheta^2)} \right]^3 \right\}.$$

The corresponding thermal capacity and thermal expansion coefficient of the system will look like

$$C_P = a_1 T + a_2 T^2 + a_3 T^3, \quad a_T = -b_1 T + b_2 T^3, \quad (15)$$

Where

$$a_1 = z\gamma^2 k_B^2 \cdot \frac{M_\alpha}{m} \left\{ e^{-Q^*} \left[\varepsilon + \frac{\beta_2 V_{02} (2+P^*)}{\lambda_2 \gamma^4} \right] - \frac{2\xi v_{01} V_{01} P^*}{3} \right\},$$

$$a_2 = \frac{M_\alpha}{m} \cdot \frac{zf(v_{01}-v_{02})P^*\gamma^3 k_B^3}{3\alpha^3},$$

$$a_3 = 3z\beta_1 k_B^4 \frac{M_\alpha}{m} \left[\frac{V_{01}(2+P^*)e^{-Q^*}}{2\lambda_1} + \frac{fP^*}{\alpha^2 \lambda_1} \right],$$

$$b_1 = \frac{2P^*\xi\gamma^2 k_B^2}{\alpha\alpha_0}, \quad b_2 = \frac{18\beta_1 k_B^4}{\alpha\alpha_0 \lambda_1}.$$

II. Results and discussions

Solutions of the self-consistent system of equations (8, 9) in the temperature region $T < 100$ K calculated numerically using the iterative method [11] (model parameters for non-crystalline solids of As(Ge)-S(Se) systems

($P^* = 0.1$, $\varepsilon = 10^{-4}$ eV, $f = (10^4 \div 10^5$ dynes/cm, $g_1/g_2 = 11 \div 15$, $\alpha \cdot \alpha_0 = 6$). The parameter Q^* is taken into account based on the results of the study of inelastic neutron scattering of chalcogenide glasses as the magnitude of the amplitude of continuum disorder: $Q^* = 0.15 \div 0.18$ [10, 11]. Linear temperature dependence of the fraction of atoms in soft atomic configurations at $T < 1^\circ K$ which the temperature rises it becomes nonlinear ($\sigma \approx T^k, k < 1$), saturated ($\sigma_{max} \approx 10^{-4} \div 10^{-2}$) about $T \approx 10 \div 10^2$ K (Fig. 1). Anomalous temperature behavior of changes in the fraction of atoms in soft atomic configurations $\partial\sigma/\partial T$ (Fig. 1) manifests itself in the temperature range ($3 \div 30$) K and correlates with the position of the frequency domain of the boson peak ν_{bp} of chalcogenide glasses [3, 8, 15]. The presence of a boson peak confirms the formation of nanoscale ordered structures in chalcogenide glasses [3, 15]. One of the variants of such an implementation is the formation through the processes of self-organization of soft atomic configurations of a fractal structure, which precisely realizes the minimum of energy dissipation [7, 14].

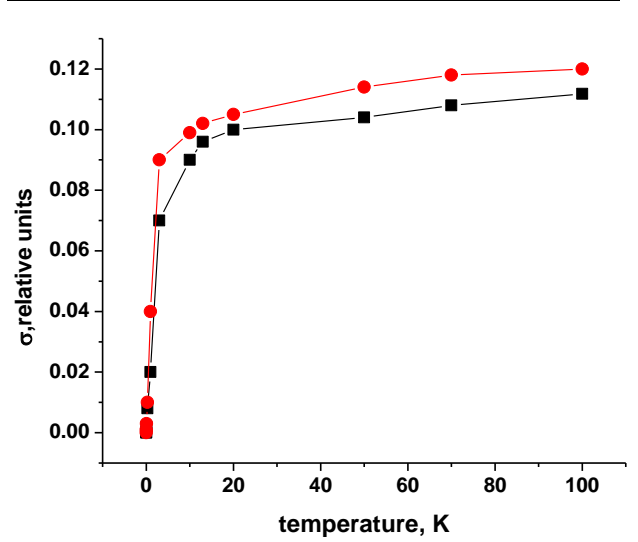


Fig. 1. Temperature dependence of the fraction of atoms in soft atomic configurations σ . $Q^*=0.15$ (black square), $Q^*=0.18$ (red circle).

Within the synergetic model, the structure of non-crystalline solids can be described as a linear contribution to thermodynamic functions C_p and a_T , caused by fluctuation transitions of atoms or their groups between soft atomic configurations, and cubic, due to phonons (Fig. 2). Since the coefficient of the linear part of the temperature dependence α_T is negative (15), then in the low temperature region a_T of the system initially decreases with heating, and then, due to the growth of the contribution of phonons ($\approx b_2 T^3$), begins to grow (Fig. 2). The nature of the effect of temperature compression of non-crystalline solids in the region $T \leq 1^\circ K$ is as follows. Structural heterogeneity at the level of the near and middle orders determines for the heterogeneity of the phonon subsystem (the anharmonicity of the atomic oscillations is most pronounced for weak links). Accumulated in the local areas of the non-crystalline system energy is stored in the form of elastic deformations $\approx P^* \exp(y_2)/(1 - \xi \sigma^2)$. With the growth of the fraction atoms in soft atomic configurations, elastic deformations will also increase, causing the sample to compress. At higher temperatures ($1 \div 10$) K cubic contribution to α_T becomes decisive (Fig. 2). The obtained result is consistent with the experimental data [4] on the temperature dependence of the Grüneisen coefficient ($G = -I$) for a number of non-crystalline solids in the temperature range $T \leq 10$ K.

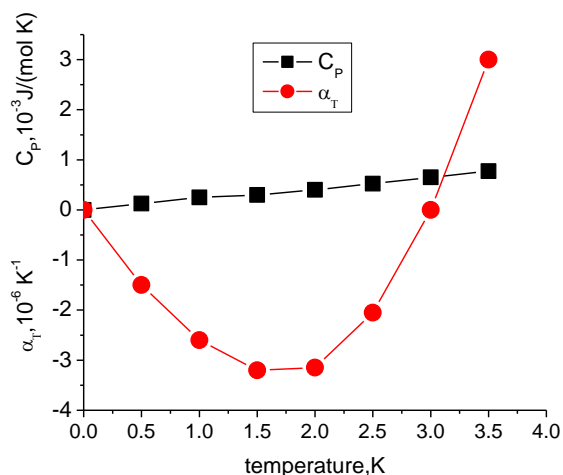


Fig. 2. Low-temperature dependences of the heat capacity C_p (black square) and the coefficient of thermal expansion α_T (red circle) for non-crystalline As_2S_3 , $Q^* = 0.18$.

This model explains the quasilinear temperature dependence of heat capacity $C_p \approx T^\delta$ ($\delta = 0.1 \div 0.4$), taking into account the self-consistent way along with the contribution of purely phonon excitations, their renormalization as a result of interaction with low-energy structural states (Fig. 3). This contribution, due to the interaction of phonons with soft configurations, has the order $a_2 \cdot T^2$ and is essential in the temperature range $T \leq (1 \div 3) \text{ K}$. The temperature dependence of the heat capacity, including not very low temperatures and calculated in accordance with (14), is presented in Fig. 3. In Fig. 3 shows experimental data of the dependence of the heat capacity C_p of glass As_2S_3 in the temperature range $T = (0 \div 12) \text{ K} \ll T_D = 161 \text{ K}$, where T_D is the Debye's temperature, which is in good agreement with the theoretically obtained ones (15). For $T = (3 \div 10) \text{ K}$ this dependence C_p/T^3 have a wide maximum ("hump"), which is due to the interaction of atoms in soft atomic configurations with the matrix of the soft atomic state, which leads to a self-consistent renormalization of the force constants, the distribution of low-energy states and its nonlinear temperature dependence.

We analyze the influence of the conditions of obtaining on the nature of low temperature anomalies of the thermal properties of non-crystalline solids. The influence of technological regimes of obtaining (changes in the temperature of synthesis T_0) was taken into account as follows: an increase in the synthesis temperature causes an increase in the continuum disorder of the framework Q^* , which is described by the dispersion of distortion of the short order and causes the change in atomic potentials (11), the frequency of oscillations of the atoms and thermodynamic functions E, V (14). So, for non-crystalline material As_2S_3 obtained by cooling the melt from the synthesis temperatures in the range $T = (870 \div 1370) \text{ K}$ (the boundaries are determined by the minimum and maximum temperature of synthesis of this composition at a rate $q = 1.8 \text{ K/s}$ [1]), there is a change in

the dispersion of interatomic distances, which is coordinated in order of magnitude with experimental data [11]. It should be noted that the variation of the conditions of synthesis does not change the universal nature of low-temperature anomalies C_p and α_T of non-crystalline solids, but causes a change in the ratios of different contributions and temperature intervals. The growth of the dispersion of the fluctuations of the interatomic distances of the material obtained at the synthesis temperature $T = 1370 \text{ K}$, causes an increase in the atomic share in soft atomic configurations and a shift in the temperature interval of saturation to a region of lower values in comparison with those for a material obtained at a temperature $T = 870 \text{ K}$. This result is consistent with the results obtained by the authors [7] in studies of the growth of the density of low-energy states with a decrease in the transition temperature to a non-crystalline state (an increase in the synthesis temperature at $q = const$ for As_2S_3 causes a decrease in the temperature interval of softening-glass transition). Correspondingly, the change Q^* affects the nature of the dependence C_p/T^3 from T , which qualitatively agrees with the experimental studies on the influence of the conditions of obtaining As_2S_3 to low-temperature heat capacity in the temperature range $(2 \div 10)^\circ\text{K}$ [16].

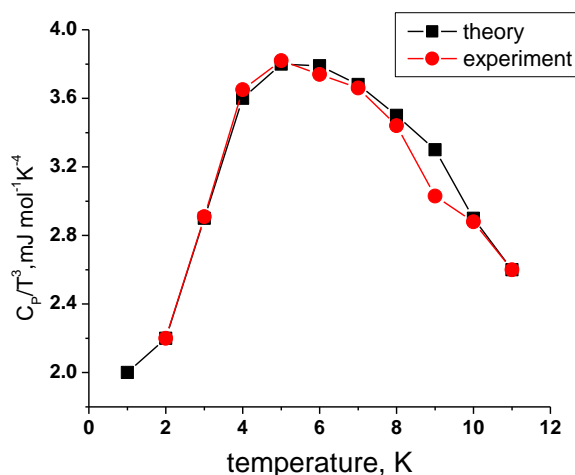


Fig. 3. Low-temperature dependence of the heat capacity of vitreous As_2S_3 . Red circle – experiment [8], black square – calculation according to (15), $Q^* = 0.18$.

It should be noted that the correlation studies of low-temperature anomalies of physical properties (mean-square displacements, heat capacity, and coefficient of thermal expansion) of non-crystalline solids with the conditions of their obtaining indicate the common features of the transition to state of the non-crystalline solids [10, 17]. The authors of [1, 5] established empirical relations between the softening temperature and the density of soft low-temperature states. In particular, it is shown that the coefficient for the linear part of the temperature dependence of the heat capacity is a function of the softening temperature $a_1 = f(T_g)$. Since the softening temperature depends on the conditions for obtaining in particular the synthesis temperature, using (15) it will be rewritten as follows:

$$a_1 = z\gamma^2 k_B^2 \cdot \frac{M_a}{m} \left\{ e^{-Q^*} \left[\varepsilon + \frac{\beta_2 V_{02}(2+P^*)}{\lambda^2 \gamma^4} \right] - \frac{2\xi v_{01} V_{01} P^*}{3} \right\} = c' + \frac{c''}{\exp(Q^*)} = c' + \frac{c''}{f(q, T_0)} = f(T_g). \quad (16)$$

That is, the ratio (16) for the coefficient of thermal dependence of the heat capacity allows us to obtain an empirical relation $a_1 = f(T_g)$, which describes the correlation between low-temperature anomalies and the transition to the state non-crystalline solids [15-18].

Conclusions

Analytical relations for the specific heat capacity, the coefficient of linear expansion of non-crystalline solids, which provide an opportunity to describe the linear and cubic contributions and their nature in the temperature range $T < 100$ K, have been obtained. The self-consistent dynamics of atomic oscillations, renormalization of local atomic potentials, and the spectrum of low-frequency excitations of non-crystalline materials are analyzed in detail from the point of view of the conditions of their

obtaining. The common features of the transition of the system into a non-crystalline state and the realization of low-temperature anomalies as a manifestation of the formation of self-organized structures are discussed.

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Низькотемпературні аномалії та нанорозмірні рівні формування самоорганізованих структур в некристалічних тілах систем As(Ge)-S(Se)

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Дискутується взаємозв'язок низькотемпературних аномалій фізико-хімічних властивостей та формування самоорганізованих структур у некристалічних тілах систем As(Ge)-S(Se). Отримані температурні залежності для питомої теплоємності та коефіцієнта лінійного розширення в області температур $T < 100^\circ K$, які деталізують лінійні та нелінійні вклади. Розглянуто вплив умов одержання некристалічних тіл на низькотемпературну поведінку фізико-хімічних властивостей та зміну співвідношень різних вкладів і температурних інтервалів. Обговорюється кореляція та спільні риси формування самоорганізованих структур некристалічних матеріалів в області низьких температур і температур розм'якшення, що проявляється у наявності нанорівнів структурування.

Ключові слова: низько-температурні аномалії, нанорозмірні ефекти, некристалічні матеріали, м'які атомні конфігурації, самоорганізовані структури, синергетика, дворівневі стани.