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Thermal properties of matter within the model of a two-phase system

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It has been established that the relations of the two-phase system model are applicable for describing the thermal properties of solids with disordered and crystalline structures. It is shown that the model adequately describes the curves of isochronous and isothermal crys-tallization of amorphous alloys and thermal changes in the volume of their samples. It is also suitable for calculating the heat capacities and thermal expansion coefficients of sub-stances that are diverse in their physical nature.

Keywords: two-phase system, thermal expansion coefficient, heat capacity, amorphous alloys, com-plex oxides.

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1. Introduction

Experimenters are currently showing a constant interest in studying the heat capacities of complex oxides [1–14]. It is due not only to their technical use (e.g., ceramics [15]), but also the uniqueness and diversity of physical properties. Smoothing of experimental data on heat capacities of complex oxides, the same as for most other solid substances, is performed using phenomenological functions of a certain type. Outside the approximation interval, they yield incorrect results. Moreover, the occurrence of polymorphous transformations, magnetic phase transitions and other phenomena in certain substances is represented on temperature dependences in the form of „peaks“ and „dips“, which are not described in the suggested models. That’s why development of models to describe the dependences of substances’ thermal properties in wide temperature ranges is one of the topical tasks of solid-state physics.

The author of [16] suggested a model of a locally-equilibrium two-phase system with an order parameter determined as the difference of volume fractions of the coexisting phases. Minimization of Gibbs energy according to this parameter has yielded a number of correlations used in earlier works and will be applied in the present paper. Therefore, the goal of the present paper is calculation and plotting (as examples) of temperature dependences of heat capacities of complex oxides and thermal expansion coefficients of simple oxides and semiconductors to demonstrate a wide application scope of the model [16].

The model [16] was used for the first time to describe phenomena and processes in amorphous alloys. Adequacy of theoretical calculations for experimental data for metal glasses makes it possible to hope that the model [16] is applicable to describing thermal properties of other systems as well.

2. Two-phase system model and its application

As distinct from point phase transitions, which occur at a certain value of the external parameter, a diffuse phase transition takes place in a certain range of change of temperature, time or other quantity [17,18]. Such transitions are observed in magnetics (the antiferromagnetic–ferromagnetic transition [17]), relaxors–magnetoelectrics [19,20], amorphous alloys [21,22] and other systems. A diffuse phase transition is accompanied with a transformation of a disordered phase (only the short-range order in the location of components is possible) into a phase with a long-range order.

1. *Isochronous and isothermal crystallizations of amorphous alloys.* An amorphous alloy is a metastable, i.e. non-equilibrium system. Non-ergodicity of metal glasses compels us to approximate them by locally-equilibrium regions in compliance with the Prigozhin’s principle [23].

Let us consider a combination of locally-equilibrium two-phase regions which contain an originating, a growing and an ordering phase (phase 1 with volume V_1) and a parent matrix (phase 2 with volume V_2) [16]. System volume V is equal to

$$V_1 + V_2 = V. \tag{1}$$

Dividing the equality (1) by volume V, we get the correlation

$$x_1 + x_2 = 1, \tag{2}$$

where $x_i = V_i/V$ — volume fraction of phase $i = 1, 2$.

Let us introduce the order parameter η using the formula

$$\eta = x_1 - x_2. \tag{3}$$

It follows from (2) and (3) that

$$x_1 = (1+\eta)/2, \qquad x_2 = (1-\eta)/2. \tag{4}$$

Formulas (4) show that the order parameter η takes on values from the range of -1 to $+1$, since volume fractions x_i vary within 0 to 1.

Gibbs energy per unit volume of a system, consisting of two non-interacting phases, is equal to

$$g = \mu_1 x_1 + \mu_2 x_2, \quad (5)$$

where μ_i — chemical potentials of phase elements are specified by formulas

$$\mu_i(P,T,t) = \mu_{i0}(P,T,t) + k_\mathrm{B} T \ln x_i(T,t), \quad (6)$$

$\mu_{i0}(P,T,t)$ — standard values of chemical potentials for each phase, P — pressure, T — temperature, k_B — Boltzmann constant.

Let us find the extremum of Gibbs energy (5) according to argument η, which corresponds to the locally-equilibrium value of the order parameter

$$(dg/d\eta)\big|_{\eta=\eta_0} = 0 \Rightarrow \eta_0(T,t) = -\mathrm{th}\big(0.5\Delta\mu_0(T,t)/(k_\mathrm{B}T)\big), \quad (7)$$

where the function $\Delta\mu_0 = \mu_{10} - \mu_{20}$. Consequently, the equilibrium fraction of the new phase at temperature T at time moment t is described by the function

$$x_1(T,t) = 0.5\big[1 - \mathrm{th}\big(\varphi(T,t)/T\big)\big], \quad (8)$$

here the argument

$$\varphi(T,t) = 0.5\Delta\mu_0(T,t)/k_\mathrm{B}.$$

Let us expand the function $\varphi(T,t)$ into a Taylor series near the point of the extreme thermal effect of the phase transition with keeping of only of the linear terms of the series

$$\varphi(T,t) = (\partial\varphi/\partial T)(T - T_x) + (\partial\varphi/\partial t)(t - t_x). \quad (9)$$

Let us consider particular cases of amorphous alloy crystallization:

– ***isochronous crystallization*** $\varphi(T,t) = f(T)$ i.e. formula (8) can be written down as follows with account of (9)

$$\varphi(T,t)/T = a(q)\big((T_x/T) - 1\big), \quad (10)$$

where the parameter $a(q) = -\partial\varphi/\partial T$, q is sample heating rate. The model parameter

$$a(q) = 2T_x(q)u_x(q) \quad (11)$$

is related to temperature $T_x(q)$, at which the maximum of the first derivative from the volume fraction of phase 1 $u_x(q) = (dx_1/dT)_{T=T_x}$ and the extremum of phase transition heat are observed.

– ***isothermal crystallization*** $\varphi(T,t) = f(t)$ at $T = \mathrm{const}$, i.e. formula (8) is as follows with account of (9)

$$\varphi(T,t)/T = b(q)\big((t/t_x) - 1\big), \quad (12)$$

where the coefficient is $b(q) = (t_x/T)\partial\varphi/\partial t$. The calculations of the volume fraction of the new phase in metal glass $Fe_{40}Co_{40}P_{14}B_6$ using the formula (8) with account of equalities (10) and (12) are shown in Fig. 1,*a* and 1,*b* respectively [24].

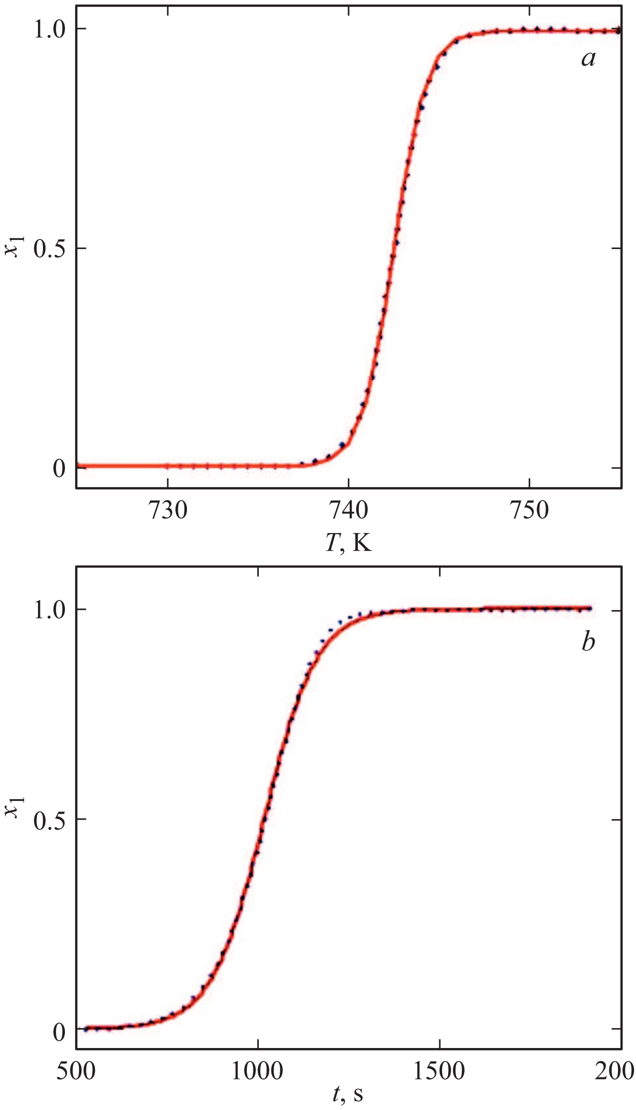

Figure 1. Functional dependences of volume fraction x_1 of the crystalline phase on temperature at $q = 40\,\mathrm{K/min}$ (*a*) and on time at $T = 619\,\mathrm{K}$ (*b*) for the amorphous alloy $Fe_{40}Co_{40}P_{14}B_6$ (the solid line shows theoretical values; dots — the experimental values [22]).

2. *Multistage nucleation in metal glasses.* The obtained correlations allow for describing not only single-stage but also multistage crystallization [25], when at least two crystals form in the alloy. Fig. 2 shows the isochronous crystallization curves for amorphous alloys $Fe_{85}B_{15}$ (Fig. 2,*a*) and $Al_{86}Ni_6Co_2Gd_6$ (Fig. 2,*b*). For instance, nucleation in the amorphous alloy $Fe_{85}B_{15}$ occurs in two stages: up to the temperature of 750 K, the α-Fe crystals are growing (phase α), and then Fe_3B crystals start nucleating (phase β) [21]. Volume fractions of $x_{1(\alpha)}$ and $x_{1(\beta)}$ crystals for each new phase were calculated using formulas (8) and (10), while the total

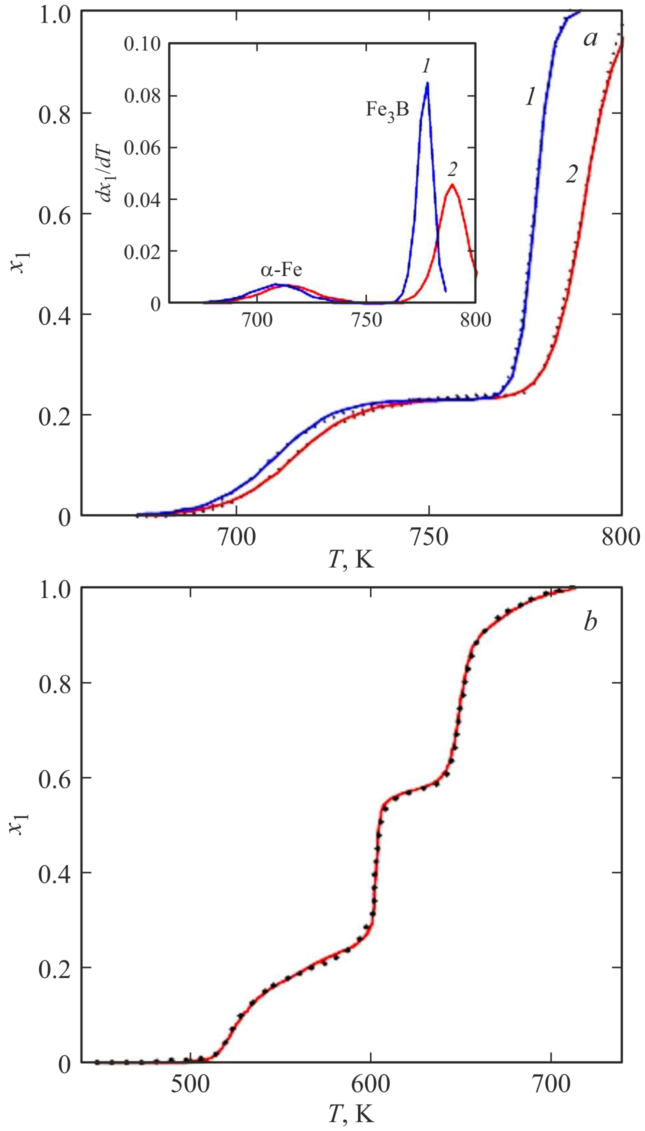

Figure 2. *a* — crystalline phase volume fraction (the inset shows a change of its first derivative) vs. temperature in the amorphous alloy $Fe_{85}B_{15}$ with the sample heating rates of $q = 10\,(1)$ and $40\,(2)$ K/min (the solid line shows theoretical values; dots — the experimental values [21]); *b* — changes of the same quantity in the amorphous alloy $Al_{86}Ni_6Co_2Gd_6$ at $q = 10$ K/min (the solid line shows theoretical values; dots — the values as per the DSC-thermogram).

volume fraction of the forming phase was determined using the formula [25]:

$$x = c_1 x_{1(\alpha)} + c_2 x_{1(\beta)}, \tag{13}$$

where the constants c_1 and c_2 do not depend on sample heating rate q (the model parameters are given in [25]).

3. Heat capacity of complex oxides

As distinct from a substantive (total) derivative which describes the function behavior at any point of the system and upon transition from one system point to another, a local (partial) derivative characterizes the function changes in the vicinity of a separate point. Let us use this fact when studying heat capacity of a locally-equilibrium region.

1. *Entropy of a locally equilibrium region.* Let us set a density of system's substantive entropy at a changing phase composition $x = x_1$ using a ratio (taken with the minus sign) of the differential of function (5) to the temperature differential

$$\sigma = -(dg/dT) = -\big[(\partial g/\partial T)_x + (\partial g/\partial x)_T (dx/dT)\big]$$
$$= \sigma_x + \varepsilon_T u, \tag{14}$$

where local entropy at a fixed phase composition x is determined by the classical formula [26,27], i.e. partial derivative of (5) with respect to temperature

$$\sigma_x = -(\partial g/\partial T)_x = \sigma_x + \sigma_d x + \sigma_s, \tag{15}$$

here the entropies are: of the original phase

$$\sigma_m = -\big(\partial \mu_{20}/\partial T\big),$$

of phase difference

$$\sigma_d = -\big[\partial(\Delta\mu_0)/\partial T\big]$$

and mixing

$$\sigma_s = -k_{\mathrm{B}}\big[x \ln x + (1-x)\ln(1-x)\big];$$

density of energy of coexistence of phases

$$\varepsilon_T = -(\partial g/\partial x)_T = -\Delta\mu_0 - k_{\mathrm{B}} T \ln\big(x/(1-x)\big), \tag{16}$$

$u = dx/dT$ is„rate" of phase composition change x at a thermal transition to a new state. Thus, locally-equilibrium entropy (15) coincides with its substantive determination (14) subject to satisfaction of equality $\varepsilon_T = 0$, which generates formula (8).

The first term in (14) describes the system entropy at a fixed phase composition of the system, and the second term — at its change, i.e. it is the „kinetic" component of substantive entropy, since it is determined by „rate" of phase composition change u.

2. *Heat capacity of locally-equilibrium substance.* Substantive heat capacity of the system depends on temperature T, phase composition x and „rate" of its change u, it is specified by the expression

$$C(T, x, u) = T(d\sigma/dT) = C_e + C_k + C_d, \tag{17}$$

where local heat capacity in the „static" thermal state

$$C_e = T(\partial\sigma/\partial T)_{x,u} = T\big[(\partial\sigma_m/\partial T)_{x,u} + (\partial\sigma_d/\partial T)_{x,u} x\big]$$
$$= k_1 T + k_2 x, \tag{18}$$

coefficients $k_1 = (\partial \sigma_m/\partial T)_{x,u}$ and $k_2 = T(\partial \sigma_d/\partial T)_{x,u}$. „Kinetic“ component of heat capacity

$$C_k = T(\partial \sigma/\partial x)_{T,u} u = \left[2\xi_T - k_B Tu/[x(1-x)]\right] Tu = f(x,u)Tu, \qquad (19)$$

where

$$\xi_T = (\partial \varepsilon_T/\partial T)_{x,u} = \sigma_d - k_B \ln\bigl(x/(1-x)\bigr).$$

The plots obtained during experimental studies of temperature dependences of heat capacities for several substances have peculiarities in the form of „peaks“ or „dips“ in the low- or high-temperature regions. According to preliminary calculations, they are described by formula (19) and arise as a reflection of phase transitions in subsystems of atoms or quasiparticles, while their manifestation in the form of „peaks“ and „dips“ depends on the sign of the transition thermal effect. „Dynamic“ component of heat capacity in formula (17) is specified by the expression

$$C_d = T(\partial \sigma/\partial u)_{T,x} w = T\varepsilon_T w \qquad (20)$$

(here $w = du/dT$). It should be noted that function (20) becomes zero when local equilibrium is established ($\varepsilon_T = 0$).

The first term in formula (18) linearly depends on temperature and, according to the classical thermodynamics [28], describes the contribution of the electron subsystem substance to heat capacity, while change of the temperature trend of the heat capacity curve is reflected by the second term. The contribution of the additional phase to heat capacity is accounted by applying formula (19), function $f(x,u)$ being constant.

Table 1. Model parameters for calculation of heat capacities of complex oxides

Oxide	$a(q)$	T_x, K	$k_1 \cdot 10^4$	k_2
$Nd_2Sn_2O_7$	0.6780	67	837	302.6
$Nd_2Ge_2O_7$	0.5571	137	271.2	362
$Tm_2O_3 \cdot 2ZrO_2$	0.3200	310	100	450
$Er_2Ge_2O_7$	0.6471	166	318.6	333
$Cu_5V_2O_{10}$	0.3300	341	15	718
$Pb_7Nd_3(GeO_4)_5(VO_4)$	0.6146	102	2304.1	1122
$Pb_9Nd(GeO_4)_3(VO_4)_3$	0.6223	94	1759.7	1165
$Tb_2Ge_2O_7$	0.5090	122	438.6	336
$LaGa_{0.5}Sb_{1.5}O_6$	0.2900	346	400	336
$FeTa_2O_6$	0.6710	187	170	276
$La_2Zr_2O_7$	0.7700	142	2096	228
$SmGaGe_2O_7$	0.6880	220	312.2	338
$Y_{0.6}Bi_{0.4}VO_4$	0.5200	215	54	205.2
$Bi_2Ti_4O_{11}$	0.4747	157	302	576
$Bi_4Ti_3O_{12}$	0.4773	174	940	616
$GdGaTi_2O_7$	0.6721	214	20.67	372

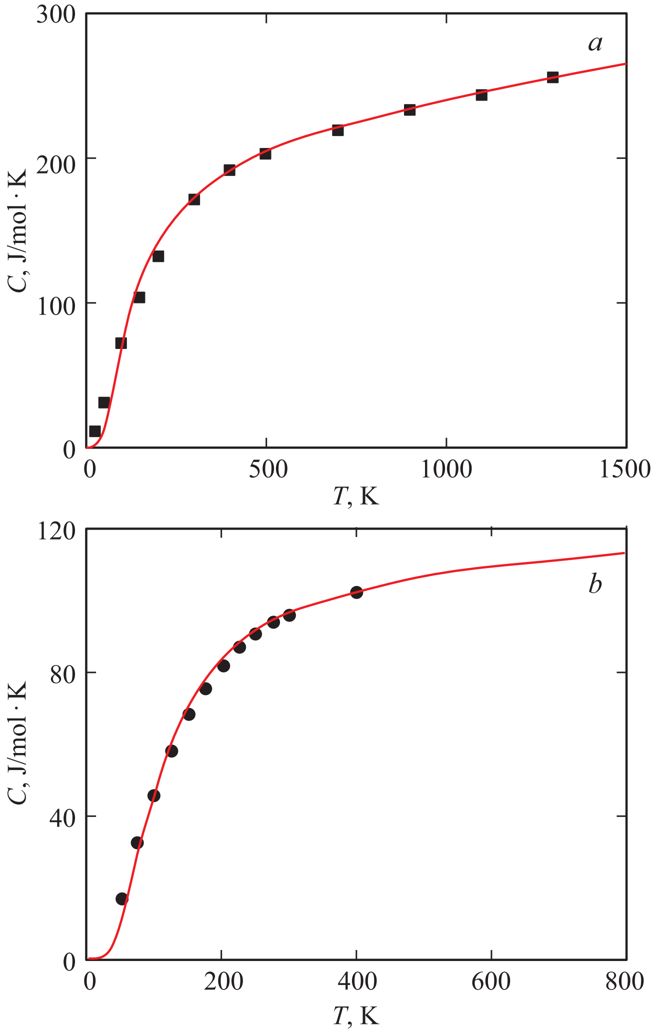

Figure 3. Temperature dependences of heat capacities of $LaGa_{0.5}Sb_{1.5}O_6$ (*a*) and $NaNbO_3$ (*b*) (the squares show the data of [8], circles — [29]).

Table 1 gives the model parameters for complex oxides discussed in [1–14]. As an example, Fig. 3 shows the experimental data and theoretical curves for $LaGa_{0.5}Sb_{1.5}O_6$ (Fig. 3, *a*) and $NaNbO_3$ (Fig. 3, *b*), calculated using formula (18) with the parameters from Table 1.

It can be seen from Fig. 3 that the thermodynamic model smooths the experimental data rather well and can be used both for single calculations and for the making of automatic process control tools for production facilities. This can be made possible by creating a database with values of coefficients of the theoretical model or by finding the relation between them and substances' individual characteristics. Model simplicity and a wide application scope indicate the universal nature of the model theoretical construction.

4. Thermal expansion of substance

An adequate description of experimental data on heat capacity of various substances by the logistic function (8) allows for using it to describe other thermal properties as well, e.g., thermal expansion coefficients.

1. *Linear (volume) thermal expansion coefficient.* According to the mixing rules (see, e.g., [30]), the thermal expansion coefficient (TEC) for a two-phase region is

$$\alpha(T, x) = \alpha_1 x + \alpha_2(1 - x) = \alpha_2 + x\Delta\alpha, \qquad (21)$$

here $x = x_1$, α_i $(i = 1, 2)$ — TEC of phase i, $\Delta\alpha = \alpha_1 - \alpha_2$ — coefficient of thermal difference of phases. Using the thermodynamic definition of heat capacity, the second Grüneisen rule (see, e.g., [31,32]) on a relation of heat capacity to TEC and taking into account equality (18), we obtain

$$\alpha = q_1 T + q_2 x, \qquad (22)$$

where the coefficients q_1 and q_2 are considered to be constant. Fig. 4 shows the charts of temperature dependences of linear thermal expansion coefficients $\alpha \cdot 10^6$ for Al_2O_3 (Fig. 4, *a*) and ZnO oxides (Fig. 4, *b*) [33]. Fig. 5 shows the trend of the temperature curves which describe the volume thermal expansion coefficients $\beta \cdot 10^5$ for Ge (Fig. 5, *a*) and Si semiconductors (Fig. 5, *b*) [34]. The calculated model parameters are given in Table 2.

2. *Effect of sudden volume change under glass transition* [35]. According to the Gay-Lussac law, when the volume thermal expansion coefficient β is constant, the system volume changes according to the law (see, e.g., [36])

$$V(T) = V_0[1 + \beta(T - T_0)], \qquad (23)$$

where $V_0 = V(T_0)$ is the melt volume at temperature T_0. Since the thermal expansion coefficient changes according to (22), attainment of temperature T_g during heating of an amorphous solid sample or during cooling of a

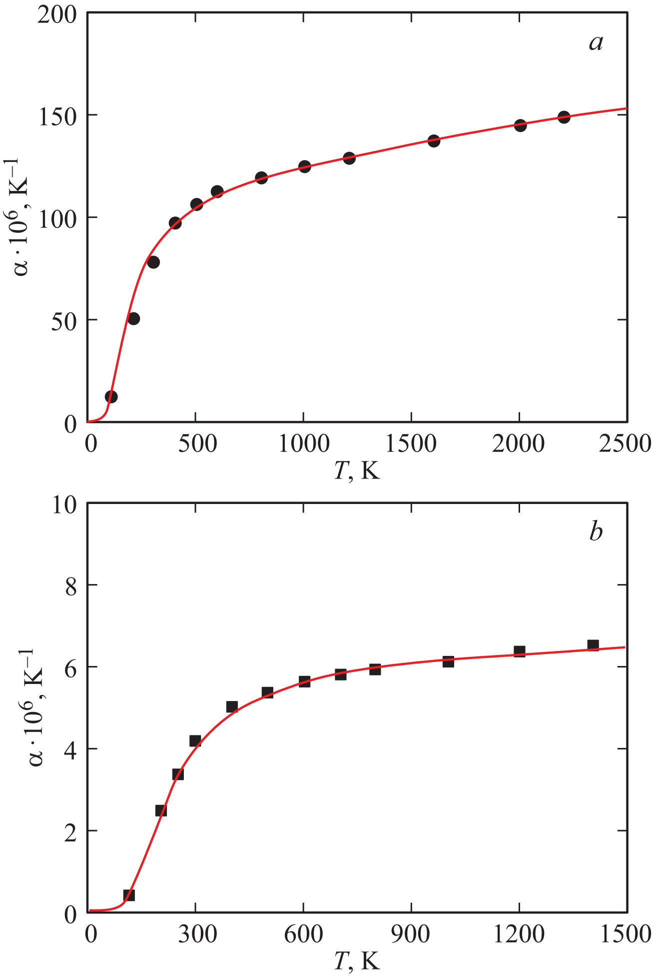

Figure 4. Change of coefficients of linear thermal expansion $\alpha \cdot 10^6$ for Al_2O_3 (*a*) and ZnO oxides (*b*) with temperature (the dots and squares show the data of [33]).

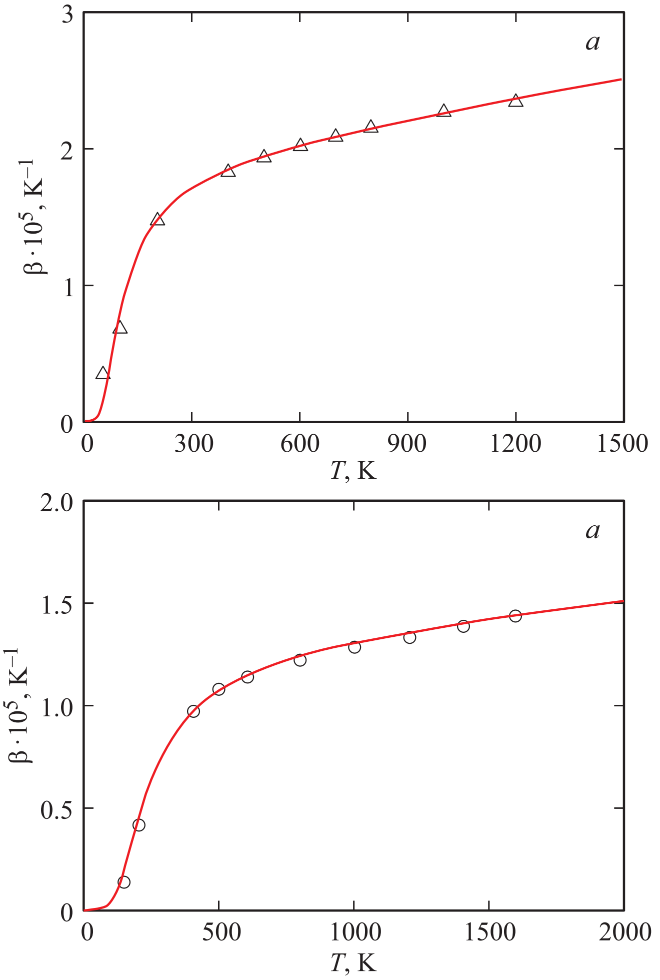

Figure 5. Change of coefficients of volume thermal expansion $\beta \cdot 10^5$ for Ge (*a*) and Si semiconductors(*b*) with temperature (the triangles and circles show the data of [34]).

Table 2. Theoretical model parameters for oxides and semiconductors

System	$a(q)$	T_x, K	$q_1 \cdot 10^4$	q_2
Al_2O_3	0.89	65	16.4	8.94
ZnO	0.92	293	1.0	7.7
Ge	0.749	145	4.5	2.3
Si	0.97	296	1.48	1.45

vitrifying melt leads to a sudden volume change (23), followed by transition to a straight line with a different angular coefficient (Fig. 6 [37]). Since glass transition temperature T_g depends on heating (cooling) rate q, the temperature range of transition to a new state is a variable quantity, while a kink of the straight line (23) is observed at point T_g [35,38]. When the glass transition rate is low enough, a kink is substituted by a smooth transition from the straight line (23) to a straight line of thermal change of glass volume. An increase of metal glass heating rate above 500 K/min results in a considerable expansion of the temperature range of the phase transition.

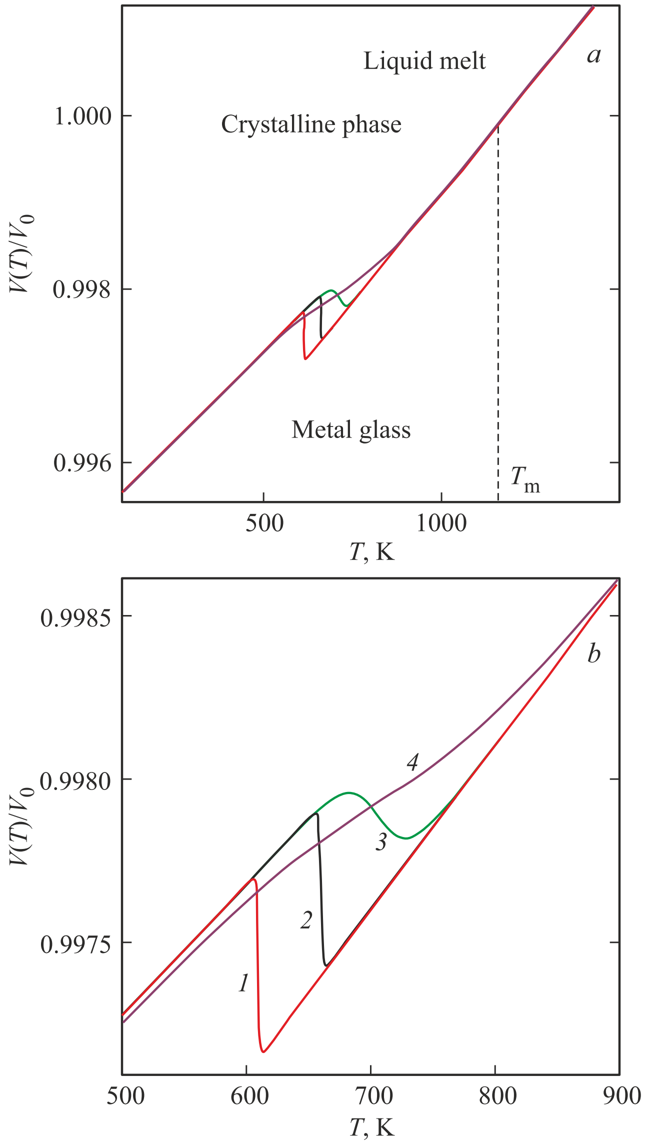

Figure 6. Temperature changes of metal glass volume (a) and increased glass-transition range (b) at heating rate q, K/min: *1* — ≈ 0, 1; *2* — 5; *3* — 200; *4* — 2500.

5. Conclusion

The performed calculations of thermal characteristics of various substances indicate the following: applicability of physical approximation of a non-equilibrium system by locally-equilibrium regions in compliance with the Prigozhin's principle; certain universalism of the model of a diffuse phase transition; similarity of thermal properties of various substances. The study has revealed that the thermal effect of phase transitions in subsystems of the main substance matrix manifests itself on the basic temperature dependence of heat capacity in the form of „peaks" and „dips" (due to different signs of transition enthalpies), which indicates their „kinetic" nature. In this respect we may suppose that the negative values of the thermal expansion coefficient for several substances in the low-temperature region are related to the manifestation of the „kinetic" effect in the parent phase.

Conflict of interest

The author declares that he has no conflict of interest.

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