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Spin and charge fluctuations in solid solutions of titanium substitution by iron group metals

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Topological electronic transitions from omega- to alpha-phase and from alpha- to beta-phase that occur with an increase in the concentration of substitution atoms in solid solutions of titanium with iron group metals are considered. It is found that due to the difference in the potentials of the Hubbard repulsion of titanium and substitution atoms, as well as temperature Bose excitations, spin and charge fluctuations of the electron density increase in the topological alpha phase. Accounting for fluctuations makes it possible to describe the concentration increase in the entropy of the phases of the titanium alloys under consideration at different temperatures. The amplification of electronic fluctuations with increasing temperature and concentration shifts the chemical potential beyond the topological area of the electronic spectrum, thereby suppressing the alpha phase and inducing an electronic transition to the beta phase. Using the example of calculations of the entropy of titanium substitution alloys with iron at different temperatures, a diagram of their structural phases is constructed, which is consistent with the available experimental data.

Keywords: solid solutions, topological electronic transitions, spin and charge fluctuations, entropy, structural phases.

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1. Introduction

It is known that the alpha phase of titanium arises from the omega phase due to a change in temperature or pressure, an energy shift of the chemical potential level to the area of topological features of the electronic spectrum [1–3]. At the same time, as it was shown in [2,3], a thermodynamic instability of spin and charge density waves associated with a change in the sign of the mode-mode parameter arises, leading to the appearance of topologically induced electronic fluctuations. The transition to the beta phase, on the contrary, is characterized by the disappearance of this instability, which leads, in accordance with the principle of maximum entropy, to the transition to the BCC structure. At the same time, attempts to explain the occurrence of a thermodynamically stable beta phase based on the ideas of an abnormally large vibrational entropy associated with lattice anharmonicity turn out to be contradictory because they lead to imaginary phonon modes [4]. Direct first-principle GGA + U calculations of the phonon and electron spectra of titanium [5] indicate a significant contribution to the thermodynamic potential of the beta phase associated with d-electrons.

In the study [6], based on the first-principle calculations of the formation heats, it was shown that when titanium atoms are replaced by transition metals, concentration electronic transformations occur between alpha, beta and omega structural phases. However, the calculations performed did not consider the topological features of the electronic structure of the alpha phase, which lead to a sharp increase in electron density fluctuations and to the emergence of a significant fluctuation contribution to entropy. Therefore, this approach describes only the metastable state of the structural alpha phase, and difficulties arise in understanding the temperature-concentration transitions between the structural phases of mutual solid solutions of titanium substitution with transition metals. In particular, there was no agreement with experimental data on the $x-T$ diagram of alloys arising from the substitution of titanium with iron impurities ($x \ll 1$) in the temperature area of the transition from alpha to beta phase.

The question remains open about the peculiarities of the influence on the entropy of the structural phases of the titanium alloys under consideration, Hubbard correlations in the system of d-electrons. At the same time, due to the difference in the parameters of the Hubbard intra-atomic repulsion (U) for titanium atoms and substitution atoms, fluctuations U should occur at the nodes, the effect of which on the thermodynamic stability of the structural phases has not yet been studied.

In this paper, a model of the electronic structure of solid solutions of titanium substitution with transition metals is developed, based on the results of DFT + U first-principle calculations of the zone structure of titanium with additional consideration of Hubbard correlations and fluctuations of Hubbard potentials. It is shown that with an increase in the concentration of (x) 3d-substitution atoms, a picture of a concentration topological electronic transition (TEP) from the omega to beta state through an intermediate alpha phase is realized in the alloys under consideration. Analysis of the concentration dependences of entropy leads to a $x-T$ diagram of the structural phases of mutual solid solutions of titanium substitution with iron, consistent with experimental data.

2. Model

Let us consider a model of an electronic system of chaotic titanium alloys with transition metals with a Hamiltonian, including the band motion term s-, p-,d- of electrons and the Hubbard correlations term d-electrons of Ti and substitution atoms

$$H = H_{s,p} + H_d. \qquad (1)$$

Here, H_{sp} — the Hamiltonian of almost free s-, p-electrons, and the subsystem of d-electrons is described by the Hubbard model, in which interelectronic correlations are taken into account along with the band motion term, related to the intra-atomic Coulomb interaction at the nodes occupied by titanium, with the value of the Hubbard interaction parameter U_1, and substitution atoms, with the value of the Hubbard repulsion parameter U_2:

$$H_d = \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}}(V) a^{+}_{\mathbf{k},\sigma} a_{\mathbf{k},\sigma} + \bar{U} \sum_{\mathbf{q}} \left(|n_{\mathbf{q}}/2|^2 - |S^{(z)}_{\mathbf{q}}|^2 \right)$$
$$+ \sum_{\boldsymbol{\nu}} \Delta U_{\boldsymbol{\nu}} \left(|n_{\boldsymbol{\nu}}/2|^2 - |S^{(z)}_{\boldsymbol{\nu}}|^2 \right), \qquad (2)$$

where

$$n_{\mathbf{q}} = \sum_{\sigma=\pm 1} n_{\mathbf{q},\sigma}, \quad S^{(z)}_{\mathbf{q}} = 2^{-1} \sum_{\sigma=\pm 1} \sigma n_{\mathbf{q},\sigma},$$

$$n_{\mathbf{q},\sigma} = \sum_{\mathbf{k},\sigma} a^{+}_{\mathbf{k},\sigma} a_{\mathbf{k}+\mathbf{q},\sigma}$$

— Fourier image of the operator of the number d-of electrons at the node $\boldsymbol{\nu}$ with the spin $\sigma/2$: $n_{\boldsymbol{\nu}} = a^{+}_{\nu,\sigma} a_{\nu,\sigma}$; $a^{+}_{\mathbf{k},\sigma}(a_{\mathbf{k},\sigma})$ — electron birth (destruction) operator with quasi-pulse $\mathbf{k}$ and spin $\sigma/2$; $\sigma = \pm 1$ — spin index; $\varepsilon_{\underline{\mathbf{k}}}(V)$ — DFT + U is the electron spectrum of d-electrons, $\bar{U} = U_1(1-x) + U_2 x$ — the average value of the Hubbard repulsion parameter at the node; $x = N_0^{-1} \sum_{\boldsymbol{\nu}} p_{\boldsymbol{\nu}}$ — atomic concentration of d-substitution impurities; $p_{\boldsymbol{\nu}}$ — projection operator equal to „+1“ if the node is occupied by a titanium ion and „−1“ — otherwise; $\Delta U_{\boldsymbol{\nu}} = (U_1 - U_2)(p_{\boldsymbol{\nu}} - x)$. The dependence of the electron spectrum on volume is described in the Heine model [7] for non-hybridized d-electrons, according to which when the volume changes from V_0 to V: $\varepsilon_{\mathbf{k}}(V) = (V/V_0)^{-5/3} \varepsilon^{(\mathrm{DFT})}_{\mathbf{k}}$.

The statistical sum of the considered electronic system is expressed in terms of the statistical sums of s-, p- and d- of electrons and is described by the expression

$$Z = Sp\exp(-H/T) = Z_d Z_{sp}.$$

Here the statistical sum of almost free s-, p-electrons is determined by their density of states $(g^{(0)}_{sp}(\varepsilon))$ calculated in the DFT + U [3] approximation and is described by a well-known expression containing the Fermi−Dirac distribution function $(f_{\mathrm{F}}(\varepsilon, \mu))$,

$$Z_{sp} = \exp\left\{ - \int d\varepsilon g^{(0)}_{sp}(\varepsilon) \ln\left(1 - f_{\mathrm{F}}(\varepsilon, \mu)\right) \right\}.$$

Statistical sum of strongly correlated d-electrons with the Hubbard Hamiltonian (2) is calculated using the Stratonovich–Hubbard transformations, which reduce the account of the Hubbard interaction, presented in the form of quadratic forms for spin and charge density operators, to the study of electron motion in exchange (ξ) fields fluctuating in space and time and charge (η) fields [8]. At the same time, using the representation of the Matsubar four pulses $q = (\mathbf{q}, \omega_{2m})$ [9] to account for dynamic fluctuations, and decomposing by fluctuating fields, we have

$$Z_d = Z^{(\mathrm{DFT})}_d$$
$$\times \int (d\eta d\xi) \exp\left\{ - \sum_{q(\neq 0)} \left(|\eta_q|^2 - |\xi_q|^2 \right) - T^{-1} \Phi_d(\xi, \eta) \right\}. \qquad (3)$$

Here

$$\Phi_d(\zeta, \eta) = T \sum_q \chi^{(0)}_q \left(|\zeta_q|^2 - |\hat{\eta}_q|^2 \right) + (4!)^{-1} \kappa(\mu, V) T$$
$$\times \sum_{q_1,q_2,q_3,q_4} \delta_{\Sigma_i q_i;0} (\zeta_{q_1}\zeta_{q_2}\zeta_{q_3}\zeta_{q_4} - 2\zeta_{q_1}\zeta_{q_2}\hat{\eta}_{q_3}\hat{\eta}_{q_4} + \hat{\eta}_{q_1}\hat{\eta}_{q_2}\hat{\eta}_{q_3}\hat{\eta}_{q_4})$$

it takes into account the space-time dependencies of the fluctuating fields described by the introduction of the Lindhard function $\chi^{(0)}_z(V)$,

$$\hat{\eta}_q = \eta_q + \Delta u \sum_q \Delta p_{q+q'} \eta_{q'}, \ \eta_{\mathbf{q}} = \eta^{(1)}_{\mathbf{q}} + i\eta^{(2)}_{\mathbf{q}},$$

$$\zeta_q = \xi_q + \Delta u \sum_q \Delta p_{q+q'} \xi_{q'}, \ \xi_{\mathbf{q}} = \xi^{(1)}_{\mathbf{q}} + i\xi^{(2)}_{\mathbf{q}},$$

$$\xi_q = r_q \exp(i\varphi_q), \Delta u = \bar{U}^{-1/2}(U_1^{1/2} - U_2^{1/2}), \Delta p_q(\sim \delta_{\omega,0})$$

— Fourier image of the projection operator,

$$(d\xi d\eta) = d\xi_0 d\eta_0 \prod_{q\neq 0, j=1,2} d\xi^{(j)}_q d\eta^{(j)}_q$$

(the index j numbers the real and imaginary parts), μ — chemical potential, the self-consistent definition of which is discussed below.

Expression (3) enables mode-mode interactions with parameter

$$\kappa(\mu, V) = \bar{U}^3 \left(\left(g^{(1)}_d(\mu)\right)^2 / g^{(0)}_d(\mu) - g^{(2)}_d(\mu) \right), \qquad (4)$$

determined by the density of the electronic states of d-electrons (DOS) of pure titanium and its l-derivatives: $g^{(0)}_d(\varepsilon)$and $g^{(l)}_d(\varepsilon)$, in DFT + U-approximation [2,3]. A similar parameter appears in the Ginzburg−Landau−Brazovsky functional [10], and in the case of changing the sign of this parameter from positive to negative, this functional describes a prolonged phase transition of the first kind induced by fluctuations. According to (4) the sign of the mode-mode parameter is determined by the derivatives of DOS, which can be related to the topological features of the electronic spectrum.

3. Free energy functional and entropy

Using the expression for the statistical sum (3), we consider the state of thermodynamic equilibrium of an electronic system, based on the principles of the minimum thermodynamic potential at constant pressure

$$G_{el} = T \ln Z + KV^2/2 + \mu N,$$

where K is the all-round compression module.

The determination of the thermodynamic minimum of free energy is reduced to calculating the statistical sum using the saddle point conditions for the variables $\eta_{\mathbf{q}}^{(j)}$, $\xi_{\mathbf{q}}^{(1,2)}$ and $r_q = |\xi_q|$ ($\xi_q = r_q \exp(i\phi_q)$), while preserving the integration at $\omega_{2m} \neq 0$, which should be supplemented with a minimization condition in volume. In the saddle point approximation, we obtain a free energy functional of the Ginzburg type–Landau

$$F_{el} = 2T \int \big(g_d(\varepsilon, x, \Delta u) + g_{s,p}(\varepsilon)\big) \ln\big(1+\exp\big((\mu-\varepsilon)/T\big)\big)$$
$$\times\, d\varepsilon + T\lambda(\mu)\big(\langle\xi^2\rangle + \langle m^2\rangle - \langle\eta^2\rangle\big) + T(4!)^{-1}$$
$$\times\, \kappa(\mu)\big(\langle\xi^2\rangle - \langle\eta^2\rangle\big)^2 + \lim_{\delta\to 0}\sum_{\mathbf{q}} \int\limits_{-\infty}^{\infty} d\omega f_B(\omega/T)$$
$$\times \ln\big[1 - \bar{U}\chi^{(0)}(\mathbf{q}, \omega + i\delta) + \lambda(\mu)\big], \tag{5}$$

describing the fluctuation phase (the prophase of which corresponds to the spin and/or charge ordering in the system of d-electrons) with spatial fluctuations of spin waves (SSW) — $\langle\xi^2\rangle$ and charge (CSW) — $\langle\eta^2\rangle$ densities implemented under conditions of negative mode-mode coupling

$$\big(\langle\eta^2\rangle - 2\langle\xi^2\rangle\big)(1 + \Delta u^2)$$
$$= -\kappa^{-1}(\mu, V)\big(1 + \bar{U} g_d^{(0)}(\mu)\big) - 2\langle m^2\rangle, \tag{6a}$$

$$\big(\langle\xi^2\rangle - 2\langle\eta^2\rangle\big)(1 + \Delta u^2)$$
$$= -\kappa^{-1}(\mu, V)\big(1 - \bar{U} g_d^{(0)}(\mu)\big) + 2\langle m^2\rangle. \tag{6b}$$

Moreover, from the conditions of the saddle point for r_q, an expression follows for the root-mean-square amplitude of thermal fluctuations of the spin density, consistent with the fluctuation-dissipative theorem

$$\langle m^2\rangle = \langle m^2\rangle_T + 2\Delta u^2 n_d^2 x(1 + x),$$
$$\langle m^2\rangle_T = (T/\bar{U})1 - \bar{U} g_d^{(0)}(\mu) + \lambda(\mu, V)^{-1},$$
$$\lambda(\mu, V) = \kappa(\mu, V)\big(\langle m^2\rangle + \langle\xi^2\rangle - \langle\eta^2\rangle\big). \tag{7}$$

The mode-mode parameter is associated with the first and second derivatives of the density of states of d-electrons at an energy equal to the energy of the chemical potential (see (4)), determined by the conditions of electroneutrality, which at $x \ll 1$ approximately has the form

$$N = \partial F/\partial\mu,$$
$$N/N_0 \approx 2\int \big(g_{s,p}^{(0)}(\varepsilon) + g_d(\varepsilon, x, \Delta u)\big) f(\varepsilon - \mu) d\varepsilon$$
$$+ \bar{U} g_d^{(1)}(\mu)\big(\langle m^2\rangle_T + \langle\xi^2\rangle - \langle\eta^2\rangle/4\big), \tag{8}$$

where

$$g_d(\varepsilon, x, \Delta u) = 2^{-1}\sum_{\alpha=\pm 1} g_d^{(0)}\big(\varepsilon + \alpha\Delta u n_d \sqrt{x(1-x)}\big)$$
$$\approx 2^{-1}\sum_{\alpha=\pm 1} g_d^{(0)}(\varepsilon) + g_d^{(2)}(\varepsilon)\Delta u n_d \sqrt{x(1-x)}.$$

In addition, we obtain that during the transition from alpha to omega or beta phase, there is a decrease in volume, which, in accordance with the condition of minimum free energy in volume at a fixed atmospheric pressure p_0, is determined by the expression

$$\Delta V \cong -|\kappa^{-1}(\mu, V)|^2 \big(\bar{U} g_d(\mu, x, V)\big)^2 (1 + \Delta U^2/U^2)$$
$$\times \big[\langle\xi^2\rangle - \langle\eta^2\rangle\big]/(3p_0).$$

Thermodynamically stable states of the system will be determined under conditions of minimum Gibbs energy, taking into account the requirement of maximum entropy. The entropy of an electronic system depends on temperature, volume and concentration x:

$$S_{el} = S_F + S_{fl} + S_{pm}, \tag{9}$$

and along with the entropy of Fermi excitations of d- and s-, p-electrons

$$S_F = \int d\varepsilon g_d(\varepsilon, x, \Delta u)\Big[f(\varepsilon - \mu)\ln f(\varepsilon - \mu)$$
$$+ \big(1 - f(\varepsilon - \mu)\ln\big(1 - f(\varepsilon - \mu)\big)\big)\Big], \tag{9a}$$

contains fluctuation and paramagnetic contributions

$$S_{fl} = -\kappa(\mu)^2\Big(\langle\xi^2\rangle - \langle\eta^2\rangle + \langle m^2\rangle_T - \Delta u^2 n_d^2 x(1-x)\Big)^2, \tag{9b}$$

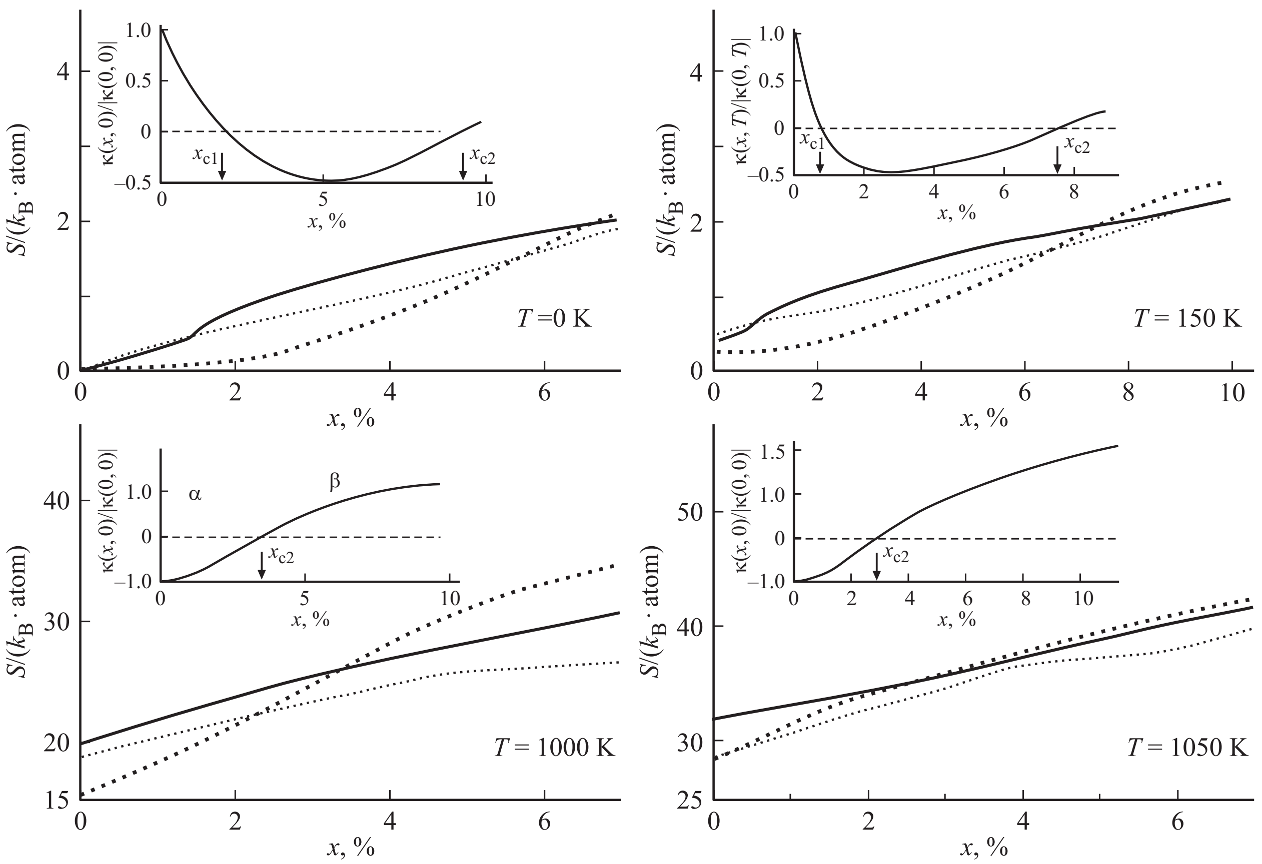

Figure 1. Dependences of the entropy of $Ti_{1-x}Fe_x$ alloys on the concentration of iron (x) at fixed temperatures, obtained taking into account the results of DFT + U calculations $g^{(0)}_{s,p,d}(\varepsilon)$ [1,2] for: α-phases (solid line), β-phases (dots), ω-phases (dotted line), counted from the entropy value ω-phases at $x = 0$ and $T = 0$ K. The inserts show the concentration dependences of the mode-mode interaction coefficient (4) at fixed temperatures.

$$S_{pm} = \lim_{\delta\to 0}\sum_{\mathbf{q}} T^{-1}\int_{-\infty}^{\infty} d\omega\, f_{\mathrm{B}}(\omega/T) \times \ln\left[1-\bar{U}\chi^{(0)}(\mathbf{q},\omega+i\delta)+\lambda(\mu)\right]. \qquad (9c)$$

In addition, to determine the concentration boundaries arising in titanium substitution alloys with $3d$-metals of structural phases, it is necessary to analyze the formulas obtained with additional consideration of the contributions of s-, p-electrons and phonons to free energy and entropy.

4. Numerical analysis

Let us consider the concentration TEP associated with the appearance (or disappearance) of the alpha phase in titanium alloys, using the example of titanium substitution with iron. At the same time, we will take into account not only the shift in the energy level of the chemical potential due to a change in the number of d-electrons, but also the energy shift of the chemical potential associated with fluctuations in the electron density (6), (7) and the difference in the Hubbard potentials of titanium ($U_1 = 1.75$ eV) and iron substitution atoms ($U_2 = 1.2$ eV). The densities of the electronic states of alpha, beta and omega phases of pure titanium calculated in the DFT + U approximation coincide with those obtained and described in [1–3]. At the same time, by approximating the DOSd- of electrons in the energy neighborhood of the chemical potential by polynomials (see [2,3]), it is possible to describe the change in the sign of the mode-mode parameters during transitions between phases.

In the expression for the Gibbs energy $G = G_d + G_{sp} + G_{ph}$, along with the term associated with the d-subsystem, the contribution of s-, p- of electrons is considered and the phonon contribution is additionally taken into account, which is considered in the Debye model: $G_{ph} = TD(T/\theta_D)$, where $D(T/\theta_D)$ — Debye integral, with characteristic temperature

$$\theta_D(x) = (1-x)\theta_D^{(\mathrm{Ti})} + x\theta_D^{(\mathrm{Fe})} \text{ [11].}$$

In addition, we take into account that, in accordance with the thermodynamic definition of entropy, its expression $S = S_{el} + S_{ph}$, along with the electronic contribution (9), includes the phonon

$$S_{Ph} = 36D(\theta_D/T) - 9(\theta_D/T)\left(\exp(\theta_D/T) - 1\right)^{-1}.$$

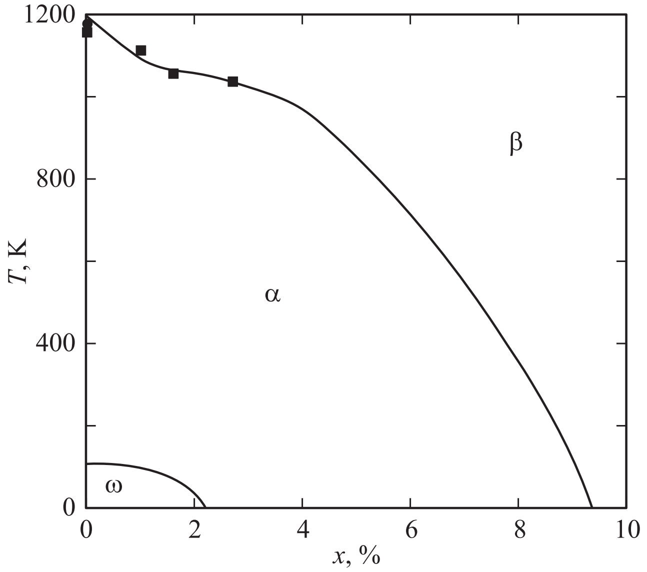

Figure 2. Solid lines — concentration-temperature boundaries of the structural phases $x_{c1}(T)$ and $x_{c2}(T)$. Experimental data: circle — [12], squares — [6].

When calculating the concentration dependences of entropy, the features of the curvature of the density of states near the energies of the chemical potential were taken into account, leading to a change in the sign of the mode-mode parameter and the appearance of spatial fluctuations (6), as well as the condition of electroneutrality (8).

Fig. 1 shows the results of calculations of the entropy of phases of solid solutions of titanium substitution with iron at different temperatures. From the results of entropy calculations, it can be seen that the transition from the GU ω-phase to the topological GPU α-phase that occurs at the values of x_{c1} is accompanied by a change in the sign of the mod-mod parameter from positive to negative (inserts on Fig. 1), and there is a gain in the entropy of the alpha phase due to the fluctuation contribution of (9b). At $x_{c2}(T)$ the GPU alpha phase loses thermodynamic stability (Fig. 1) and the BCC beta phase occurs. This phase transition is accompanied by a change in the sign of the mode-mode parameter from negative to positive. As a result, the entropy of the beta phase begins to prevail over the entropy of other phases.

Fig. 2 shows the $x-T$ diagram of the structural phases of a solid solution of substitution of titanium atoms with iron. It can be seen that the omega phase will persist near the ground state up to the concentration of $x_{c1} = 2.1\%$ (see also Fig. 1). However, with an increase in temperature, a fluctuating shift of the chemical potential occurs, towards the typologically determined features of DOS, which leads to a reduction in the concentration interval of the omega phase (Fig. 2). The alpha phase interval near the ground state is maintained up to $x_{c2} = 9.41\%$, after which the beta phase is realized (see Fig. 1, 2).

Thus, it can be seen that the concentration transition from the omega phase to the BCC beta phase, considered by the example of solid solutions of titanium substitution with iron, is carried out through the topological alpha phase induced by electron density fluctuations. At the same time, the temperature-concentration boundaries of thermodynamically stable phases found in this work and phases of metastable titanium substitution alloys with iron obtained in [6] (and apparently with other transition metals!) differ quantitatively (see table).

Values of critical concentrations $\omega-\alpha$ and $\alpha-\beta$ phase transitions x_{c1} and x_{c2} at $T \to 0$ K

	Transition	Calculation in this paper		Results calculations in [6]
		Exclusive of $\langle\Delta p^2\rangle$	Considering $\langle\Delta p^2\rangle$	
$Ti_{1-x}Fe_x$	$\omega-\alpha$	3.5	2.1	2.15
	$\alpha-\beta$	15.4	9.41	7.5

5. Conclusion

The electronic transitions considered here between the structural states of titanium alloys are associated with a change in the sign of the mode-mode interaction of electron density fluctuations. The transition from omega- to alpha-phase is accompanied by the replacement of the GU by the GPU crystal structure and is martensitic. At the same time, the sign of the mode-mode parameter changes from positive to negative, which reflects the increased interaction of fluctuation modes. As is known from the fluctuation theory of phase transitions, the latter should induce the replacement of a topological transition of the second kind with a weak transition of the first kind [10]. The transition from the GPU alpha phase to the BCC beta phase is apparently an anti-martensitic transition. In this case, the sign of the parameter of the mode-mode interaction of fluctuations changes to positive, and a topological phase transition of the second kind is realized.

Along with the topological amplification of thermodynamic fluctuations, concentration fluctuations of Hubbard potentials play an important role in the formation of the $x-T$ diagram. In particular, with an increase in the difference in the parameters of the Hubbard interaction in the substitution atom by about 1.1times, there should be suppression of the topological alpha phase and the concentration TEP already in the ground state, which is an additional mechanism for stabilizing the beta phase.

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Conflict of interest

The authors declare that they have no conflict of interest.

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