

# ELECTRONIC STRUCTURE AND ATOMIC ORDERING OF $\text{Fe}_{0.5}\text{Al}_{0.5}$ ALLOY

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The influence of the electron-phonon interaction on the density of states for  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy is studied in the model of a cluster expansion for the thermodynamic potential of a system of electrons and phonons in the coherent potential approximation. The results of calculations of the density of states for theoretically ordered ( $\eta_a = 0.9$ ) and disordered ( $\eta_a = 0$ )  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy (with and without taking into account the electron-phonon interaction), long-range atomic  $\eta_a$  and magnetic  $\eta_m$  order parameters, and local magnetic moments of constituents of the given alloy are presented. It is shown that the account of the electron-phonon interaction results in the effect of “smoothing” of a pseudogap in the density of states upon the establishment of a long-range ordering in  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy. The comparison between the experimentally obtained and theoretically calculated (with and without taking into account the electron-phonon interaction) temperature dependences of the long-range order parameter of  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy is performed.

## Introduction

Aluminides on the basis of transition metals, having a simple crystal structure such as that of CsCl, high temperature stability [1], and soft magnetic properties are of interest for both theoretical researches and practical applications. The Fe–Al system belongs to these compounds and has two stoichiometric phases: FeAl ( $B2$ ) and  $\text{Fe}_3\text{Al}$  ( $DO_3$ ). The atomic ordering considerably influences the physical characteristics and electronic structure of alloys on the basis of transition metals. The results of theoretical researches of equiatomic FeAl alloy testify to that an order-disorder phase transition occurs above the melting point [2]. It is interesting that the disordered state of this compound promotes the appearance of localized magnetic moments, and, therefore, the ferromagnetic ordering [3–5] will be energetically favorable. Calculations of the electronic structure indicate a strong hybridization of the  $s$ ,  $p$ ,  $d$  energy bands at the alloying of iron with aluminum [6]. The explanation of the phase stability through the dominant mechanism of  $d-d$  coupling between atoms of iron and the electron transfer from aluminum to iron induces the interest in studying the electronic structure and its influence on the atomic ordering

of FeAl alloy. The electronic structure of the Fe–Al system was studied both experimentally [7, 8] and theoretically [9–11]. The results of calculations of the density of electronic states testify to the presence of a pseudogap between peaks of the  $d$ -band that is responsible for the stability of the system. The energy bands of the transition metal and the nearest neighbouring interactions of aluminum and iron atoms considerably influence the full density of electronic states of FeAl alloy [12]. The temperature dependence of the long-range order parameter of  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy is experimentally investigated in work [13], but, for a better understanding of the laws of influence of the energy electron spectrum on atomic ordering, it is necessary to take into account other physical factors. The electron-electron and electron-phonon interactions have significant influence on the physical properties of the alloy. To clarify the microscopic mechanism of atomic ordering depending on the electronic structure of FeAl compound, theoretical researches should take into account localized magnetic moments and the electron-phonon interaction in equiatomic FeAl alloy, which is actual for the given work.

## 1. Results of Theoretical and Experimental Investigations of Atomic Ordering of FeAl Alloy

We consider the method of calculations of the electronic structure, long-range order parameters, and localized magnetic moments of the alloy and apply the obtained results to the FeAl alloy. Intermediate calculations are presented in [14].

In the Wannier representation, the Hamiltonian of the system of electrons and phonons of a disordered crystal

$$H = H_0 + H_{\text{int}}, \quad (1)$$

where the zero-approximation Hamiltonian

$$H_0 = \Phi_0 + H_{\text{ph0}} + H_{e0} \quad (2)$$

consist of the Hamiltonian of the subsystem of noninteracting electrons  $H_{e0}$ , Hamiltonian of the subsystem of noninteracting phonons  $H_{ph0}$ , and the energy of the electrostatic interaction of ions  $\Phi_0$  in the equilibrium positions.

The perturbation Hamiltonian in Eq. (1)

$$H_{\text{int}} = H_{ei} + H_{eph} + H_{ee} + H_{phi} \quad (3)$$

consists of the Hamiltonian of the electron-ion interaction  $H_{ei}$ , the Hamiltonian of the electron-phonon interaction  $H_{eph}$ , the Hamiltonian of the pairwise electron-electron interaction  $H_{ee}$ , the Hamiltonian of the phonon-ion interaction  $H_{phi}$ . For the system of electrons and phonons, we can get the temperature Green's function [15]. Using the Fourier transformation and the relations between the spectral representations of the temperature and time Green's functions [16], we have a set of equations for the retarded Green's functions:

$$\begin{aligned} G^{aa^+}(\varepsilon) &= G_0^{aa^+}(\varepsilon) + G_0^{aa^+}(\varepsilon) \times \\ &\times (w + \Sigma_{eph}(\varepsilon) + \Sigma_{ee}(\varepsilon)) G^{aa^+}(\varepsilon), \\ G^{uu}(\varepsilon) &= G_0^{uu}(\varepsilon) + G_0^{uu}(\varepsilon)(\Delta\Phi + \\ &+ \Sigma_{phe}(\varepsilon))G^{uu}(\varepsilon) + G_0^{uu}(\varepsilon)\Delta M^{-1}G^{PP}(\varepsilon), \\ G^{PP}(\varepsilon) &= G_0^{PP}(\varepsilon) + G_0^{PP}(\varepsilon)\Delta M^{-1}G^{PP}(\varepsilon) + \\ &+ G_0^{PP}(\varepsilon)(\Delta\Phi + \Sigma_{phe}(\varepsilon))G^{uu}(\varepsilon). \end{aligned} \quad (4)$$

where  $G^{aa^+}(\varepsilon)$  is the spectral representation of the single-particle Green's function for electrons,  $G^{uu}(\varepsilon)$  and  $G^{PP}(\varepsilon)$  are the "shift-shift" and "momentum-momentum" Green's functions for the phonon subsystem;  $\varepsilon = \hbar\omega$ ; and  $G_0^{aa^+}(\varepsilon)$ ,  $G_0^{uu}(\varepsilon)$ ,  $G_0^{PP}(\varepsilon)$  are the corresponding Green's functions in the zero approximation.

To solution to set (4) is

$$G^{aa^+}(\varepsilon) = \left[ [G_0^{aa^+}(\varepsilon)]^{-1} - (w + \Sigma_{eph}(\varepsilon) + \Sigma_{ee}(\varepsilon)) \right]^{-1}, \quad (5)$$

$$G^{uu}(\varepsilon) = \left[ [G_0^{uu}(\varepsilon)]^{-1} - \left( \frac{\varepsilon^2}{\hbar^2} \Delta M + \Delta\Phi + \Sigma_{phe}(\varepsilon) \right) \right]^{-1} \quad (6)$$

Using the diagrammatic technique [16], we represent the density operators of the Green's functions that describe many-particle interactions in the system as

$$\Sigma_{ephni\gamma, n'i'\gamma'}(\varepsilon) = \sum_{\substack{\lambda_1 n_1 i_1 \\ \lambda_2 n_2 i_2}} c_{n_1 i_1}^{\lambda_1} c_{n_2 i_2}^{\lambda_2} \Sigma_{ephni\gamma, n'i'\gamma'}^{\lambda_1 n_1 i_1, \lambda_2 n_2 i_2}(\varepsilon) \quad (7)$$

for the electron-phonon interaction, where

$$\begin{aligned} \Sigma_{ephni\gamma, n'i'\gamma'}^{\lambda_1 n_1 i_1, \lambda_2 n_2 i_2}(\varepsilon) &= -\frac{1}{4\pi i} \int_{-\infty}^{\infty} d\varepsilon' v'_{ni\gamma, n_3 i_3 \gamma_3}{}^{\lambda_1 n_1 i_1 \alpha_1} \times \\ &\times \left\{ \text{ctgh} \left( \frac{\varepsilon'}{2\Theta} \right) \left[ G_{n_1 i_1 \alpha_1, n_2 i_2 \alpha_2}^{uu}(\varepsilon') - \right. \right. \\ &- G_{n_2 i_2 \alpha_2, n_1 i_1 \alpha_1}^{uu*}(\varepsilon') \left. \right] G_{n_3 i_3 \gamma_3, n_4 i_4 \gamma_4}^{aa^+}(\varepsilon - \varepsilon') + \\ &+ \text{ctgh} \left( \frac{\varepsilon' - \varepsilon - \mu}{2\Theta} \right) G_{n_1 i_1 \alpha_1, n_2 i_2 \alpha_2}^{uu}(\varepsilon - \varepsilon') \times \\ &\times \left[ G_{n_3 i_3 \gamma_3, n_4 i_4 \gamma_4}^{aa^+}(\varepsilon') - G_{n_4 i_4 \gamma_4, n_3 i_3 \gamma_3}^{aa^+*}(\varepsilon') \right] \left. \right\} v'_{n_4 i_4 \gamma_4, n' i' \gamma'}{}^{\lambda_2 n_2 i_2 \alpha_2}. \end{aligned}$$

The phonon-electron interaction is described by the expression

$$\Sigma_{pheni\alpha, n'i'\alpha'}(\varepsilon) = \sum_{\lambda\lambda'} c_{ni}^{\lambda} c_{n'i'}^{\lambda'} \Sigma_{pheni\alpha, n'i'\alpha'}^{\lambda\lambda'}(\varepsilon), \quad (8)$$

where

$$\begin{aligned} \Sigma_{pheni\alpha, n'i'\alpha'}^{\lambda\lambda'}(\varepsilon) &= \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\varepsilon' f(\varepsilon') v'_{n_2 i_2 \gamma_2, n_1 i_1 \gamma_1}{}^{\lambda n_1 \alpha} \times \\ &\times \left\{ \left[ G_{n_1 i_1 \gamma_1, n_3 i_3 \gamma_3}^{aa^+}(\varepsilon + \varepsilon') - G_{n_3 i_3 \gamma_3, n_1 i_1 \gamma_1}^{aa^+*}(\varepsilon + \varepsilon') \right] \times \right. \\ &\times G_{n_2 i_2 \gamma_2, n_4 i_4 \gamma_4}^{aa^+*}(\varepsilon') + G_{n_1 i_1 \gamma_1, n_3 i_3 \gamma_3}^{aa^+}(\varepsilon + \varepsilon') \times \\ &\times \left. \left[ G_{n_4 i_4 \gamma_4, n_2 i_2 \gamma_2}^{aa^+}(\varepsilon') - G_{n_2 i_2 \gamma_2, n_4 i_4 \gamma_4}^{aa^+*}(\varepsilon') \right] \right\} v'_{n_3 i_3 \gamma_3, n_4 i_4 \gamma_4}{}^{\lambda' n' i' \alpha'}. \end{aligned}$$

For the electron-electron interaction, we have

$$\Sigma_{eeni\gamma, n'i'\gamma'}(\varepsilon) = \Sigma_{eeni\gamma, n'i'\gamma'}^{(1)} + \Sigma_{eeni\gamma, n'i'\gamma'}^{(2)}, \quad (9)$$

where

$$\begin{aligned} \Sigma_{een, n'}^{(1)} &= -\frac{1}{4\pi i} \times \\ &\times \int_{-\infty}^{\infty} d\varepsilon' f(\varepsilon') \tilde{v}_{n_1, n'}^{(2)n, n_2} \left[ G_{n_1, n_2}^{aa^+}(\varepsilon') - G_{n_2, n_1}^{aa^+*}(\varepsilon') \right], \end{aligned}$$

$$\begin{aligned} \Sigma_{ee\ n,n'}^{(2)}(\varepsilon) = & -\left(\frac{1}{2\pi i}\right)^2 \int_{-\infty}^{\infty} d\varepsilon_1 \int_{-\infty}^{\infty} d\varepsilon_2 \tilde{v}_{n_2,n_1}^{(2)n,n_3} \times \\ & \times \left\{ f(\varepsilon_1)f(\varepsilon_2) \left[ G_{n_5,n_2}^{aa+*}(\varepsilon - \varepsilon_1 + \varepsilon_2) G_{n_1,n_4}^{aa+}(\varepsilon_1) - \right. \right. \\ & - G_{n_2,n_5}^{aa+}(\varepsilon - \varepsilon_1 + \varepsilon_2) G_{n_4,n_1}^{aa+*}(\varepsilon_1) \left. \right] \times \\ & \times \left[ G_{n_6,n_3}^{aa+}(\varepsilon_2) - G_{n_3,n_6}^{aa+*}(\varepsilon_2) \right] + \\ & + f(\varepsilon_1)f(\varepsilon_1 + \varepsilon_2 - \varepsilon) \left[ G_{n_2,n_5}^{aa+}(\varepsilon_2) - G_{n_5,n_2}^{aa+*}(\varepsilon_2) \right] \times \\ & \times \left[ G_{n_1,n_4}^{aa+}(\varepsilon_1) G_{n_6,n_3}^{aa+}(\varepsilon_1 + \varepsilon_2 - \varepsilon) - \right. \\ & \left. - G_{n_4,n_1}^{aa+*}(\varepsilon_1) G_{n_3,n_6}^{aa+*}(\varepsilon_1 + \varepsilon_2 - \varepsilon) \right] \left. \right\} \tilde{v}_{n_6,n'}^{(2)n_4,n_5}, \\ \tilde{v}_{n_1,n'}^{(2)n,n_2} = & v_{n_1,n'}^{(2)n,n_2} - v_{n',n_1}^{(2)n,n_2}, \quad (n \equiv ni\gamma). \end{aligned} \quad (10)$$

The densities of states for the subsystems of electrons and phonons per atom calculated by averaging the relations for the Green's functions (5), (6) over the positions of atoms at the sites of the crystal lattice are written, respectively, as:

$$g_e(\varepsilon) = -\frac{1}{\pi\nu N} \text{ImSp} \langle G^{aa+}(\varepsilon) \rangle_{av}, \quad (11)$$

$$g_{ph}(\varepsilon) = -\frac{1}{\pi\nu N} 2\frac{\varepsilon}{\hbar^2} M_A \text{ImSp} \langle G^{uu}(\varepsilon) \rangle_{av}. \quad (12)$$

Angular brackets  $\langle \dots \rangle_{av}$  means the configurational averaging.

Relations (5), (6) differ from the expressions for the Green's function of the single-particle Hamiltonians of the disordered system only by the form of density operators. For the calculation of the Green's functions (5), (6), we use the known methods of the theory of disordered systems [15]. The density operators are represented as the sums of single-site operators, the Green's functions of the effective medium being chosen as the zero single-site approximation. The Green's function of the effective medium for the electron subsystem is

$$\begin{aligned} \tilde{G}^{aa+}(\varepsilon) = & \left[ G_0^{aa+}(\varepsilon) \right]^{-1} - \\ & - \left( \Sigma_{eph}^A(\varepsilon) + \Sigma_{ee}(\varepsilon) + \sigma_e(\varepsilon) \right)^{-1}, \end{aligned} \quad (13)$$

where  $\Sigma_{eph}^A(\varepsilon)$  is the density operator of the electron-phonon interaction in a pure crystal. The Green's function of the effective medium for the phonon subsystem is

$$\tilde{G}^{uu}(\varepsilon) = \left[ G_0^{uu}(\varepsilon) \right]^{-1} - \left( \Sigma_{phe}^A(\varepsilon) + \sigma_{ph}(\varepsilon) \right)^{-1}, \quad (14)$$

where  $\Sigma_{phe}^A(\varepsilon)$  is the density operator of the phonon-electron interaction in a pure crystal. In expressions (13) and (14),  $\sigma_e(\varepsilon)$  and  $\sigma_{ph}(\varepsilon)$  are the potentials of the effective medium (coherent potentials). The cluster expansion of the Green's functions allows us to write the relations for a scattering  $T$  matrix in the form of a series whose terms describe the scattering by clusters with various numbers of sites [17]:

$$T = \sum_{(n_1 i_1)} t^{n_1 i_1} + \sum_{(n_1 i_1) \neq (n_2 i_2)} T^{(2)n_1 i_1, n_2 i_2} + \dots \quad (15)$$

The configurational averaging of expression (15) and the transition to the  $\vec{k}$ -representation for the averaged scattering  $T$  matrix of the electron subsystem gives, upon neglecting the processes of scattering by clusters with three and more sites,

$$\begin{aligned} \langle T_{i\gamma, i'\gamma'} \rangle(\vec{k}, \varepsilon) = & \sum_{\substack{i_1 \\ (n_2 i_2) \neq (0i_1) \\ n, n'}} \sum_{\lambda_1, \lambda_2} \langle c_{0i_1}^{\lambda_1} c_{n_2 i_2}^{\lambda_2} \rangle \times \\ & \times T_{ni\gamma, n'i'\gamma'}^{(2)\lambda_1 0i_1, \lambda_2 n_2 i_2} e^{i\vec{k}(\vec{r}_{n'} + \vec{\rho}_{i'} - \vec{r}_n - \vec{\rho}_i)}, \end{aligned} \quad (16)$$

where  $T_{ni\gamma, n'i'\gamma'}^{(2)\lambda_1 n_1 i_1, \lambda_2 n_2 i_2}$  is the matrix element of the operator of scattering by a two-particle cluster  $T^{(2)n_1 i_1, n_2 i_2}$ , in which atoms of types  $\lambda_1$  and  $\lambda_2$  are located at the sites  $(n_1 i_1)$  and  $(n_2 i_2)$ , respectively. The expression for the scattering  $T$  matrix of the phonon subsystem is obtained from (16) by replacing the index of the energy band  $\gamma$  by the index of the projection of the vector of displacement of the atom onto the Cartesian coordinate axis  $\alpha$ . In the expression (16),  $\langle c_{n_1 i_1}^{\lambda_1} c_{n_2 i_2}^{\lambda_2} \rangle = c_{i_1}^{\lambda_1} c_{i_2}^{\lambda_2} + \varepsilon_{n_1 i_1, n_2 i_2}^{\lambda_1 \lambda_2}$  are the probabilities that the sites  $(n_1 i_1)$  and  $(n_2 i_2)$  are occupied by the atoms of type  $\lambda_1$  and  $\lambda_2$ , respectively;  $\langle c_{ni}^\lambda \rangle = c_i^\lambda$  are the probabilities that the sites of the  $i$ -th sublattice are occupied by atoms  $\lambda$ . For the cubic binary alloy, we obtain

$$c_i^A = x_i = c^A - \frac{\nu_2}{\nu} \eta, \quad c_i^B = x_i = c^B - \frac{\nu_2}{\nu} \eta$$

for  $\nu_1$  sublattices of the first type and

$$x_i = c^A + \frac{\nu_1}{\nu} \eta$$

for  $\nu_2$  sublattices of the second type, where  $\nu = \nu_1 + \nu_2$ ,  $\eta$  is the long-range order parameter,  $c^A$  and  $c^B = 1 - c^A$  are the concentrations of the components  $A$  and  $B$  of the crystal, respectively;  $\varepsilon_{n_1 i_1, n_2 i_2} = \varepsilon_{n_1 i_1, n_2 i_2}^{AA} = \langle (c_{n_1 i_1}^A - c_{i_1}^A)(c_{n_2 i_2}^A - c_{i_2}^A) \rangle$  are the parameters of pairwise interatomic correlations.

The thermodynamic potential of the system is defined as

$$\Omega = \Omega_0 + \Omega', \quad (17)$$

where  $\Omega_0$  is the thermodynamic potential of the zero approximation, and  $\Omega'$  is the thermodynamic potential of the interaction. The thermodynamic potential  $\Omega_0$  in relation (17) in the absence of interaction can be written as

$$\Omega_0 = \Omega_{\text{conf}} + \Omega_{0e} + \Omega_{0\text{ph}}, \quad (18)$$

where the thermodynamic potential of the electron subsystem is

$$\Omega_{0e} = -\nu N \Theta \int_{-\infty}^{\infty} \ln \left( 1 + e^{(\mu_e - \varepsilon)/\Theta} \right) g_{0e}(\varepsilon) d\varepsilon,$$

and that of the phonon subsystem is

$$\Omega_{0\text{ph}} = \nu N \Theta \int_{-\infty}^{\infty} \ln \left( 1 - e^{-\varepsilon/\Theta} \right) g_{0\text{ph}}(\varepsilon) d\varepsilon.$$

The density of electronic states  $g_{0e}(\varepsilon)$ ,  $g_{0\text{ph}}(\varepsilon)$  are given by (11), (12).

The configurational component of the thermodynamic potential  $\Omega_{\text{conf}}$  in the expression (18), which depends on the distribution of atoms of various types over the crystal lattice sites, reads

$$\Omega_{\text{conf}} = \langle \Phi_0 \rangle - \Theta S_{\text{conf}}, \quad (19)$$

where  $S_{\text{conf}}$  is the configurational entropy,  $S_{\text{conf}} = -\langle \ln P_{\text{conf}} \rangle$ ,  $P_{\text{conf}}(\{c_{ni}^\lambda\})$  is the distribution function of atoms over the lattice sites.

The free energy  $F$  as a function of the volume  $V$  of the system, its temperature  $T$ , the number of electrons  $N_e$  and the parameters of interatomic correlations  $(\varepsilon_{n_1 i_1, n_2 i_2}, \eta)$  which is related to the thermodynamic potential  $\Omega$  by the expression  $F = \Omega + \mu_e \langle N_e \rangle$  is

$$F = \langle \Phi_0 \rangle - \Theta S_{\text{conf}} + \Omega_{0e} + \Omega_{0\text{ph}} + \Omega' + \mu_e \langle N_e \rangle. \quad (20)$$

The configurational part of the alloy entropy  $S_{\text{conf}}$  in (20) is related to the probabilities of distribution of

atoms over the lattice sites  $P_{n_1 i_1 \dots n_\nu i_\nu}^{\lambda_1 \dots \lambda_\nu}$  by the expression [18]

$$S_{\text{conf}} = - \left[ \sum_{(n_1 i_1), \lambda_1} P_{n_1 i_1}^{\lambda_1} \ln P_{n_1 i_1}^{\lambda_1} + \frac{1}{2} \sum_{\substack{(n_1 i_1) \neq (n_2 i_2) \\ \lambda_1, \lambda_2}} P_{n_1 i_1, n_2 i_2}^{\lambda_1 \lambda_2} \ln \frac{P_{n_1 i_1, n_2 i_2}^{\lambda_1 \lambda_2}}{P_{n_1 i_1}^{\lambda_1} P_{n_2 i_2}^{\lambda_2}} + \dots \right], \quad (21)$$

where

$$P_{ni}^\lambda = \langle c_{ni}^\lambda \rangle, \quad P_{n_1 i_1, n_2 i_2}^{\lambda_1 \lambda_2} = \langle c_{n_1 i_1}^{\lambda_1} c_{n_2 i_2}^{\lambda_2} \rangle.$$

The equilibrium values of the long-range order parameter  $\eta_a$  are determined from the condition of the free energy minimum of the alloy [15]

$$\frac{\partial F}{\partial \eta_a} = 0. \quad (22)$$

The Fermi level  $\mu_e$  of the system is determined from the equation

$$\langle Z \rangle = \int_{-\infty}^{\infty} f(\varepsilon, \mu_e) g_e(\varepsilon) d\varepsilon, \quad (23)$$

where  $g_e(\varepsilon)$  is given by (11),  $\langle Z \rangle = \langle N_e \rangle / \nu N$  is the average number of electrons per atom, and  $\langle Z \rangle = c^A Z_A + c^B Z_B$ .

The obtained results allow us to study the influence of the electron-phonon interaction on the energetic spectrum of electrons, free energy, and long-range order parameters of the FeAl alloy. In addition, the influence of electron correlations on the electron structure of the alloy may be taken into account. We assume that, in the expression for the free energy of the alloy (20), the number of electrons  $Z_{ni\gamma\sigma}^{m\lambda_i}$  in the state  $(ni\gamma\sigma)$ , i.e. the number of electrons per atom at the site  $(ni)$  and in the energy band  $\gamma$  for a projection of spin  $\sigma$ , depends on the type of the atom  $\lambda$  and on the local magnetic moment  $m_{\lambda_i}$  at a given site  $(ni)$ . The magnitude of  $Z_{ni\gamma\sigma}^{m\lambda_i}$  is determined by (23), where the density of electron states  $g_e(\varepsilon)$  is replaced by the conditional partial density of states  $g_{ni\gamma\sigma}^{m\lambda_i}(\varepsilon)$  for the energy band  $\gamma$  and the spin projection  $\sigma$ , which looks as

$$g_{ni\gamma\sigma}^{m\lambda_i}(\varepsilon) = -\frac{1}{\pi} \text{Im} \langle G_{ni\gamma\sigma, ni\gamma\sigma}^{aa+}(\varepsilon) \rangle \Big|_{(ni) \in m_{\lambda_i}}. \quad (24)$$

Here, the averaging is performed under the condition that the site  $(ni)$  is occupied by an atom of type  $\lambda$  and

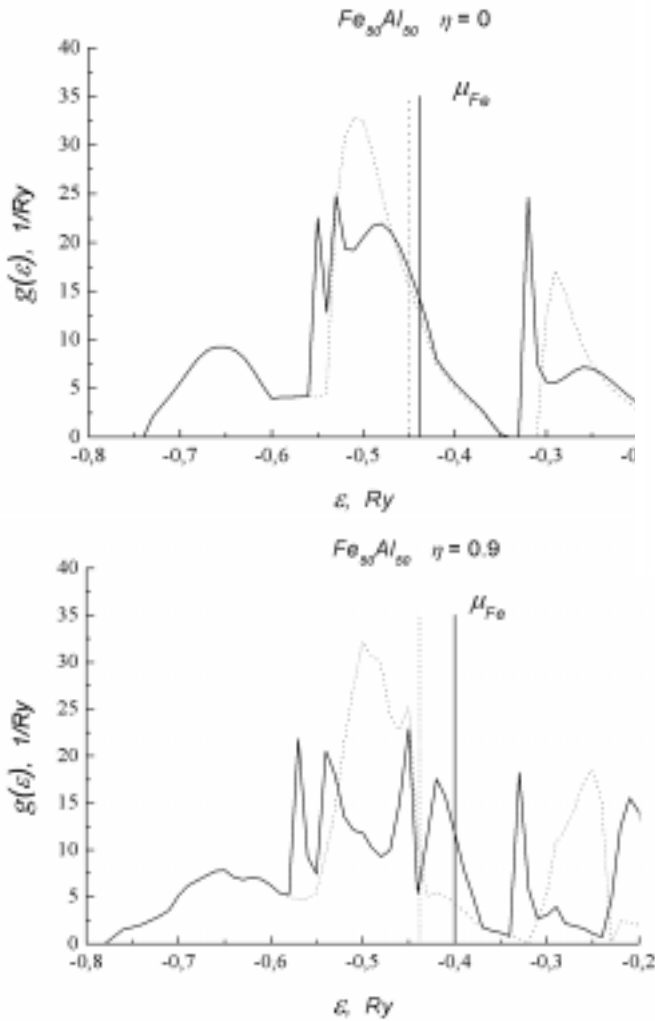


Fig.1. Density of electronic states for hypothetical disordered ( $\eta_a=0$ ) and ordered ( $\eta_a=0.9$ ) equiatomic FeAl alloy without (a dotted curve) and with taking into account (a continuous curve) electron-phonon interaction

that the projection of the localized magnetic moment of electrons is  $m_{\lambda i}$ . We also represent the relations

$$Z_{\lambda i \gamma} = Z_{ni \gamma \sigma}^{m_{\lambda i}} + Z_{ni \gamma -\sigma}^{m_{\lambda i}}, \quad m_{\lambda i \gamma} = Z_{ni \gamma \sigma}^{m_{\lambda i}} - Z_{ni \gamma -\sigma}^{m_{\lambda i}}, \quad (25)$$

where  $Z_{\lambda i} = \sum_{\gamma} Z_{\lambda i \gamma}$  and  $m_{\lambda i} = \sum_{\gamma} m_{\lambda i \gamma}$  are the number of electrons and the value of the projection of the magnetic moment per atom of type  $\lambda$  at the site ( $ni$ ), respectively.

The configurational entropy  $S_{\text{conf}}$  of the alloy in (20) is determined by the probabilities of distribution of atoms and localized magnetic moments over the lattice sites ( $ni$ ) according to expression (21), in which the

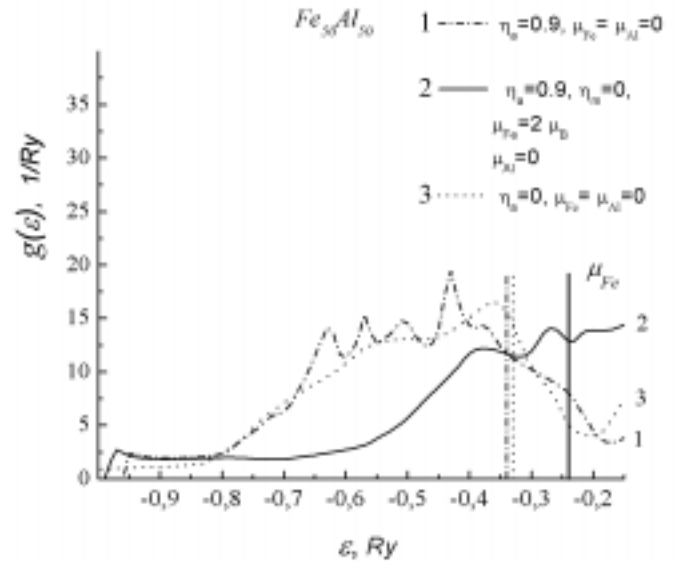


Fig. 2. Density of electronic states for hypothetical ordered ( $\eta=0.9$ ) (1 and 2) and disordered ( $\eta=0$ ) (3) of the  $\text{Fe}_{0.5}\text{Al}_{0.5}$  intermetallide without (1 and 3) and with taking into account (2) localized magnetic moments

following substitution must be performed:

$$\lambda \rightarrow m_{\lambda i}, \quad \sum_{\lambda} (...) \rightarrow \sum_{\lambda, m_{\lambda i}} (...).$$

The projections of the localized magnetic moments  $m_{\lambda i}$  and the correlation parameters for the orientations of magnetic moments at the lattice sites are found from the condition of the minimum of free energy  $F$  (20).

The densities of electronic states for hypothetical disordered ( $\eta=0$ ) and ordered ( $\eta=0.9$ )  $\text{Fe}_{0.5}\text{Al}_{0.5}$  BCC-alloys with the account (a continuous curve) and without taking into account (a dotted curve) the electron-phonon interaction at  $T = 900$  K are presented in Fig.1. According to the results of work [14], the influence of the electron-phonon interaction reduces to the effect of “smoothing” of a pseudogap in the energy electron spectrum, which is seen in the drawn plots.

In the energy electron spectrum at the long-range ordering, the arisen pseudogap is related to the splitting of the  $p$ - and  $d$ -bands. The Fermi’s level gets in the pseudogap region. At the establishment of a long-range order, the occurrence of the pseudogap results in the decrease in the free energy of the alloy, which makes the atomic ordering energetically favorable. The location of the Fermi level in the pseudogap region is the reason for the system to be an intermetallide and to exist in a completely ordered state up to the melting point [2].

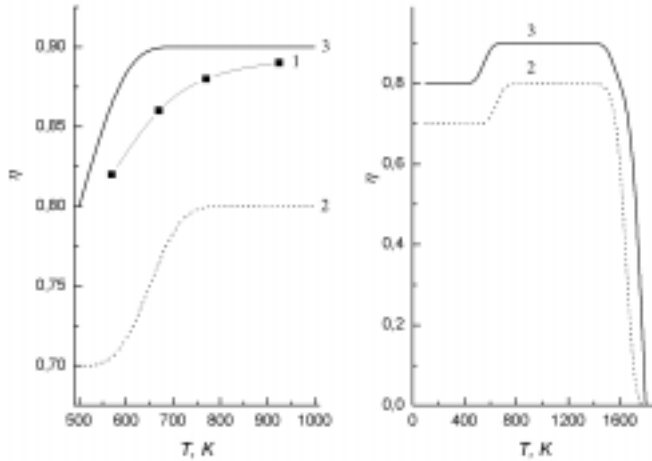


Fig. 3. Temperature dependence of long-range order parameter  $\eta_a(T)$  for  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy: 1 — experimental data; 2 — theoretical data without influence of electron-phonon interaction; 3 — with taking into account electron-phonon interaction

The results of calculations of the localized magnetic moments ( $\mu_{\text{Fe}}, \mu_{\text{Al}}$ ), parameters of atomic  $\eta_a$  and magnetic  $\eta_m$  orderings at  $T = 0$  K for the  $\text{Fe}_{0.5}\text{Al}_{0.5}$  alloy are given in Fig. 2. Curve 1 shows the density of electronic states of hypothetical ordered ( $\eta_a = 0.9$ ) FeAl alloy without influence of the localized magnetic moments, curve 2 — with account of magnetic moments at the magnetic disordering ( $\eta_m = 0$ ), and curve 3 — the density of electronic states without taking into account the magnetic moments in the atomic disordering state ( $\eta_a = 0$ ). The equilibrium maximal value of the parameter of atomic long-range order  $\eta_a$  is 0.9. As seen in Fig. 2, the calculations with electron-electron correlations indicate that the magnet disordering state is the most energetically favorable (the magnetic moments of components  $\mu_{\text{Fe}} \approx 2\mu_{\text{B}}, \mu_{\text{Al}} \approx 0$ , and the average magnetic moment of the alloy per atom  $\mu \approx 1\mu_{\text{B}}$ ). The correlations between the magnetic moments for the alloys of transition metals are weak ( $\mu \approx \mu_{\text{B}}$ ), so the ground state is paramagnetic. The deviation of the B2-structure from the ordered state results in the increase of the probabilities of occupation of sites, which are characteristic of aluminum atoms, by iron atoms. This is the reason for the appearance of localized magnetic moments on iron atoms due to the interaction between the clusters consisting of iron atoms. So, the ferromagnetic state is energetically favorable.

The experimentally derived temperature dependence of the long-range order parameter  $\eta(T)$  represented

by curve 1 is shown in Fig. 3. For comparison, the dotted curve 2 represents the theoretically calculated dependence  $\eta(T)$  without taking into account the influence of electron-phonon interactions, and curve 3 — taking into account of electron-phonon interactions. The thermodynamic characteristics of the given alloy are calculated with regard for the electron-phonon interactions. We have theoretically predicted the existence of the order — disorder phase transition near the melting point of FeAl alloy that is coordinated with the results of work [9]. In the region of temperatures  $T = 500 \div 700$  K, the significant contribution to the temperature dependence of the free energy is given by the Fermi—Dirac distribution. The influence of the Fermi factor results to a nonlinear behavior of the long-range order parameter near a temperature of 500 K. Without taking into account the electron-phonon interaction, the experimental and theoretical values of the long-range order parameter are considerably different.

## Conclusions

The derived results allow investigating the influence of multiparticle interactions on the spectrum of elementary perturbations, free energy, and parameters of interatomic correlations of the alloy. It is shown that, at the long-range ordering of the alloy, the pseudogap arises at the center of the energy band whose width grows with the long-range order parameter  $\eta$ . The establishment of a long-range order decreases the density of electronic states at the Fermi's level, which results in a reduction of the free energy of FeAl alloy, i.e. the position of the Fermi level in the pseudogap region is a reason for the temperature stability of the given system. The account of electron-electron correlations in FeAl alloy indicates that, at a long-range order, the paramagnetic state of the given compound is most favorable in energy. The influence of electron-phonon interactions results in the effect of “smoothing” a pseudogap in the energy electron spectrum of the FeAl alloy at the establishment of a long-range order.

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ЕЛЕКТРОННА СТРУКТУРА І АТОМНЕ  
ВПОРЯДКУВАННЯ СПЛАВУ Fe<sub>0,5</sub>Al<sub>0,5</sub>

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## Резюме

В моделі кластерного розкладання термодинамічного потенціалу системи електронів і фононів із застосуванням наближення когерентного потенціалу вивчено вплив електрон-фононної взаємодії на щільність електронних станів еквіатомного сплаву Fe<sub>0,5</sub>Al<sub>0,5</sub>. Наведено результати розрахунку щільності станів для гіпотетичного впорядкованого ( $\eta_a = 0,9$ ) та неупорядкованого ( $\eta_a = 0$ ) сплаву Fe<sub>0,5</sub>Al<sub>0,5</sub> (з врахуванням та без врахування електрон-фононної взаємодії), параметрів атомного  $\eta_a$  і магнітного  $\eta_m$  впорядкування та локалізованих магнітних моментів даної сполуки. Показано, що врахування електрон-фононної взаємодії приводить до ефекту замивання квазіщільни в електронному спектрі при встановленні далекого впорядкування в сплаві Fe<sub>0,5</sub>Al<sub>0,5</sub>. Проведено порівняння між експериментально отриманою та розрахованими (з врахуванням електрон-фононної взаємодії та без такої) температурними залежностями параметра далекого порядку сполуки Fe<sub>0,5</sub>Al<sub>0,5</sub>.