STRONG ELECTRON CORRELATIONS IN SHORT-RANGE MAGNETIC ORDER AND ELECTRICAL RESISTANCE OF HOMOGENEOUSLY DISORDERED BINARY CRYSTALS

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The formation of both the short-range magnetic order and the electrical conductivity in homogeneously disordered binary crystals under the influence of strong electron correlations is considered (by the example of b.c.c.-$Fe_{1-c}Co_c$ alloys). For the description of electron states in a crystal, the multiband model of a tight binding and the method of the cluster expansion for Green's functions and the thermodynamic potential of a disordered crystal are used. Strong electron correlations and the well-developed short-range order of substitutional atoms lead to the appearance of a quasigap in the electron-energy spectrum. The microscopic mechanisms of magnetic ordering and formation of the electrical resistance are concerned with both the Fermi-level position within the quasigap region and the realignment of the electron-energy spectrum with changes of the temperature or the alloying-component concentration. The parameter of pairwise magnetic correlations decreases with increase in the temperature and tends to zero in a vicinity of the Curie temperature. The nonmonotonic concentration dependence of the Fe−Co-alloy electrical resistance is investigated as well. It is caused by strong electron correlations and the magnetic order resulting from these correlations.

1. Introduction

Strong electron–electron correlations in a crystal influence significantly the formation of its magnetic and electrical properties. In Fe−Co alloy, the Coulomb interaction between *d*-electrons is strong, and sequent strong electron correlations lead to the appearance of a 'Coulomb' quasigap in the electron-energy spectrum. Herewith, the location of the Fermi level within the quasigap range is concerned with the nature of a weak temperature dependence of the electrical resistance [1, 2], appearance of spin polarized transport effects, and high saturation magnetization in Fe−Co alloy [3, 4].

In a given paper, a change of the short-range magnetic order with increase in the temperature and a role of strong electron–electron correlations in the formation of the concentration dependence of the electrical resistance for b.c.c.-$Fe_{1-c}Co_c$ alloys due to the location of the Fermi level within the 'Coulomb' quasigap range are investigated.

2. Theory

For the description of electron states in a crystal, the multiband tight-binding model and the method developed in [5] for the cluster expansion of one- and two-particle Green's functions and the thermodynamic potential of a disordered-alloy crystal are used. Within the mentioned method, the coherent potential approximation [6] is chosen as a zeroth-order one-site approximation. This approach allows one to consider the electron scattering by different-kind ion potentials and fluctuations of the spin and charge densities with regard for correlations in the atom arrangement and orientations of the localized magnetic moments at respective lattice sites. The Hamiltonian matrix elements are calculated within the scope of the Slater–Koster method of linear combination of atomic orbitals in combination with the Löwdin orthogonalization procedure [7–9].

The Hamiltonian of the electrons' and phonons' subsystems of a disordered crystal is determined by the ex-

pression

$$H = H_0 + H_{\rm int}, \tag{1}$$

where the zeroth-order approximation Hamiltonian, H_0, and the perturbation Hamiltonian, $H_{\rm int}$, have the following forms:

$$H_0 = \Phi_0 + H_{\rm ph0} + H_{e0}, H_{\rm int} = H_{ei} + H_{e\rm ph} + H_{ee}. \tag{2}$$

Here, H_0 consists of the energy of the electrostatic interaction of ions in equilibrium, Φ_0, and Hamiltonians of the subsystems of noninteracting phonons and electrons, $H_{\rm ph0}$ and H_{e0}, respectively; $H_{\rm int}$ consists of the Hamiltonians of electron–ion, electron–phonon, and pairwise electron–electron interactions, H_{ei}, $H_{e\rm ph}$, H_{ee}, respectively. Complete expressions for above-mentioned Hamiltonians within the scope of the Wannier representation are given in [5].

In our work, within the scope of the mentioned approach, the calculations of both the concentration dependence of the electrical resistance for b.c.c.-$Fe_{1-c}Co_c$ alloys and the temperature-dependent equilibrium values of the pairwise magnetic-correlation parameters of disordered b.c.c.-$Fe_{0.5}Co_{0.5}$ alloy are performed. The equilibrium values of parameters of magnetic and interatomic correlations are obtained from the condition that the free energy is minimal.

The free energy, F, as a function of the system volume (V), temperature (T), number of electrons (N_e), and parameters of both the interatomic correlations and the correlations in an orientation of localized magnetic moments is defined in terms of the thermodynamic potential, Ω, by the relation [5]

$$F = \langle\Phi_0\rangle - TS_c + \Omega_{0e} + \Omega_{0\rm ph} + \Omega' + \varepsilon_F\langle N_e\rangle. \tag{3}$$

Here, Ω_{0e} – the thermodynamic potential of noninteracting electrons; $\Omega_{0\rm ph}$ – the thermodynamic potential of noninteracting phonons; Ω' – the thermodynamic potential contribution which is caused by the processes of electrons' and phonons' scatterings in a disordered crystal; ε_F – the chemical potential of electrons; $\langle\Phi_0\rangle$ – the average value of electrostatic interaction energy of ions; S_c – the configuration entropy of ions' subsystem. The configuration contribution, S_c, is explicitly determined by parameters of a short-range atomic order [5]. Moreover, the contribution of the electron subsystem depends on the parameters of short-range atomic and magnetic orders by means of the one-particle Green's function of the electron subsystem. This function determines the electron density of states as well.

Within the two-center cluster approximation, the electron density of states has the form [5]

$$g_e(\varepsilon) = -\frac{1}{N\pi}{\rm ImSp}\langle G(\varepsilon)\rangle =$$

$$= \frac{1}{\nu}\sum_{i,\gamma,\sigma,\lambda,m_{\lambda 0i\gamma}} P_{0i}^{\lambda,m_{\lambda 0i\gamma}} g_{0i\gamma\sigma}^{\lambda,m_{\lambda 0i\gamma}}(\varepsilon), \tag{4}$$

where ε – the energy parameter; $\langle G(\varepsilon)\rangle$ – the configuration averaged Green's function of electrons; ν – the number of sites within a conditional unit cell; N – the total number of lattice sites; $P_{0i}^{\lambda,m_{\lambda ni\gamma}}$ – the probability to find a λ-kind atom with the localized magnetic moment $m_{\lambda ni\gamma}$ at the site (ni), where the first index, n, in a bracket numbers a conditional cell in a crystal, and the second index, i, indicates the number of a sublattice in this conditional cell; $g_{0i\gamma\sigma}^{\lambda,m_{\lambda 0i\gamma}}(\varepsilon)$ – the conditional partial electron density of states for the energy band γ with the projection of spin σ, which is defined by one-particle Green's functions and by means of the conditional probabilities to find localized magnetic moments and different-kind atoms at the nearest sites of a crystal lattice under condition that, at the site $(0i)$, there is a λ-kind atom with the localized magnetic moment of electrons $m_{\lambda 0i\gamma}$ [2].

The Fermi level, ε_F, of electrons in a crystal is defined by the formula [5]

$$\langle Z\rangle = \int\limits_{-\infty}^{\infty} f(\varepsilon,\varepsilon_F) g_e(\varepsilon) d\varepsilon. \tag{5}$$

Here, $\langle Z\rangle$ is the average number of electrons per atom; and $f(\varepsilon,\varepsilon_F)$ is the Fermi function.

The localized magnetic moment $m_{\lambda ni}$ is defined by the difference between conditional partial electron densities of states with different spin projections at the given site:

$$m_{\lambda ni} = \sum_{\gamma} m_{\lambda ni\gamma},$$

$$m_{\lambda ni\gamma} = \int\limits_{-\infty}^{\infty} f(\varepsilon,\varepsilon_F)(g_{ni\gamma\sigma}^{\lambda,m_{\lambda ni\gamma}}(\varepsilon) - g_{ni\gamma-\sigma}^{\lambda,m_{\lambda ni\gamma}}(\varepsilon))d\varepsilon. \tag{6}$$

Let the localized magnetic moment have only two projections on a quantization axis: $m_{\lambda ni} = \mu_{\lambda ni}^{\uparrow}$ (up), $\mu_{\lambda ni}^{\downarrow}$ (down), and let their values be determined by both the electron–electron interaction and the kind of an atom at

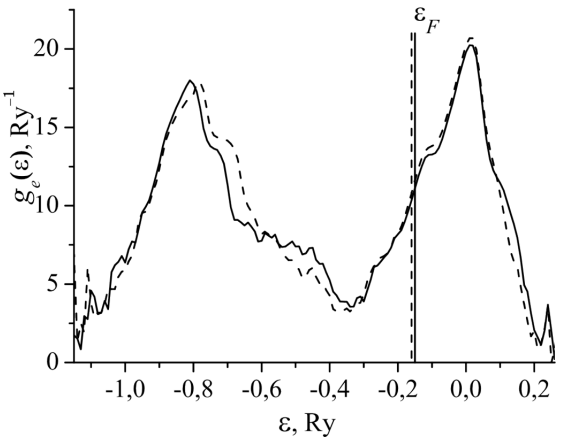

Fig. 1. Electron density of states of b.c.c.-$Fe_{0.5}Co_{0.5}$ alloy with equilibrium parameters of atomic and magnetic orders at 300 K (—) and 500 K (- - -)

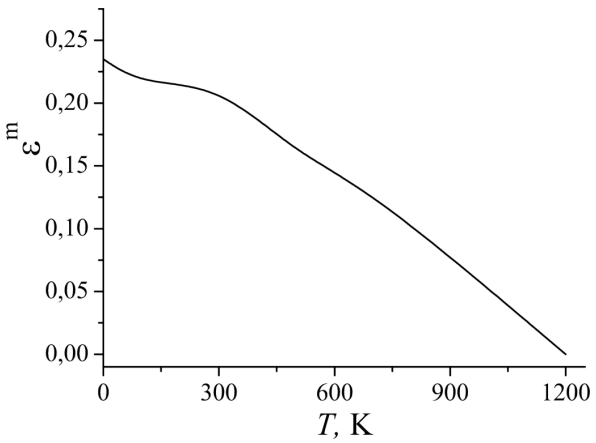

Fig. 2. Temperature dependence of equilibrium values of the pairwise magnetic-correlation parameter ε^m for b.c.c.-$Fe_{0.5}Co_{0.5}$ alloy

the given site. Equilibrium values of the local characteristics of the magnetic structure are determined by means of the minimization of the free energy by the parameter of pairwise magnetic correlations, $\varepsilon^{m\mu^{\uparrow}_{\lambda n_1 i_1}\mu^{\uparrow}_{\lambda' n_2 i_2}}_{n_1 i_1 n_2 i_2}$, in the whole definition interval of it. The parameter of pairwise magnetic correlations is concerned with the conditional probability, $P^{\mu^{\uparrow}_{\lambda n_1 i_1}/\mu^{\uparrow}_{\lambda' n_2 i_2}}_{n_1 i_1 n_2 i_2}$ (*i.e.* the probability to find a localized magnetic moment $\mu^{\uparrow}_{\lambda n_1 i_1}$ at the site $(n_1 i_1)$ under condition that a magnetic moment $\mu^{\uparrow}_{\lambda' n_2 i_2}$ is localized at the site $(n_2 i_2)$), by the formula [10]

$$\varepsilon^{m\mu^{\uparrow}_{\lambda n_1 i_1}\mu^{\uparrow}_{\lambda' n_2 i_2}}_{n_1 i_1 n_2 i_2} = P^{\mu^{\uparrow}_{\lambda' n_2 i_2}}_{n_2 i_2}(P^{\mu^{\uparrow}_{\lambda n_1 i_1}/\mu^{\uparrow}_{\lambda' n_2 i_2}}_{n_1 i_1 n_2 i_2} - P^{\mu^{\uparrow}_{\lambda n_1 i_1}}_{n_1 i_1}). \quad (7)$$

Expressions (4)–(7) describe the interrelation between the magnetic-order formation in an alloy and its electronic structure.

The exact expression for the static electrical conductivity of an alloy, $\sigma_{\alpha\beta}$, was derived in [2] as follows:

$$\sigma_{\alpha\beta} = -\frac{e^2\hbar}{N\Omega_0}\int\limits_{-\infty}^{\infty} d\varepsilon(-\frac{\partial f(\varepsilon,\varepsilon_F)}{\partial\varepsilon})\times$$

$$\times \mathrm{Sp}\langle \upsilon_\alpha(G(\varepsilon^+) - G(\varepsilon^-))\upsilon_\beta(G(\varepsilon^+) - G(\varepsilon^-))\rangle. \quad (8)$$

Here, $G(\varepsilon^{\pm}) \equiv G(\varepsilon \pm i\delta) = (\varepsilon^{\pm} - H)^{-1}$ is the retarded ('+') or advanced ('−') Green's function of an alloy crystal, H – the one-electron Hamiltonian of an alloy, $\delta \to +0$; $G = \tilde{G} + \tilde{G}\hat{T}\tilde{G}$, where $\tilde{G}$ – the Green's function for the effective medium, $\hat{T}$ – the matrix of electron scattering by the random potential; υ_α – the α-component of the electron velocity vector operator; Ω_0 – the primitive unit-cell volume; e – the electron charge; $\hbar$ – the Planck's constant; the brackets $\langle ... \rangle$ denote the configuration averaging.

3. Numerical Results

The Hamiltonian matrix elements calculated for the Fe–Co alloy within the scope of the Slater–Koster method determine values of the atomic scattering potentials for d-electrons which are equal to 0.04 (in units of a respective band width, w_d) by order of magnitude. At the same time, the estimation of the parameter, U_d, of the Coulomb interaction between d-electrons at the one site shows that the strong electron-correlation regime takes place in the given system, because $U_d/w_d \sim 0.55$. In this case, the splitting of energy bands (for d-electrons) in the Fe–Co alloy is caused by the Coulomb interaction between electrons more than by its scattering on the crystal potential.

Phase transitions of alloys with strong electron correlations are caused by changes of an electronic structure. The realignment of the electron-energy spectrum occurs with changing the temperature (Fig. 1). An increase in the temperature leads to a 'tailing' of the 'Coulomb' energy quasigap on the curve of the electron density of states due to the scattering of electrons by vibrations of the crystal lattice (*i.e.* electron–phonon interaction). An increase in the temperature leads also to the damping of the correlations in an orientation of the magnetic moments or, in other words, to a decrease of the param-

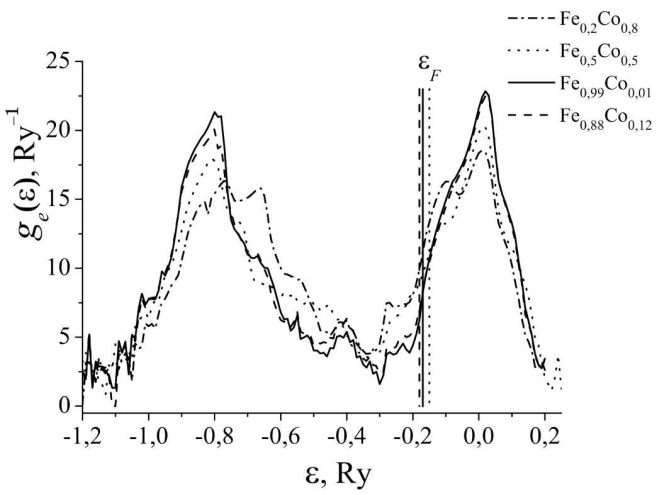

Fig. 3. Electron density of states of b.c.c.-$Fe_{1-c}Co_c$ alloys with equilibrium values of atomic and magnetic orders at different values of concentration, c

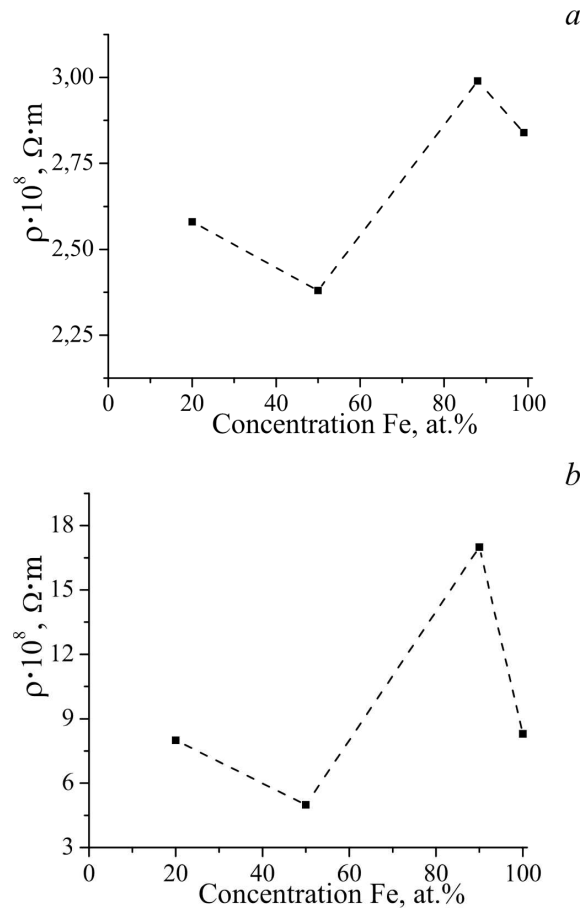

Fig. 4. Concentration dependences of resistivity, $\rho = 1/\sigma_{zz}$, for b.c.c.-$Fe_{1-c}Co_c$ alloys at $T = 300$ K: a – computation results according to (5); b – experimental values [13]

eter of pairwise magnetic correlations (Fig. 2) up to zero at a temperature of $\cong 1200$ K, which is close to the experimental values of the Curie temperature ($T_C \cong 1250$ K) for FeCo alloy [11, 12]. In the presented results of numerical calculations, the short-range magnetic order is determined by the parameter of pairwise correlations in an orientation of magnetic moments at the first coordination sphere only, $\varepsilon^m \equiv \varepsilon^{m\mu^{\uparrow}_{\lambda 01}\mu^{\uparrow}_{\lambda' m2}}_{01m2}$. Note that the equilibrium values of ε^m correspond to the ferromagnetic phase (*i.e.* $\varepsilon^m > 0$).

A change of the chemical composition of an alloy leads to a change of the partial contributions of alloy's components to the electron density of states (Fig. 3). At the same time, according to expression (6), the values of localized magnetic moments and the density of states at the Fermi level within the energy quasigap change that influences the formation of conductivity (8) of a binary alloy with strong electron correlations.

The concentration-dependent electrical resistance of b.c.c.-$Fe_{1-c}Co_c$ alloys at a temperature of 300 K is presented in Fig. 4,*a*. For the comparison, the experimental values of b.c.c.-$Fe_{1-c}Co_c$ resistivity [13] are given in Fig. 4,*b*.

The results of computations conform qualitatively to the experimental data [13]. The lack of a quantitative coincidence of computed electrical-resistance values and respective experimental ones may be explained in part by the concentration heterogeneity of the investigated alloys [14] and their multiphase structure [15] which are not accountable yet in the presented numerical computations. Moreover, excepting the above-mentioned imperfections of a crystal structure, there is the additional factor which is not taken into account in our theoretical model and, nevertheless, is crucially important for the atomic ordering of alloys into the $B2$-type superstructure. This is the tendency of the investigated alloys to their decomposition into the antiphase domains of the $B2$-type atomic order. Boundaries of such domains cause the additional scattering of conduction electrons and the corresponding enhancement in values of the experimentally measured electrical resistance of a real sample in comparison with ones for a homogeneously (dis)ordered alloy.

As revealed by means of the analysis of the concentration dependence of the b.c.c.-$Fe_{1-c}Co_c$ electrical resistance in the view of the electron-energy spectrum realignment, a change of the electrical conductivity has a weak dependence on the atomic-order degree. This is confirmed by the presence of minima in the experimental concentration dependences of resistance for

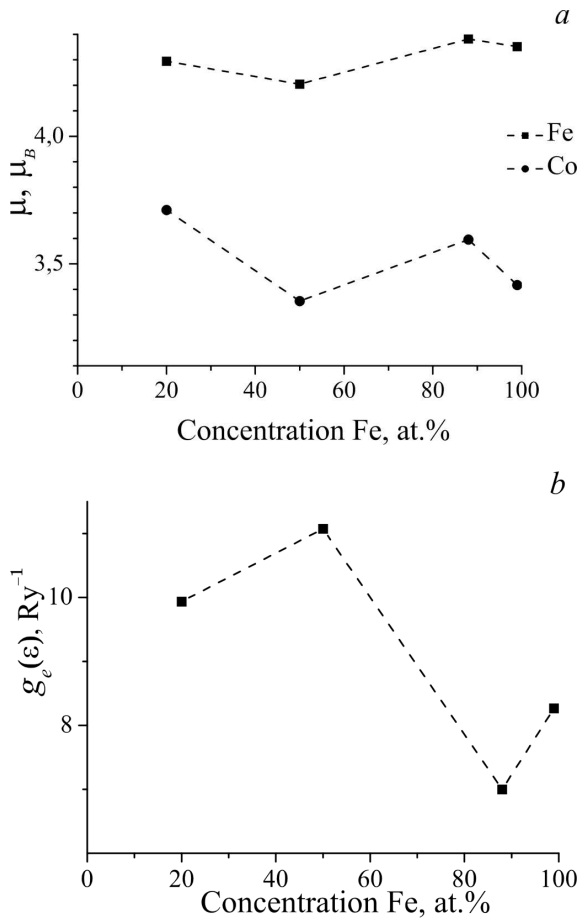

Fig. 5. Concentration dependences of the averaged localized magnetic moments (a) and the electron density of states at the Fermi level (b) for b.c.c.-$Fe_{1-c}Co_c$ alloys

the equiatomic composition at high temperatures (*i.e.* higher than the order–disorder transition point), as well as at low temperatures [16]. At the same time, the magnetic-subsystem state is a distinct governing factor. The nonmonotonic concentration dependence of the Fe−Co-alloy electroresistance is caused by both the strong electron correlations and the magnetic order resulting from these correlations. Strong correlations between electrons lead to the appearance of the 'Coulomb' quasigap in the electron-energy spectrum of a binary alloy. The Fermi-level position in the quasigap range determines the electrical conductivity of an alloy. A decrease of the averaged localized magnetic moments (Fig. 5,a) and an increase of the electron density of states at the Fermi level in the energy spectrum (Fig. 5,b) lead to a decrease of the electrical resistance of an alloy with strong electron correlations. The opposite change of above-mentioned parameters of state leads to an increase of the electrical resistance.

4. Conclusions

The microscopic mechanisms of magnetic ordering in a $Fe_{0.5}Co_{0.5}$ alloy and formation of the electrical resistance of b.c.c.-$Fe_{1-c}Co_c$ alloys concern with both the Fermi-level position within the 'Coulomb' quasigap range in the electron-energy spectrum and the realignment of this spectrum.

The values of parameters of the pairwise magnetic correlations decrease with increase in the temperature that allows one to estimate the Curie temperature of the alloy under study (for $Fe_{0.5}Co_{0.5}$, $T_C \cong 1200$ K).

The nonmonotonic concentration dependence of the Fe–Co-alloy electroresistance is conditioned by strong electron correlations and the magnetic order resulting from these correlations to a greater extent than by the atomic-order degree.

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СИЛЬНІ ЕЛЕКТРОННІ КОРЕЛЯЦІЇ В БЛИЗЬКОМУ МАГНІТНОМУ ПОРЯДКУ ТА ЕЛЕКТРООПОРІ ОДНОРІДНО НЕВПОРЯДКОВАНИХ БІНАРНИХ КРИСТАЛІВ

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Р е з ю м е

Розглянуто формування близького магнітного порядку та електропровідності в однорідно невпорядкованих бінарних кристалах під впливом сильних електронних кореляцій (на прикладі сильно корельованої електронної підсистеми сплаву ОЦК-$Fe_{1-c}Co_c$). Для опису електронних станів у кристалі використано багатозонну модель сильного зв'язку та метод кластерного розкладання для грінових функцій і термодинамічного потенціалу невпорядкованого кристала. Сильні електронні кореляції та "розвинутий" близький порядок атомів заміщення приводять до появи квазіщілини в енергетичному спектрі електронів. Мікроскопічні механізми магнітного упорядкування і формування електроопору пов'язані з розташуванням рівня Фермі в області квазіщілини, а також з перебудовою енергетичного спектра електронів внаслідок змін температури або концентрації легуючої компоненти. Параметр парних магнітних кореляцій зменшується зі зростанням температури і прямує до нуля в області температури Кюрі. Досліджено також немонотонну концентраційну залежність електроопору сплаву Fe–Co. Вона визначається сильними електронними кореляціями та зумовленим ними магнітним порядком.