Spin fluctuations in disordered metallic ferrimagnetic alloys

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Keywords: itinerant electron magnetism, thermal spin fluctuations, Fe-Al disordered alloys, atomic magnetic moments, magnetic phase transitions.

Abstract. We propose a theory for the quantitative description of the thermodynamic, magnetic and electronic properties of disordered alloys of transition and normal metals, for example, the system Fe-Al, as obtained from melting, and in the form of metastable states produced by mechanical grinding. System of itinerant *d*-electrons with two constants of the intra-atomic exchange interaction is investigated on the basis of the fluctuating field approach. Fundamentally new feature of the system is the presence of several ferrimagnetic states, transitions between them generate a variety of temperature dependences of the magnetic characteristics in disordered alloys. The magnetization vs temperature curves are calculated for the disordered alloy models.

Introduction

A successful description of magnetic properties of ferromagnetic metals at finite temperatures essentially requires consideration the fluctuations of the electron spin density (see, e.g., [1-4]). Although all of the spin-fluctuation theories are formulated on the first-principle level, however their final realizations have significant differences. For example, our approach is quite different from the Kakehashi's dynamical version of the single-site spin-fluctuation [3]. First, we do not restrict ourselves to a single band model but consider all five *d*-states of a transition atom. Unified fluctuating exchange field acts on the full spin moment produced by total electron density. Second, we use not the one-dimensional Ising model for the exchange field and consider the 3D Heisenberg like representation with fluctuations around the all three-dimensional space. Third, the essential physical principle of our theory is the separation of spin fluctuations on the zero-point (high frequency) and thermal (low-frequency) fluctuations in the equation for the coherent potential. The zero-point fluctuations is taken into account in the DOS ab initio calculations for the studied real compound and not a single-band model. Electron scattering on the low thermal fluctuations almost does not change their energy therefore the original integral equation for the coherent potential is replaced by an algebraic equation with fixed energy. The microscopic treatment of the spin fluctuations, based on the functional-integral method, allowed the Curie–Weiss susceptibility to be explained and a reasonable estimate for the Curie temperature to be obtained [5, 6].

Magnetization of the practical interest alloys of iron with aluminum, silicon, tin has a complex temperature dependence often wearing a no monotonic character [7, 8]. The source of such behavior is the nonequivalence of the magnetic atoms due to different nearest neighborhoods. Therefore, generalization of the theory of spin fluctuations on the class of disordered metallic magnetic materials is a very important problem. To solve it we consider a system of itinerant 3*d*-electrons with several intra-atomic exchange interaction constants u_j. Our goal is to calculate the thermodynamic characteristics of the electronic subsystem (entropy, heat capacity) at an arbitrary temperature, find the values of the local magnetic moments on the atoms of different types, the resulting magnetization and its variation in the magnetic field, describe the magnetic phase transitions. The problem is solved by the dynamical theory of electron spin density fluctuations [5, 9]. Its generalization gives a few

ferrimagnetic states, transitions between them generate a variety of temperature dependences of magnetic properties in disordered alloys.

Brief formalism

As an application of the theory of spin fluctuations are usually considered ferromagnetic models with identical magnetic atoms. Below we give a very brief and very simplified summary of the theory describing the alloys with two or more types of magnetic atoms. Consider a system of itinerant 3*d*-electrons, which is characterized by one-electron density of states DOS ν(*e*) (see, e.g., Fig. 2 (a)) and the strong intra-atomic exchange interaction $H = -u_j \mathbf{s}_j^2$, where $\mathbf{s}_j$ stands for spin of electrons at the atom number *j* (summation over repeated indices meant). Let the constants u_j take two values $u_j = \{u_1, u_2\}$ with probabilities $c_j = \{c_1, c_2\}$ depending on the atom type is determined by its nearest neighbors. Our approach can be easily extended to a larger number of atom types, but we restrict ourselves to only two types for the simplicity.

Using the Stratonovich-Hubbard transformation [1] for the partition function we reduce our problem to calculation of the characteristics of itinerant electrons moved in the fluctuating exchange field $\bar{V}_j + \Delta V_j = (\bar{V}_{jz} + \Delta V_{jz})\tau_z + \Delta V_{jx}\tau_x + \Delta V_{jy}\tau_y$, where $\bar{V}_j$ stands for the mean value of the field V_j at the site *j*, ΔV_j denote its deviations and **τ** are the 2×2 Pauli spin matrices. Fig. 1 (a) shows the distribution of the mean fields and fluctuations on different atoms.

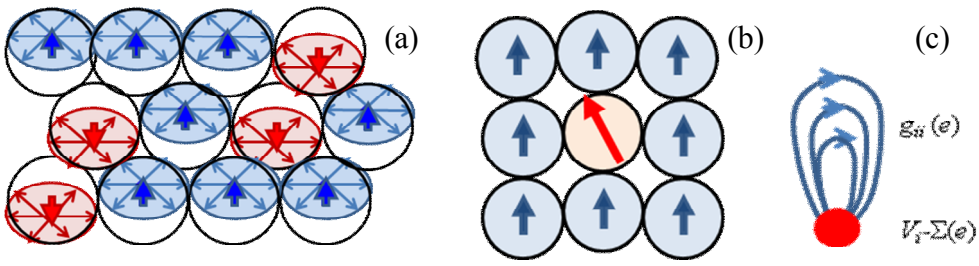

***Fig. 1**. (a) A distribution pattern of mean values (up down arrows) and fluctuations of the exchange field on atoms of the two types. (b) A central atom with the field **V** embedded in the coherent potential Σ(e) (blue up arrows) and (c) a diagram for the multiple scattering on the atom.*

A brief description of the basic equations of the spin fluctuation theory in ferromagnetic alloysis given below. The mean field values on the two types of atom are described by the simple recognizable equalities

$$\bar{V}_j = -u_j < s_j > . \tag{1}$$

The right-hand side contains the mean spin $< s_j >$ (angle brackets denote averaging over the thermal fluctuations of the exchange field ΔV_j) which depends on both V_j and the distribution function of fluctuations, therefore Eqs. (1) form a system of two coupled equations. The spin is determined by the integral over energy *e* of the imaginary part of the single-site ("impurity") Green function and the Fermi function *f*(*e*)

$$< s_j > = \int \mathrm{Im}\left\langle g[1-(\bar{V}_j + \Delta V_j - \Sigma)g]^{-1} \right\rangle f de/\pi \,; \quad g(e) = \int \nu(e')[e - \Sigma(e) - e']^{-1} de' . \tag{2}$$

The average single-site Green function *g*(*e*) is calculated for a given density of states ν(*e*) using a homogeneous coherent potential Σ(*e*). Basic field configurations and approximations used in our approach are illustrated in the figure. Fig. 1 (a) shows a distribution pattern for fluctuating field. Fig. 1 (b) shows the atom with field *V* embedded in the effective crystal described by the coherent potential Σ(*e*) as well as (c) a diagram for the multiple scattering process for calculation of the single-site Green function and the coherent potential.

The coherent potential provides the best crystal-like approximation for the disordered alloy in fluctuating field. It provides minimum for the alloy free energy. The energy dependent complex function $\Sigma(e)$ is found by solving the equation

$$\Sigma = \left\langle\left\langle V[1-g(V-\Sigma)]^{-1}\right\rangle\right\rangle \tag{3}$$

which contains full averaging over fluctuations ΔV_j and the atom types with concentrations c_j.

Field fluctuations have a Gaussian distribution. The mean squared fluctuation values ς_j^2 are defined by integrals of the Bose function with the dynamical single-site spin susceptibilities $\chi_j(e)$ which are calculated using the local Green function introduced in Eqs. (2). Evaluation of the integrals leads to the simple equalities

$$\varsigma_j^{\,2} \approx \frac{u_j T}{\pi\lambda_j}\operatorname{atan}\frac{\pi^2 u_j T\varphi_j}{6\lambda_j};\ \lambda_j = 1-u_j\chi_j(0);\ \ \varphi_j = \operatorname{Im}\frac{d\chi_j}{de}(0) \quad . \tag{4}$$

Practically all dependence on the temperature T is provided by spin fluctuations ς as the temperature effect from the Fermi distribution $f(e)$ is negligible.

Results and analysis

Note, if we drop out fluctuations and assume that all the atoms are the same, the Eqs. 1–4 reduce to the equations of the Stoner-Wohlfarth theory of ferromagnetism [10]. If we take into account spin fluctuations, but we assume the exchange interaction u_j are identical on all atoms, we arrive at the equations of the theory of spin fluctuations of itinerant ferromagnetism [2, 5].

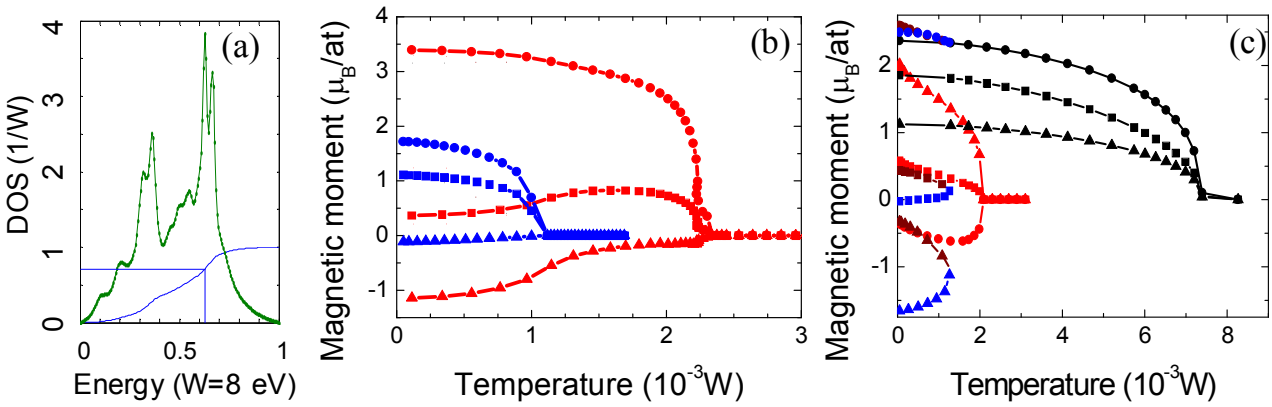

Fig. 2. *(a) The nonmagnetic density of states for disordered Fe_3Al alloys (blue line shows integrated DOS). (b) The magnetic moments versus temperature calculated for the system with two types of atoms: coupling constants u = {0.066, 0.086}W and the concentrations of c = {0.67, 0.33}. Two kinds of solutions are highlighted, the points and triangles indicate the atomic magnetic moments, squares shows their average value. (c) Magnetic moments of the two type magnetic atoms diluted with nonmagnetic atoms: interactions u= {0.132, 0.120, 0}W, concentrations c = {0.2, 0.3, 0.5}. Four kinds of states are highlighted.*

The principal new feature of our approach compared with the standard fluctuating field theory of the ferromagnetic materials is the presence of two (or more) types of atoms with different interactions $u_j = \{u_1, u_2\}$. Instead of the single equation, the system of equations Eqs. 1 arises for determining the mean field values and the electron spin densities values on different sites. This system has several ferrimagnetic solutions, transitions between them give a variety of the temperature dependences of magnetic properties in disordered alloys.

We illustrate the novel properties of the theory on the example of the model for the disordered alloy of the Fe_3Al type. Its normalized density of states is shown in Fig. 2 (a) where the integrated

DOS and the Fermi energy also given. For the DOS calculation, we used a unit cell containing eight Fe(I) atoms, four Fe(II) atoms and four Al atoms [11]. Averaging over the permutations of atoms was performed to account for the alloy disorder. The full width of the *d*-bands W = 8 eV.

Now we concentrate on the study of the possible types of magnetic states in the model by specifying different values for the coupling constants and the concentration of atoms. Let the system has two types of iron atoms are characterized by the two exchange constants $u_1 = 0.066\ W$ and $u_2 = 0.086\ W$ randomly distributed on the lattice sites with concentrations $c_1 = 0.67$ and $c_2 = 0.33$, respectively. In this case Eqs 1-4 have two solutions presented in Fig. 2 (b). In the first state (blue lines), the atomic magnetic moments at zero temperature are equal to $m_1 = 1.7\ \mu_B$, $m_2 = -0.1\mu_B$ (in the opposite direction) and net magnetization $m = 1.1\ \mu_B$. With increasing temperature, the average value of these moments is reduced to zero at the Curie temperature $k_BT_{C1} = 0.0011W$ or $T_{C1} = 102$ K. The second state (red lines) has large antiparallel magnetic moments: $m_1 = -1.1\ \mu_B$, $m_2 = 3.4\ \mu_B$, and net magnetization $m = 0.4\ \mu_B$ at zero temperature. With increasing temperature the mean magnetic moment of atoms with a small interaction u_1 decreases quickly, trying to go to the paramagnetic state about $k_BT = 0.00125\ W$, which is prevented due to the magnetization from the strong subsystem having magnetic moment about 3 μ_B. The Curie temperature of paramagnetic transition for this state $k_BT_{C1} = 0.0022W$ twofold higher than for the first state. Pay attention to the nonmonotonic temperature dependence of the net magnetization. Magnetization increases at low temperatures, reaches a maximum at 0.0015 and then decreases. The curve $m(T)$ may look even more bizarre if at low temperatures the system is in fist state (blue line), and on further heating it goes to the second state (red). In this case, as seen in Fig. 2 (b), the function $m(T)$ will have two maxima and a minimum. Incidentally, this behavior is not rarely observed in systems Fe-Al, Fe-Si, Fe-Sn systems [7, 8].

As a second example, consider a diluted magnets with the three interactions values u= {0.132, 0.120, 0}W with probabilities c = {0.2, 0.3, 0.5}. The magnetic moments vs temperature curves are shown in Fig. 2 (c). We can see two familiar states with the parallel zero-temperature magnetic moments $m_1 = 1.13\ \mu_B$, $m_2 = 2.37\ \mu_B$ (net magnetization $m = 1.85\ \mu_B$ and the highest Curie temperature $k_BT_{C1} = 0.0074W$, black lines) and antiparallel $m_1 = 1.95\ \mu_B$, $m_2 = -0.35\ \mu_B$, net $m = 0.55\ \mu_B$ and low $k_BT_{C2} = 0.0021\ W$ (red lines). There are two more states (wine and blue lines) not turning into the paramagnetic state with increasing temperature. These states are going to meet each other with increasing temperature and annihilate at $T^* = 0.0013$ where $m_1 = 2.34\ \mu_B$, $m_2 = -1.12\ \mu_B$ and the net moment $m = 0.26\ \mu_B$. Itinerant magnets with different constants of the intra-atomic exchange interactions demonstrate a set of different magnetic states, especially at low temperatures. Transitions between these states generate non-monotonic temperature dependence of magnetic characteristics observed in metallic alloys.

Conclusions

We have developed a method of spin fluctuations to describe the thermodynamic and magnetic properties of disordered alloys in a wide temperature range from zero to the paramagnetic transition temperature and higher. The alloy is modeled by introducing several types of atoms with different constants of the intra-atomic exchange interaction. A self-consistent equations are written for calculating characteristics of itinerant electrons moving in the equilibrium field and in an alternating dynamical field generated by thermal fluctuations of the electron spin density. Unlike pure iron the equations have not one but several solutions. Thus electronic system may be located in several metastable states, transitions between them provide a variety of temperature dependence of magnetic properties of the metal alloys.

Support by RFBR grants 14-02-00080, 14-02-00260 and UD RAS 12-U-2-1002, 12-П-2-1041 is acknowledged.

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