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Magnetic properties of Fe-Ge with low atomic content of Ge: First-principles modeling

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In this paper, the magnetic properties of the disordered $A2$ phase of $Fe_{100-x}Ge_x$ alloys were studied using methods of density functional theory. The distribution of the magnetic exchange interaction parameters J_{ij} obtained in the study exhibits nonlinear behavior. The strongest ferromagnetic interaction in the first coordination sphere is ≈ 23.3 meV. In the third and fourth coordination spheres, an antiferromagnetic exchange interaction is observed, reaching a value of ≈ -2.5 meV. The magnetic transition temperatures, calculated using the Mean-field approximation based on the experimental values of a and the parameters a_0 calculated within the framework of density functional theory, indicate the presence of a concentration range of $4 \leq x \leq 8$ at.%, in which T_C increases. The cross-section of the calculated Curie temperature distribution shows that for the studied Fe-Ge alloys with lattice parameters less than 2.74 Å and greater than 2.94 Å it is possible to construct a dependence $T_C(x)$ that is in quality agreement with the experimental one.

Keywords: Fe-Ge alloys, density functional theory, exchange interaction, Curie temperature.

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1. Introduction

The interest of the scientific community in the study and design of multifunctional iron-based materials is always growing. The demand for the green materials makes these alloys especially attractive. The unique combination of mechanical, magnetic, electric characteristics of Fe-Me (Me = Al, Ga, Ge) alloys with high values of magnetostriction in low magnetic fields [1,2] causes a special interest of researchers and makes them promising for design of sensors and actuators and other magnetomechanical devices. The ability to convert magnetic energy into mechanical and back makes it possible to use these alloys in the industry, power engineering, motor equipment and other fields. Therefore, the studies of Fe-Me (Me=Al, Ga, Ge) alloys remain relevant both from the point of view of the fundamental science and practical application, which is confirmed by the continuous growth of publications in this area.

The experimental studies show that in phase diagrams of $Fe_{100-x}Me_x$ (Me = Al, Ga, Ge) systems in the range of concentrations from 0 to 10 at.% there is a disorderd BCC structure $A2$ (symmetry group № 229, Im-$3m$, prototype α-Fe). Besides, a similar behavior of Curie temperature is observed. In the single-phase area $A2$ there is a gradual decrease of the Curie temperature T_C, which becomes sharper when the $D0_3$ structure is formed. This is accompanied by the appearance of the two-phase area that influences the magnetic characteristics of the material [3].

The comprehensive approach using computations *ab initio* and simulation with Monte Carlo method in Curie temperature calculation presented in papers [4–7], showed an interesting feature of the obtained results. In the $x > 12$ at.% concentration area there is a descending behavior of dependence in $T_C(x)$ curves of alloys $Fe_{100-x}Al_x$, $Fe_{100-x}Ga_x$, and $Fe_{100-x}Ge_x$. However, in the field of lower concentration the behavior of the experimental curve may not be reproduced: the studies show the opposite trend — Curie temperature increases as the concentration Me = Al, Ga, Ge changes from zero to 12 at.%. The authors explain this behavior by enhanced exchange interaction J_{ij} between the iron atoms and the increase of the magnetic moment of Fe atoms μ_{Fe}. Therefore, the obtained results show a complicated interconnection between the structure, composition and magnetic properties of $Fe_{100-x}Me_x$ (Me = Al, Ga, Ge) alloys, which opens new prospects for the fundamental study of the mechanisms to form the magnetic properties in these materials.

This paper provides the analysis of the crystal lattice parameter and concentration of non-magnetic Ge atoms that influence the exchange interaction, and also the Curie temperature of $A2$ structure in $Fe_{100-x}Ge_x$ alloy using the first-principle simulation.

2. Calculation details

To simulate the magnetic properties in $Fe_{100-x}Ge_x$ ($0 \leq x \leq 14$ at.%.) system the Korringa-Kohn-Rostoker Green's function method implemented in the SPRKKR (a Spin Polarized Relativistic Korringa-Kohn-Rostoker

code) program package [8]. The paper studied the disordered structure $A2$. For this phase Fe and Ge atoms are distributed randomly in the Wyckoff position $2a$ (0, 0, 0). To develop nonstoichiometric compositions, the single-site coherent potential approximation (CPA) was used, which makes it possible to built the averaged potential for the lattice site occupied with several atom types [8]. It should be noted that this approximation successfully describes the properties of many compositionally disordered (nonstoichiometric) alloys.

At the first stage the constants of Heisenberg magnetic exchange interaction J_{ij} were calculated as specified by A. Liechtenstein et al. [9]. The calculations included using spin-polarized scalar-relativistic Dirac Hamiltonian with orbital moment $l_{\max} = 2$ cut. To conduct both self-consistent calculations and J_{ij} calculations, 4495 k-points with grid of $57 \times 57 \times 57$ were generated. The accounting for interactions was carried out up to 49th coordination sphere and limited to the value multiple of 6 lattice parameters. The exchange-correlation potential used the generalized gradient approximation as specified by Perdew-Burke-Ernzerhof (PBE) [10]. The obtained constants of magnetic exchange interaction were used to estimate the Curie temperature T_C in the Mean-field approximation (MFA) [11]. The crystal lattice parameter varied in the range of $2.7 \leq a \leq 3.0$ Å.

3. Results and discussion

At the first stage the parameters of magnetic exchange interaction were calculated for the $A2$ structure of $Fe_{98}Ge_2$ alloy with the experimental value of $a_{\exp} = 2.87$ Å lattice parameter [12]. Figure 1 shows the values J_{ij} between Fe atoms depending on the distance between the atoms (in units of the crystal lattice parameter). The exchange interaction between the pairs of atoms Ge-Ge and Fe-Ge does not exceed the value of 0.1 meV and is excluded from the consideration. As you can see in the figure, J_{ij} demonstrate oscillating damping behavior. The highest ferromagnetic (FM) interaction is observed in the first coordination sphere of Fe atoms and is ≈ 22.3 meV. Let us consider further how values J_{ij} change depending on the parameter of the crystal lattice and concentration of Ge atoms.

Figure 2 shows the distribution of constants of the magnetic exchange interaction of $Fe_{100-x}Ge_x$. alloy series on the crystal lattice value and concentration of Ge atoms for the first four coordination spheres (at the distance of $\sqrt{3}/2a$ — Figure 2, a, at the distance of a — Figure 2, b, at the distance of $\sqrt{2}a$ — Figure 2, c and at the distance of $\sqrt{11}/2a$ — Figure 2, d).

The distribution J_{ij} has a non-linear behavior. Between the nearest Fe atoms (Figure 2, a), depending on the Ge concentration, the change is not more than 6.5 meV, from ≈ 18.5 meV for pure Fe to ≈ 25 meV in case of $Fe_{86}Ge_{14}$. Note that such increase corresponds to the

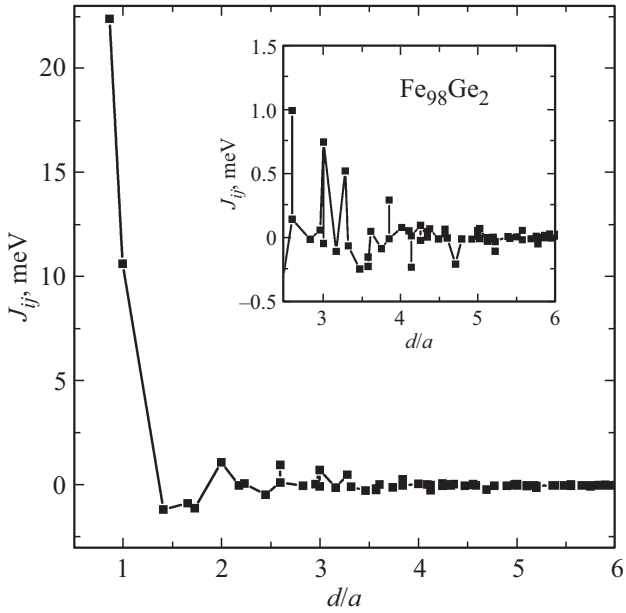

Figure 1. Constants of magnetic exchange interaction J_{ij} depending on distance calculated using the experimental value of crystal lattice parameter $a_{\exp} = 2.87$ Å [12]. The insert shows the area $d/a > 2.5$ at the scale enlarged along the vertical line.

area of the $a = 2.85-2.86$ Å lattice parameters close to the experimental value. In the area of values $a \leq 2.72$ Å J_{ij} demonstrate weak dependence on Ge concentration, changing by 2 meV ($\approx 21- \approx 23$ meV), whereas in the area of values $a \geq 2.92$ — change J_{ij} exceeds 4 meV ($\approx 20- \approx 24.5$ meV).

In the second coordination sphere (at the distance of a, Figure 2, b) the distribution of exchange parameters differs, and two areas may be identified in the distribution depending on the crystal lattice parameter. In the first area with small values of lattice parameters ($a \leq 2.82$ Å) J_{ij} hardly change with the growth of Ge concentration, increasing practically linearly with the increase of the crystal lattice parameter at fixed Ge concentration in the alloy. In this area the parameter values vary from ≈ 4.5 to ≈ 10.5 meV. The second area in Figure 2, b is characterized by reduction of the magnetic exchange with the increased concentration of Ge at invariable values of a parameter and fixed Ge concentration in the alloy. In this area the change of the magnetic exchange interaction parameters is observed from ≈ 6.0 to ≈ 14.0 meV.

In case of interaction between Fe atoms at the distance of $\sqrt{2}a$ (Figure 2, c, the third coordination sphere) the distribution demonstrates an area of antiferromagnetic (AFM) interaction. This area is observed for the lattice parameters $2.8 \leq a \leq 3.0$ Å, and concentration range $0 \leq x \leq 8$ at.%. In general you can note weak (3 meV max.) change of magnetic exchange interaction constants in the third coordination sphere.

The area of AFM exchange is also observed in the distribution of parameters of magnetic exchange interaction

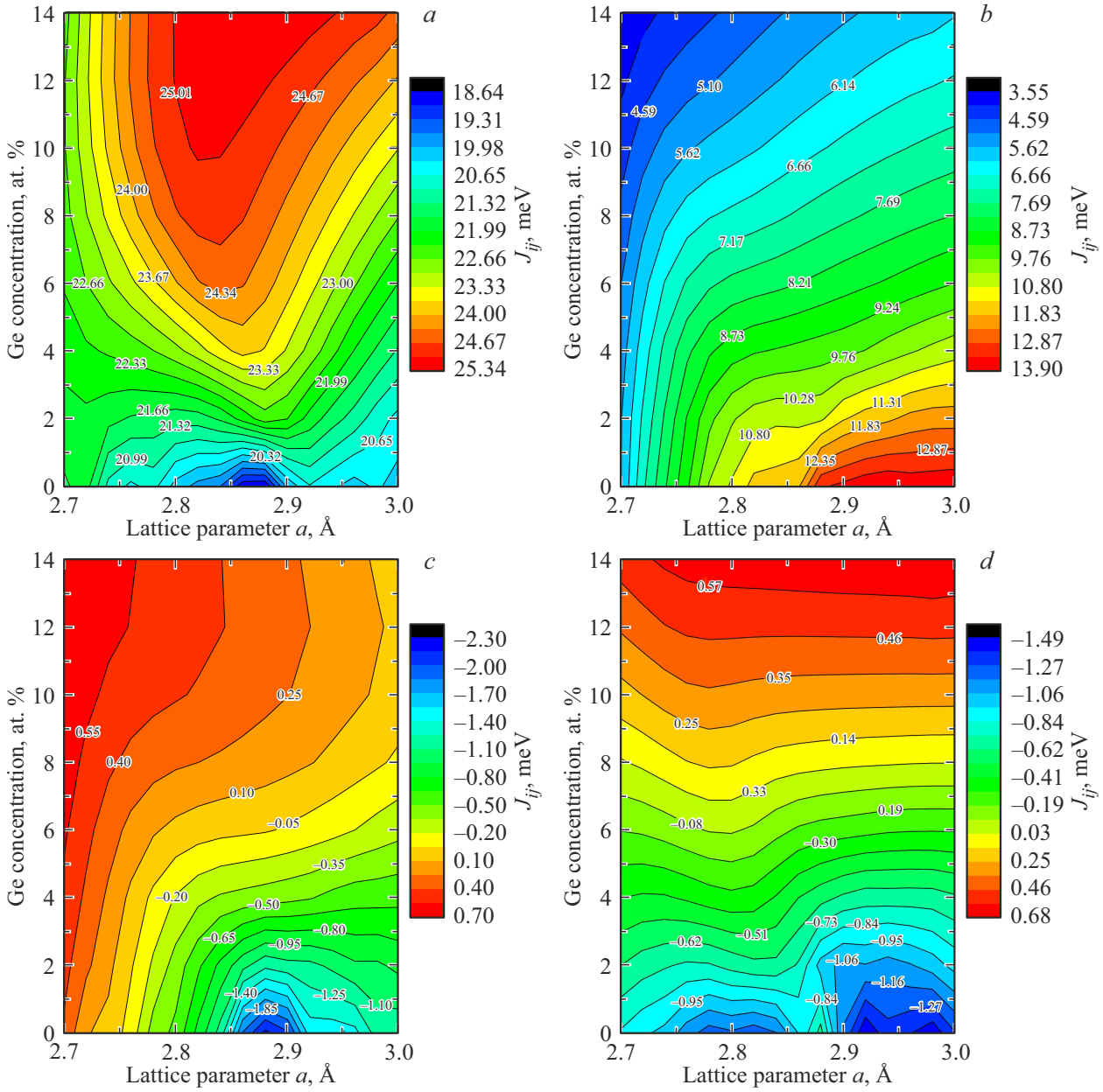

Figure 2. Distribution of the magnetic exchange interaction constant of $Fe_{100-x}Ge_x$ alloys between Fe atoms depending on the crystal lattice parameter a and content of Ge x in (a) the first, (b) second, (c) third and (d) fourth coordination spheres.

between Fe atoms, at the distance $\sqrt{11}/2a$ (Figure 2, d, fourth coordination sphere). In this case you can identify two areas in the distribution. In the first area, at concentration of Ge ≤ 8 at.% in the entire considered range of crystal lattice parameters the exchange interaction of -1.5 meV minimum is observed. At Ge concentrations, also for all the crystal lattice parameters, the exchange interaction is FM with the values of 0.6 meV max.

In papers [5–7,13–15] the authors demonstrated that when simulation by Monte Carlo method is used, the estimated values of Curie temperature depend substantially on J_{ij} value. Taking into account this fact, this paper analyzed the impact of the crystal lattice parameter value and concentration of non-magnetic atoms in $Fe_{100-x}Ge_x$ alloys on the Curie temperature value.

T_C values given in Figure 3 from the crystal lattice parameter and concentration of Ge atoms are obtained in MFA, which, as the results of papers [4,13] show, provides overestimated values compared to the experimental values, but makes it possible to qualitatively reproduce the experimentally observed trends [4,16].

As you can see from Figure 3, for the considered system, 3 areas may be identified in the distribution. The first area ($a \leq 2.74$ Å) demonstrates the descending behavior of Curie temperature. In the second area ($2.74 \leq a \leq 2.94$ Å) there is maximum Curie temperature in the concentration

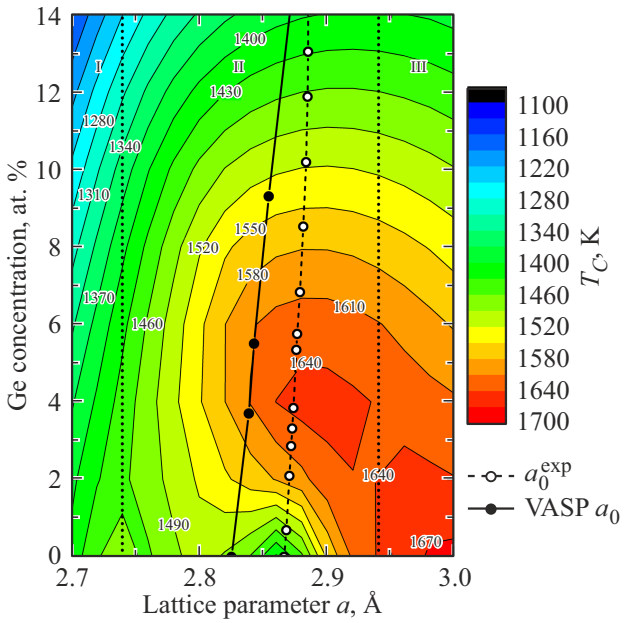

Figure 3. Distribution of Curie temperature of $Fe_{100-x}Ge_x$ alloys, depending on crystal lattice parameter a and content of Ge x.

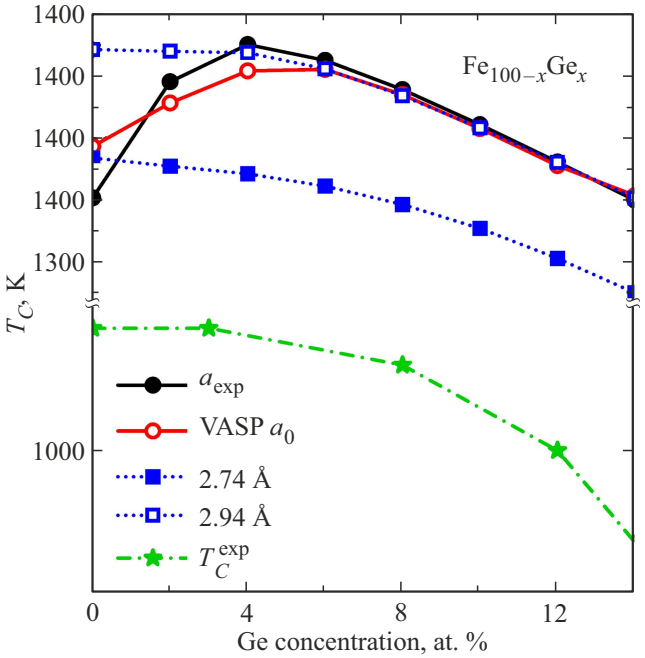

Figure 4. Calculated values of Curie temperature in $Fe_{100-x}Ge_x$ alloys obtained at experimental, theoretical (from *ab initio* geometric optimization) crystal lattice parameters, and at parameters of 2.74, 2.94 Å, together with experimental values of Curie temperature. The experimental data was taken from [12].

range of $2 \leq x \leq 4$ at.%. In this case the concentration dependence of Curie temperature does not match the Curie temperature observed with descending behavior in the experiment [12]. In the third area ($a \geq 2.94$ Å) there is also descending behavior of Curie temperature. Figure 3 shows cross sections $T_C(x)$, corresponding to the experimental values of crystal lattice parameter [12] and crystal lattice parameters obtained using *ab initio* geometric optimization of the crystal lattice [7]. Note that these sections are in the second area of Curie temperature distribution.

For more clarity, Figure 4 shows the Curie temperatures depending on the concentration of Ge atoms along the experimental and theoretical dependences of the lattice (Figure 3), and also the Curie temperature at crystal lattice parameters of 2.74 and 2.94 Å.

As you can see, the accounting for both experimental and theoretical values a_0 when calculating T_C does not make it possible to obtain dependences corresponding to the experimental $T_C(x)$. From the figure you can see that maximum Curie temperature corresponds to concentration interval of $4 \leq x \leq 8$ at.%. Curie temperature to this interval increases, and decreases after. At parameters of 2.74 and 2.94 Å lattice parameter Curie temperature changes from the higher to the lower value with the increase of the doped element concentration and will agree qualitatively with the experimental dependence.

4. Conclusion

Therefore, this paper, using the density functional theory methods, conducted magnetic properties of the disordered phase $A2$ in $Fe_{100-x}Ge_x$ ($0 \leq x \leq 14$ at.%) alloys.

The distribution of constants J_{ij} obtained in the paper has a non-linear behavior. The highest interaction is observed between the nearest Fe atoms, is ferromagnetic and amounts to ≈ 24 meV. The second coordination sphere maintains the ferromagnetic interaction between Fe atoms, and J_{ij} values vary within 4–14 meV. In the third and fourth coordination spheres the antiferromagnetic exchange interaction is observed, which reaches the value of ≈ -2.5 meV. Estimation of the Curie temperature using Mean-field approximation yields overestimated values compared to the experimental results. Curie temperatures obtained at experimental parameters of the crystal lattice have concentration dependences that differ from the ones observed experimentally, namely they have maximum Curie temperature in the concentration range of $4 \leq x \leq 8$ at.%. Using the obtained distributions of Curie temperature depending on the constant crystal lattice and content of Ge in $Fe_{100-x}Ge_x$ alloys, it is possible to obtain concentration dependences that qualitatively describe the decreasing behavior $T_C(x)$, observed experimentally. The conducted study shows that the value of Curie temperature is in complex non-linear dependence both on the constant of the crystal structure lattice and on the parameters of the magnetic exchange interaction.

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Conflict of interest

The authors declare that they have no conflict of interest.

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