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Magnetostriction of the $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) alloys: calculation by the magnetic torque method

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This work presents an *ab initio* study of the effect of a small addition of the third element of III and IV groups on the elastic and magnetoelastic properties of $Fe_{75}Ga_{25}$ alloy. The dependencies of the tetragonal elastic modulus C', magnetoelastic constant $-b_1$, and the tetragonal magnetostrictive constant λ_{001} on the concentration of the Z-element in the cubic crystal structures A2 and $D0_3$ were obtained with the help of the density functional theory and the magnetic torque method in $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) alloys ($0 \le x \le 6$ at.%). It is shown that the addition of Al and Si atoms leads to an increase in the tetragonal elastic modulus compared to the $Fe_{75}Ga_{25}$ binary alloy. A correlation was established in the dependence equilibrium lattice constants $a_0(x)$ and $\lambda_{001}(x)$ in the studied ternary alloys for the A2 structure.

Keywords: tetragonal magnetostriction constant, magnetoelastic constant, tetragonal shear modulus, magnetic torque method.

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1. Introduction

Iron-based magnetically-ordered alloys have been the subject of studies more than two decades. A small addition of non-magnetic semimetallic and post-transient elements of the groups III and IV, such as Ga, Ge, Al and Si into the structure α-Fe enable producing prospective soft magnetic materials with high values of magnetostriction in weak magnetic fields. Alloys Fe-(Ga, Ge, Al, Si) within the range of content of added element up to 30 at.% have similar phase diagram in the area of room temperatures, and feature the presence of totally disordered structure A2 with further formation of totally and/or partly ordered structures $D0_3$ and/or B2 [1–4]. For all four alloys the first peak of tetragonal magnetostriction λ_{001} is associated with the presence of the phase A2 and pertains to the boundary of the area A2/($D0_3$/B2). That peak also is associated with the limit of solubility of the added element in the structure α-Fe. For the alloys with elements of the group IV Fe-Ge and Fe-Si the limit of solubility corresponds to ≈ 10 at.% and ≈ 5 at.% of the content of Ge and Si, accordingly, meanwhile further increase of concentration of these elements in alloys results in change of the magnetostriction sign, from positive to negative. For the alloys with the elements of the group III Ga and Al behavior of the curves $\lambda_{001}(x)$ is similar to each other, and the first peak of magnetostriction associated with disordered phase A2 is observed with the content of the added atoms of about 19 at.% and further concentration increase does not result in the sign change λ_{001} [5]. In the alloys Fe-Al and Fe-Si magnetostriction reaches only one peak unlike the systems Fe-Ga and Fe-Ge, where the second peak of magnetostriction is associated with the single-phase composition of the alloys with totally ordered structure $D0_3$ in compositions with the concentration $x^{Ga} \approx 27-28$ at.% and $x^{Ge} \approx 19$ at.% [5–7]. Only in alloys Fe-Ga in the area between two peaks, with simultaneous existence of the structures A2 and $D0_3$ different ratio of these phases results in the minimum at $x \approx 25$ at.%, and then in increase λ_{001} up to the second peak, where, as it has already been mentioned above, only one phase $D0_3$ is observed. It was proven by experiments that addition of Al, Si, Ga and Ge atoms reduces tetragonal modulus of shift as far as the concentration is increased, however, in the alloys Fe-Ga and Fe-Ge there is significant „softening“ C' [5,6].

The purpose of this work is to study the impact of replacement of Ga atoms in alloy $Fe_{75}Ga_{25}$ with low number of atoms of the groups III and IV in order to establish possible correlations of change of the equilibrium parameter of the lattice with changes of the magnetostriction value. The work deals with properties of A2 and $D0_3$ cubic symmetry crystal structures of alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) ($0 \le x \le 6$ at.%).

2. Calculation details

The first-principles SPR-KKR (spin polarized relativistic Korriga–Kohn–Rostoker) software package based on the Korriga–Kohn–Rostoker Green's function [8], was chosen to perform te study, which allows to efficiently solve problems related to impurities in the crystal without

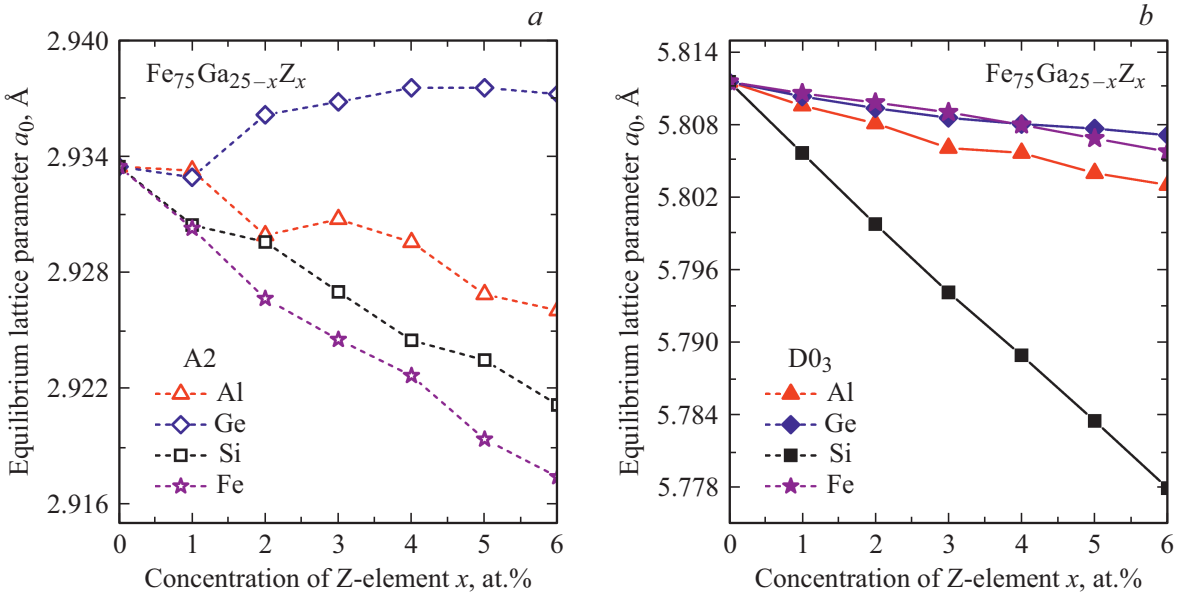

Figure 1. Dependence of equilibrium parameters of the lattice on the concentration of Z-element in alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) ($0 \le x \le 6$ at.%) for structures (*a*) A2 and (*b*) D0$_3$. The values for alloys Fe-Ga are taken from the work [13] indicated by open and filled asterisks.

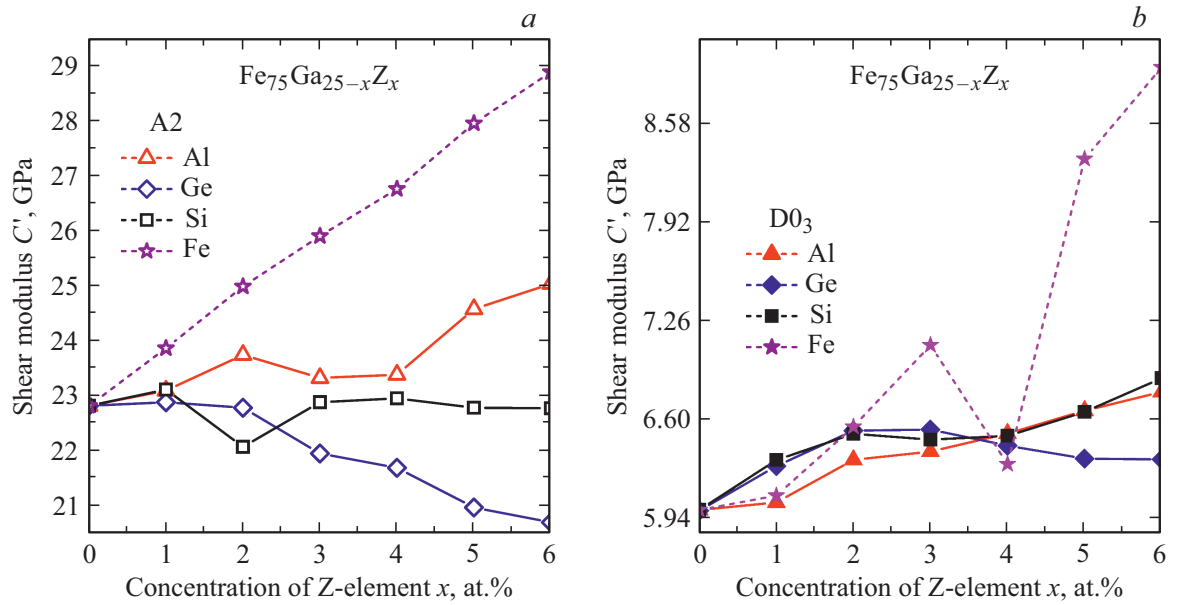

Figure 2. Dependence of shear modulus C on the concentration of Z-element in alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) ($0 \le x \le 6$ at.%) for structures A2 and D0$_3$. The values for alloys Fe-Ga are taken from the work [13] indicated by open and filled asterisks.

using additional geometry related to the formation of a finite cluster or supercell. To create non-stoichiometric compositions we used Coherent potential approximation (CPA). The maximum number of CPA steps was 30; the convergence limit of self-consistent functions was 10^{-5} Ry. The exchange-correlation effects were considered in the generalized gradient approximation (GGA), according to Perdew–Burke–Ernzerhof (PBE) [9]. For self-consistent cycles the scattering operator was derived by integration of the Brillouin zone on the k-grid $45 \times 45 \times 45$ with 2300 k-points. The following lattices were chosen for geometric optimization of cubic structures: $Im\bar{3}m$ (№ 229) symmetry group for A2 with Wyckoff positions $2a$ (0; 0; 0) occupied by Fe, Ga, Z (Z = Al, Ge, Si); $Fm\bar{3}m$ (№ 225) symmetry group for D0$_3$ with Wyckoff positions $4a$ (0; 0; 0) occupied by Ga, Z (Z = Al, Ge, Si), Fe atoms located at $4b$ (0.5; 0.5; 0.5) and $8c$ (0.25; 0.25; 0.25). Equilibrium parameters of the

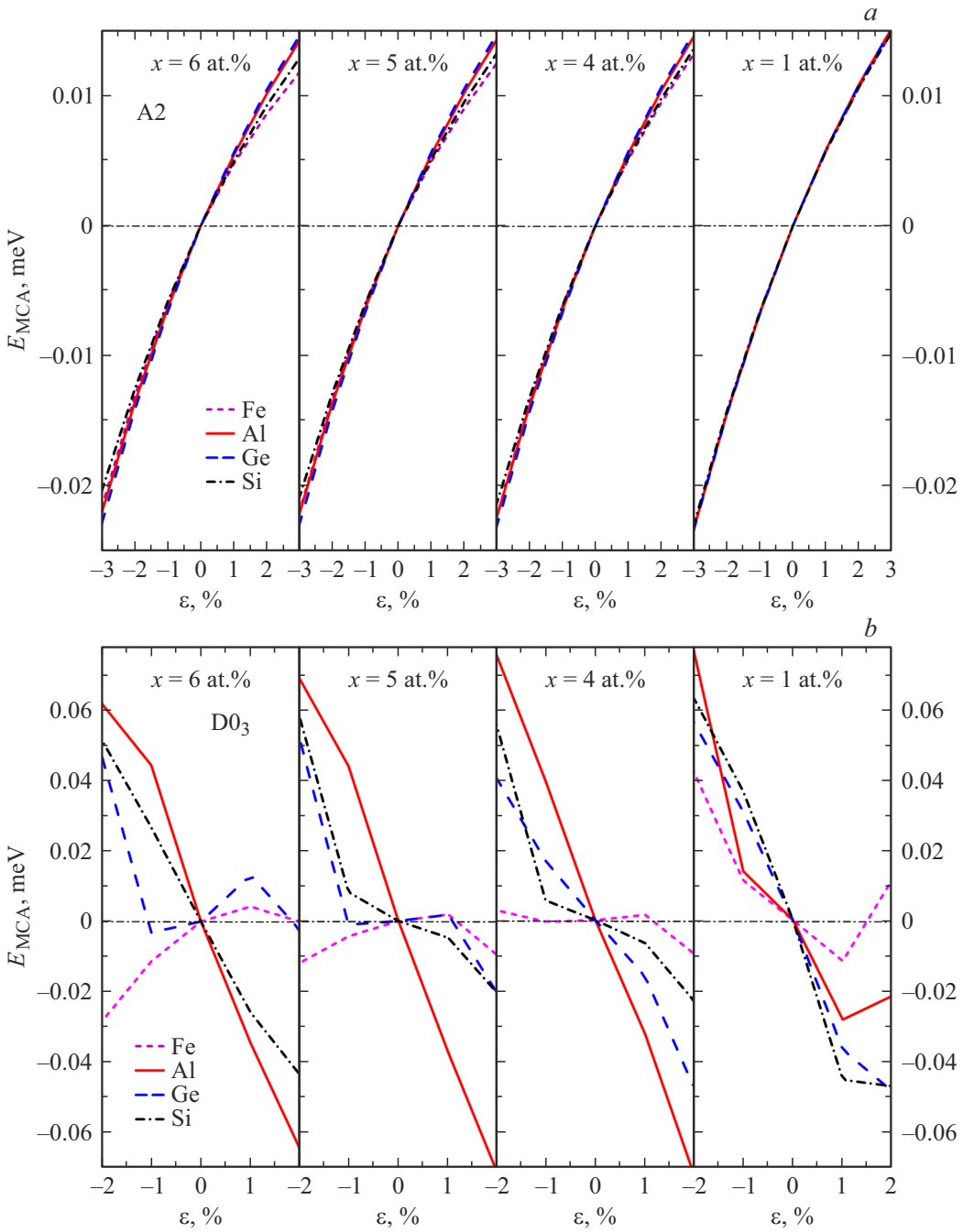

Figure 3. Dependence of the magnetocrystalline anisotropy energy E_{MCA} on the degree of low deformations ε in alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) ($0 \le x \le 6$ at.%) for the structures (*a*) A2 and (*b*) $D0_3$. Values for alloys Fe-Ga are taken from the work [13] (short dashes).

structures a_0 were determined from the curves of the energy dependence on the lattice parameter by using the Birch–Murnaghan equation of state. Based on the obtained a_0 for original cubical structures the systems were created with low degree of tetragonal distortion $\varepsilon \pm 3\%$ with persistence of the volume ($\varepsilon_x = \varepsilon_y = -1/2\varepsilon_z$). The next groups of symmetry were selected for initial cubic lattices: *Immm* (№ 71) for A2 with 2*a* (0; 0; 0) Wyckoff positions; *Fmmm* (№ 69) for $D0_3$ with Wyckoff positions 4*a* (0; 0; 0) occupied by Ga, *Z* (*Z* = Al, Ge, Si) and Fe atoms located at 4*b* (0; 0; 0.5) and 8*f* (0.25; 0.25; 0.25) sites. Tetragonal elastic modulus was determined from the ratio $C' = (d^2E_{total}/d\varepsilon^2)/3V$ [10] based on the obtained dependences of the values of total energy E_{total} on the degree of small distortions.

The second stage of calculations included determination of magnetoelastic constants $-b_1$ and tetragonal magnetostriction constants λ_{001}. Magnetoelastic constants are

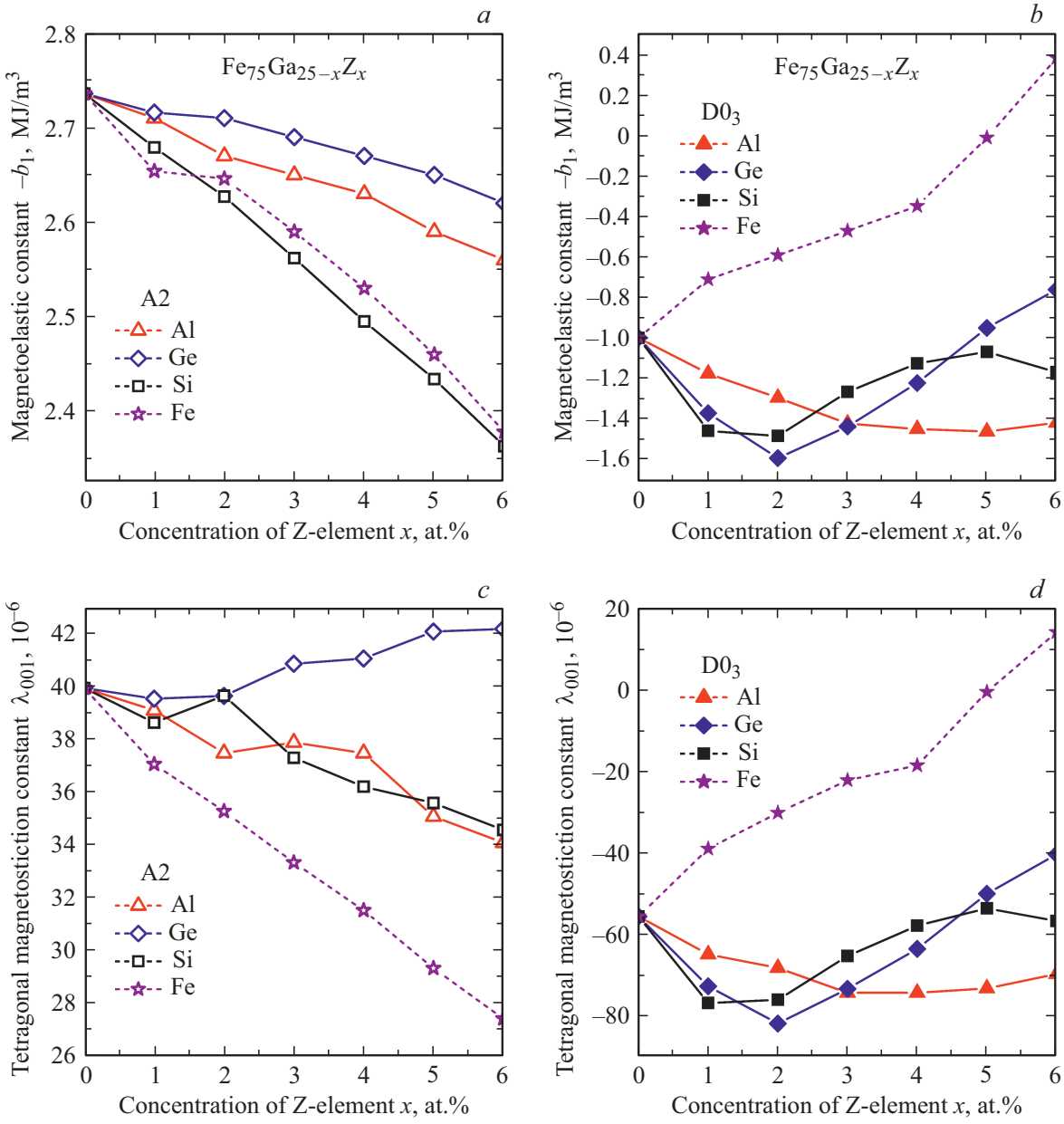

Figure 4. Dependence (a, b) of magnetoelastic constant $-b_1$ and (c, d) tetragonal magnetostriction constant λ_{001} on the concentration of Z-element in the alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) $(0 \le x \le 6$ at.%) for the structures A2 and D0$_3$. The values for alloys Fe-Ga are taken from the work [13] indicated by open and filled asterisks.

associated with the magnetocrystalline anisotropy energy E_{MCA} with the ratio $-b_1 = 2(dE_{MCA}/d\varepsilon)/3V$ [10]. For the calculation of E_{MCA}, the magnetic torque method implemented in the SPR-KKR software package was used. Its main advantage is that E_{MCA} can be calculated in one certain magnet orientation and perform integration in k-space with that orientation [11,12]. The potentials obtained by performing self-consistent calculations of the total energy of systems from the degree of small tetragonal distortions and by setting the magnetization axis along the [001] direction were used to calculate the magnetocrystalline anisotropy energy.

3. Results and discussion

Fig. 1 shows the results of calculating the equilibrium lattice parameters for the A2 and D0$_3$structures. For comparison, the values of equilibrium lattice parameters obtained for binary Fe-Ga alloys in [13] were plotted, meanwhile e.g. in terms of $Fe_{75}Ga_{25-x}Z_x$ ternary systems, the concentration of added element in the volume of 6 at.% corresponds to a substitution of Ga atoms by Fe atoms and the composition $Fe_{81}Ga_{19}$ ($Fe_{75}(Ga_{19}Fe_6)$). In general, it can be noted that in the case of the D0$_3$ phase a_0 decreases insignificantly with an increase in the number of Ge and

Al atoms, as opposed to addition of Si atoms. Such behavior is explained by the least atomic radius of Si among all atoms forming part of the alloys. In case of the structure A2, dependence $a_0(x)$ in the alloys Fe-Ga-Ge demonstrates the rising trend, while for binary alloys and the alloys with added Al and Si lower content of Ga in the system corresponds to the lower value of the lattice parameter.

Dependence of shear modulus on the content of the third element is shown in Fig. 2. As we can see in Fig. 2, *a* in the phase A2 behavior of dependence $C'(x)$ is opposite to behavior of $a_0(x)$. For the phase $D0_3$ modulus of elasticity is increased by 10% in compositions $Fe_{75}Ga_{19}(Al, Si)_6$ versus the alloy $Fe_{75}Ga_{25}$. In the $Fe_{79}Ga_{21}$ and $Fe_{75}Ga_{21}Ge_4$ compositions, the 21 at.% Ga content leads to an inflection point on the $C'(x)$ curve.

At the final stage of studies, by using the magnetic torque method, magnetoelastic constants and tetragonal magnetostriction constants, which are interrelated by the ratio $\lambda_{001} = -b_1/3C'$ [10] were determined. Fig. 3 shows dependence of the magnetocrystalline anisotropy energy on the degree of small deformations for the content of additional atom in the volume of 1, 4, 5 and 6 at.%. In the A2 phase, the slope of the $E_{MCA}(\varepsilon)$ curve remains positive, and as the number of Al, Ge, and Si atoms increases, there is a change in the slope angle in the strain range 1−3%. In the phase $D0_3$ $E_{MCA}(\varepsilon)$ demonstrates complex non-linear behavior (see Fig. 3, *b*). The considered alloys are characterized by a strong change in the slope angle of the $E_{MCA}(\varepsilon)$ curves with increasing concentration of adding elements. The maximum change of the magnetostriction energy is observed in the area of deformations ±1% in compositions close to the alloy $Fe_{75}Ga_{25}$ ($x = 1$ at.%). In binary systems with the content Ga≥ 21 at.% there is change of easy and hard axes; in case of ternary alloys, the easy axis corresponds to the direction [111].

Fig. 4 shows dependencies $-b_1(x)$ and $\lambda_{001}(x)$ on concentration of the atoms Al, Ge and Si in the structures A2 and $D0_3$ of the alloys $Fe_{75}Ga_{25-x}Z_x$ (Z = Al, Ge, Si) ($0 \le x \le 6$ at.%). In case of phase A2 higher content of Gallium corresponds to higher value of $-b_1$ and changes are virtually linear. Also it should be noted that the curves $-b_1(x)$ of Fe-Ga and Fe-Ga-Si alloys are close to each other, the same as the dependencies $E_{MCA}(\varepsilon)$. Such similarity can be explained by small difference in the Fe and Si atom sizes. Dependencies obtained for the phase A2 $\lambda_{001}(x)$ reproduce behavior $a_0(x)$ (see Fig 1, *a*, 4, *c*). Similar relationship was obtained during the study of the structure of alloys $Fe_{100-x}Ga_x$ ($15 \le x \le 30$ at.%) by means of diffraction of neutrons in the work [14]. Authors established that behavior of concentration dependence of equilibrium parameters of the lattice of A2 and $D0_3$ phases is similar to that of the magnetostriction constant. For the structure $D0_3$ the dependence profiles $\lambda_{001}(x)$ reproduce the curve profiles $-b_1(x)$, and the obtained values of tetragonal magnetostriction are negative for all ternary alloys. By absolute value the least value of λ_{001} in ternary alloys was obtained in the composition $Fe_{75}Ga_{23}Ge_2$ (-82×10^{-6}), the least in $Fe_{75}Ga_{19}Ge_6$ (-40×10^{-6}).

4. Conclusion

This work presented a comprehensive study of impact of small addition of Ge, Al and Si atoms to structural, elastic and magnetostriction properties of the $Fe_{75}Ga_{25}$ alloy. All calculations were performed within the framework of the density functional theory and the magnetic torque method implemented in the first-principles SPR-KKR software package. Increase of the lattice parameter was obtained only for the structure A2 in $Fe_{75}Ga_{25-x}Ge_x$ alloys within the whole studied range. Addition of Al and Si atoms results in continuous increase of the tetragonal modulus of elasticity by the value about 10% versus composition $Fe_{75}Ga_{25}$. It is shown that the nature of change of dependence of the constant of tetragonal magnetostriction in the phase A2 corresponds to the profile of equilibrium parameters change curves in that phase for all studied compositions. It was found that the crystall structure $D0_3$ makes a negative contribution into the value of tetragonal magnetostriction.

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Conflict of interest

The authors declare that they have no conflict of interest.

This article does not contain any researches with participation of a human as the object of studies.

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