STRUCTURAL AND MAGNETIC STATES OF MAGNETOSTRICTIVE ALLOYS Fe_3Me, Me = Al, Ga, Ge IN A WIDE TEMPERATURE RANGE

© 2025 A. M. Balagurov[a,b,c], I. S. Golovin[a,b], B. Yerzhanov[a,d,e], K. V. Kalugin[a,c,*], S. V. Sumnikov[a], D. Yu. Chernyshov[f]

[a]*Joint Institute for Nuclear Research, Dubna, Russia*
[b]*National Research Technological University MISiS, Moscow, Russia*
[c]*Lomonosov Moscow State University, Moscow, Russia*
[d]*Kazan Federal Universit, Kazan, Russia*
[e]*Institute of Nuclear Physics of the Ministry of Energy of the Republic of Kazakhstan, Almaty, Kazakhstan*
[f]*SNBL at ESRF, France, Grenoble*

[*]*e-mail:* kirivale@yandex.ru

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Abstract. A series of diffraction experiments on a number of alloys similar in composition to stoichiometric Fe_3Me with Me = Al, Ga, Ge were performed on X-ray, synchrotron, and neutron radiation sources. In the temperature range (20–1100 K), the structural, magnetic and microstructural characteristics of alloys were determined and their temperature evolution during continuous slow heating and subsequent cooling was studied. The information available in the literature on metastable and equilibrium states of alloys at elevated temperatures is clarified and specified, and their comparative analysis is performed. The identity of the temperature behavior of the alloys was observed at $T < 100$ K. The search for the tetragonal $L6_0$ phase, the formation of which in Fe-Ga alloys is considered as the main reason for the sharp increase in the magnetostriction constant, did not lead to a positive result.

Keywords: *Magnetostriction; Fe–Al, Fe–Ga, Fe–Ge alloys; structural phase transitions; magnetic structure; X-ray, synchrotron and neutron diffraction*

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INTRODUCTION

Interest in the study of materials science and structural properties of various binary iron-based alloys has recently been associated mainly with determining their similarities or differences with the properties of Fe alloys.–Ga, in which the giant magnetostriction effect was discovered in the early 2000s [1]. A detailed quantitative comparison of the magnetostrictive properties of Fe alloys–Ga, Fe–Al и Fe–Ge at the level of tetragonal magnetostriction constants, $\lambda_\gamma = (3/2)\lambda_{100}$, and the energies of magnetoelastic coupling are performed in [2]. From the data presented in this article, it follows that in Fe–Ga и Fe–Al constant $\lambda_\gamma$is positive, and in its dependence on the concentration of the element replacing iron, there is a clearly defined maximum in the region of the transition from a disordered to an ordered state (about 19 at.% Ga or Al). At this maximum $\lambda_\gamma$approximately 10 or 20 times more for Fe alloys–Al и Fe–Ga than in α-Fe, respectively. In the Fe-Ga alloy, in addition, a second magnetostriction maximum is observed at ~27 at.% Ga with a similarly large (~400 ppm) λ value.$_\gamma$. In the Fe-Ge alloy, magnetostriction is at its maximum at ~18 at.% Geby absolute valueapproximately

the same as in Fe–Al, but has a negative sign. Magnetostriction is a structurally determined property, and since understanding the listed features of Fe alloys–Me, where Me = Al, Ga, Ge has not yet been achieved, then, accordingly, an in-depth study of the details of their structural states is required.

From the existing descriptions of equilibrium phase diagrams of these alloys (see, e.g., [3 – 6]), it follows that states of compositions close to stoichiometric $Fe_{75}Me_{25} = Fe_3Me$ correspond to structural phases $D0_3$ (Fe_3Al), $L1_2$ (Fe_3Ga) or $D0_3 + B8_2$ (Fe_3Ge). However, metastable states arising at high (>30 K/min) cooling rates differ significantly from equilibrium ones, which has also been demonstrated in a number of works (see, e.g., [7, 8]). In particular, it was shown that Fe_3Al can exist in the $B2$ phase, Fe_3Ga - in the $D0_3$ phase, Fe_3Ge - in a two-phase ($D0_{19} + L1_2$) state with a small (<10%) admixture of $B8_2$ phase. These states were confirmed in a series of neutron diffraction experiments [9 – 11], which suggests that they are a bulk property of the alloys, not distorted by possible surface effects. From the results of these same experiments, it can be concluded that some both equilibrium and metastable states of Fe_3Al, Fe_3Ga and Fe_3Ge alloys need clarification, including because the type of atomic structure resulting from cooling the alloy from $T \approx 1100$ K to room temperature (RT) directly depends on its rate [12]. The behavior of the magnetic structure of alloys under temperature influences is also non-trivial, with a complex temperature dependence of magnetization due to occurring structural phase transitions. Additionally, in the hexagonal phase $D0_{19}$, found in Fe_3Ga and Fe_3Ge, besides the standard ferro-paramagnetic transition, an orientational (spin-flip) transition is also observed [11, 13].

In addition to analyzing the structural and magnetic states of Fe_3Al, Fe_3Ga, and Fe_3Ge alloys during heating and cooling to room temperature, their comparative behavior when cooled to helium temperatures is of interest. Another relevant task is to search for effects that could confirm or refute the presence of a metastable tetragonal phase $L6_0$ in these alloys. Its formation in Fe –Ga alloys was suggested in work [14] as a possible cause for the increase in the magnetostriction constant when iron is alloyed with gallium due to elastic anisotropic deformations arising in $A2$- or $D0_3$-structures. Up to the present time, the main models for explaining the giant magnetostriction effect have been based on the formation of the $L6_0$ phase in the alloy with different morphologies. It is assumed that it may be present both as regions of short-range order with a coherence length of ~10 Å and as well-ordered regions with sizes up to ~800 Å. However, the presence of this phase, at least in the form of bulk regions with long-range crystalline order, still remains in doubt. Among possible other non-standard phases, only the formation of a phase with a still unknown structure (phase $Ga_{73}Ga_{27}$, the lattice of which can be represented as hexagonal with X) has been reliably established in the composition Fe $a \approx \sqrt{8}\, a_0$, $c \approx \sqrt{12}\, a_0$, where $a_0 \approx 2.87$ Å is the lattice parameter of α-Fe [15].

This paper presents new information about structural and magnetic transformations in cast alloys with compositions close to stoichiometric Fe_3Me, Me = Al, Ga, Ge. Experimental data were obtained using neutron diffraction, X-ray and synchrotron radiation in a wide temperature range from 20 to 1100 K during slow heating and subsequent cooling. From the diffraction spectra, the temperature dependences of the position, width, and intensity of characteristic diffraction peaks of various structural phases were extracted and analyzed. The physical and material properties of these alloys are not addressed, as they are discussed in detail in many publications, particularly in review [16].

SAMPLES, EXPERIMENT AND DATA PROCESSING

The main results presented in this work were obtained on cast samples of Fe –Al, Fe –Ga and Fe –Ge, melted from corresponding mixtures of pure Fe, Al, Ga, and Ge in an induction furnace under argon atmosphere with subsequent rapid crystallization in a copper mold. More details on the sample

preparation procedure are provided in works [9 – 11]. The alloy with Ge was additionally annealed at 873 K for 48 hours and quenched in water. The chemical compositions of the ingots were determined using energy-dispersive spectroscopy with an accuracy of 0.2% and were close to the expected stoichiometric composition, namely: $Fe_{74.0}Al_{26.0}$, $Fe_{74.5}Ga_{25.5}$ and $Fe_{74.1}Ge_{25.9}$. For uniformity and brevity, they will be denoted hereafter as Fe_3Me, where Me = Al, Ga, or Ge. Some additional results were obtained on the composition $Fe_{73.4}Ga_{26.6}$, prepared in a similar manner. For neutron experiments, parallelepiped-shaped samples with dimensions of 4×8×50 mm were cut from the ingots. For synchrotron experiments, needle-shaped samples obtained by cutting and polishing were used. They were etched with a mixture of HNO_3 with ethanol to reveal individual monocrystalline grains with a typical size of ~100 μm. X-ray diffraction spectra measurements were performed on samples whose surface was previously cleaned with abrasive materials of various grain sizes, removing a surface layer of material approximately ~300 μm thick. Then the surface was etched with a mixture of HNO_3 with ethanol to relieve internal stresses introduced during grinding.

Neutron diffraction spectra measurements were performed on a high-resolution Fourier diffractometer (HRFD) at the pulsed reactor IBR-2 at JINR (Dubna) [17]. HRFD is a time-of-flight correlation diffractometer with a fast Fourier chopper and the ability to switch between high-resolution mode ($\Delta d / d \approx 0.0015$) and high-luminosity mode with medium resolution ($\Delta d / d \approx 0.015$). Neutron diffraction patterns measured with high resolution were used for the analysis of diffraction peak profiles. For diffractometer normalization and verification of the correctness of peak profile descriptions, a spectrum measured on $La^{11}B_6$ powder (NIST standard) was used. In the second mode, the complete diffraction spectrum was measured with the necessary statistics in ~1 min, and it was used for continuous temperature scanning up to ~1100 K and back to RT at a rate of ± 2 K/min. Diffraction spectra of the samples measured with high resolution in the initial state and after heating – cooling are shown in Fig. 1. The spectra clearly show weak but quite observable superstructure peaks of the phases *B* 2 (200, 222, etc.), *D* 0_3 (111, 200, 311, etc.), *L* 1_2 (100, 110, etc.), *D* 0_{19} (101, 110, etc.), which indicates their high degree of ordering.

Fig. 1. Neutron diffraction spectra of alloys in the initial state (left) and after slow heating to 1100 K and cooling to RT (right). Miller indices of several first Bragg peaks are indicated. Vertical dashes represent calculated peak positions. For Fe_3Al, peak positions are indicated for the unit cell of the *D* 0_3 phase ($a_{D03} \approx 2\, a_{B2}$). In the Fe_3Ge spectrum (c) at $d \approx 2$ Å, two weak peaks of the *B* 8_2 phase are visible. In the Fe_3Ga spectrum (e) at $d \approx 2.05$ Å, a weak 110 peak of the *A* 2 phase is visible. In the Fe_3Ge spectrum (f) at $d \approx 1.85$ Å, a weak 200 peak of the *A* 1 phase is visible.

The designation shown in Fig. 1d *B* 2/ *D* 0_3 refers to the complex microstructural state discussed further: a *B* 2 matrix with dispersed regions (clusters) of the *D* 0_3 phase distributed within it. A more detailed description of the neutron research methodology is contained in review [18], which describes a number of physical problems solved for Fe –Ga and Fe –Al alloys using experiments performed on FDNR. In addition to analyzing the evolution of structural phases, they also included determining the temperature dependence of the directions and magnitude of iron magnetic moments.

Synchrotron diffraction experiments were performed at ESRF (Grenoble) at beamlines BM01A [19] ($\lambda_0 = 0.6867$ Å) and ID28 [20] ($\lambda_0 = 0.6968$ Å). Both beamlines are equipped with multi-pixel Pilatus detectors that allow detailed scanning of large volumes of reciprocal space. Data from the BM01A beamline allowed quantitative analysis of Bragg peak intensities. Data from the ID28 beamline, which has approximately 100 times higher photon flux, were used for analysis of diffuse scattering and weak superstructure diffraction peaks. The small cross-section of the photon beam at this beamline (0.02×0.04 mm^2) allows collecting diffraction information from a single monocrystalline grain with minimal contribution from neighboring grains.

X-ray diffraction spectra measurements were carried out on a PANalytical Empyrean laboratory diffractometer equipped with Cu and Co X-ray tubes and a highly efficient position-sensitive Pixel3D detector. Scanning was performed in Bragg-Brentano geometry in the scattering angle range from 20° to 100° with a step of 0.013° and exposure times from 200 to 4000 seconds per step. A Ni filter was used to suppress the contribution of the CuK K_β line. Filtering of the CuK $K_{\alpha 2}$ line was not performed, so the X-ray diffraction peaks had the form of a $K_{\alpha 1}$ - $K_{\alpha 2}$ doublet.

Diffraction data analysis was carried out using FullProf [21] and Fityk [22] software packages. FullProf was used for spectrum analysis by Rietveld method, while Fityk was used to extract the main geometric characteristics of diffraction peaks: amplitude, area, position, and width. The high symmetry of the alloys' crystal lattices, relatively small unit cell parameters, and sufficiently high resolution of the diffractometers used determined that a large number of diffraction peaks were single, and geometric characteristics were determined individually for each peak. Peak profiles were described by the Voigt function, which is a convolution of Gaussian and Lorentzian functions. Conclusions about the microstructure of alloys were made based on the functional dependencies of diffraction peak widths (full width at half maximum was analyzed) on interplanar spacing. The resolution level of high-resolution X-ray diffraction allowed determining the characteristic sizes of coherent scattering regions (CSR) if $L_{coh} < 3000$ Å, and microstrains in crystallites (static fluctuations of unit cell metric parameters, $\varepsilon \approx \Delta a / a$), if $\varepsilon > 3 \cdot 10^{-4}$.

Fig. 2. 2D representation of the evolution of diffraction spectra for the $Fe_{74.5}Ga_{25.5}$ composition, measured during its heating to 1100 K (+2 K/min) and subsequent cooling to RT (− 2 K/min). Temperature axis – bottom to top, interplanar spacing axis – left to right. During heating, the following transitions are observed: $D0_3 \rightarrow L1_2 \rightarrow D0_{19} \rightarrow A2$. During cooling, the following transitions are observed: $A2 \rightarrow D0_3 \rightarrow L1_2$. The measurement time for one spectrum was 1 min, and the entire 2 D -map contains about 850 spectra.

Phase transformations occurring in samples during heating – cooling were tracked using 2D maps of diffraction peak intensity changes. The map for Fe_3Ga is shown in Fig. 2, maps for Fe_3Al and Fe_3Ge are presented in [10, 11]. The transition temperatures between different structural phases were determined by the disappearance or appearance of characteristic peaks. The changes in volume fractions of structural phases and their unit cell parameters were determined from the temperature dependencies of intensities and positions of the main peaks.

FEATURES OF DIFFRACTION IDENTIFICATION OF ORDERED PHASES Fe_3Me

At element concentrations corresponding to the formula Fe_3Me, the phases $D0_3$, $L1_2$ and $D0_{19}$ can be ordered at low temperatures. In this case, the Bragg peaks in the diffraction spectrum can be divided into two groups. The intensities of the first group of peaks (commonly called fundamental) do not depend on the degree of ordering of Fe and Me atoms, i.e., they do not disappear if a transition to a disordered state occurs, for example, $D0_3 \rightarrow A2$. The intensities of the second group of peaks (commonly called superstructure peaks) are determined by the degree of structural ordering (ξ), which depends on the alloy preparation method and temperature: $I(T) \sim \xi(T)^2$, $0 \le \xi(T) \le 1$. The structure factors, normalized to the number of formula units, for both fundamental (F_F) and superstructure (F_S) peaks are identical for the ordered phases $D0_3$, $L1_2$ and $D0_{19}$ and are given by the expressions:

$$F_F = 3b_{Fe} + b_{Me}, \qquad F_S = b_{Fe} - b_{Me}, \tag{1}$$

Table 1. Basic structural characteristics of ordered phases observed and presumed in Fe_3Me, where Me = Ga, Al, Ge. The columns indicate the symmetry of the crystal lattice, space group, unit cell parameters, number of atoms in the cell, and atomic volume (cell volume per 1 atom) for Fe_3Ga. *X* means the phase detected in the work [15]. At the end of the table, for reference purposes, are given the characteristics of the element structures

Phase	Symm.	Space group	Fe_3Ga, Å	Fe_3Al, Å	Fe_3Ge, Å	*N*	V_a, Å^3
B 2	cub.	$Pm\bar{3}m$	$a \approx 2.91$	$a \approx 2.93$		2	12.32
D 0 $_3$	cub.	$Fm\bar{3}m$	$a \approx 5.81$	$a \approx 5.79$		16	12.27
L 1 $_2$	cub.	$Pm\bar{3}m$	$a \approx 3.68$		$a \approx 3.66$	4	12.46
D 0 $_{19}$	hex.	*P* 6 $_3$ / *mmc*	$a \approx 5.20$ $c \approx 4.23$		$a \approx 5.18$ $c \approx 4.22$	8	12.40
B 8 $_2$	hex.	*P* 6 $_3$ / *mmc*			$a \approx 4.03$ $c \approx 5.03$	6	11.79
L 6 $_0$	tet.	*P* 4/ *mmm*	$a \approx 4.11$ $c \approx 2.90$			4	12.25
D 0 $_{22}$	tet.	*I* 4/ *mmm*	$a \approx 3.70$ $c \approx 7.20$			8	12.32
X	hex.	?	$a \approx 8.12$ $c \approx 9.94$			48	11.82
α-Fe	cub.	$Im\bar{3}m$	$a \approx 2.867$			2	11.78
Ga	ortho.	*Cmca*	$a \approx 4.517$ $b \approx 7.645$ $c \approx 4.511$			12	12.98
Al	cub.	$Fm\bar{3}m$	$a \approx 4.050$			4	16.61
Ge	cub.	$Fd\bar{3}m$	$a \approx 5.660$			8	22.67

where b_{Fe} and b_{Me} – are atomic scattering factors in the case of X-ray or synchrotron radiation or coherent scattering lengths in the case of neutron diffraction (the Debye –Waller factor is omitted for brevity). In photon scattering, for estimating structural factors of peaks with large d_{hkl} instead of *b* one can use atomic numbers of elements: $Z_{Fe} = 26$, $Z_{Al} = 13$, $Z_{Ga} = 31$, $Z_{Ge} = 32$. For the ratio of X-ray structural factors of superstructural and main peaks for Fe-Ga, we get: $F_S / F_F = (b_{Fe} - b_{Ga}) / (3 b_{Fe} + b_{Ga}) \approx 0.046$. Since $I \sim |F|^2$, the ratio of peak intensities under otherwise equal conditions is: $I_S / I_F \approx 0.002$. In the case of neutrons, the following values should be used for calculations: $b_{Fe} = 9.45$, $b_{Al} = 3.45$, $b_{Ga} = 7.29$, $b_{Ge} = 8.18$ fm (1 fm = 10^{-13} cm) [23]. It can be verified that neutron superstructural peaks are somewhat more intense than X-ray ones, and they are most intense in the case of Fe_3Al due to the greatest contrast between the scattering factors of Fe and Al: $F_S / F_F = (b_{Fe} - b_{Al})/(3 b_{Fe} + b_{Al}) \approx 0.189$, $I_S / I_F \approx 0.036$.

From these estimates, it follows that the task of registering superstructure peaks of Fe_3Me alloys is difficult but quite feasible, as can be seen from the spectra shown in Fig. 1 and Fig. 2. For compositions with slightly disturbed stoichiometry, for example, for $Fe_{74}Me_{26} \approx Fe_{2.96}Me_{1.04}$, the overall diffraction pattern remains unchanged. The intensities of superstructure peaks slightly decrease due to reduced contrast, and the incoherent background somewhat increases. Significant changes can occur only with a stronger violation of stoichiometry (± 1 atom per cell and more). In

this case, with ordered substitution, for example, of an Me atom with an excess Fe atom, the prohibition rules outlined above are violated, and peaks forbidden for the ordered composition Fe_3Me may appear in the diffraction spectrum.

The main structural characteristics of the listed ordered phases are given in Table 1. If disordering occurs, the phases $B2$ and $D0_3$ transition to the BCC-phase $A2$ (the cell parameter of $D0_3$ decreases by a factor of 2), $L1_2$ - to the FCC-phase $A1$, $D0_{19}$ - to the HCP-phase $A3$ (the cell parameter a decreases by a factor of 2). In addition to the mentioned phases, the tetragonal phase $D0_{22}$ is included in the table, which in some models, like $L6_0$, is considered as a possible cause of increased magnetostriction of Fe –Ga alloys. Data for cell parameters are taken from review papers [16, 18], data for the phase $D0_{22}$ - from [24]. The parameter values are approximate (depending on the measurement temperature).

STRUCTURAL STATES OF ALLOYS Fe_3Me

Structural states upon heating to 1100 K. In the quenched state of the Fe_3Al alloy, its diffraction spectrum (Fig. 1a) corresponds to the $B2$ phase, for which the indices of superstructure peaks satisfy the condition $h + k + l = 2n$, but $\neq 4n$. Since strict ordering of the structure according to the $B2$ type is only possible for the composition Fe_1Me_1, the structure of Fe_3Al contains a significant number of defects that disrupt the long-range order. Indeed, as follows from the analysis of diffraction peak profiles and from the functional dependence of their width on the interplanar spacing, the characteristic size of the coherent scattering regions (L_{coh}) does not exceed 600 Å. When the alloy is heated in the temperature range of (570-820) K, diffraction peaks of the ordered $D0_3$ phase appear. But as follows from the analysis of the diffraction peak widths [10], this phase does not fill the entire volume of the sample, but exists in the form of clusters with characteristic sizes of ~(50-200) Å. Further heating leads first to the disappearance of $D0_3$ clusters and the formation of $B2$, and then to the transition to a completely disordered $A2$ state. Thus, during slow heating of the quenched Fe_3Al alloy, transitions occur: $B2 \rightarrow B2/D0_3$ (at 570 K) $\rightarrow B2$ (at 820 K) $\rightarrow A2$ (at 1090 K).

Fig. 3. Parameters of the elementary cells of the matrix (triangles, a_F, doubled value) and clusters (rhombuses, a_S) in the Fe_3Ga alloy, determined from the interplanar distances of individual (main and superstructure) diffraction peaks.

The diffraction spectrum of the Fe_3Ga alloy in the as-cast state (Fig. 1b) corresponds to the metastable phase $D0_3$. In work [9], its microstructure was described as homogeneous with characteristic CSR sizes at the level of $L_{coh} \approx 2000$ Å. However, new diffraction data obtained in the present work better correspond to the same microstructure model as for the Fe_3Al alloy – clusters of the $D0_3$ phase embedded in a matrix with a $B2$ structure. This follows from the analysis of the diffraction peak width using the Williamson- –Hall method and from the small but evident difference in the unit cell parameters of the matrix and clusters (Fig. 3). The unit cell parameter of the more ordered $D0_3$ phase was determined from the positions of superstructure peaks allowed only in this phase. It turned out to be ≈0.0004 Å smaller than the unit cell parameter of the $B2$ phase, which is dominant in the sample. The effect of a small decrease in the unit cell parameter during ordering is well known for intermetallic compounds (see, e.g., review [25]) and has been previously observed in various compositions of Fe –Al and Fe –Ga, including during temperature scanning [26]. In this case, the small magnitude of the difference between a_F e a_S ($\Delta a / a \approx 8 \cdot 10^{-5}$) is associated with the proximity of the structures of the $B2$ and $D0_3$ phases and the incomplete level of ordering of these phases. During slow heating of the alloy (2 K/min), structural phase transitions occur: $B2/D0_3 \rightarrow L1_2$ (at 710 K) $\rightarrow D0_{19}$ (at 890 K) $\rightarrow A2$ (at 950 K).

In the Fe_3Ge alloy sample, the phases $L1_2$ and $D0_{19}$ are present in approximately equal proportions. During slow heating, the disordering of the $L1_2$ phase (transition to $A1$) occurs at $T \approx 710$ K, and at $T \approx 980$ K this phase transitions to $A3$. The proportion of the $D0_{19}$ phase in the sample remains unchanged up to $T \approx 1070$ K, above which a transition to the disordered state $A3$ occurs. Thus, when heating the two-phase Fe_3Ge, the transitions $L1_2 \rightarrow A1$ (at 710 K) $\rightarrow A3$ (at 980 K) and $D0_{19} \rightarrow A3$ (at 1070 K) occur.

Types of crystal lattices are the same for phases $A2$, $B2$ and $D0_3$ (BCC [1]), $A1$ and $L1_2$ (FCC), $A3$ and $D0_{19}$ (HCP) and structural transitions within these groups, for example, $B2 \leftrightarrow D0_3$, are second-order transitions of the order – disorder type. Their basis – diffusive movements of individual atoms, with the distances they move being comparable in magnitude to the size of the unit cell. Transformations between phases with different types of lattices are first-order transitions, occurring with a volume jump; they are based on cooperative movements, with atomic displacements being small, and the environment of specific atoms changes little or not at all. According to the IUPAC classification [27], order-disorder transitions should be designated as diffusive, while transitions with a volume jump as displacive. A detailed analysis performed in [28] allowed establishing a more complex (combined) nature of transitions between phases with different types of lattices ($D0_3 \leftrightarrow L1_2 \leftrightarrow D0_{19}$). Specifically, they include both displacive and diffusive components of atomic displacements. The diffusive stage is necessary for the formation of intermediate, disordered states, the transition between which leads to a change in the lattice type, and, for example, the transition from $D0_3$ to $L1_2$ should be written as $D0_3 \rightarrow A2 \rightarrow A1 \rightarrow L1_2$. In [28], it is suggested that this scheme has a general character, namely: the transition between structurally ordered phases belonging to different types of crystal lattices must include a stage of structural disorder.

Fig. 4. Temperature dependences of atomic volumes: Fe_3Al alloys (right scale) and Fe_3Ge (left scale) (a); pure iron and Fe_3Ga alloy, determined during their heating at a rate of 2 K/min (b). In the initial state of the Fe_3Ge alloy, phases $L1_2$ (upper curve) and $D0_{19}$ (lower curve) are present in approximately equal proportions. Vertical lines indicate 1st order structural transitions for Fe and Fe_3Ga and 2nd order for Fe_3Al and Fe_3Ge alloys. Approximate values of volumetric thermal expansion coefficients (in units of 10^{-5} 1/deg), determined from sections with linear change in cell volume, are indicated.

The differences between these two types of transitions - purely diffusional and combined - are well manifested in the dependences of atomic volume on temperature (Fig. 4). In Fe_3Al and Fe_3Ge alloys during transitions ($L1_2 \rightarrow A1$ and $D0_{19} \rightarrow A3$), there is no change in the lattice type, respectively, for them the dependences $V_a(T)$ are continuous. In contrast, in Fe_3Ga all observed transitions are combined and, just like the α-Fe ↔ γ-Fe transition, occur with a volume jump. The magnitudes of the jumps differ significantly, amounting to $\Delta V_a / V_a \approx 0.010$ for $D0_3 \rightarrow L1_2$, 0.002 for $L1_2 \rightarrow D0_{19}$, 0.0015 for $D0_{19} \rightarrow A2$, but, in principle, they are comparable with $\Delta V_a / V_a \approx 0.005$ during the α-Fe → γ-Fe transition.

With slow cooling to RT, alloys transition to an equilibrium (or near-equilibrium) state. In this process, Fe_3Al undergoes transitions: $A2 \rightarrow B2$ (at 1090 K) $\rightarrow B2/D0_3$ (at 790 K), in Fe_3Ga: $A2 \rightarrow D0_3$ (at 930 K) $\rightarrow L1_2$ (at 810 K), in Fe_3Ge: $A3 \rightarrow D0_{19}$ (at 1070 K). At RT, traces of $_3Ga$, and traces of $A2$ and $A3$ phases are observed in Fe_3Ge. The corresponding high-resolution neutron diffraction spectra are shown in Fig. 1. $A1$ and $B8_2$ are observed in Fe

[1] Formally, the structure of the phase $D0_3$ is described within the FCC group, which is related to the ordering of Ga atoms.

Structural states during cooling to 20 K. Cooling below RT (down to helium temperatures) does not lead to changes in the structural phase state of these alloys fixed at RT. The only effect is the temperature change in atomic volume, which by definition is represented as

$$V_a(T) = V_a(T_0)\left[1 + \int_{T_0}^{T} \beta(T)dT\right], \quad (2)$$

where $\beta(T)$ is the volumetric coefficient of thermal expansion (CTE). For metallic crystals with cubic symmetry $\beta(T) \sim aC_e(T) + bC_v(T)$, where a and b are some coefficients, C_e and C_v are contributions to the crystal heat capacity from conduction electrons and atomic dynamics (phonons). It is known that at low temperatures, approximately at $T < 0.1\Theta_D$, where Θ_D is the Debye temperature, $C_e(T) \sim T$, $C_v(T) \sim T^3$ (see, e.g., [29]), and integration in (2) leads to the dependence:

$$Va(T)/Va(T0) = [1 + a/2\cdot(T - T0)^2 + b/4\cdot(T - T0)^4]. \quad (3)$$

According to the data presented in [30] for Fe, the volumetric CTE at low temperatures is: $\beta(T) = 3\alpha(T) = 3(32\cdot10^{-10}T + 0.8\cdot10^{-11}T^3)$, where $\alpha(T)$ is the linear CTE, from which it follows that the electronic and lattice contributions are approximately equal at $T = 20$ K. At low temperatures, the atomic volumes of Fe_3Me alloys, obtained from our X-ray data, are shown in Fig. 5. The absolute values of atomic volumes at 20 K differ significantly: $V_a(Fe) \approx 11.71$ Å^3, $V_a(Fe_3Al) \approx 12.06$ Å^3, $V_a(Fe_3Ga) \approx 12.18$ Å^3, $V_a(Fe_3Ge) \approx 12.23$ Å^3, but, as can be seen from Fig. 6, their temperature behavior at $T < 100$ K is similar.

Fig. 5. Temperature dependence of atomic volumes of Fe_3Me alloys for Me = Al, Ga, Ge in the low temperature range.

Fig. 6. Normalized at 20 K values of atomic volumes of Fe_3Me alloys in the low temperature range. The curve (dashed) for Fe is plotted using data from [30].

Modified *D* 0$_3$ -phase. In work [14], it was shown that the exchange of positions Fe ↔ Ga in two pairs of Fe –Ga atoms in the $D0_3$ phase in positions (8c) of group Fm3m leads to a structure that became known as modified $D0_3$ (*m*-$D0_3$)-phase. By transforming the coordinate system, it can be represented as a tetragonally distorted $L1_2$ structure (phase $L6_0$, prototype $CuTi_3$, space group $P4/mmm$). Assumptions about the presence of this phase in Fe –Ga alloys in the form of structurally ordered regions, which are a transitional metastable structure, are presented in a large number of experimental works. Mainly, they are based on two effects: deformation of the profiles of the main diffraction peaks observed in X-ray and synchrotron experiments, and the presence of specific superstructure diffraction spots recorded in electron diffraction experiments. Such evidence began to appear shortly after the publication of work [14] (see, e.g., [31]), continues to appear at present (see, e.g., [32]), and so far relates only to Fe –Ga alloys. However, in paper [33] it was already pointed out that the reliability of this evidence is not absolute. Indeed, recent X-ray diffraction experiments [34] have shown that the observed splitting of the main diffraction peak profiles occurs due to the formation of *A* 2- and *B* 2-structures in the near-surface layers at depths up to 10 μm and is not related to the tetragonal phase $L6_0$. Moreover, simple model calculations show that the main peaks should split into three components, not two, as seen from the above-mentioned articles with X-ray data. Superstructure diffraction peaks, resolved in the $L6_0$ phase, were detected only in SAED (Selected –Area –Electron –Diffraction) experiments. Attempts to register them using other types of radiation have not yet been successful.

The structure of the ordered phase $L6_0$ of composition Fe_3Me can be described in the primitive tetragonal group $P4/mmm$, where the Ga atom is located at the site (0, 0, 0) and Fe atoms are at the centers of the faces. With complete coherence between the unit cells of phases $D0_3$ and $L6_0$, the

unit cell parameters of the latter should be: $a = a(D0_3)/\sqrt{2} \approx 4.11$ Å, $c = a(D0_3)/2 \approx 2.90$ Å, $c/a = 1/\sqrt{2} = 0.707$, 4 atoms per cell, $V_a \approx 12.25$ Å^3. For the main peaks of the $L6_0$ phase, the FCC-cell selection rule applies (111, 200, etc.), while the superstructure peaks have mixed Miller indices, and their structure factors are calculated using formulas (1). The total energy of the $L6_0$ phase is related to its degree of tetragonality [24], which can vary from $c/a = 0.707$ to 1, corresponding to the transition $L6_0 \rightarrow L1_2$. Data in the literature on the unit cell vary significantly, for example, $a = 4.10$ Å, $c = 2.98$ Å, $c/a = 0.727$ in the composition $Fe_{73}Ga_{27}$ [24] and $a = 4.05$ Å, $c = 2.87$ Å, $c/a = 0.709$ in the composition $Fe_{81}Ga_{19}$ [35].

The volume fraction of the phase $L6_0$ according to data from different sources ranges from (3-5)% [36, 37], up to ~15% in $Fe_{81}Ga_{19}$ composition [38]. Of course, with such quantities of the phase, its specific superstructural diffraction peaks should be confidently observed in X-ray, synchrotron, or neutron experiments, but for some reason they cannot be registered. Based on the level of background fluctuations at the locations of the superstructural peaks of the phase $L6_0$, in work [39] for the composition $Fe_{81}Ga_{19}Tb_{0.1}$ upper estimates of the possible content of this phase in the sample volume were obtained: 2% according to neutron data and 0.2% according to synchrotron data, which is significantly less than the values given in [36 – 38].

Fig. 7. *Hkk* -layer of reciprocal space of the $Fe_{73}Ga_{27}$ alloy, reconstructed from diffraction data obtained at 20°C at the ID28 station (ESRF). The lattice and Miller indices are given for the cubic cell $D0_3$ with parameter $a \approx 5.81$ Å. In $D0_3$ reflections with all even or all odd Miller indices are allowed. Reflections with mixed indices belong to the phase X. The intensity distribution in the highlighted direction [2 *kk*] is shown in Fig. 8.

Fig. 8. Intensity distribution in the direction [2 *kk*] in the layer *hkk* shown in Fig.7.

For compositions with a high Ga content, it is not possible to obtain such estimates, since they contain the phase X mentioned in the introduction. The hexagonal lattice of this phase can be represented as cubic with parameter $a \approx 3\,a(D0_3) \approx 17.43$ Å, and all possible reflections of the $L6_0$ phase overlap with reflections of the X phase (Fig. 7). The amplitudes of the X phase peaks do not exceed 10^3 counts (Fig. 8), whereas the amplitudes of the main peaks of the $D0_3$ phase are $>10^7$. Nevertheless, the peaks of the X phase in the $Fe_{73}Ga_{27}$ alloy are confidently registered.

Fig. 9. *Hhl* -layer of reciprocal space of the $Fe_{73}Al_{27}$ alloy, reconstructed from diffraction data obtained at 20°C at the ID28 station (ESRF). The lattice and Miller indices are given for the cubic cell $D0_3$ with parameter $a \approx 5.78$ Å. For the main (strong) reflections, the Miller indices satisfy the condition $h + k + l = 4n$ (-2-2-4, 00-4, etc.). The remaining reflections are superstructural. The intensity distribution in the highlighted direction [*hhl*] is shown in Fig. 10.

Fig. 10. Intensity distribution in the direction [*hhl*] in the *hhl* layer shown in Fig. 9.

In other Fe_3Me alloys, this phase has not yet been found. For example, from the reciprocal space section of the $Fe_{73}Al_{27}$ alloy shown in Fig. 9, it follows that all observed reflections correspond to the symmetry of the $D0_3$ phase. The intensity distribution in one of the directions in this section is shown in Fig. 10. The main peaks (-2-20 and 22-4), whose intensities are greatly underestimated due to count losses, and superstructural peaks are clearly visible, but there are no traces of non-standard phases X and $L6_0$.

MAGNETIC STATE OF ALLOYS

Fe_3Al, Fe_3Ga and Fe_3Ge alloys are ferromagnetic at room temperature and transition to paramagnetic state (FM → PM) when heated above 820, 910 and 730 K respectively. For Fe_3Ga and Fe_3Ge, a complex dependence of magnetization on temperature $M(T)$ is observed due to structural

transitions occurring during heating [9, 11]. With slow cooling and formation of stable structural phases in Fe_3Al, Fe_3Ga and Fe_3Ge, the dependencies $M(T)$ become more regular, typical for PM → FM transitions at $T_C \approx 800$, 720 and 630 K respectively (Fig. 11). Visible irregularities in $M(T)$ are associated with the formation of FM states of various phases. For example, for Fe_3Ge, the irregularity at 470 K is associated with the formation of the FM state of the $B8_2$ phase.

Fig. 11. Temperature dependencies of magnetization of Fe_3Me alloys, measured during cooling at a rate of 6 K/min. The temperatures of transitions to the ferromagnetic state are marked with vertical lines.

In cubic phases $D0_3$ and $L1_2$ the magnetic moments of Fe are directed along one of the unit cell axes. A feature of the hexagonal phase $D0_{19}$ of the Fe_3Ge alloy is the aforementioned presence of an orientation transition of magnetization. When the alloy is cooled, the magnetic moments of iron first align along the 6th order rotation axis in the range of (630–380) K, and then rotate into the basal plane of the hexagonal structure (see [11] for details). The temperature dependencies of the ordered iron moment magnitude were determined using the Rietveld method (FullProf package). For the Fe_3Ge alloy, examples of spectra processing and temperature dependencies of magnetic moment components (μ_x and μ_z) in the $D0_{19}$ phase during heating and cooling are given in [11]. The parametric description of the $\mu(T)$ dependencies was carried out using a phenomenological power function:

$$\mu(T) = \mu_0[1 - (T / T_C)^{\alpha}]^{\beta}, \quad (4)$$

where μ_0 is the magnetic moment at $T = 0$, T_C is the Curie temperature, α and β are refinable parameters. This formula allows for an accurate reproduction of the experimental dependence over a wide temperature range. At temperatures close to T_C, equation (4) transforms into the standard formula $\mu(T) \sim [1 - T / T_C]^{\beta}$, which at $\beta = 0.5$ is valid for the mean field model. Figure 12 shows how function (4) reproduces the experimental dependence of the ordered moment of the $L1_2$ phase in the $Fe_{73}Ga_{27}$ composition with parameters: $\mu_0 = (2.2 \pm 0.1)\ \mu_B$, $T_C = (760 \pm 3)$ K, $\alpha = (10 \pm 1)$, $\beta = (0.64 \pm 0.09)$. For the stoichiometric composition Fe_3Ga, the dependence is similar, with a small shift in temperature ($T_C \approx 720$ K).

Fig. 12. Temperature dependence of the ordered magnetic moment of iron in the $L1_2$ phase of $Fe_{73}Ga_{27}$ alloy during its slow cooling. The line corresponds to the phenomenological formula (4) with parameters specified in the text.

DISCUSSION AND CONCLUSIONS

The presented data on the structural states of cast Fe_3Al, Fe_3Ga and Fe_3Ge alloys before and after slow heating – cooling and their changes during heating – cooling clarify and specify the information available in the literature. A schematic representation of the states registered in these alloys at elevated temperatures is shown in Fig. 13.

Fig. 13. Phase states of Fe_3Al, Fe_3Ga and Fe_3Ge alloys at elevated temperatures during slow heating and subsequent cooling. In the initial state of the Fe_3Ge alloy, the phases $D0_{19}$ and $L1_2$ are present in approximately equal volume fractions.

The structural transitions in Fe_3Al look the simplest. However, even in this case, there are noticeable differences from the information presented on the equilibrium phase diagram of this alloy [3]. In addition to the initial stage ($B2$ up to 570 K and $B2/D0_3$ up to 820 K), the final state at RT is also non-standard. Judging only by the diffraction spectrum (Fig. 1d), it corresponds to the $D0_3$ phase with well-defined superstructural peaks (111, 200, 311, etc.). From a more detailed analysis, including consideration of the diffraction peak widths, it follows that, just as in the initial heating

stage, this state represents a matrix with the structure of a partially ordered $B2$ phase and dispersed areas (clusters) of the $D0_3$ phase distributed within it. The real morphology of the clusters is unknown; computer modeling predicts the presence of interpenetrating regions with blurred boundaries and complex topology. Nevertheless, from the perspective of radiation diffraction, the clusters of the $D0_3$ phase can be considered as connected regions with a different type and degree of order than in the matrix. A characteristic size of the coherent scattering regions (CSR) can be assigned to such a set of regions, which can be determined from diffraction data using the Scherrer or Williamson- –Hall approximations (for more details, see [18]).

Fig. 14. Dependencies $(\Delta d)^2$ of d^2 for the Fe_3Al alloy in states before heating (triangles) and after the heating-cooling cycle (crosses and rhombuses). The widths of the peaks allowed in the initial $B2$ phase follow a quadratic relationship corresponding to an average CSR size $L_{coh} \approx 580$ Å. The peak widths after heating-cooling are described by a quadratic relationship for the superstructural peaks of the $D0_3$ phase ($L_{coh} \approx 650$ Å) and a linear relationship for the $B2$ phase. The values of $(\Delta d)^2$ are multiplied by 10^6.

Example of Williamson –Hall plot for diffraction peak width determined from neutron diffraction patterns of initial and final states of Fe_3Al, measured on HRFD, is shown in Fig. 14. The linear character of the dependence $(\Delta d)^2$ on d^2 means that size effect in peak width is absent (large CSRs). Parabolic character of this dependence means that characteristic sizes of CSRs are small. From Fig. 14 it follows that in the initial state, the alloy structure represented a homogeneous but highly defective $B2$ phase (average CSR size $L_{coh} \approx 580$ Å). After the heating – cooling procedure, the defect degree of the $B2$ phase significantly decreased, and the $D0_3$ phase clusters formed in it as a matrix have a characteristic size $L_{coh} \approx 650$ Å. Thus, the equilibrium state of Fe_3Al is the $B2/D0_3$ phase, not $D0_3$, as indicated on phase diagrams. Furthermore, it should be noted that all structural transitions in Fe_3Al are second-order transitions occurring without jumps in atomic volume.

The microstructure of the initial state of Fe_3Ga, just like Fe_3Al, represents a matrix of $B2$ with $D0_3$ clusters distributed in it. The difference consists in a significantly lower degree of defectiveness of both phases, with characteristic CSR (coherent scattering region) sizes of ~2500 Å. Another distinctive feature of this alloy is the single-phase equilibrium state obtained at a cooling rate of 2 K/min, representing a $L1_2$ structure. A slight increase in Ga content to 27 at.% leads to the appearance of a significant, up to 50%, proportion of the $D0_{19}$ phase after heating – cooling of the alloy at the same rate of 2 K/min [9]. Unlike Fe_3Al, structural transitions in Fe_3Ga include first-order transitions between phases with different types of crystal lattices. In reality, these transitions are combined, including both shear and diffusion components of atomic displacements. The resulting volume jumps are comparable in magnitude to the jump ($\Delta V_a / V_a \approx 0.005$) during the α-Fe → γ-Fe transition.

The initial state of cast Fe_3Ge is two-phase, $L1_2 + D0_{19}$, with approximately equal volume fractions of phases. Their structural transformations during heating occur independently of each other, in areas initially occupied by specific phases. The $D0_{19}$ phase transitions to a disordered $A3$ state at $T \approx 1100$ K. In areas occupied by the $L1_2$ phase, $A3$ also appears, but through an intermediate $A1$ state and at a noticeably lower temperature, $T \approx 980$ K. In contrast to the published phase diagrams [3, 6], which predict a chain of transitions $A3 \rightarrow D0_{19} \rightarrow L1_2 \rightarrow D0_3 + B8_2$ during slow cooling from 1100 K, according to our data, only a single transition $A3 \rightarrow D0_{19}$ is realized, while the $B8_2$ phase is constantly present, with its fraction (~10%) being practically independent of temperature.

All ordered structural phases present in Fe_3Al, Fe_3Ga and Fe_3Ge are ferromagnetic in a certain temperature range. In Fe_3Ga, the temperature dependence of magnetization, shown in [9], is irregular due to structural transitions $B2/D0_3 \rightarrow L1_2 \rightarrow D0_{19}$, with different T_C for the phases that replace each other. The orientational magnetic transition in the $D0_{19}$ phase, observed in Fe_3Ge, is also

possible in Fe_3Ga, but reliable experimental data for its analysis are currently lacking. The highest T_C during heating (920 K) was recorded for the $D0_{19}$ phase in Fe_3Ga. During cooling, the Fe_3Al alloy transitions to the ferromagnetic state earlier than the others ($T_C \approx 800$ K), while for Fe_3Ge, T_C is almost 200 K lower. After cooling to room temperature, the magnitudes of the ordered magnetic moments are approximately the same for all three alloys and close to the moment of pure iron, $\mu_{Fe} \approx 2.2\ \mu_B$.

The listed results were obtained in neutron diffraction experiments using high-resolution modes and thermo – diffractometric scanning with high data acquisition rates. Their important feature is the bulk nature of the obtained information, not distorted by surface effects or structural inhomogeneities. The synchrotron diffraction experiments performed at ESRF were primarily aimed at searching for the tetragonal phase $L6_0$, the formation of which in Fe –Ga alloys is considered as the main cause of the sharp increase in the magnetostriction constant. It turned out, however, that in compositions close to Fe_3Ga, possible superstructure reflections of the $L6_0$ phase completely overlap with reflections of the hexagonal phase X, first discovered in work [15]. The structure of this phase is not yet reliably known, although it can be assumed that it is similar to the structure of $B8_2$. The basis for this is the coincidence of crystal systems and good correspondence of the refined lattice parameters of the X phase ($a \approx 8.29$ Å, $c \approx 10.14$ Å) to the doubled parameters of $B8_2$ ($a \approx 4.03$ Å, $c \approx 5.03$ Å). The structure of $B8_2$ has been repeatedly found in different variants of Fe –Ge alloys, in particular, its detailed analysis was performed in work [40], where neutron diffraction was used to obtain data on the defect composition of $Fe_{3.34}Ge_2$.

Calculations of the total energy of the phase $L6_0$ showed [24] that it only slightly exceeds the energy of phases $D0_3$ and $L1_2$, and can form and persist during quenching and certain annealing regimes. However, the data presented in this work and in [39] lead to the conclusion that if $L6_0$ is present, its proportion in the sample is too small to affect such a bulk property as magnetostriction. Indeed, according to synchrotron data, the intensity of reflections that could be identified with reflections of the $L6_0$ phase in $Fe_{73}Ga_{27}$ is approximately 10^5 times weaker than the main peaks of the $D0_3$ phase. The same applies to the Fe_3Al alloy, whose magnetostriction constant is almost 10 times higher than the value for pure iron, but no traces of $L6_0$ can be found in it, including using electron diffraction.

In this regard, it is necessary to pay attention to other variants of modification of $A2$- or $D0_3$-structures, which could lead to an increase in the magnetostriction constant of the alloy. One possible variant is the formation of " $B2$-clusters" (two $B2$-cells connected by a common face), during which pairs of Ga –Ga atoms are formed along certain directions in the crystal. Model calculations performed in works [41, 42, 43] showed that the anisotropy introduced into the structure by Ga-Ga pairs may be sufficient to explain the increased magnetostriction of Fe-Ga alloys. This is also evidenced by the correlation of Zener relaxation parameters (reorientation of Ga –Ga pairs in the field of external stresses) with the level of magnetostriction, noted in work [44] (for more details, see reviews [16, 45]).

The possibility of forming a significant volume fraction of $B2$-clusters in Fe –Ga alloys was confirmed by the results of diffraction studies and diffuse X-ray scattering on $Fe_{82}Ga_{18}$ single crystals [46]. The same $B2$-clusters with Al –Al pairs were found when investigating several compositions of $Fe_{1-x}Al_x$ using Mössbauer spectroscopy [47]. It should also be noted that recently, studies have emerged that draw attention to the importance of accounting for the relationship between alloy atomic ordering and magnon dynamics to explain its physical properties (see, e.g., [48]). The combination of this information along with the data presented in this work suggests that the explanation for the

enhanced magnetostriction of Fe –Al, Fe –Ga, and Fe –Ge alloys should be sought without invoking the hypothesis of the presence of $L6_0$ phase inclusions in their crystalline matrix.

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CONFLICT OF INTERESTS

The authors of this work declare that they have no conflict of interest.

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