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Comparative study of structural phase transitions in bulk and powdered Fe-27Ga alloy by real-time neutron thermodiffractometry

Anatoly M. Balagurov,^a Igor S. Golovin,^b Ivan A. Bobrikov,^a* Valeria V. Palacheva,^b Sergej V. Sumnikov^a and Victor B. Zlokazov^c

^aFrank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, 141980 Dubna, Russian Federation, ^bNational University of Science and Technology 'MISIS', Leninsky avenue 4, 119049 Moscow, Russian Federation, and ^cLaboratory of Information Technologies, Joint Institute for Nuclear Research, 141980 Dubna, Russian Federation. *Correspondence e-mail: bobrikov@nf.jinr.ru

Phase transformations in an iron-gallium alloy have been analyzed by in situ real-time neutron diffraction in the temperature range from 293 to 1223 K. Two compositionally identical samples were studied: the first was in the as-cast bulk state, and the second was ground into a powdered state. In both samples, the same sequence of structural transitions was recorded on heating with a constant heating rate (D0₃ \rightarrow A2 \rightarrow L1₂ \rightarrow D0₁₉ \rightarrow A2), and the same structural state $(D0_3 + L1_2)$ was recorded after slow cooling to room temperature. Owing to strong texture in the bulk sample, only diffraction patterns of the powdered sample were treated with the Rietveld method to determine the volume fractions of the coexisting phases, the coefficients of thermal expansion, and the thermal and static atomic disorder parameters. The occupancy of Ga positions and the ordered iron magnetic moment were refined at selected temperatures. The level of microstrain in the crystallites in the initial as-quenched state is small, but it sharply increases in the course of phase transitions when the alloy is heated. The microstrains are high and strongly anisotropic after slow cooling. Generally, phase transformations occur similarly in the powdered and bulk samples, but with a noticeable difference in details. The fulfilled analysis of the bulk and powdered samples allowed the real possibilities of the quantitative neutron diffraction analyses of phase transitions in ferromagnetic ordered alloys to be assessed.

1. Introduction

Engineering properties of steels and iron-based functional alloys are largely dependent on their specific atomic structure, the volume content of different structural phases and their microstructural state. A direct method to determine these characteristics is Bragg diffraction of X-rays, synchrotron radiation or neutrons. From diffraction patterns, one can determine the space group symmetry and parameters of the unit cell for a particular phase state, the occupancies of crystallographic sites in the ordered alloys, and the phase fractions in the multiphase state. Along with structural data, information about microstresses in the crystallites, typical sizes of coherently scattering domains and the texture of the bulk samples can be obtained as well. In a neutron diffraction experiment, the data concerning the magnetic structure of the alloy can be determined.

For studies of phase transformations in a wide temperature range, an *in situ* real-time neutron diffraction experiment is most informative. In this case one can easily determine temperature ranges of phase transitions, coefficients of thermal expansion and changes in sample microstructure. This knowledge is necessary for understanding proposed physical mechanisms of transformations and for the formation of predetermined physical and mechanical properties. One issue in such experiments is a possible dependence of phase transformations on a particular microstructure of the material, and especially on the type of a sample: whether it is in a powdered state or in an as-cast bulk state. As-cast alloys usually are composed of relatively large crystallites and are highly textured. These features make a structural analysis by means of X-rays or synchrotron radiation with their narrow beams and small penetration depth practically impossible. Finegrained powders do not have such limitations; however, the extent to which phase transformations occur identically in ascast and powdered samples has not yet been analyzed in detail.

In the present study, this issue has been investigated using the example of the Fe-27.0Ga alloy during an in situ experiment in which as-cast and powder samples are heated from room temperature to ~1223 K with subsequent cooling. Neutron diffraction data were accumulated in real-time mode (real-time neutron thermodiffractometry). The composition Fe-27Ga is known for its exceptional magnetostriction compared to other binary iron-based alloys (Clark et al., 2003), and the analysis of the phase diagram and the properties of Fe-Ga alloys have been actively conducted by a variety of methods (see, for instance, Summers et al., 2007, and references therein). Neutron diffraction studies of Fe-Ga alloys are practically absent from the literature. We are aware only of the work of Zhao et al. (2008) and Mellors et al. (2012), where phase transformations of these alloys were studied as a function of the Fe/Ga ratio in the Ga concentration range from 15 to 22.5%. In addition, Zarestky et al. (2005, 2007) used inelastic neutron scattering to measure the dispersion curves as a function of the composition.

For a simultaneous analysis of the changes in the atomic structure and microstructure of the material in the course of phase transitions, it is necessary to repeatedly switch between high-intensity and high-resolution diffraction modes. This condition is fulfilled at the high-resolution Fourier diffractometer (HRFD), operating at the IBR-2 pulsed reactor at JINR (Dubna). In our previous diffraction studies of as-cast Fe-Ga alloys (Golovin, Balagurov, Bobrikov et al., 2016; Golovin, Balagurov, Palacheva et al., 2016), new data about phase transformations upon heating and cooling were obtained, and their comparison with magnetostriction properties was performed. However, the strong texture of the studied samples prevented a detailed analysis of their atomic and magnetic structures. In this paper, we present new results obtained with HRFD for both as-cast and powdered Fe-27Ga samples. It was found that the sequence of phase transitions and variations of the unit-cell parameters for both types of samples are similar, though some details are different. Only in the study of the powder sample did we succeed in drawing conclusions concerning the long-range magnetic structure and the degree of order of the gallium atoms. In addition, we defined the atomic displacement parameters, for which we managed to separate the temperature and static contributions.

Some new and interesting details were found through the analysis of microstrain in the crystallites for both types of sample. This comparison allows estimation of the horizons of the quantitative neutron diffraction analyses of phase transitions in ferromagnetic alloys with atomic ordered structures.

2. Samples

An Fe-27.0Ga alloy was produced by directional solidification using pure Fe and Ga by induction melting under the protection of a high-purity inert argon gas in an Indutherm MC-20V mini furnace. Using energy dispersive spectroscopy, the chemical composition of the buttons was confirmed with accuracy within $\pm 0.2\%$. Two as-cast parallelepipeds of $4 \times 8 \times$ 50 mm size were prepared, one of which was milled into powder. High-resolution diffraction patterns were measured for both samples: as-cast bulk (SB hereafter) and milled (SP hereafter) (see §3). Both patterns could be described in the cubic space group with $a \simeq 5.812$ Å, *i.e.* the samples were in the D0₃ phase. While the diffraction peaks from SB were only slightly broadened in comparison with the diffractometer resolution function, the peaks from SP were \sim 2.5 times wider, *i.e.* the milling led to a strong level of microstrain in the crystallites. Therefore, SP was annealed at 1173 K and then



Figure 1

Neutron diffraction patterns of Fe–27.0Ga in bulk (a) and powdered (b) states, measured at room temperature in high-resolution mode ($\Delta d/d \simeq 0.0015$). The vertical bars indicate peak positions of the D0₃ phase. The Miller indices of several first peaks are specified. Small negative dips around the peak profiles are associated with special features of the high-resolution correlation mode of data acquisition at HRFD.



Magnetization curve of the as-cast Fe–27.0Ga sample, showing the existence of ordered magnetism up to 923 K on heating.

rapidly cooled (quenched). After this procedure, the diffraction patterns of the two samples were almost identical (Fig. 1); some difference was observed only in the ratio of the intensities of the peaks with the same Miller indices, which is associated with the presence of texture in the bulk sample. For instance, the difference in the ratio of the intensities of the 400 and 220 peaks for SB and SP samples is twofold, which means that the texture is strong in the bulk sample. A magnetization curve of the as-cast sample (Fig. 2) clearly shows the ordered magnetism for the first two structural states, which were identified as D0₃ (up to \sim 723 K) and L1₂ (723–923 K) phases.

The phase transitions in the Fe-27.0Ga compound (as-cast SB sample) during slow ($\sim 2 \text{ K min}^{-1}$) heating from room temperature up to 1223 K were studied in our previous paper (Golovin, Balagurov, Palacheva *et al.*, 2016), where the following sequence was established: D0₃ \rightarrow L1₂ \rightarrow D0₁₉ \rightarrow A2. The main structural data of these phases (together with

the B2 and A3 phases, which also could appear if another heating rate is used) are as follows:

D0₃, *Fm3m* (N225), $a \simeq 5.834$ Å, Ga, Fe1, and Fe2 atoms are partially ordered in (4*a*), (4*b*) and (8*c*) positions, respectively, according to Wyckoff notation; the total number of atoms in the unit cell, *N*, is equal to 16.

L1₂, *Pm3m* (N221), $a \simeq 3.717$ Å, Ga and Fe atoms are partially ordered in (1*a*) and (3*c*) positions; N = 4.

 $D0_{19}$, $P6_3/mmc$ (N194), $a \simeq 5.280$ Å, $c \simeq 4.283$ Å, Ga and Fe atoms are partially ordered in (2c) and (6h) positions; N = 8.

B2, Pm3m (N221), $a \simeq 2.917$ Å, Ga and Fe atoms are partially ordered in (1*a*) and (1*b*) positions; N = 2.

A2, Im3m (N229), $a \simeq 2.917$ Å, disordered structure, atoms are randomly distributed between (2*a*) positions; N = 2.

A3, $P6_3/mmc$ (N194), $a \simeq 2.645$ Å, $c \simeq 4.283$ Å, disordered structure, atoms are randomly distributed between (2*d*) positions; N = 2.

Nearly complete ordering in the D0₃, L1₂ and D0₁₉ phases is possible in the Fe–27Ga composition if thermal disorder is neglected. In the B2 phase, ordering can be only partial as the full order is possible for the Fe–50Ga composition alone, whereas in the A2 and A3 phases, there is no order in Ga atoms positions in Fe-based α and γ lattices. The appearance of atomic order in the A2 phase of Fe–Ga alloys leads to the formation of the B2 or D0₃ phases, and order in the A3 phase leads to the D0₁₉ structure.

Order in Fe–Ga alloys gives rise to extra or 'superlattice' peaks in the neutron diffraction patterns, which have reduced intensity because intensity is proportional to $(b_{\rm Fe} - b_{\rm Ga})^2$, where $b_{\rm Fe} = 0.945$ and $b_{\rm Ga} = 0.729$ (in 10^{-12} cm units) are the coherent neutron scattering lengths for iron and gallium. Owing to the relatively small difference between $b_{\rm Fe}$ and $b_{\rm Ga}$, the intensity of the superlattice peaks is about ten times less than that of regular ones. There is some magnetic



Figure 3

A two-dimensional visualization of the diffraction pattern evolution of the Fe–27.0Ga samples in the as-cast (*a*) and powdered (*b*) states measured upon heating to 1223 K in the real-time mode. The temperature (and time) axis goes from bottom to top; the *d*-spacing axis goes from left to right. At room temperature both samples are in the D0₃ state, which is characterized by the 111, 200 and 220 diffraction lines. After that the phases L1₂ with the 100, 110, 111 and 200 lines, D0₁₉ with the 002 and 201 lines, and A2 with the 110 line appear. No other structural phases, *e.g.* A1, D0₂₂, are seen in these diagrams. The temperature of both samples was increased linearly with a rate of 2.25 K min⁻¹.

contribution, modulated by the magnetic form factor of iron, to all of the diffraction peaks, including the superlattice ones. Both magnetic and nuclear contributions to the superlattice peaks disappear if disordering of Ga and Fe takes place. The magnetic contrast between Fe and Ga is higher than the nuclear contrast as gallium atoms have no magnetic moment. Nevertheless, the intensity of superlattice peaks remains low because of the small value of the ordered magnetic moment of iron ($\mu_{\rm Fe} \leq 2 \mu_{\rm B}$).

3. Neutron diffraction experiment

Neutron diffraction patterns were measured at HRFD. A description of the diffractometer can be found in the article by Balagurov (2005) and more details are given by Balagurov et al. (2015). HRFD is a correlation time-of-flight (TOF) spectrometer with several detectors placed at fixed scattering angles from 30 to 152°. As mentioned above, one of the features of HRFD is its ability to switch easily between highintensity and high-resolution modes of operation, which is important for in situ studies of changes in both atomic structure and crystal microstructure. If a correlation analysis is used, the HRFD $\Delta d/d$ resolution is determined by the maximum rotational speed of a fast Fourier chopper. In routine operation ($V_{\text{max}} = 4000 \text{ rmin}^{-1}$), 'high-resolution' patterns ($\Delta d/d \simeq 0.001$ for d = 2 Å) are measured with backscattering detectors ($2\theta = 152^\circ$, *d*-spacing range is 0.6– 3.6 Å). In parallel, conventional (without correlation analysis) TOF patterns of 'medium' resolution ($\Delta d/d \simeq 0.01$ -0.02 up to $d_{hkl} = 4.5$ Å) are measured, their intensity being about ten times higher than that of the high-resolution patterns. Furthermore, if the Fourier chopper is moved out of the neutron beamline, only 'medium'-resolution patterns are measured with four times higher intensity.

High-resolution diffraction patterns were measured before heating and after slow cooling to room temperature. During the heating process, medium-resolution patterns were measured continuously with 1 min exposure time. The average intensity of the diffraction peaks was around several tens of thousands of counts and this is easily sufficient for the analysis of time dependencies. The heating of the samples up to 1223 K was carried out in a special high-temperature furnace (ILL standard). The temperature was increased linearly with a rate of 2.25 K min⁻¹, which was maintained with good accuracy. Cooling proceeded linearly with a rate of -2 K min⁻¹ down to $T \simeq 573$ K.

An analysis of diffraction data by the Rietveld method was carried out using the *MRIA* (Zlokazov & Chernyshev, 1992) and *FullProf* (Rodriguez-Carvajal, 1993) software packages. For the analysis of *in situ* data – to obtain the time dependencies of intensities, positions and widths of the diffraction peaks – the specialized program *SPEVA* (Zlokazov *et al.*, 2016) was used. One feature of HRFD connected with the correlation method of data acquisition in the high-resolution mode is the presence of small negative dips (visible in Fig. 1) at one or both sides of the diffraction peaks. For the description of such profiles in the *MRIA* package, there is the possibility of

introducing a two-sign model that can be specified numerically. The *FullProf* package has well developed algorithms for accounting of the magnetic structure and was used to determine the value of the ordered magnetic moment. The *SPEVA* program is designed to handle large volumes of diffraction data, measured when transition processes in crystals are studied in real-time mode. It has the ability to take into account the physical process flow, namely, the structural characteristics of the crystal at the current step of the evolution can be taken as initial values for the parameters of the next step. In the absence of sudden jumps in the structural evolution of a crystal, this allows one to fully automate the data processing and obtain structural characteristics of the phases (volume fraction, unit-cell parameters, atomic



Figure 4

Volume phase fractions in the SB (*a*) and SP (*b*) samples heated with a rate of 2.25 K min⁻¹ during the $D0_3 \rightarrow A2 \rightarrow L1_2 \rightarrow D0_{19} \rightarrow A2$ transitions. The temperature range with significant changes in the sample states is shown.

coordinates, occupancy factors *etc.*) as a function of time or of temperature.

Thus, the characteristics of the HRFD instrument, in particular, the possibility of changing its operating modes, the available sample environment and the existing software, fully meet the conditions required for *in situ* real-time studies of phase transformations in metallic alloys over a wide temperature range.

4. Structural phase transitions in bulk (SB) and powdered (SP) samples under heating

Upon continuous heating to 1223 K in both samples, bulk (SB) and powdered (SP), several phase transitions occur. Fig. 3 compares two-dimensional views of phase transitions $D0_3 \rightarrow$ $A2 \rightarrow L1_2 \rightarrow D0_{19} \rightarrow A2$ in SB and SP. In both samples, superlattice peaks corresponding to the ordered phases D0₃ and L_{1_2} are clearly visible. In particular, the disappearance of the 111 peak allowed only for the $D0_3$ phase and the 200 peak allowed for both D03 and B2 phases is simultaneous. This means that the B2 state is not really formed in the sample, and the disordered phase A2 appears immediately after $D0_3$, coexisting with L1₂ in a certain temperature range. Similarly, the $L1_2 \rightarrow D0_{19}$ transition occurs without any intermediate phases, i.e. the superlattice peaks 100 and 110 belonging to the $L1_2$ phase disappear simultaneously with the main peaks of this phase. The superlattice peak 101, which characterizes the ordered phase D0₁₉, is rather weak in the SB patterns and is barely visible in the SP patterns. This means that the degree of ordering of Fe and Ga in this phase is small and the crystal state can be actually described as a disordered phase A3 rather than a $D0_{19}$ phase for the used regime of constant heating. The ordering of $D0_{19}$ depends on the heating rate; in our preliminary test with the SB sample with a lower heating



Figure 5

Volume phase fractions in the SP sample heated with a rate of 2.25 K min⁻¹ as a function of time (relative). The subsequent transitions (1) A2 (blue diamonds) \rightarrow L1₂ (red triangles) and (2) L1₂ (purple oblique crosses) \rightarrow D0₁₉ (green upright crosses) are shown. The rate of the first transition is about three times longer than that of the second one. For the SB bulk sample these dependences are identical.

rate of 1.1 K min⁻¹ the 101 superlattice peak is more clearly seen, indicating the role of annealing time on A3 \rightarrow D0₁₉ ordering.

Normalized changes in volume fractions of different phases are shown in Fig. 4, and the temperature ranges of their existence and overlapping are listed in Table 1. The sequence of phase transitions is almost identical for both bulk and powdered samples. The main difference is that the entire volume of the SB sample is completely converted from the D0₃ through the A2 state into the L1₂ phase and then to the D0₁₉ phase. In contrast, in SP the D0₃ through the A2 to the L1₂ transition takes place only partially: some limited fraction of the volume (~12%) remains in the A2 disordered state. The kinetics of the A2 \rightarrow L1₂ and the L1₂ \rightarrow D0₁₉ transitions are the same in both samples, but the rate of these two transitions is very different. Namely, L1₂ \rightarrow D0₁₉ takes about 6 min (twofold decrease), while the A2 \rightarrow L1₂ transition takes about



Figure 6

Intensities of the several superlattice diffraction peaks in the $D0_3$ and $L1_2$ phases in the SB (*a*) and SP (*b*) samples measured upon heating. Experimental points are shown with statistical uncertainties.

15 min (Fig. 5) for the same heating rate $\Delta T/\Delta t = 2.25 \text{ K min}^{-1}$. Possible reasons for this are discussed in §7.

The analysis of changes with temperature in the intensity of superlattice peaks allows us to draw several conclusions about disordering of Fe and Ga atoms in the D0₃ phase and their ordering in the L1₂ phase during the D0₃ \rightarrow A2 \rightarrow L1₂ transition. For the unpolarized neutron beam, the nuclear and magnetic intensities are summed (the interference term vanishes), and thus

$$I_{\rm hkl} \simeq |F_{hkl,\rm nuc}|^2 + |F_{hkl,\rm mag}|^2, \qquad (1)$$

where $F_{hkl,nuc}$ and $F_{hkl,mag}$ are the nuclear and magnetic structure factors for a particular set of Miller indices. Both structure factors can be easily calculated for some degree of Fe, Ga and magnetic order. The calculation implies that the strongest magnetic contribution should be observed in the 111 peak of the D0₃ phase compared with the nuclear contribution. Fig. 6 shows that the intensity of the 111 peak reduces rapidly with an increase in temperature in both SB and SP samples, while the intensity of the 200 peak remains nearly constant. One can conclude that the long-range ferromagnetic (FM) order is gradually destroyed with increasing temperature and practically disappears at $T \simeq 693$ K. This temperature Table 1

Temperature intervals and existing phases found during heating of the SE	3
and SP samples up to \sim 1223 K.	

Bulk sample, SB		Powder sample, SP		
T (K)	Phases	<i>T</i> (K)	Phases	
<703	$D0_3$	<693	$D0_3$	
703–763	$A2 + L1_2$	693-888	$A2 + L1_2$	
763-898	$L1_2$			
898-923	$L1_2 + D0_{19}$	888-918	$A2 + L1_2 + D0_{19}$	
923–953	$D0_{19}$			
953-1003	$D0_{19} + A2$	918-976	$D0_{19} + A2$	
>1003	A2	>976	A2	

agrees well with the magnetization curve as shown in Fig. 2. Conversely, a high degree of gallium atom ordering is preserved up to this temperature, and then the degree of order decreases sharply to zero. The growth of the 100 peak intensity of the L1₂ phase indicates a gradual increase in the degree of Ga and magnetic ordering, reaching a maximum and sudden disappearance at $T \simeq 893$ K.

5. Structural analysis of various phase states

For a structural analysis of the phase states with the Rietveld method by using diffraction data measured for SP, the



Figure 7

Diffraction patterns (medium resolution) of the SP sample measured upon heating for several characteristic temperatures and processed by the Rietveld method. Experimental points and calculated and difference lines are shown. The vertical bars indicate peak positions of the indicated phases (from top to bottom).

temperature range was divided into several parts with various combinations of the coexisting phases. A prior calibration of the experimental conditions (flight pass, scattering angle *etc.*) was performed with the Al₂O₃ reference sample (NIST standard). Preliminary data processing, including data measured with high resolution, showed that simultaneous refinement of Ga occupancy (n_{Ga}), atomic displacement parameter (B) and Fe magnetic moment (M_{Fe}) leads to strong correlations between them, and accordingly some deviations from the real values are possible. The reasons are quite clear – the functional dependences of the Debye–Waller factor and the magnetic form factor on the momentum transfer are very similar. In addition, the value of n_{Ga} mainly depends on the intensities of weak superlattice peaks at large d_{hkl} , which have a substantial magnetic contribution. Therefore, in further data



Figure 8

Atomic volumes (the unit-cell volume per atom) for structural phases $D0_3$, $L1_2$, $D0_{19}$ and A2 for the SB (*a*) and SP (*b*) samples as a function of temperature. Near the curves the volumetric thermal expansion coefficients (in 10^{-5} K⁻¹ units) determined over the intervals with linear dependence are indicated.

processing, the changes of n_{Ga} , B and M_{Fe} relative to their values at room temperature ($n_{\text{Ga}} = 1$, $B = 0.5 \text{ Å}^2$, $M_{\text{Fe}} = 2 \mu_{\text{B}}$) were determined. The calculation showed that, in the D0₃ phase, the magnetic contribution to the intensity of the first two superlattice peaks is 4–5 times higher than the nuclear contribution, but at smaller d_{hkl} the difference is around a few percent. Accordingly, the data processing was performed over the interval $d_{hkl} < 2.5 \text{ Å}$, where magnetic contribution can be neglected.

Examples of the Rietveld refinement patterns for several particular phase states are shown in Fig. 7, and the data obtained are presented in Figs. 8–10 and in Table 2. It is evident that the processing quality is quite good. The variations of χ^2 values and R_{ω} , describing the correspondence between experimental and calculated data, are 2.2–4.3 and 3.8–5.2%, respectively, at the statistical factor $R_e = 2.7-6.0\%$.

With the Rietveld method, the unit-cell parameters of the coexisting phases are reliably and accurately determined. Fig. 8 shows how atomic volumes (the unit-cell volume per atom) are changed on heating. The highest atomic volume jump occurs at the $D0_3 \rightarrow A2 \rightarrow L1_2$ transition, whereas at the $L1_2 \rightarrow D0_{19}$ transition there is little discontinuity. This factor and the difference in volumetric thermal expansion coefficients ($\beta = 1/V dV/dT$) for different phases lead to deformation microstresses that affect the rate of kinetic processes during phase transitions. Appearing micro deformations manifested themselves in diffraction patterns as some additional (with respect to the resolution function) broadening of the peaks (see §6). A lattice shear deformation occurs at the $D0_3 \rightarrow A2 \rightarrow L1_2$ transition and is revealed as a peak of internal friction (for details see Golovin, Balagurov, Bobrikov et al., 2016).

A Debye–Waller factor was included in the refinement as $D_{hkl}(T) = \exp[-B(T)/4d_{hkl}^2]$, *i.e.* in the conventional isotropic approximation, for which the displacement parameter *B* is considered to be independent of a particular set of Miller



Figure 9

Displacement parameter *B* as a function of temperature for structural phases D0₃, L1₂, D0₁₉ and A2. The solid line shows the thermal contribution to the *B* value determined for the 473–673 K range and extrapolated up to 1173 K.

indices (*hkl*). The parameter *B* includes both the ubiquitous mean square thermal (dynamic) and possible local static displacements of atoms from their equilibrium positions, *i.e.* $B = 8\pi^2 [\langle u_T^2 \rangle + \langle u_S^2 \rangle]$, where u_T and u_S are the thermal and static displacements, respectively. For the monatomic cubic structure, the dependence of $\langle u_T^2 \rangle$ on temperature is well described by the Debye approximation, with B(T) increasing almost linearly with temperature, if $T > \Theta_D$, where Θ_D is Debye temperature.

The parameter *B* refined by the Rietveld method as a function of temperature is shown in Fig. 9. For pure iron, $\Theta_D = 464$ K, and we can assume that at T > 473 K the dependence B(T) should be close to linear. It is seen in Fig. 9 that the linear dependence is realized up to $T \simeq 673$ K. After the appearance of the L1₂ phase (in the mixed state), *B* increases dramatically, which is obviously connected with the emergence of large static displacements, comparable in magnitude to the thermal



Figure 10

Temperature dependence (up to 573 K) of the Fe-ordered magnetic moment (*a*) and Ga occupancy factor for the (4*a*) site in the $Fm\overline{3}m$ space group (*b*). These values were normalized at T = 293 K as $M_{\rm Fe} = 2 \mu_{\rm B}$ and $n_{\rm Ga} = 1$, respectively.

Table 2

Structural data of the SP sample at several temperatures obtained from Rietveld refinements.

The unit-cell parameters and atomic volumes are listed in the same order as the phases in the first column. The displacement parameter *B* was common for all atoms in the coexisting phases. The standard R_{ω} and $R_{\rm e}$ factors, and the value of χ^2 , are indicated in the second column.

T, phase(s)	χ^2, R_ω, R_e	Lattice parameters (Å)	$V(\text{\AA}^3)$	B (Å ²)
293 K, D0 ₃	4.3, 4.4, 2.6	<i>a</i> = 5.8113 (2)	12.27	0.92 (8)
501 K, D0 ₃	3.5, 5.2, 4.9	a = 5.8338 (2)	12.41	0.88 (9)
723 K, L1 ₂ + A2	2.2, 4.1, 5.7	a = 3.7064 (2) a = 2.9294 (2)	12.73 12.57	1.93 (4)
893 K, L1 ₂ + A2 + A3	3.3, 5.3, 7.2	a = 3.7215 (4) a = 2.9370 (3) a = 2.6335 (4) c = 4.261(2)	12.89 12.70 12.88	2.18 (4)
951 K, A2 + A3	2.2, 4.5, 7.2	a = 2.9391 (4) a = 2.6343 (2) c = 4.269 (1)	12.70 12.90	2.00 (5)
1133 K, A2	3.2, 3.8, 2.7	a = 2.9593 (2)	12.96	1.8 (1)

ones. The gradual relaxation of static displacements begins above 973 K, and at temperatures higher than 1123 K the dependence B(T) becomes linear again.

We succeeded in refining the dependence on temperature of the iron ferromagnetic moment and the degree of gallium and iron ordering with acceptable accuracy only in the range up to \sim 573 K (Fig. 10). In general, both these parameters correspond to the observed dependence of the magnetization on temperature (Fig. 2) and changes of intensity of the 111 and 200 peaks (Fig. 6). At higher temperatures, the correlations between these parameters are too strong, resulting in large fluctuations and nonphysical values.

6. Sample microstructure before, during and after heating

Diffraction data processing by the Rietveld method allows some microstructural characteristics of the material to be obtained. In particular, the lattice microstrain $\varepsilon \simeq \Delta d/d$ (static variance of metric parameters) and the finite coherently scattering domain size L_{coh} (CSDS) can be calculated from the dependence of the width of the diffraction peaks on the scattering vector. The estimated microstrains can be transformed, if necessary, into microstresses in crystallites by using the generalized Hooke's law. However, by the Rietveld method only averaged data can be obtained, which does not allow revelation of possible anisotropy of internal stresses. An independent estimation of the widths of the individual diffraction lines and use of the Williamson-Hall method for their analysis makes it possible to identify the effects of anisotropy and, furthermore, to differentiate between strain and size effects on the peak broadening.

Earlier, it was shown for HRFD (Balagurov *et al.*, 2015) that for a sample with negligibly small ε and very large L_{coh} the dependence of the peak width on the *d* spacing is reproduced with good accuracy by the expression $(\Delta d_R)^2 = C_1 + C_2 d^2$, where C_1 and C_2 are constants relating to the experimental setup. In the presence of microstrain and for finite $L_{\rm coh}$, an additional broadening of diffraction peaks occurs. According to the Williamson–Hall analysis (see *e.g.* Mittemeijer & Welzel, 2008), this additional broadening can be expressed as $(\Delta d_\delta)^2 \simeq (2\varepsilon d)^2 + (d^2/L_{\rm coh})^2$, if a Gaussian approach for the distribution functions is used. Combining $(\Delta d_R)^2$ and $(\Delta d_\delta)^2$, the following expression can be obtained:

$$(\Delta d)^2 = C_1 + (C_2 + C_3)d^2 + C_4 d^4, \qquad (2)$$

where $C_3 \simeq (2\varepsilon)^2$ and $C_4 \simeq (1/L_{\rm coh})^2$. The dependence of $(\Delta d)^2$ on d^2 is linear if there is no size effect (large crystallites) and parabolic otherwise. By employing this dependence over a sufficiently wide range of the measured d_{hkl} , it is possible to determine both ε and $L_{\rm coh}$. The HRFD resolution allows one to reliably determine deformations in crystallites at a level of $\varepsilon \simeq 8 \times 10^{-4}$ or higher and the mean CSDS at a level of $L_{\rm coh} \simeq 3500$ Å or lower. In addition, anisotropic peak broadening effects can be determined with confidence. For determining all these effects the weak dependence of the HRFD resolution on d_{hkl} is an important factor, making possible the use of a large number of diffraction peaks for analysis of the $(\Delta d)^2$ function.

To find the peak width (FWHM), the description of the peak profile by the experimentally measured model function with the *UPEAK* (Zlokazov, 1975) program was used. In §2, we already mentioned that the bulk alloy milling resulted in a dramatic increase in the widths of the peaks, *i.e.* to the emergence of strong microstresses in the crystallites. However, after the SP sample was annealed at 1173 K and quenched, the level of microstress returned to its initial (before milling) value. The corresponding dependences of $(\Delta d)^2$ on d^2 are shown in Fig. 11. As clearly seen in the figure, the corresponding dependences are linear, and hence there is no size effect. In the initial state and after annealing, the microstrain level is very low ($\varepsilon \simeq 0.001$) and the slope of the lines is only slightly larger than for the resolution function. Milling has led



Figure 11

The $(\Delta d)^2$ over d^2 dependences (Williamson–Hall plots) for SB (red squares), SP just after milling (green diamonds), and SP after annealing at 1173 K and quenching (blue crosses). The $(\Delta d)^2$ values are multiplied by 10³. The statistical errors of experimental points are about the same size as the symbols. The bottom line corresponds to the diffractometer resolution function measured with a standard sample.

not only to an increase of microstrain by 2.6 times but also to the noticeable effects of anisotropic broadening, manifesting itself as a deviation of experimental points from the straight line.

After heating and relatively slow cooling (the furnace was turned off and slowly cooled), both SB and SP samples were found to be in a two-phase $D0_3 + L1_2$ state with approximately 50% sample volume for each, and a dramatic broadening of the diffraction peaks (Fig. 12) took place. Moreover, the broadening of the peaks in both samples appeared strongly anisotropic, *i.e.* the widths were heavily dependent on a particular set of Miller indices. Fig. 13(*a*) vividly shows that the widths vary irregularly. Another feature of these dependencies is that for both SB and SP samples the widths in the $L1_2$ phase closely agree, whereas for the $D0_3$ phase their values markedly differ.

In the frame of the Williamson–Hall method the effects of anisotropic broadening of the peaks in crystals with a cubic lattice can be accounted for by introducing into equation (2) a dislocation-related contrast factor Γ (Ungár & Borbély, 1996):

$$(\Delta d)^2 = C_1 + C_2 d^2 + C_3 (A - B\Gamma) d^2 + C_4 d^4, \qquad (3)$$

where $\Gamma = (h^2k^2 + h^2l^2 + k^2l^2)/(h^2 + k^2 + l^2)^2$, and A and B are constants dependent on the density and the relative content of the edge and screw dislocations. If the size effect is absent and the resolution function is taken into account, the contribution to the width of the peaks associated with microstress is

$$\left(\Delta d_{\varepsilon}\right)^{2} = (K_{1} - K_{2}\Gamma)d^{2}, \qquad (4)$$

where new constants K_1 and K_2 are introduced instead of the products C_3A and C_3B , through which the anisotropic deformation ε_{hkl} can be expressed as $\varepsilon_{hkl} = 0.5(K_1 - K_2\Gamma)^{1/2}$. Using the peaks with different Γ factors, which range from 0 to 1/3, and selecting coefficients K_1 and K_2 , it is possible to minimize the deviation of $(\Delta d)^2$ values from the linear dependence on d^2 . If the peak broadening is really connected





Neutron diffraction pattern (high resolution) of SP measured after its slow cooling from 1223 K to room temperature. The sample is in the mixed $L1_2 + D0_3$ state, the peak positions of which are indicated (top and bottom lines, respectively). The Miller indices of several lines are shown. The widths of the peaks are different owing to anisotropic broadening. The diffraction pattern for the SB bulk sample measured after its slow cooling from 1173 K to room temperature is similar.

with the deformation anisotropy, the $[(\Delta d_{\varepsilon})^2 + K_2 \Gamma d^2]$ value as a function of d^2 should be approximated by a straight line passing through the origin of coordinates. These functions for both coexisting phases, D0₃ and L1₂, are shown in Fig. 13(*b*), and one can see that, indeed, they are linear with good accuracy. Using these estimated K_1 and K_2 constants the anisotropic deformations ε_{hkl} can be calculated as well as the average deformation $\langle \varepsilon \rangle = \sum \varepsilon_{hkl} j_{hkl} / \sum j_{hkl}$, where j_{hkl} is the reflection multiplicity for the given (*hkl*) set. The calculation of these values leads to $\langle \varepsilon \rangle = 0.0027$ and 0.0026 for the L1₂ and D0₃ phases, respectively, in the SB bulk sample and to $\langle \varepsilon \rangle =$ 0.0027 and 0.0016 for the L1₂ and D0₃ phases, respectively, in the SP powdered sample.

During heating, the diffraction patterns were measured only with medium resolution, and the peak broadening effect is not expressed clearly. However, as seen in Fig. 14, the peak widths vary in quite a natural way. Namely, at low and very high temperatures the widths are only slightly higher than the values corresponding to the HRFD resolution function,





(a) The $(\Delta d)^2$ over d^2 dependences for the SB and SP samples after the heating-cooling cycle. The points for SB are connected by green solid lines (for clarity) for the two phases D0₃ (green squares) and L1₂ (green diamonds). The points for SP are connected by red dashed lines for the two phases D0₃ and L1₂ (red oblique and straight crosses). (b) The same experimental points after correction for the dislocation anisotropy factor. The points of the SP, L1₂ phase are not shown because they coincide closely with the points of the SB, L1₂ phase. The $(\Delta d)^2$ values are multiplied by 10³. The statistical errors of experimental points are about the same size as the symbols. The Miller indices of several points are shown.

whereas at the phase transitions and in the temperature ranges with coexisting phases the diffraction peaks broaden considerably. Significant microstresses arise in the $L1_2$ phase, which can be estimated to be about five times higher than in the initial (D0₃) state.

7. Discussion

The comprehensive analysis of neutron diffraction data obtained in the course of heating from 293 to 1173 K of the ascast bulk (SB) and powdered (SP) samples of the Fe–27.0Ga alloy reveals an identical picture of phase transformations. Namely, the sequence of structural transitions was found to be $D0_3 \rightarrow A2 \rightarrow L1_2 \rightarrow D0_{19} \rightarrow A2$ in both samples. The coincidence of the temperatures for the disappearance of the





Temperature dependence (upon heating) of some particular diffraction peak widths of the $D0_3$, $L1_2$, $D0_{19}$ and A2 phases for SB (*a*) and SP (*b*). The dashed line indicates the peak width corresponding to the diffractometer resolution function.

Anatoly M. Balagurov et al. • Structural phase transitions in Fe–27Ga alloy **207**

 $D0_3$ ordered phase (its transition into the disordered A2 state) and the appearance of the $L1_2$ ordered phase could mean that a direct transition between the two ordered states, $D0_3 \rightarrow L1_2$, is forbidden (or severely hampered) and, in reality, the $D0_3 \rightarrow$ $A2 \rightarrow L1_2$ transition is realized.

This question has been open for discussion since the publication of the articles by Khachaturvan & Viehland (2007) and Boisse *et al.* (2011). They proposed that the $D0_3$ to $L1_2$ transition cannot occur only by an atomic exchange between the sites, and a diffusionless displacive body-centered cubic $(b.c.c.) \rightarrow$ face-centered cubic (f.c.c.) reconstruction takes place. The atomic order inherited from the D0₃ phase corresponds to a transformed tetragonal (D0₂₂ structure) lattice, and the corresponding Bain strain transforms the b.c.c. lattice further into a face-centered tetragonal (f.c.t.) lattice, bringing Fe and Ga atoms to their new positions in an f.c.c.-based lattice without an atomic interchange between these sites. The Bain orientation relations between the parent b.c.c.-based D0₃ and f.c.t. $D0_{22}$ phases are $[001]_{D03} || [110]_{D022}$, $(100)_{D03} ||$ $(110)_{D022}$ and $(010)_{D03} || (1\overline{10})_{D022}$. The orientation relations produce a tetragonal elastic extension (ε_c) along $a_{D03}/2 \rightarrow$ $a_{\text{D022}}/2(2)^{1/2}$ and an elastic contraction (ε_a) along the two perpendicular axes. Finally, the orientation relations for the diffusionless $D0_3 \rightarrow L1_2$ transition can be written as

$$a_{\text{L}1_2} = \frac{a_{\text{D}0_3}}{2}(1+\varepsilon_c)$$
 and $a_{\text{L}1_2} = \frac{a_{\text{D}0_3}}{2^{1/2}}(1+\varepsilon_a),$ (5)

with the corresponding values of the elastic deformation being estimated by Zhao *et al.* (2008) as $\varepsilon_c \simeq 0.265$ and $\varepsilon_a \simeq -0.105$. From our measurements of the lattice constants at T = 713 K $(a_{\rm D03} \simeq 2a_{\rm A2} = 5.8574$ Å and $a_{\rm L12} = 3.7017$ Å) it follows that $\varepsilon_c \simeq 0.266$ and $\varepsilon_a \simeq -0.106$ are in good agreement with Khachaturyan's estimation. Most recently, a similar scheme of transition was confirmed using a transmission electron microcopy study of an Fe-27Ga alloy (Lina & Lin, 2015). The D0₃ nanoclusters were reported to undergo a phase transformation to an intermediate tetragonal phase (L10-like martensite) via Bain distortion, and finally the L12 structure is formed. According to our presented data, the scheme of the $D0_3 \rightarrow L1_2$ transition should be modified, at least, by introducing a preliminary transition $D0_3 \rightarrow A2$ that continues with the A2 \rightarrow L1₂ transition. The fact that the D0₂₂ tetragonal structure does not manifest itself in the diffraction patterns can be explained by its highly dispersed form.

The $D0_3 \rightarrow A2 \rightarrow L1_2$ transition occurs in the entire volume of the SB bulk sample: no traces of the A2 phase were observed in the 773–943 K temperature range. In contrast, as seen in Fig. 4, a significant fraction of SP volume (~12%) remains in a disordered A2 state, and this fraction does not change substantially in the range of 763–913 K, *e.g.* up to the transition $D0_{19} \rightarrow A2$. A possible reason for this discrepancy is a somewhat higher level of defects in SP (preserved in spite of annealing), as follows from the widths of the diffraction peaks shown in Fig. 14. In the SP sample, the temperatures of all the transitions – the appearance of a new phase and the disappearance of a previous one – are slightly (5–15 K) lower than in the SB bulk sample. This is another notable difference in the as-cast and powder samples and might be also related to different dislocation densities in the studied samples.

The huge difference (about 2.5 times) in the rate of the $A2 \rightarrow L1_2$ and the $L1_2 \rightarrow D0_{19}$ transitions (Fig. 5) is certainly connected with a relatively large difference between the atomic volumes of the A2 and L1₂ phases, and, in contrast, their numerical equality for the $L1_2$ and $D0_{19}$ phases. A strong lattice deformation appears at the A2 \rightarrow L1₂ transition and slows it down. The same situation arises for the $D0_{19} \rightarrow A2$ transition, whose rate is comparable to that for the A2 \rightarrow L1₂ transition. For both of them, the atomic volume jump is about 1.5%, whereas for the transition between the close-packed phases $L1_2 \rightarrow D0_{19}$ it is almost 20 times lower. The temporal resolution (1 min) for the diffraction data acquisition seems to be optimal for the chosen heating rate of 2.25 K min⁻¹. Even for a relatively fast $L1_2 \rightarrow D0_{19}$ transition, it allows us to obtain about ten experimental points for the fractions of the coexisting phases, which is sufficient to analyze the kinetics of the transition.

Powder neutron diffraction made it possible to perform a detailed structural analysis of the emerging phase states. Unfortunately, owing to the strong correlations between n_{Ga} and M_{Fe} the calculations failed to follow these values over the entire temperature range, but the correct trends of their changes were determined up to 573 K, which, together with the analysis of the intensities of individual peaks (Fig. 6), led us to important conclusions about a gradual decrease of the long-range magnetic order and, conversely, a relatively abrupt change in the degree of gallium atom ordering in the D0₃ \rightarrow A2 transition. From this fact it follows that a complete correlation between these two parameters is absent. The reasons for this feature are not yet clear: naturally synchronous temperature dependence would be expected.

In addition, the structural analysis allowed us to find very accurate data for the phase fractions and the unit-cell parameters for the whole temperature range. Fig. 8 shows that, over fairly large temperature intervals, the unit-cell parameters vary linearly, *i.e.* the coefficients of thermal expansion are constant, and at the same time in SP the unit-cell parameters are slightly less than those in SB for all structural phases. During transitions between the phases the behavior of thermal expansion coefficients is strongly nonlinear, which requires additional analysis in future. The displacement parameter B is another important value, whose changes with temperature can be determined only by a structural analysis of a powder sample. Its behavior during heating of the sample (Fig. 9) reveals the appearance of strong static atomic displacements in the $D0_3 \rightarrow A2 \rightarrow L1_2 \rightarrow D0_{19} \rightarrow A2$ transitions. For the range of 723-973 K, both thermal and static displacements are of about the same magnitude ($u_{\rm T} \simeq$ $u_{\rm S} \simeq 0.1$ Å).

High-resolution neutron diffraction provides detailed information about the microstructure of the crystalline state, in particular about the level of microstrain in crystal grains and the finite coherently scattering domain size. Currently there are several approaches to extracting microstructural data by analysis of diffraction patterns, including, for instance, whole powder pattern modeling (Scardi *et al.*, 2010). However, for the initial evaluation of the microstructure parameters, the Williamson–Hall analysis [relation (2) for TOF data] continues to be of high importance and the present study demonstrates that TOF neutron diffraction can provide very accurate data for the microstructure of the studied materials. In the present study, the important factors were the possibility of measurements in a high-resolution mode and a weak dependence of the resolution function on d_{hkl} .

The Williamson–Hall plots show that the $(\Delta d)^2$ lines are linear (no size effect) for the initial state (Fig. 11). This means that $L_{\rm coh}$ is not less than 3500 Å, which is the limit of sensitivity for HRFD. In contrast, the effects associated with microstress in the crystallites are expressed very clearly. In particular, it was found that residual microstrains are quite low, not higher than $\varepsilon \simeq 0.001$ in the quenched Fe–27.0Ga samples (regardless of whether it is an as-cast piece or powdered). Under heating, the microstrain gradually grows and the broadening of the diffraction peaks is observed even in the medium-resolution mode. At the $D0_3 \rightarrow A2 \rightarrow L1_2$ transition the microstrain increases to $\varepsilon \simeq 0.006$ and at subsequent transitions $L1_2 \rightarrow D0_{19} \rightarrow A2$ it gradually decreases to the initial state (Fig. 14). However, after slow cooling, a mixture of $D0_3$ and $L1_2$ phases appear in both SB and SP, and highly anisotropic microstrains increase again (Fig. 13). This deformation anisotropy, which manifests itself in the strong dependence of the peak widths on a particular set of Miller indices, has been taken into account in the frame of the modified Williamson-Hall method by introducing the dislocation anisotropy factor. It has been shown that this approach does allow the linear dependences of $(\Delta d)^2$ on d^2 to be obtained, indicating an adequate modeling of the physical reasons for anisotropic broadening.

After slow cooling, both samples achieve the same mixed $D0_3 + L1_2$ state, and in that state the widths of the diffraction peaks of the $L1_2$ phase (and, accordingly, microstrains) are practically the same in both samples. However, for reasons still unclear, in the $D0_3$ phase the microstresses in the SP sample are 1.6 times lower than they are in the SB bulk sample. Perhaps, this difference is connected with the presence of the texture in the as-cast sample.

8. Conclusions

In this work, we have presented an analysis of phase transformations in the Fe–27Ga alloy by *in situ* real-time mediumresolution neutron TOF diffraction, which was combined with a high-resolution diffraction study of the initial and final states of the alloy before and after heat treatment. Two compositionally identical samples were studied: the first was in a strongly textured as-cast bulk state, and the second was ground into a powdered state. The data from the bulk and powder samples are in good agreement with the general picture of phase transformations during heating and cooling in the 293–1223 K range, though some details are noticeably different. The powder neutron diffraction patterns have been refined using the Rietveld method, thus allowing the determination of the behavior of the phase fractions, lattice parameters (coefficients of thermal expansion) and mean square atomic displacements in the entire temperature range. The occupancy factors in the ordered phases and the Fe magnetic moment due to the strong correlations between them were identified only at selected temperatures up to T = 573 K. The most interesting new structural facts directly related to the physical processes which take place in this alloy are as follows.

The percentage of the sample volume involved in the $D0_3$ (A2) $\rightarrow L1_2$ phase transition is different in the cast (100%) and powder (~88%) samples, which seems to be due to different levels of microstrain in the crystallites. In both samples, the transition between the ordered states (from $D0_3$ to $L1_2$) goes through the disordered phase A2, *i.e.* the order– disorder–order type transition is realized.

The dynamics of the temperature-induced transitions $D0_3$ (A2) $\rightarrow L1_2$ and $L1_2 \rightarrow D0_{19}$ are significantly (by a factor of 2.5) different, which is certainly connected with the special features of the b.c.c. \rightarrow f.c.c. and f.c.c. \rightarrow hexagonal close packed transformations. The same features are the reason for very different atomic volume jumps in the course of these transformations.

The temperature dependences of the changes in the degree of atomic and magnetic order in the $D0_3$ phase are significantly different, and therefore the proposed correlation between them is not realized.

The Debye-like temperature behavior of the atomic displacement parameter is greatly disturbed in the range of structural phase transitions by the appearance of strong static displacements, whose amplitude is comparable to the thermal displacements.

The anisotropic broadening of diffraction peaks that appeared after slow heating and cooling could be almost perfectly described by introducing a dislocation factor.

All the results presented in this paper were obtained by using a combination of very high resolution neutron diffraction and real-time neutron thermodiffractometry with good ($t_s = 1 \text{ min}$) temporal resolution.

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