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Neutron Diffraction Study of Fe₂MnGa Heusler Alloys

Yu. V. Kudryavtsev, A. O. Perekos, I. N. Glavatskyy*, J. Dubowik**,
and Yu. B. Skirta***

*G. V. Kurdyumov Institute for Metal Physics, N. A. S. of Ukraine,
36 Academician Vernadsky Blvd.,
UA-03680 Kyiv, Ukraine*

**Helmholtz Centre Berlin for Materials and Energy,
1 Hahn Meitner Platz,
14109 Berlin, Germany*

***Institute of Molecular Physics, PAS,
17 Mariana Smoluchowskiego,
60-179 Poznań, Poland*

****Institute of Magnetism, N.A.S. of Ukraine,
36-b Academician Vernadsky Blvd.,
03142 Kyiv, Ukraine*

Effect of temperature and magnetic field on the structure and magnetic properties of Fe_{50.1}Mn_{22.7}Ga_{27.2} and Fe_{51.6}Mn_{17.8}Ga_{30.6} alloys is investigated in a temperature range 100 K < T < 580 K by using elastic neutron diffraction (ND) and magnetometry. The degree of atomic order as well as magnetic moments localized at the Mn and Fe sites in the investigated Fe₂MnGa alloys are experimentally evaluated using the ND data. Some disagreement between the experimental and calculated values of magnetic moments localized at the Mn and Fe sites can be explained by noticeable atomic disorder in the prepared Fe₂MnGa alloys. If antiferromagnetic order really exists in L1₂-phase containing Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy, this order has not a collinear character.

Key words: neutron diffraction, magnetic properties, atomic order, structural transformations.

Методами пружної дифракції нейтронів (ДН) та магнетометрії в температурній області 100 K < T < 580 K було досліджено вплив температури й

Correspondence author: Yuri Vladimirovich Kudryavtsev
E-mail: kudr@imp.kiev.ua

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магнетного поля на структуру та магнетні властивості стопів $\text{Fe}_{50,1}\text{Mn}_{22,7}\text{Ga}_{27,2}$ і $\text{Fe}_{51,6}\text{Mn}_{17,8}\text{Ga}_{30,6}$. З використанням даних ДН в роботі було експериментально визначено як ступінь атомового порядку в стопах Fe_2MnGa , так і величини магнетних моментів, що локалізовані на вузлах Mn та Fe. Деяка невідповідність між експериментальними та розрахованими теоретично величинами магнетних моментів, що локалізовані на вузлах Mn та Fe, пояснюється значним ступенем атомового безладу у виготовлених стопах Fe_2MnGa . Якщо антиферромагнетний порядок дійсно формується в $L1_2$ -фазі стопу $\text{Fe}_{50,1}\text{Mn}_{22,7}\text{Ga}_{27,2}$, цей порядок має неколінеарну структуру.

Ключові слова: дифракція нейтронів, магнетні властивості, атомовий порядок, структурні перетворення.

Методами упругой дифракции нейтронов (ДН) и магнитометрии в температурной области $100 \text{ K} < T < 580 \text{ K}$ было проведено исследование влияния температуры и магнитного поля на структуру и магнитные свойства сплавов $\text{Fe}_{50,1}\text{Mn}_{22,7}\text{Ga}_{27,2}$ и $\text{Fe}_{51,6}\text{Mn}_{17,8}\text{Ga}_{30,6}$. С использованием данных ДН были экспериментально определены как степень атомного порядка в сплавах Fe_2MnGa , так и величины магнитных моментов, локализованных на узлах Mn и Fe. Некоторое несоответствие между экспериментальными и теоретически рассчитанными значениями магнитных моментов, локализованных на узлах Mn и Fe, объясняется существенной степенью атомного беспорядка в приготовленных сплавах Fe_2MnGa . Если антиферромагнитный порядок действительно формируется в $L1_2$ -фазе сплава $\text{Fe}_{50,1}\text{Mn}_{22,7}\text{Ga}_{27,2}$, то этот порядок имеет неколлинеарную структуру.

Ключевые слова: дифракция нейтронов, магнитные свойства, атомный порядок, структурные превращения.

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1. INTRODUCTION

Significant interest to the stoichiometric and off-stoichiometric Fe_2MnGa alloys is based on the discovery in these alloys of martensitic transformation [1–5], metamagnetic transformation from antiferromagnetic (AFM) to ferromagnetic (FM) phase [6–9] and giant exchange bias [6–8].

Similarly to the temperature dependence of magnetization under metamagnetic transformation (*i.e.* growth of magnetization with temperature at low magnetic fields), it may be also observed for spin-glass systems. The authors made the conclusion on the formation of AFM magnetic order in the disordered (melt spun ribbons) $\gamma\text{-Fe}_2\text{MnGa}$ alloys on the basis of analysis of $M(H)$ dependences taken at different temperatures after various heat treatments [6, 7]. However, no direct evidence of kind of AFM order (if any) in such alloys has yet been presented.

According to Zhu *et al.*, body-centred cubic (b.c.c.) phase in

Fe_{50.0}Mn_{22.5}Ga_{27.5} alloy is not ferromagnetically ordered at room temperature (*RT*). The FM order in alloy is induced by martensitic transformation from b.c.c. phase to tetragonal one, which takes place at $T = 150$ K (on cooling) [1]. Contrary to this statement Shih *et al.* have shown that melt spun two phases b.c.c. + f.c.c. (face-centred cubic) ribbon of Fe₅₀Mn₂₄Ga₂₆ alloy which does not exhibit martensitic transformation is ferromagnet due to b.c.c. phase with the Curie temperature of $T_c = 190$ K [9]. Thus, ‘Is ferromagnetism of Fe₅₀Mn₂₄Ga₂₆ alloy an entire property of b.c.c. phase or result of martensitic transformation?’ still is an open question.

This work aims to study the effect of temperature and magnetic field on the structure and magnetic properties of nearly stoichiometric Fe₂MnGa alloys by employing mainly elastic neutron diffraction and magnetometry.

2. EXPERIMENTAL DETAILS

Two bulk polycrystalline Heusler Fe₂MnGa alloys have been prepared by melting of the Fe, Mn and Ga pieces of 99.99% purity together in an arc-furnace with water-cooled Cu hearth under 1.3 bar Ar atmosphere. The Ar gas in the furnace before melting was additionally purified by multiple remelting of Ti₅₀Zr₅₀ alloy getter. To promote the volume homogeneity, the ingot was remelted 5 times. Miserable weight losses after ingot meltings were observed. X-ray dispersion spectroscopy analysis revealed alloy compositions as Fe_{50.1}Mn_{22.7}Ga_{27.2} and Fe_{51.6}Mn_{17.8}Ga_{30.6}. Additionally as cast alloys were annealed at $T = 1220$ K during 96 h.

The structural characterization of the samples was carried out at 100, 300 and 580 K for magnetic fields of 0 and 50 kOe by employing elastic neutron diffraction (ND) at Helmholtz Centre Berlin for Materials and Energy using Flat-Cone Diffractometer (E2) with PG (002) monochromator to produce monochromatic unpolarised neutron beam of $\lambda = 0.239$ nm wavelength. Bulk Fe₂MnGa alloy samples were mounted into aluminium container. Therefore, experimental ND spectra may contain diffraction peaks related to Al. Besides ND, the crystalline structure of alloys was also analysed using X-ray diffraction (XRD) in a Θ - 2Θ -geometry with CuK $_{\alpha}$ -radiation ($\lambda = 0.15406$ nm).

The magnetic properties of bulk Fe₂MnGa alloy samples were investigated for a temperature $2 < T < 400$ K and magnetic field $0 < H < 70$ kOe, by using PPMS-14T Quantum Design magnetometer, and for $80 < T < 825$ K, temperature range by measuring DC-magnetic susceptibility.

3. RESULTS AND DISCUSSION

According to the results of XRD study, Heusler Fe_{51.6}Mn_{17.8}Ga_{30.6} alloy has the disordered A2-type of structure (only principle $[(h + k + l)/2 =$

$= 2n]$ diffraction peaks 220, 400, and 422 are seen) with a lattice parameter of $a = 0.5844$ nm. At the same time, XRD spectrum for $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy reveals that it is a mixture of the ordered $L1_2$ -type phase with a lattice parameter $a = 0.3703$ nm and the disordered $A2$ -type phase with a lattice parameter $a = 0.5844$ nm (see Fig. 1). Taking into account the intensities of the most intense fundamental reflections for $L1_2$ - and $A2$ -phases, their relative volume content in $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy may be estimated as 71 vs. 29%, respectively.

At the same time, ND results for $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloy reveal that, at room temperature (*RT*), it has an ordered crystalline structure of $L2_1$ -type with a lattice parameter of $a = 0.5882$ nm—additionally to fundamental diffraction (220) peak at $2\Theta = 70.15^\circ$, (111) and (200) superstructure reflections are clearly seen. The diffraction peaks at $2\Theta = 61.2^\circ$ and 71.9° may be definitely ascribed to (111) and (200) reflections of $A1$, respectively (see Fig. 2).

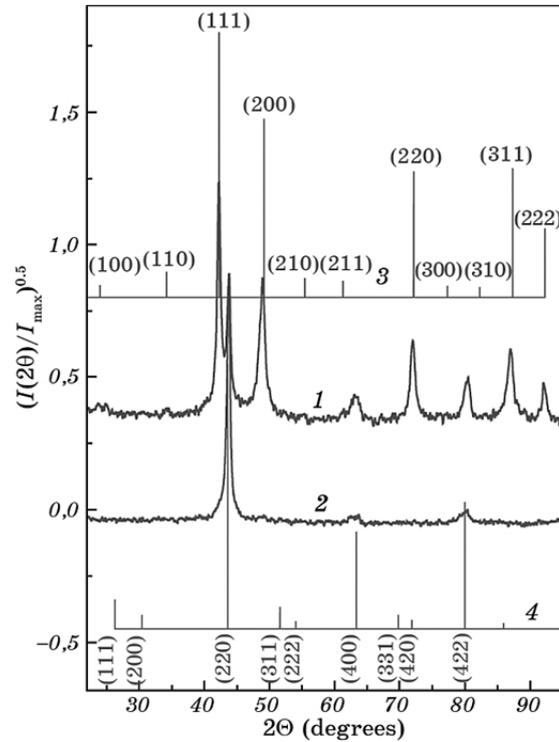


Fig. 1. Experimental XRD spectra for $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ (1) and $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ (2) alloys together with calculated stroke-diagrams for perfectly ordered $L1_2$ (3) and $L2_1$ (4) structures with lattice parameters of $a = 0.3703$ nm and $a = 0.5844$ nm, respectively. XRD spectra are shifted relatively each other for convenience of observation.

Similarly to XRD results, ND spectrum for Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy at RT shows that the alloy is a mixture of the ordered $L1_2$ -type phase with the lattice parameter of $a = 0.3720$ nm [see (100), (110), (210) superlattice diffraction lines] and the ordered $L2_1$ -type phase with the lattice parameter of $a = 0.5875$ nm [(111) and (311) superlattice diffraction lines satisfy the condition that h, k, l are all odd] (see Fig. 3). The co-existence of $L2_1$ - and $L1_2$ -phases in the off-stoichiometric Fe₂MnGa alloys was also found in a quite large range of Fe₂MnGa alloy compositions [1, 9].

$L2_1$ -structure of full Heusler X_2YZ alloys can be comprised as four interpenetrating f.c.c. sublattices $A, B, C,$ and D [10]. In this view, A and C sites of full Heusler Fe₂MnGa alloy entirely occupied by X (Fe) atoms, but B and D sites occupied by Y (Mn) and Z (Ga) atoms, respectively. For such a structure, permitted Bragg reflections are those for which Miller indices are unmixed, *i.e.* all odd or all even. They have the following structure amplitudes F :

$$F(111) + 4 \left| \sqrt{(f_A - f_C)^2 + (f_B - f_D)^2} \right| = 4 \left| f_{\text{Mn}} - f_{\text{Ga}} \right|, \quad (1)$$

$$F(200) = 4 \left| f_A - f_B + f_C - f_D \right| = 4 \left| 2f_{\text{Fe}} - f_{\text{Mn}} - f_{\text{Ga}} \right|, \quad (2)$$

$$F(220) = 4 \left| f_A + f_B + f_C + f_D \right| = 4 \left| 2f_{\text{Fe}} + f_{\text{Mn}} + f_{\text{Ga}} \right|, \quad (3)$$

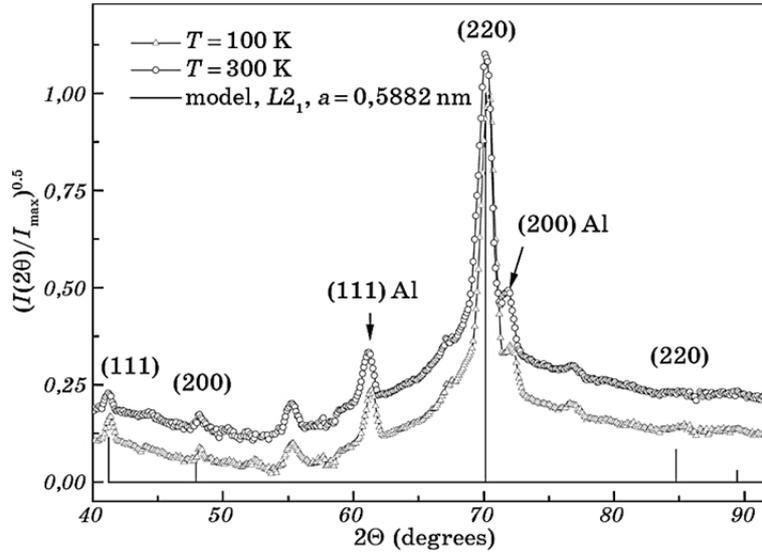


Fig. 2. Experimental ND spectra of Fe_{51.6}Mn_{17.8}Ga_{30.6} alloy taken at different temperatures and $H = 0$ kOe (symbols) together with simulated stroke-diagrams for $L2_1$ ($a = 0.5882$ nm, solid line). ND spectra are shifted relatively each other for convenience of observation.

where f_A , f_B , f_C , and f_D are the average scattering factors for atoms at the sites A , B , C , and D , respectively [10]. Factors f_{Fe} , f_{Mn} and f_{Ga} are equal to $+9.52$, -3.73 and $+7.29 \cdot 10^{13}$ cm, respectively [11, 12]. Thus, for perfectly ordered Fe_2MnGa alloy with $L2_1$ -type of atomic order, the ratio of the superlattice diffraction peak intensities to main principle one in the ND spectrum should be equal to:

$$\frac{F^2(111)}{F^2(220)} = 0.2378, \quad \frac{F^2(200)}{F^2(220)} = 0.4692. \quad (4)$$

Experimental ratios of superstructure lines to the most intense ones for $L2_1$ - and $L1_2$ -phases of $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ and $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloys at different temperatures and magnetic fields are shown in Tables 1 and 2.

If the atomic disorder appears randomly in ternary alloys (like Fe_2MnGa), all the superstructure lines will be reduced in the intensity by factor η^2 , where η is the degree of long-range order [13]. However, if preferential disorder occurs only between certain sites, then 2 groups of superlattice reflections will be affected differently, and it is not possible to describe the state of order in terms of a single ordering parameter [10, 14]. In such case, the total state of order may be represented by two factors S and α [14].

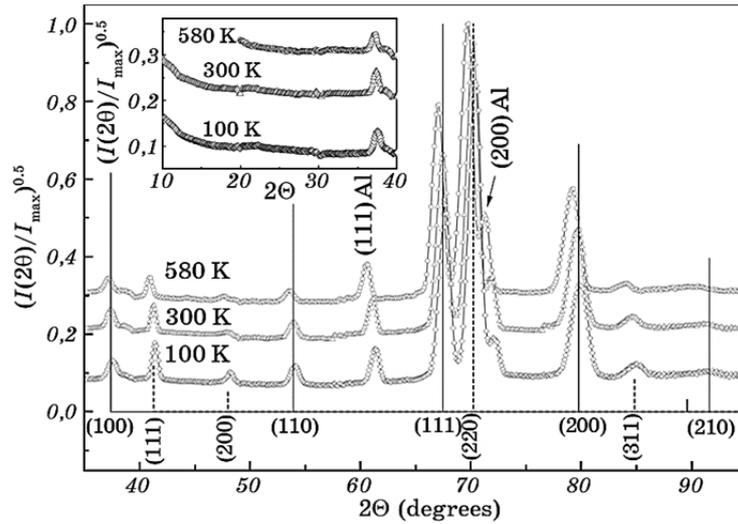


Fig. 3. Experimental ND spectra of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy taken at different temperatures and $H = 0$ kOe (symbols) together with simulated stroke-diagrams for $L2_1$ ($a = 0.5875$ nm, dashed line) and $L1_2$ ($a = 0.3727$ nm, solid line) types of structure. Inset shows the small-angle parts of the experimental ND spectra. ND spectra are shifted relatively each other for convenience of observation.

TABLE 1. Effect of temperature and magnetic field on the relative intensities of superlattice diffraction lines in the experimental ND spectra for Fe_{51.6}Mn_{17.8}Ga_{30.6} alloy.

$I_{\text{super}}/I_{\text{fund}}$	$T = 300 \text{ K},$ $H = 0 \text{ kOe}$	$T = 100 \text{ K},$ $H = 0 \text{ kOe}$	$T = 100 \text{ K},$ $H = 50 \text{ kOe}$
$I(111)/I(220)$	0.0100	0.0190	0.048
$I(200)/I(220)$	0.0037	0.0046	0.005
$I(311)/I(220)$	0.0030	0.0040	0.009

TABLE 2. Effect of temperature and magnetic field on the relative intensities of superlattice diffraction lines in the experimental ND spectra for Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy.

$I_{\text{super}}/I_{\text{fund}},$ structure	$T = 580 \text{ K},$ $H = 0 \text{ kOe}$	$T = 300 \text{ K},$ $H = 0 \text{ kOe}$	$T = 100 \text{ K},$ $H = 0 \text{ kOe}$	$T = 300 \text{ K},$ $H = 50 \text{ kOe}$	$T = 100 \text{ K},$ $H = 50 \text{ kOe}$
$I(111)/I(220), L2_1$	0.035	0.042	0.054	0.043	0.082
$I(200)/I(220), L2_1$	0.007	0.007	0.015	0.007	0.023
$I(311)/I(220), L2_1$	0.013	0.014	0.017	0.017	0.023
$I(100)/I(111), L1_2$	0.051	0.071	0.088	0.093	0.101
$I(110)/I(111), L1_2$	0.031	0.050	0.055	0.063	0.068
$I(200)/I(111), L1_2$	0.023	0.036	0.039	0.034	0.049

In the case of preferential $Y \leftrightarrow Z$ (or $\text{Mn} \leftrightarrow \text{Ga}$ in our case) disorder, the intensities of odd superlattice diffraction lines are reduced by factor $(1-2\alpha)^2$, where α is an occupation parameter defined as portion of Mn or Ga atoms at improper sites. Thus, α can increase from 0 to 0.5 upon increase in $Y \leftrightarrow Z$ disordering of perfectly ordered Heusler alloy. Comparing experimental intensity ratios of (111) reflection to (220) one for Fe_{50.1}Mn_{22.7}Ga_{27.2} and Fe_{51.6}Mn_{17.8}Ga_{30.6} alloys taken at $T = 100 \text{ K}$ and $H = 0 \text{ kOe}$ with those calculated for perfectly ordered Fe₂MnGa alloy, the experimental degrees of Mn \leftrightarrow Ga disorder was estimated and found to be $\alpha = 0.262$ and 0.359 , respectively.

The even superlattice lines are unaffected by $Y \leftrightarrow Z$ disorder; they are only reduced by factor S^2 , which characterizes $X \leftrightarrow Y$ disorder. The corresponding experimental value of S for Fe_{50.1}Mn_{22.7}Ga_{27.2} and Fe_{51.6}Mn_{17.8}Ga_{30.6} alloys calculated for $T = 100 \text{ K}$ and $H = 0 \text{ kOe}$ is equal to $S = 0.179$ and 0.099 . S is changed from 1 to 0 upon disordering from perfectly ordered to completely disordered state. Thus, it is seen that both investigated Fe₂MnGa alloys are noticeably disordered with $X \leftrightarrow Y$ as well as $Y \leftrightarrow Z$ types of disorder.

Figures 4 and 5 present magnetic properties of investigated

Fe_2MnGa alloys. The temperature dependence of DC magnetic susceptibility for mainly $L2_1$ -phase containing $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloy looks typical of ferromagnets with the Curie temperature of $T_c \cong 200$ K— increase in temperature from $T = 80$ K causes decrease in χ value. A minute increase in susceptibility near $T = 500$ K may be related to the traces of FM at these temperatures $L1_2$ phase in the alloy (see inset in Fig. 4). The $\chi(T)$ dependence for $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy is more complicated. Indeed, observed near $T \cong 250$ K decrease (on warming) in $\chi(T)$ is concerned with the FM-to-PM transformation in $L2_1$ phase of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy (see Fig. 4). Further increase in temperature causes rapid growth of $\chi(T)$ above $T \cong 400$ K. Similar temperature dependence is also observed for magnetization of Fe_2MnGa alloys measured at weak ($H = 50$ Oe) polarizing magnetic field (see Fig. 5).

Any structural transformation as a origin of such changes in $\chi(T)$ and/or $M(T)$ at $T = 300$ – 400 K is unlikely because of lack of any visible qualitative changes in ND spectra with temperature (see Fig. 3). The growth with temperature of $\chi(T)$ or/and $M(T)$ above $T = 300$ – 400 K was observed earlier in γ - Fe_2MnGa alloys of similar composition and explained mostly by metamagnetic phase transition from AFM to FM magnetic order in $L1_2$ -phase of alloy [6–9, 18].

Application of the external magnetic field of $H = 50$ kOe drastically changes $M(T)$ dependences for both alloys. Significant polarizing magnetic field suppresses the AFM order in $L1_2$ -phase of

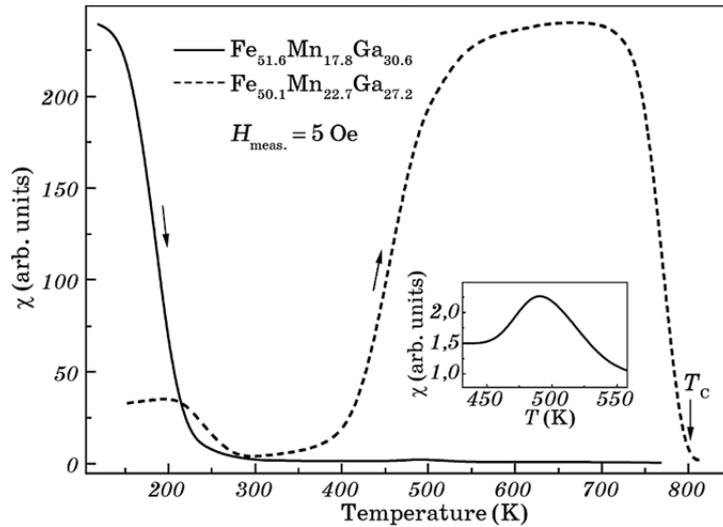


Fig. 4. Temperature dependences of the DC magnetic susceptibility of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ and $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloys taken at weak measuring magnetic field. Inset shows enlarged view of the $\chi(T)$ plot for $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloy in restricted temperature range.

Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy making $M(T)$ dependence for this alloy typical for ferromagnets (see Fig. 5). At the same time, significant magnetization above the Curie temperature is also observed for Fe_{51.6}Mn_{17.8}Ga_{30.6} alloy due to alignment of magnetic fluctuations.

Hence, we conclude that, at $400 < T < 800$ K and a low magnetic field, Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy contains only FM $L1_2$ -phase with $T_C = 800$ K and PM $L2_1$ -phase. Near RT , it has AFM $L1_2$ - and PM $L2_1$ -phases, and at $T < 200$ K, Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy comprises of FM $L2_1$ -and AFM $L1_2$ -phases.

Current opinion on AFM-to-FM transition as the reason of $\chi(T)$ and/or $M(T)$ behaviour at low magnetic fields is supported mainly by the measurements of $M(H)$ hysteresis loops at different temperatures [6, 7]. However, to the best of our knowledge, any direct evidence of AFM order existence at low magnetic fields was presented.

The formation of the AFM order with antiparallel alignment of magnetic moments (if any) should be accompanied with doubling of the plane spacing of ‘magnetic lattice’ commensurate with $L1_2$ -type lattice and hence should lead to an appearance of additional diffraction peaks in the small-angle region. However, it can be seen that no additional reflections appear in the small-angle region of ND spectra taken at $T = 300$ K and even $T = 100$ K without external magnetic field (see Fig. 3). Consequently, if AFM order is really formed in Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy, it has not collinear but probably more complicated (like in rare-earth metals) character.

The coherent neutron diffraction peaks of ferromagnetic alloys con-

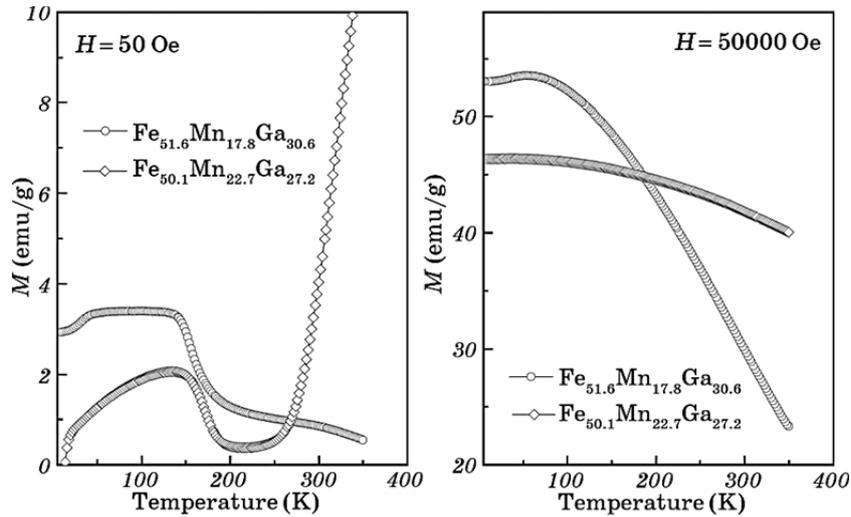


Fig. 5. Effect of temperature and polarizing magnetic field on magnetization of Fe_{50.1}Mn_{22.7}Ga_{27.2} and Fe_{51.6}Mn_{17.8}Ga_{30.6} alloys.

tain both nuclear and magnetic contributions to the resulting structure factor of elastic neutron scattering. For unpolarised neutrons, the magnetic and nuclear scattering intensities are additive, and the total structure factor F is given by the equation:

$$F_{\text{tot}}^2 = F_{\text{nucl}}^2 + q^2 F_{\text{magn}}^2, \quad (5)$$

where q is magnetic interaction vector given by:

$$q^2 = |1 - (\mathbf{E} \cdot \mathbf{M})| = \sin^2 \gamma, \quad (6)$$

where γ is the angle between magnetization \mathbf{M} and scattering vector \mathbf{E} . For polycrystalline cubic samples in the absence of external magnetic field only an average value of q^2 is equal to $q_{\text{av}}^2 = 2/3$ for all reflections [10]. If, however, the magnetization is aligned along the scattering vector then $q^2 = 0$; the magnetic scattering is effectively extinguished and $F_{\text{tot}}^2 = F_{\text{nucl}}^2$. In our case, magnetic field was directed normally to scattering vector. Therefore, $q^2 = 1$. Thus, one can write:

$$F_{\text{tot}}^2(\mathbf{H} = \mathbf{0}) = F_{\text{nucl}}^2 + 2F_{\text{magn}}^2/3, \quad (7)$$

$$F_{\text{tot}}^2(\mathbf{H} \perp \mathbf{E}) = F_{\text{nucl}}^2 + F_{\text{magn}}^2, \quad (8)$$

$$\Delta(F_{\text{tot}}^2) = F_{\text{tot}}^2(\mathbf{H} \perp \mathbf{E}) - F_{\text{tot}}^2(\mathbf{H} = \mathbf{0}) = F_{\text{magn}}^2/3. \quad (9)$$

The intensities of the odd peaks depend only upon the magnetic moments at the Mn sites whereas the intensities of the even superlattice peaks depend upon the difference in the magnetic moments localized at the Mn and Fe sites [14].

Figures 6 and 7 show the effect of polarizing magnetic field on the ND spectra of $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ and $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloys taken at $T = 100$ K. It is seen that magnetic ordering due to external magnetic field causes increase in the intensities of superlattice diffraction peaks related to $L2_1$ -phase of $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloy as well as to $L2_1$ - and $L1_2$ -phases of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy.

For magnetic scattering, the structure amplitudes in Eqs. (7)–(9) are written in terms of magnetic scattering length p . The p is related to the atomic moment μ (in Bohr magnetons μ_B) by the equation:

$$p = 0.269\mu f_{\Theta}, \quad (10)$$

where f_{Θ} is the magnetic form factor correction at the angle of reflection [14]. The magnetic form factor corrections f_{Θ} for the Fe and Mn atoms at $2\Theta = 41.3^\circ$ ($4\pi\sin\Theta/\lambda = 1.85 \text{ \AA}^{-1}$) are equal to 0.8 and 0.75, respectively [15, 16].

Taking into account the magnetic contribution to (111) superlattice reflection of the ND spectra of Fe₂MnGa alloy at $T = 100$ K and $H = 50$ kOe [$\Delta I(111)/I(220) = 0.0278$ for Fe_{50.1}Mn_{22.7}Ga_{27.2} and 0.0294 for

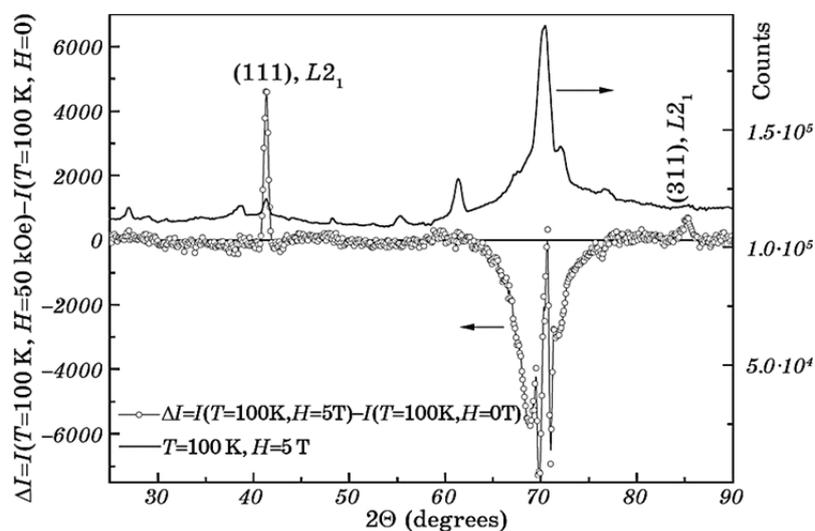


Fig. 6. Magnetic contribution to ND spectrum of Fe_{51.6}Mn_{17.8}Ga_{30.6} alloy taken at $T = 100$ K and $H = 50$ kOe (circles, left scale). ND spectrum at $T = 100$ K and $H = 0$ T is shown by line (right scale).

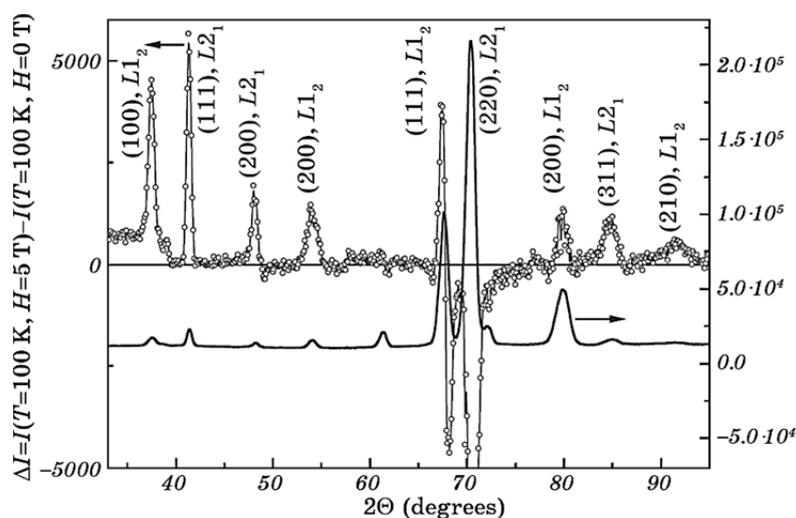


Fig. 7. Magnetic contribution to the ND spectrum of Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy taken at $T = 100$ K and $H = 50$ kOe (circles, left scale). ND spectrum at $T = 100$ K and $H = 0$ Oe is shown by line (right scale).

$\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$], the magnetic moment located at the Mn sites of $L2_1$ -phase of these alloys was evaluated: $\mu_{\text{Mn}} = 1.43\mu_B$ and $\mu_{\text{Mn}} = 1.47\mu_B$, respectively. Considering magnetic contribution to (200) superlattice reflection in the ND spectrum of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy ($\mu_{\text{Mn-Fe}} = 0.84\mu_B$) the magnetic moment located at the Fe sites of $L2_1$ -phase was also estimated: $\mu_{\text{Fe}} = 0.59\mu_B$. The obtained values of μ_{Mn} and μ_{Fe} somewhat differ from calculated ones ($\mu_{\text{Mn}}^{\text{calc.}} = 2.48\mu_B$ and $\mu_{\text{Fe}}^{\text{calc.}} = 0.23\mu_B$) [17]. Furthermore, the experimental practically saturation magnetization value for $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy at $T = 4.2$ K and $H = 50$ kOe (see Fig. 5) was found to be $M_{\text{sat.}} = 46.4$ emu/g or $2.00 \mu_B/\text{f.u.}$ Taking into account the content of the $L1_2$ and $L2_1$ phases in $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy as well as calculated magnetization values of these phases ($\mu_{L2_1}^{\text{calc.}} = 2.011 \mu_B/\text{f.u.}$ and $\mu_{L1_2}^{\text{calc.}} = 6.11 \mu_B/\text{f.u.}$ [17]) perfectly ordered $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy should have $\mu_{L2_1+L1_2}^{\text{calc.}} = 3.96 \mu_B/\text{f.u.}$ The observed difference between experimental and calculated magnetic properties of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy can be concerned with significant atomic disorder in it.

Some of slightly off-stoichiometric Fe_2MnGa alloys, which contain $L2_1$ (or $B2$) phase, exhibit a martensitic transformation in the temperature range $50 < T < 250$ K. This phase transition is accompanied with significant changes in resistivity and magnetization [1–5]. However, variation of the temperature within $100 < T < 580$ K range does not lead to any qualitative changes in the ND spectra of $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ alloy—the ordinary temperature shift of the diffraction peaks is observed (see Figs. 2 and 3). Lack of any visible peculiarities in the temperature dependences of resistivity for the $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ and $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ (see Fig. 8) alloys in a $80 < T < 400$ K temperature range (besides usual temperature dependence with positive temperature coefficient of resistance) together with temperature dependence of ND spectra allows us to conclude that the martensitic transformation does not take place in these alloys.

At the same time, both $\text{Fe}_{50.1}\text{Mn}_{22.7}\text{Ga}_{27.2}$ and $\text{Fe}_{51.6}\text{Mn}_{17.8}\text{Ga}_{30.6}$ alloys contain $L2_1$ -phase. Thus, one can conclude that FM order of this phase at low temperatures (see Figs. 4 and 5) is an intrinsic property of $L2_1$ -phase and not caused by any martensitic transformation.

4. CONCLUSIONS

1. The degree of atomic order in the investigated Fe_2MnGa alloys was experimentally estimated. As shown, a long-term annealing did not result in formation of well-ordered alloys.
2. Magnetic moments localized at the Mn and Fe sites of Fe_2MnGa alloys were experimentally evaluated from neutron scattering data.

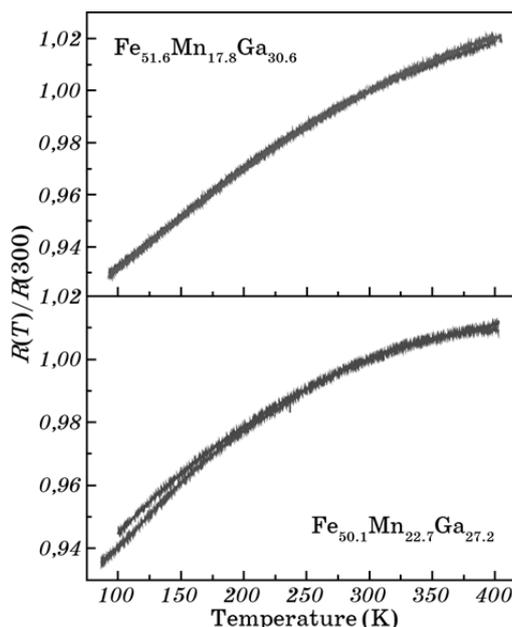


Fig. 8. Temperature dependences of the $R(T)/R(T = 300 \text{ K})$ dependences normalized with respect resistance at $T = 300 \text{ K}$ for Fe₂MnGa alloys obtained for cooling and warming regimes.

Some disagreement between experimental and calculated values of magnetic moments located at the Mn and Fe sites can be explained by noticeable atomic disorder in the prepared Fe₂MnGa alloys.

3. If antiferromagnetic order is really formed in $L1_2$ -phase containing Fe_{50.1}Mn_{22.7}Ga_{27.2} alloy, this order has not collinear character.

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