Annealing effects on the structural and magnetic properties of off-stoichiometric Fe-Mn-Ga ferromagnetic shape memory alloys

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Abstract:

Annealing plays an important role in modifying structures and properties of ferromagnetic shape memory alloys (FSMAs). The annealing effect on the structures and magnetic properties of off-stoichiometric Fe₄₅Mn₂₆Ga₂₉ FSMA has been investigated at different elevated temperatures. Rietveld refinements of neutron diffraction patterns display that the formation of the γ phase in Fe₄₅Mn₂₆Ga₂₉ annealed at 1073 K increases the martensitic transformation temperature and reduces the thermal hysteresis in comparison to the homogenized sample. The phase segregation of a Fe-rich cubic phase and a Ga-rich cubic phase occurs at the annealing temperature of 773 K. The atomic occupancies of the alloys are determined thanks to the neutron's capability of differentiating transition metals. The annealing effects at different temperatures introduce a different magnetic characteristic that is associated with distinctive structural changes in the crystal.

Graphical abstract

Key words: ferromagnetic shape memory alloy; annealing; γ phase; phase segregation.

Introduction

Ferromagnetic shape memory alloys (FSMAs) exhibit both reversible martensitic and ferromagnetic transformations in response to the external magnetic, temperature and stress fields. The shape memory effect that is based on those integrated transformations thus becomes controllable, and it makes FSMAs viable for potential applications as actuators, sensors and electromagnetic devices. Since a large magnetically induced strain was confirmed in Ni_2MnGa alloys [1,2], extensive studies have been conducted in developing new ferromagnetic shape memory alloys, such as Ni_2FeGa [3,4], Co_2NiGa [5,6], **CuAlMn** [7,8] and Fe_2MnGa [9,10] alloys.

Among those, the Fe-Mn-Ga is a new system of FSMA since the martensitic transformation from paramagnetic cubic austenite phase to the ferromagnetic tetragonal martensite phase was firstly studied in $\text{Fe}_{43}\text{Mn}_{28}\text{Ga}_{29}$ in 2009 [9]. The Fe-Mn-Ga alloys

gradually attracted great interests due to the exhibiting large difference in magnetization between the parent austenite and martensite phases and therefore a large shape memory strain up to 3.6 % [10], in comparison with 1.5 % in NiMnGa polycrystalline [11] and 0.1 % in NiFeGa alloy [12]. The study of the Fe-Mn-Ga alloys has been extended to the magnetic, magnetotransport and optical properties [13,14] and then the correlation to the martensitic transformation [15,16]. The evolution of the magnetic domain structure in single crystals of a near the composition Fe_2MnGa was observed during phase transition by photoelectron emission microscopy [15]. A recent study of $\text{Fe}_{50}\text{Mn}_{23}\text{Ga}_{27}$ alloy found that the defects-induced local symmetry break suppressed the martensitic transformation without affecting the magnetic ordering [16].

However, most of the studies above had the sample prepared only through the process of homogenization treatment. The annealing treatments following, which are commonly used in the various alloy systems to tune the structures and the magneto mechanical properties in the development of FSMA were barely reported in Fe-Mn-Ga alloys. In fact, although the annealing effects from structures to properties were intensively reported in other FSMA, the results and the mechanisms were not consistent. The martensitic transformation temperature was found to increase with elevating the annealing temperature in $\text{Ni}_{55}\text{Fe}_{18}\text{Ga}_{27}$ [17], $\text{Ni}_{51}\text{Fe}_{22}\text{Ga}_{27}$ [18] and the $\text{Ni}_{50.6}\text{Mn}_{28}\text{Ga}_{21.4}$ microwires [19], while it could be remarkably decreased with longer annealing time in the $\text{Ni}_{53}\text{Mn}_{23.5}\text{Ga}_{18.5}\text{Ti}_5$ alloy [20]. In terms of the magnetic properties under annealing effects, the Curie temperature was increased in $\text{Ni}_{50.6}\text{Mn}_{28}\text{Ga}_{21.4}$ [19] but decreased in $\text{Ni}_{51}\text{Fe}_{22}\text{Ga}_{27}$ [20] and the saturation magnetization was enhanced in both $\text{Ni}_{49.5}\text{Mn}_{29.3}\text{Ga}_{21.2}$ [21] and $\text{Ni}_{50.6}\text{Mn}_{28}\text{Ga}_{21.4}$ [19]. For whatever the trend is, the authors claimed the correlation with the atomic ordering in the ternary systems [18, 19, 22]. Moreover, the precipitation may form after the thermal treatment to alter the phase structure. The $\text{Ni}_{49.5}\text{Mn}_{29.3}\text{Ga}_{21.2}$ alloy after annealing at 1173 K with presence of γ

phase precipitation possesses good ductility and enhanced saturation magnetization [21]. Increased annealing time results in the formation of Ni_3Ti precipitates as observed in the $\text{Ni}_{53}\text{Mn}_{23.5}\text{Ga}_{18.5}\text{Ti}_5$ alloy, and after annealing, the formed five-layer martensitic structure of the alloy improves its ductility at room temperature [23]. Additionally, the decomposed Co-rich precipitates in the $\text{Co}_{46}\text{Ni}_{27}\text{Ga}_{27}$ martensite [24] and the secondary phase in the $\text{Ni}_{55}\text{Fe}_{18}\text{Ga}_{27}$ alloy [25] were respectively observed after annealing treatment. Apparently, the properties of the FSMA have been tuned through the structure modification, as many reports claimed. However, the annealing effects may differ from case to case, the specific study of the phase structure evolution and the complex atomic ordering in a ternary Fe-Mn-Ga system under the annealing treatment is essential to understand the structural mechanism of the material behaviors that are closely related to the FSMA performance.

The neutron diffraction is a suitable technique for the phase structure investigation of Fe-Mn-Ga alloys. The large cross-section of all components make it possible to differentiate multiple phases, and the large penetration depth of neutrons further reflect the behavior of the bulk instead of the surface. Neutron diffraction enables deciphering the site occupancies in the crystal structure of a complex FSMA system, owing to the differentiable scattering contrasts of transition metals elements [26-29]. Utilizing the advantages of the neutron diffraction for the material structure characterization, in this paper, we select the $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$, an off-stoichiometric Fe_2MnGa , as the model material to report the significant temperature-dependence of the phase composition and the atomic ordering under the annealing. The annealing effects on the martensitic transformation and the magnetic properties of this FSMA system is to be revealed.

Sample preparation and measurements

The polycrystalline button ingot with a nominal composition of $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ was prepared

with high-purity iron (99.99%), manganese (99.99%) and gallium (99.99%) by melting three times in an arc-melting furnace under a high-purity argon atmosphere. According to the process reported before [9,10,13], the ingot was homogenized at 1273 K for 24 h and quenched in water and then cut into three sections (named as Sample I~ III). Sample I was homogenized at 1273 K, Sample II and III were annealed 2 h at temperatures of 1073 K and 773 K, respectively. All the three samples were water quenched after thermal treatment to explore crystal structure evolutions and magnetism behaviors.

The bulk magnetic properties were examined using a superconducting quantum interference device magnetometer with heating and cooling rates of 2 Kmin⁻¹. The magnetization measurements were performed in the temperature range of 4 K - 300 K with an applied magnetic field up to 6.5 T. The temperature dependence of the magnetization was measured in zero field cooling (ZFC) and in field cooling (FC) modes. The crystal structures of Sample I were investigated by neutron diffraction at the POWGEN diffractometer at 300 K and 20 K. The neutron diffraction measurements of Sample II and III were collected at the VULCAN instrument [30] at the Spallation Neutron Source (SNS). The neutron diffraction under 3 T magnetic field were measured at the HB2a powder diffractometer at High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory. The neutron diffraction patterns were analyzed by performing Rietveld refinement using the GSAS software [31] and EXPGUI interface [32], which extract the refined lattice parameters, site occupancies and weight fraction of each phase.

Results

The influence of annealing on microstructure of Fe₄₅Mn₂₆Ga₂₉

The crystal structures were obtained by performing Rietveld analysis on the neutron diffraction patterns. For example, Fig. 1 shows the refined patterns of Sample I at 20 K and

300 K. The $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ has a pure austenitic cubic phase ($\text{Fm}\bar{3}\text{m}$ space group) at 300 K [14], while at 20 K both the martensite phase ($\text{I4}/\text{mmm}$ space group) [9] and the austenite phase co-exist in it. The lattice parameter a of the austenite is $\sim 5.874 \text{ \AA}$ at 300 K and $\sim 5.842 \text{ \AA}$ at 20 K. The lattice parameters of the martensite are $\sim 3.790 \text{ \AA}$ for a and $\sim 7.083 \text{ \AA}$ for c at 20 K, respectively.

In order to reveal the chemical ordering of Fe, Ga and Mn in the austenite lattice and in the martensite lattice, their occupancies at the three lattice sites were calculated with Rietveld refinement. In the refinement, the arbitrary exchanges of two elements between two sites were allowed with the full site occupancy constraint and the composition constraint, and the process was repeated throughout the three elements and all three sites until the stable minimum χ^2 was reached. The determined atom occupancies in the austenite (300 K) and martensite (4 K) are presented in Table 1 and Table 2.

At 300 K, the results show that in the austenite unit cell, the Fe, Ga and Mn atoms mainly occupy the Wyckoff Position 8c ($1/4, 1/4, 1/4$), 4b ($1/2, 1/2, 1/2$), and 4a ($0, 0, 0$), respectively. However, as an off-stoichiometric composition, the structure is not fully ordered because there are multiple species of atoms in each site. The excess Ga atoms (Ga_{Mn}) occupy the Mn sites and some Mn atoms (Mn_{Fe}) shift to the free Fe sites as the result of the defect pair of Ga_{Mn} and Mn_{Fe} . The Fe and Mn atoms also have a certain degree of exchange between 8c and 4a sites. Therefore, the $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ has a partial ordered atomic arrangement in the cubic austenite at 300 K.

Figure 1. Neutron diffraction patterns of Sample I homogenized at 1273 K for 24 h (a) the austenite ($\text{Fm}\bar{3}\text{m}$) phase at 300 K and (b) the martensite (I4/mmm) and austenite phases co-exist at 20 K.

The martensite and austenite phases co-exist at 20 K. The weight fractions of martensite and austenite phases are 69% and 31%, respectively, as extracted from the Rietveld refinement using GSAS. The martensite phase is not fully ordered as shown by the atom site occupancy in Table 2. The low-temperature austenite phase lattice monotonically shrinks upon cooling, not showing any anomalous changes.

Figure 2. Neutron diffraction pattern at 300 K; (a) Sample II and (b) Sample III; the graphs on the right side of (b) show the peak splitting that indicate co-existence of two cubic phase.

Table 1. Structural details of Sample I (austenite phase) at 300 K; $Fm\bar{3}m$ space group; $a=5.86608(2)$ Å. The U_{iso} values of the atoms at the same site are constrained the same.

Atom	Wyckoff	x	y	z	Site occupancy	$100*U_{iso}$
Fe/Mn/Ga	8c	0.25	0.25	0.25	0.616(4)/0.266(4)/0.118(5)	0.984(7)
Fe/Mn/Ga	4b	0.5	0.5	0.5	0.183(4)/0.114(4)/0.703(5)	1.00(1)
Fe/Mn/Ga	4a	0	0	0	0.386(4)/0.393(4)/0.221(5)	0.78(2)

Table 2. Structural details of Sample I (martensite phase) at 20 K; $I4/mmm$ space group; $a=3.7900(1)$ Å, $c=7.0721(3)$ Å. The U_{iso} of all atoms are constrained the same.

Atom	Wyckoff	x	y	z	Site occupancy	$100*U_{iso}$
Fe/Mn/Ga	4d	0	0.5	0.25	0.589(8)/0.233(8)/0.178(9)	0.213(4)
Fe/Mn/Ga	2b	0	0	0.5	0.229(8)/0.155(8)/0.616(1)	0.213(4)

Fe/Mn/Ga	2a	0	0	0	0.393(7)/0.419(7)/0.188(9)	0.213(4)
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For the tetragonal martensite phase at 20 K, the Fe, Ga and Mn atoms prefer to occupy the Wyckoff Position 4d (0, 1/2, 1/4), 2b (0, 0, 1/2), and 2a (0, 0, 0) in decreasing order (Table 2). The 4d site is not fully occupied by Fe atoms, with nearly half of it occupied by Mn and Ga atoms. The 2b site is mainly occupied by Ga atoms. The Mn and Fe atoms share most of the 2a site. Similar to the cubic austenite phase, the atoms have different degrees of hybridization at the three transition metal sites.

Table 3. Structural details of Sample II (austenite phase) at 300 K; $Fm\bar{3}m$ space group; $a=5.8615(5)$ Å. The U_{iso} values of the atoms at the same site are constrained the same.

Atom	Wyckoff	x	y	z	Site occupancy	100* U_{iso}
Fe/Mn/Ga	8c	0.25	0.25	0.25	0.664(7)/0.241(0)/0.095(3)	1.38(2)
Fe/Mn/Ga	4b	0.5	0.5	0.5	0.208(4)/0.158(4)/0.634(2)	0.65(3)
Fe/Mn/Ga	4a	0	0	0	0.411(5)/0.355(3)/0.234(2)	0.74(3)

After annealing at 1073K and being quenched in water, there are strong peaks corresponding to the austenite $Fm\bar{3}m$ phase and three weak peaks resulting from the γ phase [32-34]. The major phase is indexed as a disordered face-centered cubic structure with the lattice parameter $a \sim 3.71211(1)$ Å (Fig. 2 (a)). The refined stoichiometry based on the neutron results is $Fe_{48.68}Mn_{24.87}Ga_{26.45}$ (Table 3), which deviated from the composition of Sample I. The weight fractions of austenite and γ phases are 96.10% and 3.90%, respectively based on the Rietveld refinement. The dissimilar room temperature crystal structure between Sample I and Sample II indicates that the martensitic transformation temperature for Sample II is different from Sample I. This is due to the compositional change resulting from the

formation of the γ phase. The detailed effect of annealing on the magnetic transformation will be discussed later.

Table 4. Structural details of the two segregated cubic phases in Sample III at 300 K; $Fm\bar{3}m$ space group; Fe-rich phase: $a = 5.8465(1)$ Å; Ga-rich phase: $a = 5.8896(3)$ Å. The U_{iso} values of the atoms at the same site are constrained the same.

Atom	Wyckoff	x	y	z	Site occupancy	100* U_{iso}
Fe/Mn/Ga	8c	0.25	0.25	0.25	0.437(16)/0.183(2)/0.380(14)	1.95(2)
Fe/Mn/Ga	4b	0.5	0.5	0.5	0.733(3)/0.267(3)/0	0.98(3)
Fe/Mn/Ga	4a	0	0	0	0.538(3)/0.462(3)/0	0.12(4)
Fe/Mn/Ga	8c	0.25	0.25	0.25	0/0.439(4)/0.561(4)	0.9(1)
Fe/Mn/Ga	4b	0.5	0.5	0.5	0.104(58)/0.199(8)/0.697(50)	6.4(2)
Fe/Mn/Ga	4a	0	0	0	0.542(90)/0.079(13)/0.379(77)	2.2(1)

After annealing at 773 K and being quenched in water, Sample III has two cubic phases (Fe-rich phase and Ga-rich phase) due to compositional segregation. This manifests as a splitting of a family of peaks in the neutron diffraction pattern (Fig. 2 (b)). Both phases have a cubic symmetry ($Fm\bar{3}m$ space group), and the difference of the lattice parameters is about 0.043 Å. With allowing variations of the composition in the two phases, the refinement using GSAS suggested a possible structure model of site occupancies in the segregating phases (Table 4), which resulted in a good fitting of the neutron diffraction pattern (Figure 2). Based on this proposed model, the composition of Fe-rich and Ga-rich phases are calculated as $Fe_{53.625}Mn_{27.375}Ga_{19}$ and $Fe_{16.15}Mn_{28.9}Ga_{54.95}$, and the weight fractions are 70% and 30%, respectively. As one can see in Table 4, the composition segregation alters the atomic

occupancies of the two phases in comparison with the purely austenite phase in Sample I (Table 1). In the Fe-rich phase, a reduced number of Ga atoms are all gathered at the 8c site, while the 4a site, which is the Ga site in Sample I, is occupied by the excessive Fe atoms and the Mn atoms shifted from the 8c and 4b sites. In the Ga-rich phase, most of the Fe atoms take over the 4a site, while the 4b site is filled by the excessive Ga atoms. The Mn concentration in the two segregated phases remains same.

In addition, there is no trace of the γ phase found in Sample III after being annealed at 773 K, but the single cubic phase becomes metastable and segregates into the two phases, which may be the reason for the suppression of the formation of martensite phase even when the sample was annealed at 773 K temperature. A further establishment of Fe–Mn–Ga phase diagram is required. Nevertheless, the phase segregation is a common phenomenon in alloys, and it has been reported in other systems such as NiTiHf [36], NiMnAl [37] and CoNiAl [38] after the relatively low-temperature annealing.

Figure 3. (a) Neutron diffraction patterns of Sample I heated from 20 K to 280 K, field cooled with an applied magnetic field of 3 T; (b) temperature dependence of the martensite and austenite weight fractions during the heating process.

In order to understand the structure evolution and the correlation to the magnetic behaviors at the temperature region that we are interested in, the cryo neutron diffraction with

applied magnetic field of 3 T was carried out. After field cooling at 3 T, Sample I shows the martensitic transformation upon heating from 20 K to 280 K as shown Fig. 3(a). The crystal structure of $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ at 300 K under 3 T magnetic field is $\text{Fm}\bar{3}\text{m}$ austenite cubic phase, while at 20 K both martensite phase (I4/mmm) and the austenite phase co-exist, the austenite phase and martensite phase structures are same as the structures without the magnetic field. In the heating process, the martensite weight fraction kept unchanged at ~70% until the temperature reached 140 K, around which the phase transition happens (Fig. 3(b)). In the temperature range from 140 K to 190 K, the martensite weight fraction reduced significantly almost to 0 and the austenite weight fraction rose accordingly.

Based on the neutron phase analysis, we summarize the sequence of phase transition and reaction in $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ alloy with different annealing treatments in Figure 4. In detail, the influences of annealing on the phase transition are displayed.

Figure 4. Scheme of the phase evolutions in the $\text{Fe}_{45}\text{Mn}_{26}\text{Ga}_{29}$ alloy prepared by different heat treatments.

Effect of annealing on magnetic properties

Figure 5. The ZFC and FC thermomagnetization curves are measured at 5 T for Sample I, Sample II and Sample III. Sample I is denoted as black open-square curves; Sample II is denoted as red open-circle curves; Sample III is denoted as blue open-up-triangle curves. Sample III remains paramagnetic throughout the temperature range below room temperature while Sample I and II display ferromagnetic hysteretic behaviors.

In the magnetic field of 5 T, the magnetization of the three samples at 5 K are 91 emu/g, 102 emu/g and 59 emu/g, respectively (Fig. 5). According to the results of previous studies of $\text{Fe}_{50}\text{Mn}_{22.5}\text{Ga}_{27.5}$, the magnetization at 5 K in 5 T magnetic field is about 94 emu/g, and the magnetization of $\text{Fe}_{43}\text{Mn}_{28}\text{Ga}_{29}$ at 4.2 K in the same magnetic field is about 83 emu/g [9,10]. This behavior corresponds to a coupled magneto structural phase transition between the ferromagnetic martensite and paramagnetic austenite, which indicates the chemical compositions affect the magnetic properties on Fe-Mn-Ga alloys. In our cases, the annealing treatment results in similar changes on magnetic properties in the same component.

Figure 6. Magnetic field dependence of the magnetization of Sample I, II, and III measured from 0 T to 6.5 T; (a) the magnetization curves at 300 K; (b) the magnetization curves at 4 K.

The magnetization curves of the samples at 300 K and 4 K are shown in Fig. 6 (a) and (b), respectively. At both temperatures, the saturated magnetization agree with the results in the $M(T)$ data in Figure 5. At 300 K, Sample I that is a pure austenite phase shows a linear behavior with increasing magnetic field, as it is paramagnetic (Fig. 6 (a)). The behavior in Sample II does not deviate too much in comparison with Sample I, because the paramagnetic austenite is the major phase (96.1 wt. %), as indicated by neutron diffraction. The slight increase in magnetization may be attributed to the small amount of the coexisting γ phase, which is ferromagnetic at room temperature [33,34]. In Sample III, with the coexistence of the Fe-rich phase and Ga-rich phase, although the segregated phases have the same lattice symmetry and similar lattice parameters as the austenite, a significant ferromagnetic moment is observed at 300 K. Based on the current experimental results, one cannot distinguish the contributions from the two cubic phases or conclude the mechanism for the ferromagnetism. Nevertheless, it is indicated that the ferromagnetism in Fe-Mn-Ga system has significant sensitivity to the rearrangement of different species of atoms in the $Fm\bar{3}m$ cubic lattice.

It should be noted that the difference of magnetic behaviors is not caused by the dependence of the sample orientation or texture. There are not significant textures as shown in the diffraction patterns. We have also carried out the magnetic measurements with the samples cut from different positions of the ingot. The samples were randomly placed in the magnetometer and the measurements were repeated over twice. The results turned out to be consistent, and any texture formed in the thermal treatment should not be responsible for the distinctive magnetic behaviors.

Discussion

Based on the results of the magnetic properties, one can observe the distinct difference in the three samples. In Sample I (homogenized at 1273 K), the magnetization at 5 T shows an abrupt onset of ferromagnetism at 100 K during cooling and decay to the paramagnetic phase at 160 K during heating. The large hysteresis behavior of the magnetization suggests a first-order transition that can be correlated to the reversible austenite to martensite structural transition in response to the temperature change under the external magnetic field. Above 190 K, Sample I becomes paramagnetic completely. The result of the thermomagnetization curve Sample I is supported by the neutron diffraction results with an applied magnetic field showing the structural phase transitions at temperatures when bulk magnetic phase transitions were observed. Fig. 3 suggests that in the Fe-Mn-Ga system, the change of magnetic properties between ferromagnetic and paramagnetic phases is correlated with a similarly reversible structural transformation.

In contrast to Sample I, the formation of the disordered face-centered cubic γ phase in Sample II (annealed at 1073 K) causes a remarkable increase of the martensitic transformation temperatures. Meanwhile, unlike other FSMAs, where the γ phase increases the thermal hysteresis regions [35,36], in Sample II, the apparent thermal hysteresis decreases

due to the introduction of the γ phase. The possible reasons for this are: (i) the γ phase promotes the growth of the martensite nuclei [35], which is ascribed to the local order heterogeneity; and (ii) the γ phase may bring the local variations in composition of the sample. Therefore, considering the presence of the small amount of γ phase which can narrow the phase transition hysteresis and increase the martensitic transformation temperature, one can control the martensitic transformation temperatures by tuning the phase composition via the purposive annealing. The structural change may further impact the thermomagnetization behavior of the Fe-Mn-Ga alloy.

Distinct from the two samples above, Sample III (annealed at 773 K) does not exhibit the first-order ferromagnetic transition. The magnetization gradually increases as the temperature goes down towards 5 K, without showing any anomalous behavior. This suggests that the Fe/Ga segregation in the two cubic phases due to the annealing at 773 K hinders the structural transition. Moreover, the change in the atomic composition and distribution in the lattice sites also alters the ferromagnetism of the material. This follows from the fact that Fe-rich and Ga-rich phases are ferromagnetic at 300 K, different from the paramagnetic austenite phase as in Sample I.

When the temperature decreases to 4 K, the paramagnetic austenite phase in Sample I and Sample II mostly transforms to the ferromagnetic martensite phase. The martensite phase dominates the magnetization at the low field, and it leads to much higher saturated magnetizations at 4 K than those values at 300 K in both Sample I and II (Fig. 6 (b)). The existence of the ferromagnetic γ phase in Sample II is the possible reason for the discrepancy from Sample I at the saturation, which is consistent with the situation at 300 K. Moreover, one can see that the magnetization of Sample II reached saturation at a lower applied magnetic field than that of Sample I, and the γ phase is also thought to be responsible for that. In Sample III, the behavior at 4 K is consistent with that at 300 K despite showing

slightly higher magnetization at the saturation. This agrees with the fact that the cubic $Fm\bar{3}m$ lattice has avoided the structure transformation toward a martensite lattice after the atomic rearrangement during the phase segregation.

As the results of the quenching, dislocations may be expected besides the observed structure changes. But the quantities or dislocation density are expected to be comparable and will not show distinctive changes of the magnetic behaviors that is discussed in the following. If significant different amount of dislocations developed in each sample, the neutron diffraction patterns would show different peak broadenings, which is, however, not observed obviously. Therefore, we believe the magnetic properties are associated more with the distinctive structural changes.

Conclusion

In summary, the annealing effects on the structures and magnetic properties in $Fe_{45}Mn_{26}Ga_{29}$ were investigated. The annealing at different temperatures on Fe-Mn-Ga alloys introduces changes in phase structures and magnetism. Annealing at 1073 K leads to the increase of the martensitic transformation temperature and reduces the thermal hysteresis in comparison to the as homogenized sample, which could be ascribed to the formation of the small amount of γ phase. Furthermore, one can optimize the material composition to attain advanced FSMA by annealing at a temperature that controls the formation of γ phase. In addition, the Fe-rich phase and Ga-rich phase have similar cubic structures when annealed at 773 K. In this phase, both have no structural transformation and display robust ferromagnetism in the temperature range from 4 K to 300 K.

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