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# A method based on magnetic moment measurement to identify the structural transition of quenched $Fe_{1-x}Ga_x$ ($x = 0.15$–$0.30$) alloys*

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A method based on the measurement of Fe average atomic magnetic moment to identify the structural transition caused by the increase of Ga content in quenched $Fe_{1-x}Ga_x$ alloys ($0.15 \leq x \leq 0.30$) is proposed. The quenched $Fe_{1-x}Ga_x$ alloys show a change of the Fe average atomic magnetic moment from $2.25\mu_B$ to $1.78\mu_B$ and then to $1.58\mu_B$, which corresponds to the structural transition from A2 to $D0_3$ and then to B2. The relationship between the structure and the magnetostriction is clarified, and the maximum magnetostriction appears in the A2 phase. The variation tendency of the magnetostriction is well characterized, which also reflects the structural transition.

**Keywords:** magnetic moment, structure, magnetostrictions, Fe–Ga alloy

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## 1. Introduction

The addition of non-magnetic Ga enhances the magnetostriction of Fe over tenfold,[1] and has recently attracted a lot of attention.[2–9] The alloy is a new class of actuator materials. The composition dependence of the magnetostriction of $Fe_{1-x}Ga_x$ ($0.04 \leq x \leq 0.35$) was intensively studied, showing two magnetostriction peaks along the $\langle 100 \rangle$ direction ($\lambda_{100}$) at Ga contents of 19% and 27.5%.[10,11] A large decrease of the magnetostriction was observed at Ga contents higher than 30%.[10] Previous studies showed that the complicated structures depend on the Ga content and the thermal history.[2,10] Several different models were presented for the relationship between the suggested crystal structures and the large magnetostriction of the Fe–Ga alloys. Mössbauer spectroscopy,[12,13] transmission electron microscopy,[14] neutron diffraction,[15,16] and conventional X-ray diffraction[17,18] were used to determine the crystal structure. The structural determination of Fe–Ga alloys by X-ray diffraction is troublesome due to the similarity in the atomic scattering factors of Fe and Ga atoms, which leads to an extremely weak peak intensity of super lattice diffusion scattering.

In this paper, based on the Fe atomic magnetic moment measurement, combined with the determined lattice parameter and the measured Curie temperature, we explore another approach to identify the structural transition with the increase of the Ga content in quenched $Fe_{1-x}Ga_x$ alloys ($0.15 \leq x \leq 0.30$). The corresponding magnetostrictions connected with the structures are also reported for clarifying the relationship between the structures and the magnetostrictions.

## 2. Experiment

$Fe_{1-x}Ga_x$ ingots with $x = 0.15$, 0.17, 0.19, 0.21, 0.25, 0.275 and 0.30 were prepared by arc-melting pure gallium (99.999%) and electrolytic iron (99.99%) in an arc-melting furnace four times under an argon atmosphere. The ingots were remelted and drop cast into a copper mould to form rods with a diameter of 7 mm and a length of 100 mm. The rods were annealed at 1000 °C for 24 h to achieve composition homogenization and then were quenched by water. The X-ray diffraction examination was performed on a Regaku Japan D/max2200PC X-ray diffractometer using Cu $K_\alpha$ radiation. The magnetization measurement was carried out using a Lakeshore-MTS 7307/XP vibrating sample magnetometer. The phase transformation and magnetic transition were checked by using the

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thermal gravity (TG) method with a STA 449C thermal analysis system. The magnetostriction was measured by standard strain gauges.

## 3. Results and discussion

### 3.1. Structure

Figure 1 shows the X-ray diffraction (XRD) patterns of the quenched $Fe_{1-x}Ga_x$ ($x = 0.15$, 0.17, 0.19, 0.21, 0.25, 0.275, 0.30) alloys. The substitution of Fe by Ga shifts the diffraction peaks to smaller $2\theta$ values compared with that of pure $\alpha$-Fe in bcc phase. The lattice parameters $a$ of the $Fe_{1-x}Ga_x$ alloys are determined from the X-ray diffraction patterns, which are summarized in Table 1. The density is measured and is also summarized in Table 1. For quenched $Fe_{1-x}Ga_x$ alloys, the lattice parameter $a$ and the density do not increase monotonically with the Ga content, in spite of the fact that the Ga atom is larger and heavier than the Fe atom. The non-linear changes of the lattice parameter and the density indicate the structural transition of the quenched $Fe_{1-x}Ga_x$ alloys with the increase of Ga content.

The X-ray diffraction patterns show that the observed structures in the quenched $Fe_{1-x}Ga_x$ alloys are of a bcc structure, including the disordered A2 and the ordered $D0_3$ phases.[2] In order to investigate the structure in detail, high-energy X-ray diffraction analyses (48 kV, 120 mA) of $Fe_{0.7}Ga_{0.3}$ and $Fe_{0.83}Ga_{0.17}$ are taken. The diffraction peaks for $Fe_{0.7}Ga_{0.3}$ alloy shown in Fig. 2 correspond to the superlattice reflections originating from the B2 ordering. The $\langle 111\rangle$ reflection at 54° is the figure print of the B2 phase.[19]

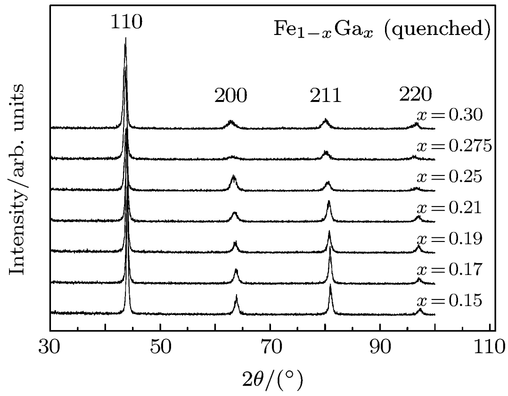


**Fig. 1.** The X-ray diffraction patterns of quenched $Fe_{1-x}Ga_x$ ($x = 0.15$–0.30) alloys.

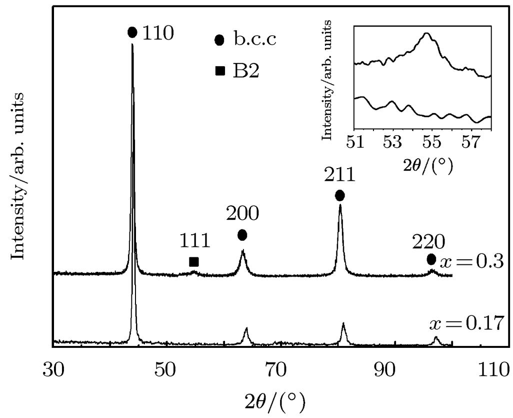


**Fig. 2.** High-energy X-ray diffraction patterns of quenched $Fe_{0.83}Ga_{0.17}$ and $Fe_{0.7}Ga_{0.3}$ alloys. The insert shows the patterns around 54°.

**Table 1.** Densities, saturation magnetizations, lattic parameters and magnetic moments of $Fe_{1-x}Ga_x$ alloys.

| Ga doping/% | 15 | 17 | 19 | 21 | 25 | 27.5 | 30 |
|---|---|---|---|---|---|---|---|
| $a$/Å | 2.9073 | 2.9108 | 2.9164 | 2.9173 | 2.9302 | 2.9343 | 2.9375 |
| Density/g·cm$^{-3}$ | 7.896 | 7.955 | 8.037 | 8.008 | 7.995 | 7.892 | 7.964 |
| $M_s$/emu·g$^{-1}$ | 182 | 176 | 169 | 144 | 123 | 120 | 102 |
| $\mu_{Fe}/\mu_B$ | 2.24 | 2.25 | 2.25 | 1.96 | 1.78 | 1.78 | 1.58 |

However, it is impossible to determine the fine structure from the X-ray diffraction patterns because of the similar atomic scattering factors of Fe and Ga, which cause the disappearance of the super lattice diffraction peaks. The thermal gravity method is used to check the phase transformation and the magnetic transition in the bcc structure.

Figure 3 shows the thermal gravity curves measured under a low magnetic field for the $Fe_{1-x}Ga_x$ alloys. The changes of the weight with the applied magnetic field indicate a magnetic transition.[20] The magnetic transitions are marked by arrows in Fig. 3. Only one magnetic transition is detected for $x \leq 0.19$. For $x = 0.21$, a slight decrease at 560 °C and an abrupt decrease at 680 °C are observed. For $x = 0.25$ and 0.275, the TG curves are similar, which undergo dropping down, then rising up, and followed by dropping down again. For $x = 0.30$, another decline is also observed.

Combined with the metastable phase diagram for the Fe–Ga alloys, for $x \leq 0.19$, the magnetic transition should be from a ferromagnetic (f) A2 phase to paramagnetic (p) A2 phase. For $x = 0.25$ and 0.275, the first magnetic transitions occur at 481 °C and 394 °C, respectively, which agrees well with the magnetic transition temperature of the $D0_3$ phase presented by Ikeda.[21] The rising stages of the TG curves between 500 °C and 670 °C for $x = 0.25$ and 0.275 can be regarded as the phase transition from $D0_3$ to B2, and the last drop in the curve should be connected with the Curie transformation of the B2 phase. The magnetic transition sequences for $x = 0.25$ and 0.275 should be $D0_3(f)\rightarrow D0_3(p)\rightarrow B2(f)\rightarrow B2(p)$, and the structure of the samples is determined to be in the $D0_3$ phase at room temperature. Similar magnetic transitions are observed for $x = 0.3$.

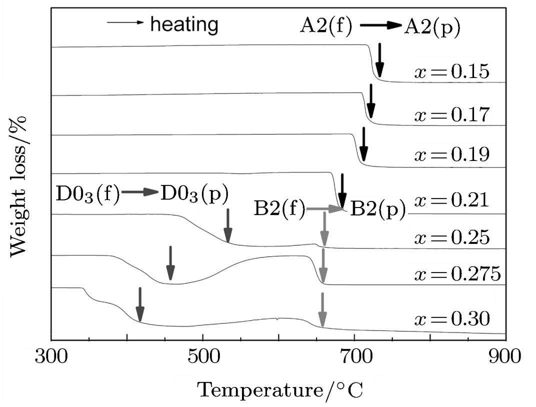


**Fig. 3.** TG curves measured under an applied magnetic field for quenched $Fe_{1-x}Ga_x$ alloys.

## 3.2. Magnetic moment

We assume that the magnetic moment of Fe is not unchanged in the same structure and ignore the magnetic moment of the non-magnetic Ga atom. A method based on the measurement of the Fe average atomic magnetic moment to identify the structural transition caused by the increase of Ga content is then proposed.

Figure 4 shows the initial magnetization curves of the quenched $Fe_{1-x}Ga_x$ alloys. The saturation magnetizations are also summarized in Table. 1. The saturation magnetization decreases with the increasing Ga content. It is reasonable that the substitution of Fe by non-magnetic Ga atom decreases the saturation magnetization. The average magnetic moment of Fe, $\mu_{Fe}$, in the quenched $Fe_{1-x}Ga_x$ alloys is calculated, regarding Ga as the dilute atom. The result alone with the lattice parameter is shown in Fig. 5. It is worth noting that there are two flat stages in the curve of the average magnetic moment of Fe. The first flat stage is for $x \leq 0.19$, where $\mu_{Fe}$ is equal to $2.25\mu_B$, which is comparable to the magnetic moment of a pure Fe atom. The second flat stage is for $x = 0.25$ and 0.275, where $\mu_{Fe}$ is equal to $1.78\mu_B$. The $\mu_{Fe}$ for $x = 0.30$ is $1.58\mu_B$. Therefore, we preliminarily suggest that the alloys in the first stage with $x \leq 0.19$ have a kind of single phase structure, the alloys in the second stage with $x = 0.25$ and 0.275 have another kind of single phase structure, and the two phases should coexist for alloys with $x = 0.21$.

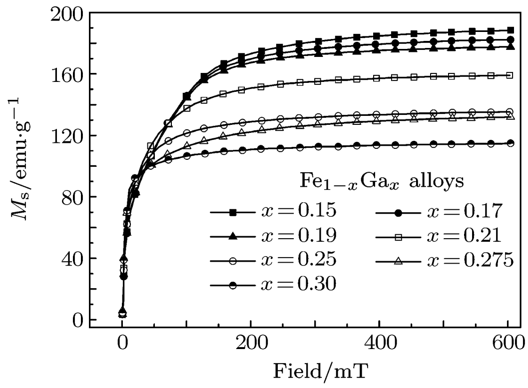


**Fig. 4.** The initial magnetization curves of quenched $Fe_{1-x}Ga_x$ ($x = 0.15$–$0.30$) alloys.

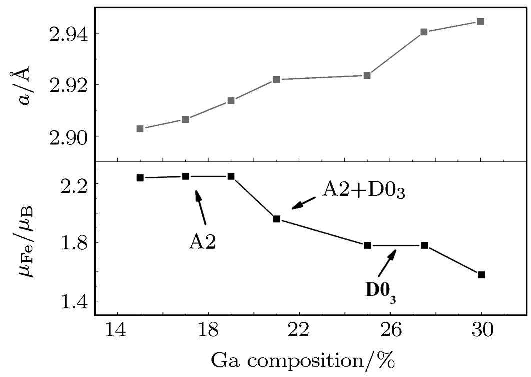


**Fig. 5.** (a) Lattice parameters and (b) Fe average atomic magnetic moments of quenched $Fe_{1-x}Ga_x$ alloys.

The magnetic moments of the supposed structures A2, $D0_3$ and B2 for the Fe–Ga alloys are calculated using the full potential linearized augmented plane wave method. The calculated average magnetic moments of Fe for A2, $D0_3$, and B2 phases are $2.1624\mu_B$, $2.0954\mu_B$ and $1.9366\mu_B$, respectively. Here the values for $D0_3$ and B2 phases are lower than that for the A2 phase. The result agrees with the expectation that the structures of the quenched $Fe_{1-x}Ga_x$ alloys change from A2 to $D0_3$ and then to B2 with the increasing Ga content. The calculated and measured values are not completely the same due to different external conditions, such as temperature and lattice perfection. It is not clear whether there are any other factors that can cause the difference between the calculated and the measured values, such as the lower Curie temperature of $D0_3$ and B2 phases. However, the variation tendency of the average magnetic moment of Fe for A2, $D0_3$ and B2 phases is credible.

We now can briefly summarize above results. The X-ray diffraction patterns show that only the bcc structures, such as A2, $D0_3$ and B2, can be observed in the quenched samples. For $x \leq 0.19$, the structures should be identical, since the measured average magnetic moments of Fe are a constant. In the TG curves, only one magnetic transition is observed, which matches well with the Curie transition of the A2 phase. In addition, the measured average magnetic moment of Fe is consistent with the calculated value for the A2 phase. Combining the measured and the calculated magnetic moments with the TG result, a single A2 phase can be confirmed for $x \leq 0.19$. For $x = 0.25$ and 0.275, the alloys should be in another single phase structure. As suggested by the TG measurement, the $D0_3$ phase definitely exists in those quenched samples. The measured average magnetic moment of Fe for the second flat stage is reduced compared with that for the first stage, which is consistent with the decreasing tendency of the calculated magnetic moment for $D0_3$ phase compared with that for A2 phase. Hence, the $D0_3$ phase is inferred for $x = 0.25$ and 0.275. For $x = 0.21$, the quenched sample should be in the mixed phase of A2 and $D0_3$, as the average magnetic moment lies between the two plateaus and the complex transitions are observed in the TG curve. It is possible that the appearance of the B2 phase further reduces the average magnetic moment and makes the TG curve more complicated for $x = 0.3$.

## 3.3. Magnetostriction

Based on the Fe atomic moment measurement, combined with the determined lattice parameter and the measured Curie temperature, the structural transition is identified. The relationship between the structures and the magnetostrictions should be clarified.

Figure 6 shows the magnetostriction of the quenched $Fe_{1-x}Ga_x$ alloys, which is measured parallel to the applied magnetic field. In Fig. 7, two peaks of the saturation magnetostriction for the quenched Fe–Ga alloys are observed with the increasing Ga content, which were also reported in the work of Lograsso *et al.*[2] The magnetostriction monotonically increases with the increasing Ga content up to $x = 0.19$, reaching a maximum value of $1.73\times10^{-4}$ at $x = 0.19$. The magnetostriction decreases when the Ga content is higher than 19%. The other peak of the magnetostriction is also monitored with the value of $1.16\times10^{-4}$ at $x = 0.275$. The same phenomenon was also reported by Clark *et al.*[10] As the Ga content increases to higher than 27.5%, the magnetostriction further decreases.

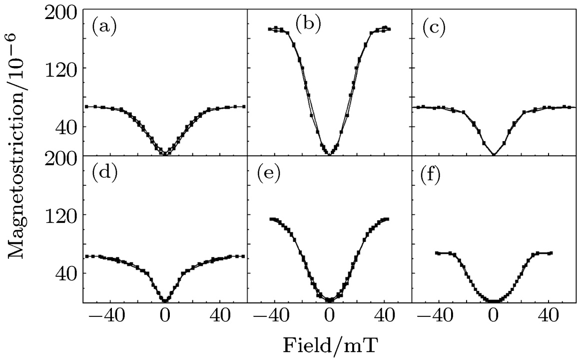


**Fig. 6.** The magnetostriction of quenched $Fe_{1-x}Ga_x$ ($x$ = 0.17–0.30) alloys. The Ga contents are (a) 0.17, (b) 0.19. (c) 0.21, (d) 0.25, (e) 0.275, and (f) 0.3.

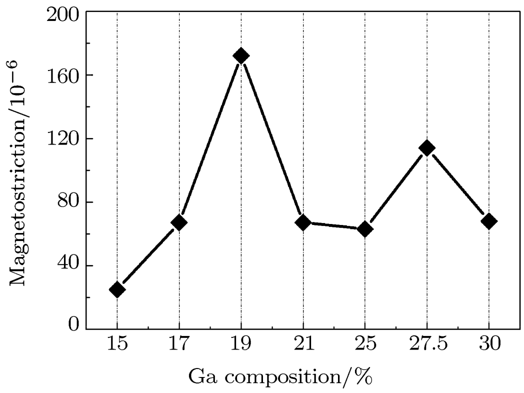


**Fig. 7.** Magnetostriction under the magnetic field of 40 mT for the quenched $Fe_{1-x}Ga_x$ alloys.

As discussed above, different regimes of magnetostriction correspond to different phases. For $x \leq 0.19$, the quenched samples have an A2 single phase; as a result, excellent magnetostriction can be obtained. The magnetostriction increases with an increase in Ga content, suggesting that the non-magnetic Ga atom doped in the A2 phase can enhance the magnetostriction. In the A2 phase, the magnetic anisotropy changes with the Ga local environment, which leads to the distortion of the lattice and results in a large magnetostriction. The maximum magnetostriction can be obtained at $x = 0.19$, and then the magnetostrictions reduces quickly, which should be connected with the change of the structure. As clarified above, the $D0_3$ phase appears as the Ga content is higher than 19%, which deteriorates the magnetostriction. Wu has predicted the lower magnetostriction for the $D0_3$ phase by using the first-principle calculation.[22] The other peak of the magnetostriction at $x = 0.275$ was reported to be connected with the softening of the shear modulus,[23] which is not connected with any structural transition. Based on the present result, the magnetostriction further decreases with the presence of the B2 phase.

## 4. Conclusion

A method based on Fe atomic magnetic moment measurement is used to identify the structural transition with the increase of Ga doping in quenched $Fe_{1-x}Ga_x$ alloys ($0.15 \leq x \leq 0.30$). The structural transition of quenched $Fe_{1-x}Ga_x$ alloys, which is from A2 to $D0_3$ and then to B2 in the experimental Ga content range, is clarified. The variation tendency of the magnetostriction is well characterized, which reflects the structural transition. The maximum of the magnetostriction appears in the A2 phase.

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