Magnetostriction and electrical resistivity of Mn doped Fe₈₁Ga₁₉ alloys

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Abstract

The paper reports on the effect of substituting Mn for Fe on the magnetostrictive and related properties of Fe_{81-x}Mn_xGa₁₉ alloys. Samples of composition were prepared in polycrystalline form and were all single phase BCC. It is shown that the magnetostriction observed in Fe₈₁Ga₁₉ can be enhanced up to about 11% by Mn substitution with a maximum effect at $x = 8$. The change in the magnetostrictive coefficient of Fe₈₁Ga₁₉ with Mn substitution is accompanied by a monotonic increase in resistivity, being increased by almost a factor of 2 for $x = 8$. We also found that the lattice parameter of the alloys increased with the increase in Mn concentration and the Curie temperature decreased monotonically at the same substitution.

1. Introduction

Magnetostriction is the effect observed when the dimensions of a specimen change with the application of a magnetic field, which has been utilized for many applications such as ultrasonic generation and detection, vibration control and magnetostrictive filters [1, 2]. Fe–Ga alloys (Galfenol) have larger magnetostriction coefficient, λ , than that of Ni and Fe and have the advantages of a low temperature coefficient for λ , low cost compared with rare earth intermetallic compounds, and have the advantage of being operational at low magnetic fields because of their negligible magnetic hysteresis [3–6]. Moreover, unlike many current active materials, such as Terfenol-D and cobalt ferrite, which are brittle and unable to withstand any substantial amount of tension, Galfenol has an appreciable tensile strength and good ductility. Besides being used as actuator materials, Galfenol can also serve as energy harvesting materials for converting mechanical vibrations into voltage signals or electric power. However, the Fe–Ga alloys show a relatively low electrical resistivity and eddy current occurs even at modest frequency, which limits the application range.

Previously, many efforts were made to increase the electrical resistivity in the Terfenol-D magnetostrictive alloys [7–9]. Co, Ni or Al elements were doped, but high content of the substituting elements is required (such as about 50% increase with the addition of 10% Al), which leads to a poor magnetostriction. But to our knowledge, works on increasing the electrical resistivity of Galfenol have rarely been reported. Study on intrinsically increasing the electrical resistivity without magnetostriction greatly deteriorated in Fe–Ga alloys currently keeps open and attractive.

Mn has much higher resistivity than that of other magnetic metals. Therefore, it is possible to achieve higher resistivity by doping Mn in Fe–Ga alloys [10]. In this work, we present the results of the effects of Mn doping on the structure, electrical resistivity (ρ), the magnetostriction and Curie temperature (T_C) of Fe–Ga alloys. Both high electrical resistivity and even enhanced magnetostrictive properties were obtained in the FeMnGa alloys.

2. Experimental

The ingots of Fe_{81-x}Mn_xGa₁₉ ($x = 0-16$) were prepared by arc melting the constituent elements, Fe and Ga of purity 99.99% and Mn of purity 99.95%, in an arc furnace under an

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Figure 1. X-ray diffraction patterns of $\text{Fe}_{81-x}\text{Mn}_x\text{Ga}_{19}$ ($x = 0\text{--}16$).

argon atmosphere. Since Mn has very high vapour pressure leading to the loss of the element, usually 3% of Mn was added in excess to the total mixture to compensate for the loss [11]. The melting was carried out four times for good homogeneity and the weight loss after the final melting was less than 0.5%. X-ray diffraction measurements were performed by using Cu $K\alpha$ radiation in a Rigaku D/MAX2200PC diffractometer. The lattice parameters were evaluated by least-squares refinement from powder x-ray diffractograms. The magnetic transition temperatures were measured by ac magnetic susceptibility measurements. The magnetostriction was measured by standard strain gauge on the samples with dimensions of $10 \times 10 \times 2 \text{ mm}^3$. In this open magnetic circuit measurement, the demagnetizing effect has not been deducted. Strip specimens for electrical resistivity measurements were cut by using a spark cutting with dimensions of $1 \times 1 \times 15 \text{ mm}^3$. The electrical resistivity was measured by using the traditional four-probe technique in the temperature range from 100 to 300 K. In order to reduce the possible measuring errors all specimen surfaces were polished and the dimensions were precisely controlled.

3. Results and discussion

Powder x-ray diffraction patterns of Mn doped $\text{Fe}_{81-x}\text{Mn}_x\text{Ga}_{19}$ ($x = 0\text{--}16$) arc-melted samples are presented in figure 1, exhibiting mainly a disordered BCC A2 structure and Mn addition does not change the BCC structure in FeMnGa. According to the phase diagram [12], $\text{Fe}_{81}\text{Ga}_{19}$ alloy is in the disordered BCC A2 phase region at temperatures above 570 °C. On slow cooling to room temperature, ordered DO_3 phase was also reported for $\text{Fe}_{81}\text{Ga}_{19}$ alloy [13, 14]. However, no superlattice reflection peaks of DO_3 were detected and A2 structure is believed for the present arc-melted FeMnGa samples. The substitution of Fe by Mn atoms changed the peak positions to smaller values of 2θ by comparing with the corresponding BCC A2 structure of $\text{Fe}_{81}\text{Ga}_{19}$ alloy. So the lattice parameters are seen to slightly increase with increasing Mn concentration in accordance with the larger ionic radius of Mn compared with Fe as shown in figure 2(a).

Figure 2. Variation of lattice parameter (a), Curie temperature (b) and resistivity at room temperature (c) for $\text{Fe}_{81-x}\text{Mn}_x\text{Ga}_{19}$ with x .

Figure 3. A typical magnetostriction measured parallel and perpendicular to the magnetic field for the composition of $\text{Fe}_{73}\text{Mn}_8\text{Ga}_{19}$ at room temperature.

The Curie temperatures were determined from ac magnetic susceptibility measurements at high temperature. It is found that the Curie temperatures decrease with the substitution of Mn for Fe as expected, which is attributed to the smaller exchange interaction of Mn–Fe than that of Fe–Fe [15]. The general trend is depicted in figure 2(b).

Figure 3 shows a typical magnetostriction graph for composition $\text{Fe}_{73}\text{Mn}_8\text{Ga}_{19}$, measured at room temperature, parallel and perpendicular to the applied magnetic field, respectively. The magnetostriction strain is positive for the parallel orientation (λ_{par}) and a negative value is obtained for the perpendicular orientation (λ_{per}). The magnetostriction values measured in these two directions are not very exactly consistent with the general expression ($\lambda_{\text{par}} = -2\lambda_{\text{per}}$) for them, probably due to slightly preferential orientation of the grains in the samples. Additionally, there is negligible

Figure 4. Mn content dependence of the parallel, perpendicular and total magnetostriction of Fe_{81-x}Mn_xGa₁₉ ($x = 0-16$).

hysteresis in the magnetostriction against H when Mn atoms are added, resulting in a reversible magnetostriction.

The equilibrium strain state of a crystal is determined by minimization of the sum of a crystal's anisotropic magnetic, magnetostrictive and elastic energies [16]. So in polycrystalline cubic materials, the general expression of $\lambda = 3/2\lambda_s(\cos^2\theta - 1/3)$ is applicable. Here θ is the angle between the direction of the magnetic field and the measuring direction. λ_s is the saturation magnetostriction coefficient, which can be obtained by measuring λ in a saturation field in parallel and perpendicular to the direction of measurement of magnetostriction. The total change obtainable in length caused by the magnetic field is then given by $\lambda_{\text{total}} = \lambda_{\text{par}} - \lambda_{\text{per}} = 3/2\lambda_s$, independent of the initial domain distribution. In our samples the highest obtainable value of the magnetostriction in a magnetically saturated state was obtained for the composition of Fe₇₃Mn₈Ga₁₉, reaching a value of 102 ppm at an applied field $H = 80 \text{ kA m}^{-1}$.

The variation of the parallel, perpendicular and total magnetostriction as a function of Mn concentration is shown in figure 4. A composition dependence of magnetostriction on the Mn concentration is observed along both directions. As can be seen, the largest magnetostriction improvement was obtained for Fe₇₃Mn₈Ga₁₉, reaching a value of 11% larger than the value of Fe₈₁Ga₁₉. However, with a further increase in Mn concentration from $x = 8$ to $x = 16$, λ_{total} decreases and finally it is about 12% less than the undoped sample. It can be considered that the magnetostriction performance in polycrystalline Fe–Ga alloys can be improved by the substitutions of Mn for Fe. This improvement can be understood based on the dependence of the magnetostriction on the Akulov model [17].

The modelling of Akulov approximation has been used in the past to estimate the saturation magnetostriction λ_s of polycrystalline materials, which assumes no grain interaction and is equivalent to an average magnetostriction. As given in equation of $\lambda_s = 2/5\lambda_{100} + 3/5\lambda_{111}$, λ_s is represented as a combination of the cubic magnetostriction constants. The recent study of Clark's results have indicated that the addition of 3d transition elements of Mn reduce the saturation magnetostriction of λ_{100} , but the saturation magnetostriction

Figure 5. Temperature dependence of electrical resistivity of Fe_{81-x}Mn_xGa₁₉ ($x = 0-16$).

λ_{111} increases with Mn doping [18]. So the increase in the magnetostriction resulting from Mn doping in $x < 8$ can be attributed to the increased magnetostriction in the λ_{111} constant, which compensates for the decreased effect of λ_{100} . But with Mn doping more than $x = 8$, the observation means that the decreasing of λ_{100} becomes dominated and thus the saturation magnetostriction λ_s will decrease with the increase in Mn concentration. This further indicates that the Mn composition dependence of λ_{100} and λ_{111} are not the linearity, which results in a non-linear peak relation of λ_s in our Mn-doped samples.

Temperature dependence of the electrical resistivity for Mn-doped Fe–Ga samples was measured in the temperature range from 100 to 300 K, which is shown in figure 5. For all samples, the electrical resistivity exhibits a monotonic increase and shows a metallic behaviour. At room temperature, the electrical resistivity of arc-melted Fe₈₁Ga₁₉ is $88 \mu\Omega \text{ cm}$, which is basically in the same order of magnitude as Terfenol-D [7–9]. The resistivity rises to $159 \mu\Omega \text{ cm}$ for the $x = 8$ and $211 \mu\Omega \text{ cm}$ for the $x = 16$ sample, respectively, which is 81% and 140% larger than Fe₈₁Ga₁₉ alloy. The variation of resistivity at room temperature for Fe_{81-x}Mn_xGa₁₉ with x is shown in figure 2(c).

Apparently, Mn addition is responsible for the increase in the electrical resistivity in our FeMnGa alloys. Intuitively, the random distributions of Mn atoms act as scattering centres for conduction electrons, and the electronic structure variation due to the addition of Mn with fewer conduction electrons than Fe is also considered to affect the electrical resistivity. Therefore, the electrical resistivity shows a positive composition dependence on Mn. The physical mechanism should be complex and detailed work for that is required in the future.

Increasing ρ is good for reducing the eddy current in the magnetic materials. Furthermore, achieving this by doping Mn, we also found the effective increase in the magnetostriction of Fe–Ga alloys in quite a large range of Mn concentration. Our work suggests that Mn doped Fe–Ga alloys may become an attractive candidate material for practical applications.

4. Conclusion

In conclusion, the effects of Mn substitution for Fe on the magnetostriction and electrical resistivity of $\text{Fe}_{81}\text{Ga}_{19}$ have been investigated. An obvious increase in electrical resistivity, up to 140%, is achieved for the $\text{Fe}_{81-x}\text{Mn}_x\text{Ga}_{19}$ ($x = 0-16$) alloys without a great magnetostriction deterioration. We found that the substitution of Fe by Mn in the concentration of $x = 0-8$ not only increases the electrical resistivity but also enhances the magnetostriction, about 11% larger than undoped alloys. It may make the Fe–Ga alloys become very attractive candidate materials for magnetic actuator applications. The related physics is complex and require a further fundamental investigation.

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