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Effect of interstitial additions on magnetostriction in Fe–Ga alloys

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Abstract

The additions of trace amounts of small interstitial atoms (carbon, boron, and nitrogen) to Fe–Ga (Galfenol) alloys have a small but beneficial effect on the magnetostriction of Fe–Ga alloys especially at high Ga compositions. The saturated magnetostrictions $[(3/2)\lambda_{100}\text{'s}]$ of both slow cooled and quenched single crystal Fe–Ga–C alloys with Ga contents >18 at. % are about 10%–30% higher than those of the comparable binary Fe–Ga alloys. For boron and nitrogen additions, the magnetostrictions of slow cooled alloys with Ga content >18 at. % were approximately 20% higher than those of the binary Fe–Ga alloys. We assume that these small atoms enter interstitially into the octahedral site as in pure α-Fe and inhibit chemical ordering, resulting in increased λ_{100}. Thermal analysis of the Fe–Ga binary alloys and Fe–Ga–C ternary alloys indicates that the addition of C into the Fe–Ga system decreases the formation kinetics of $D0_3$ and extends the disordered region beyond the maximum for slow cooled binary samples.

Keywords

IPRT, gallium alloys, interstitials, iron alloys, magnetostriction, thermal analysis

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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The additions of trace amounts of small interstitial atoms (carbon, boron, and nitrogen) to Fe–Ga (Galfenol) alloys have a small but beneficial effect on the magnetostriction of Fe–Ga alloys especially at high Ga compositions. The saturated magnetostrictions [$(3/2)\lambda_{100}$'s] of both slow cooled and quenched single crystal Fe–Ga–C alloys with Ga contents >18 at. % are about 10%–30% higher than those of the comparable binary Fe–Ga alloys. For boron and nitrogen additions, the magnetostrictions of slow cooled alloys with Ga content >18 at. % were approximately 20% higher than those of the binary Fe–Ga alloys. We assume that these small atoms enter interstitially into the octahedral site as in pure α-Fe and inhibit chemical ordering, resulting in increased λ_{100}. Thermal analysis of the Fe–Ga binary alloys and Fe–Ga–C ternary alloys indicates that the addition of C into the Fe–Ga system decreases the formation kinetics of $D0_3$ and extends the disordered region beyond the maximum for slow cooled binary samples.
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INTRODUCTION

Clark and co-workers[1–6] have discovered that Fe–Ga (Galfenol) alloys possess the highest single crystalline magnetostrictive strain for a binary alloy (~13 times that of pure Fe) and unlike other magnetostrictive alloys retain significant ductility. Our previous study[7] indicates that additions of 2 at. % or greater transition elements reduce the saturation magnetostriction of single crystals Fe–Ga samples. The decrease in the magnetostriction in ternary Fe–Ga alloys containing transition elements is due to the increased stability of the $D0_3$ structure.[8] In contrast, additions of small amounts of C (0.07, 0.08, and 0.14 at. %) significantly increased the magnetostriction of the Fe–Ga alloys in the slow cooled condition implying that small atoms such as C, B, and N, when added into the Fe–Ga alloy, may have an opposite effect on the magnetostriction from the transition metals.

Previous studies of B additions in polycrystalline Fe–Ga alloys have found that B can have a beneficial effect on magnetostriction. Gong *et al.*[9] investigated the effect of B addition on the magnetostriction of Fe–Ga alloys with composition of $(Fe_{0.81}Ga_{0.19})_{100-x}B_x$. Their results showed that, when $x=1$, as-cast Fe–Ga–B alloy has higher magnetostriction than the Fe–Ga binary; $(Fe_{0.81}Ga_{0.19})_{100-x}B_x$ with $x=1$, 5, and 10 have higher magnetostriction than the oil quenched binary alloy $Fe_{81}Ga_{19}$. Kubota and Inoue[10] investigated the magnetostriction of melt-spun amorphous alloy and crystallized alloy with a composition of $(Fe_{0.82}Ga_{0.18})_{85}B_{15}$. Their results indicated that the crystallized Fe–Ga–B alloy had higher magnetostriction (60 ppm) than the melt-spun amorphous alloy ribbon (30 ppm). Moreover, the crystallized polycrystalline $(Fe_{0.82}Ga_{0.18})_{85}B_{15}$ alloy had better response compared with the polycrystalline binary $Fe_{82}Ga_{18}$ alloy. However, in these two studies, the high level of B additions induced the formation of borides and the effect of the boride second phase on the magnetostriction was not investigated.

In this paper, we extend our previous studies of C additions to both higher and lower Ga compositions and examine whether B and N additions have similar effects on the magnetostriction of Fe–Ga single crystals. The solubility limits of C, B, and N in disordered bcc Fe–Ga alloys are unknown although they are not expected to be large since the solubility of these light elements in α-Fe is small. Therefore, in order to avoid the formation of boride, carbide, and precipitates, we limit our additions to trace levels (<500 ppm by weight). In order to further elucidate the role of interstitials, we present thermal analysis data, which suggest that the addition of C suppresses the formation of $D0_3$, thereby effectively extending the disordered bcc region beyond the maximum for slow cooled binary samples.

EXPERIMENT

Appropriate quantities of electrolytic grade iron (99.99% purity) and electronic grade gallium (99.9999% purity) and master alloys of Fe–C, Fe–B and Fe–N were arc-melted several times into buttons under an argon atmosphere. The buttons were then remelted and the alloy drop cast into a copper chill cast mold to ensure compositional homogeneity throughout the ingot. Single crystal growth was done using the Bridgman technique in a resistance furnace as described elsewhere.[1,11] Oriented (001) disks, 6 mm in diameter and

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TABLE I. Composition and magnetostriction of Fe–Ga–C, Fe–Ga–B, and Fe–Ga–N alloys.

Composition	$3/2\ \lambda_{100}$ (ppm) Slow cooled	Quenched
$Fe_{90.14}Ga_{9.7}C_{0.16}$	152	147
$Fe_{87.84}Ga_{11.9}C_{0.16}$	202	204
$Fe_{83.7}Ga_{16.2}C_{0.08}$ [a]	268	279
$Fe_{82.4}Ga_{17.6}C_{0.07}$ [a]	322	298
$Fe_{81.4}Ga_{18.6}C_{0.08}$ [a]	369	334
$Fe_{81.3}Ga_{18.6}C_{0.17}$ [a]	342	369
$Fe_{80.38}Ga_{19.6}C_{0.03}$	311	396
$Fe_{78.92}Ga_{20.9}C_{0.18}$	307	432
$Fe_{77.1}Ga_{22.7}C_{0.20}$	258	317
$Fe_{85.48}Ga_{14.5}B_{0.02}$	247	253
$Fe_{81.22}Ga_{18.7}B_{0.08}$	350	383
$Fe_{84.59}Ga_{15.4}N_{0.01}$	270	266
$Fe_{80.49}Ga_{19.5}N_{0.01}$	334	370

[a]Data published in Ref. 7.

2–3 mm thick, were sectioned from the ingot and marked with the in-plane [100] and [110] directions for strain gage alignment. Strain gages aligned along the [100] direction were attached to each disk. Magnetostriction was measured by rotating the instrumented disk in a 20 kOe magnetic field applied parallel to the face of the disk. The resulting strain was fitted to the lowest order magnetostriction term which follows a $\cos^2\theta$ angular dependence. The next higher order magnetostriction term, which follows a $\cos^4\theta$ angular dependence, was generally less than 10 ppm. Magnetostriction was also measured on the disks after being annealed in a sealed quartz tube at 1000 °C for 4 h and quenched into water.

Composition measurements for the disks were done by energy-dispersive spectrometers (EDSs) in a Jeol 840A scanning electron microscope (SEM) using two phase Fe–Ga alloys with known composition as an internal standard. Carbon and nitrogen content was analyzed using combustion gas analyzer method and boron content was analyzed using the inductively coupled plasma (ICP) technique. Differential scanning calorimetry (DSC) measurements on water quenched samples with a weight of about 50 mg were done on a Perkin-Elmer DSC 7.

RESULTS AND DISCUSSION

The magnetostriction and measured compositions of the C, B, and N doped Fe–Ga alloys are summarized in Table I. These data together with the fitted binary Fe–Ga data are shown in Figs. 1 and 2 for comparison. Figure 1 shows that the magnetostriction for low Ga content (<12 at. %) is consistent with our previous study,[7] i.e., the magnetostrictions of the quenched and the slow cooled ternary alloys with C addition are comparable to those of the binary Fe–Ga alloys and quenching does not improve the magnetostriction over the slow cooled state. For Ga contents >18 at. %, our new data confirm that C additions increase magnetostrictions of the slow cooled Fe–Ga–C over slow cooled Fe–Ga binary alloys up to Ga compositions of 22.7 at. %. Further quenching improves the magnetostriction significantly as shown in Fig. 1 in the ternary alloys, as in the binary alloy.[5] For example, the magnetostriction of $Fe_{80.38}Ga_{19.6}C_{0.03}$ and $Fe_{78.92}Ga_{20.9}C_{0.18}$ alloys increased by quenching from 311 to 396 ppm and from 307 to 432 ppm, respectively. In fact, the magnetostriction of the quenched $Fe_{78.92}Ga_{20.9}C_{0.18}$ (432 ppm) is the largest strain reported for single crystalline Fe-based alloys. The addition of B and N acts in a similar fashion as C additions, as shown in Fig. 2. Noticeably, the magnetostriction of slow cooled Fe–Ga–B alloy with a composition of 18.7 at. % Ga is comparable to the quenched binary alloys with a similar composition and the quenched sample is ~20% higher than the quenched Fe–Ga binary alloys.

The addition of small atoms such as C, B, and N has a considerably different effect than that of transition metals such as Ni, Rh, Co, Cr, Mo, and V.[7,12] The different effect on

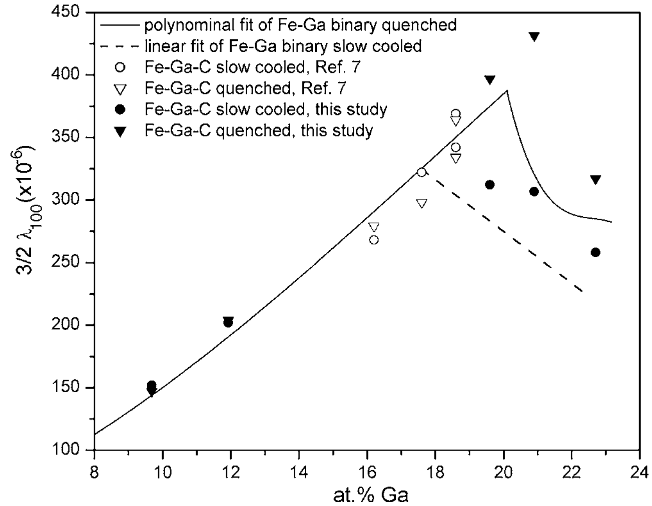

FIG. 1. (3/2) λ_{100} strain measurements as a function of Ga content for Galfenol with C additions together with the fitted binary data for comparison.

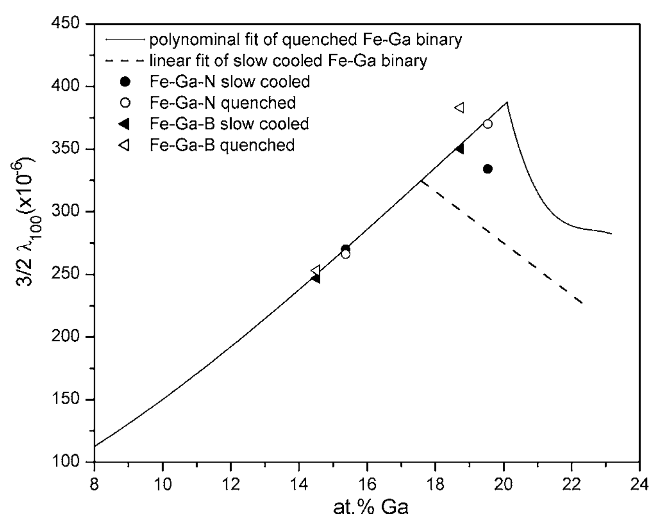

FIG. 2. (3/2) λ_{100} strain measurements as a function of Ga content for Galfenol with B and N additions together with the fitted binary data for comparison.

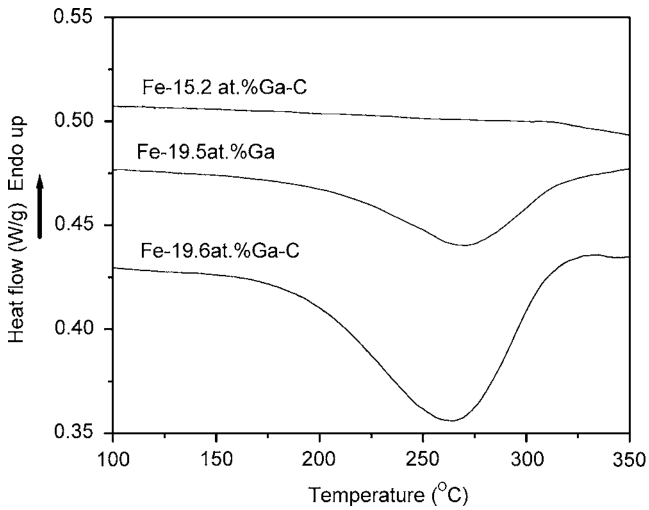

FIG. 3. Results of DSC scans of binary Fe–Ga and Fe–Ga–C quenched alloys with compositions as indicated. The released heats are 0.39 and 1.02 kJ/mol atom for Fe–Ga binary and Fe–Ga–C ternary alloys, respectively.

the magnetostriction of Fe–Ga alloys by the additions of transition metals and interstitial atoms may be due to their occupation site within the crystal lattice. Experimental lattice constant data[13] show that the radius of the octahedral site in Fe–Ga alloys is larger than those in the pure α-iron because of the substitution of Fe by larger Ga, and therefore, it is reasonable to assume that small atoms such as C, B, and N will enter into the octahedral site as they do in pure α-iron.[14–17] Further, interstitial atoms in α-iron are known to produce a tetragonal distortion,[14] and by analogy, Fe–Ga alloys will also be tetragonally distorted. Such tetragonal distortions are thought to contribute to increased magnetostriction.[3] It is reported that interstitial C in FeAl prefers octahedral sites to tetrahedral sites and induces a large tetragonal distortion.[18]

Figure 3 shows the DSC results for Fe-19.5 at. %Ga, Fe-15.2 at. %Ga-C, and Fe-19.6 at. %Ga-C alloys quenched from 1000 °C into water. For Fe-15.2 at. %Ga-C alloy, no exothermal peak was observed during heating indicating no precipitation of $D0_3$ and that this alloy is within the solubility limit of bcc Fe–Ga.[19] However, an exothermal peak is observed at Fe ~ 19.5 at. %Ga with and without C additions. Note that the amounts of released heat are 0.39 kJ/mol for the binary alloy and 1.02 kJ/mol for the Fe–Ga–C ternary alloys. This release of heat has been directly associated with the formation of $D0_3$.[20] The heat released for the Fe–Ga–C ternary alloy is more than double that of the binary alloy (see Fig. 3), indicating that a larger quantity of $D0_3$ phase has precipitated and therefore implying that C additions inhibit the formation of $D0_3$ phase in ternary alloys. More detailed phase analysis using transmission electron microscopy is currently underway to verify the phase distribution indicated by our thermal analysis results.

SUMMARY

We have investigated the effect of ternary additions of trace amounts of small atoms of C, B, and N on the magnetostriction of Fe–Ga alloys. When Ga > 18 at. %, the saturation magnetostriction $[(3/2)\lambda_{100}]$ of slow cooled Fe–Ga–X (X=C, B, and N) alloys are much larger than those of the comparable binary Fe–Ga alloys. The saturation magnetostriction $[(3/2)\lambda_{100}]$ of quenched Fe–Ga–X (X=C, B, and N) are slightly larger than those of the comparable binary Fe–Ga alloys; for furnace-cooled binary and ternary alloys with Ga < 18%, no significant change in $(3/2)\lambda_{100}$ was observed between the slow cooled and quenched conditions. DSC results indicate that the interstitial atoms of C, B, and N can inhibit the undesirable $D0_3$ ordering by slow kinetics, apparently causing the increase of the magnetostriction observed.

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[1] A. E. Clark, J. B. Restorff, M. Wun-Fogle, T. A. Lograsso, and D. L. Schlagel, IEEE Trans. Magn. **36**, 3238 (2000).
[2] G. Petculescu, K. B. Hathaway, T. A. Lograsso, M. Wun-Fogle, and A. E. Clark, J. Appl. Phys. **97**, 10M315 (2005).
[3] J. R. Cullen, A. E. Clark, M. Wun-Fogle, J. B. Restorff, and T. A. Lograsso, J. Magn. Magn. Mater. **226–230**, 948 (2001).
[4] T. A. Lograsso, A. R. Ross, D. L. Schlagel, A. E. Clark, and M. Wun-Fogle, J. Alloys Compd. **350**, 95 (2003).
[5] A. E. Clark, K. B. Hathaway, M. Wun-Fogle, J. B. Restorff, T. A. Lograsso, V. M. Keppens, G. Petculescu, and R. A. Taylor, J. Appl. Phys. **93**, 8621 (2003).
[6] A. E. Clark, M. Wun-Fogle, J. B. Restorff, T. A. Lograsso, and J. R. Cullen, IEEE Trans. Magn. **37**, 2678 (2001).
[7] A. E. Clark, J. B. Restorff, M. Wun-Fogle, K. B. Hathaway, T. A. Lograsso, M. Huang, and E. Summers, J. Appl. Phys. **101**, 09C507 (2007).
[8] N. Kawamiya and K. Adachi, J. Magn. Magn. Mater. **31–34**, 145 (1983).
[9] Y. Gong, C. Jiang, and H. Xu, Acta Metall. Sin. **42**, 830 (2006).
[10] T. Kubota and A. Inoue, Mater. Trans. **45**, 199 (2004).
[11] Single crystals synthesisized at the Materials Preparation Center, Ames Laboratory US DOE, Ames, IA (www.mpc.ameslab.gov).
[12] J. B. Restorff, M. Wun-Fogle, A. E. Clark, T. A. Lograsso, A. R. Ross, and D. L. Schlagel, J. Appl. Phys. **91**, 8225 (2002).
[13] J. M. Borrego, J. S. Blazquez, C. F. Conde, A. Conde, and S. Roth, Intermetallics **15**, 193 (2007).
[14] G. K. Williamson and R. E. Smallman, Acta Crystallogr. **6**, 361 (1953).
[15] R. R. Hashiguchi and G. Kamoshita, J. Phys. Soc. Jpn. **9**, 646 (1954).
[16] W. R. Thomas and G. M. Leak, Nature (London) **176**, 29 (1955).
[17] M. E. Hermant, Acta Metall. **16**, 1 (1968).
[18] B. V. Reddy and S. C. Deevi, Mater. Sci. Eng., A **A329–A331**, 395 (2002).
[19] O. Ikeda, R. Kainuma, I. Ohnuma, K. Fukamichi, and K. Ishida, J. Alloys Compd. **347**, 198 (2002).
[20] S. Datta, M. Huang, J. Raim, T. A. Lograsso, and A. B. Flatau, Mater. Sci. Eng., A **A435–A436**, 221 (2006).