Magnetostrictive Properties of Galfenol Alloys Under Compressive Stress

Arthur E. Clark¹, Marilyn Wun-Fogle², James B. Restorff² and Thomas A. Lograsso³

¹Clark Associates, Adelphi, MD 20783, USA
²Naval Surface Warfare Center, Carderock Division, Code 645, W. Bethesda, MD 20817, USA
³Ames Laboratory, Ames, IA 50011, USA

Fe–Ga alloys, in which the α-Fe structure is maintained, are rich sources of high strength, low cost magnetostrictive alloys for transducer and vibration reduction applications. Although the magnetostriction of Fe itself is very low, when a relatively small fraction of the Fe atoms are replaced by Ga, the magnetostriction, \( \lambda \), increases greatly. Until recently, the highest magnetostriction was found with the replacement of Fe by Al (Alfenol). In this paper, we present our measurements of magnetostriction on Fe\(_1-x\)Ga\(_x\) alloys at room temperature and the temperature dependence of the saturation magnetizations remain high, \( M_s \approx 1.7 \, \text{T} \), and the Curie temperatures are far above room temperature, \( T_C \approx 700 \, \text{°C} \). In most alloys studied, the magnetostrictions and magnetizations are fully saturated in fields less than 24 kA/m, even under compressive stresses >100 MPa. For \( x = 0.24 \) (near Fe\(_3\)Ga), an anomalous increase in magnetostriction with temperature occurs with a peak magnetostriction above room temperature. Small additions of Ni and Mo to the binary Fe–Ga alloys decrease the room temperature value of \( \lambda_{100} \).

(Received September 20, 2001; Accepted December 6, 2001)

Keywords: magnetostriction, Galfenol, iron-gallium alloys

1. Introduction

It was recently recognized that a substantial increase in the magnetostriction, \((3/2)\lambda_{100}\), of Fe occurs with the substitution of small amounts of Ga for Fe.\(^1,2\) This is true as long as the bcc α-Fe phase is maintained.\(^3\) This phase is not the equilibrium phase in Fe\(_1-x\)Ga\(_x\) at room temperature for \( x > 0.15 \).\(^4\) By rapid quenching into water from temperatures >800°C, the disordered α-Fe structure can be extended to larger values of \( x \) and the magnetostriction further increased.\(^5\) The phenomenal increase in Fe magnetostriction with Ga is remarkable, since Ga is non-magnetic.

This paper is divided into two parts. In the first part we present: (1) the field dependence of the magnetostriction, \((3/2)\lambda_{100}\), of Fe\(_{0.81}\)Ga\(_{0.19}\) under high compressive stresses at room temperature and (2) the temperature dependence of \((3/2)\lambda_{100}\) from -269°C to 42°C. Magnetostriction measurements show that quenching from 800°C and 1000°C increases the magnetostriction over 40% to values ~400 ppm at room temperature and ~420 ppm at -269°C. This slight increase in magnetostriction as the temperature is decreased from 315 to ~269°C is consistent with the small increase of magnetization reported over the same temperature range.\(^3,5\) We find for Fe\(_{0.81}\)Ga\(_{0.19}\) that the magnetostriction has a well-behaved temperature dependence and an intrinsic magnetostriction that is very large. For Fe\(_{0.76}\)Ga\(_{0.24}\) (near Fe\(_3\)Ga), this is no longer true. In the second part of the paper we report the effect of small amounts of Ni and Mo on the saturation magnetostriction constants, \( \lambda_{100} \) and \( \lambda_{111} \), of the Fe–Ga alloys. In all cases the large positive magnetostriction constant, \( \lambda_{100} \), decreases in value.

2. Sample Preparation

To prepare single crystal samples of Fe\(_{1-x}\)Ga\(_x\), as-cast ingots containing Ga (99.999% pure) and Fe (99.99% pure) were inserted into alumina crucibles and heated to 1650°C. The ingot/crucible was stabilized for 1 h and then withdrawn at a rate of ~2 mm/h. Following crystal growth, the alloys were annealed at 1000°C for 168 h and furnace cooled at a rate of 10°C/min. The large single crystals were oriented and rods (~2.5 cm x 0.6 cm dia.) and discs (~0.3 cm x 0.6 cm dia.) of the proper crystalline directions were extracted. Samples were examined for homogeneity and Fe/Ga ratio. Initial magnetostriction and magnetization measurements were made on these furnace-cooled alloys. Following these measurements, some alloys were then reheated to 800°C and 1000°C in evacuated quartz tubes for 1 h and finally rapidly cooled by quenching into a water bath.

3. Experimental Methods

A conventional dead-weight apparatus was used to apply compressive loads to the Fe\(_1-x\)Ga\(_x\) alloys at room temperature.\(^6\) Magnetic fields up to 80 kA/m were applied to the samples from a solenoid energized by a constant current source. Magnetic hysteresis loops were calculated from the emfs generated by a small pick-up coil surrounding the center of the samples. Displacements as a function of magnetic field at compressive stresses up to 120 MPa were determined from the output of three linear variable differential transformers (LVDT’s). See Fig. 1. In order to obtain the intrinsic \( (T=0) \) values of the saturation magnetostriction, measurements were made in high fields from room temperature to ~269°C on oriented single crystals. Disc samples of the appropriate alloys were affixed with special low temperature non-magnetoresistive strain gages (Kyowa K-19-1S1). (The temperature dependence of the gage factors was taken from Gersdorf.\(^7\)) The small disc samples were mounted on a rotating fixture and inserted into a liquid He cryostat positioned between the poles of a large electromagnet. Strains along the [100] direction were measured as a function of an-
4. Magnetostriiction of Binary Fe$_{1-x}$Ga$_x$ Alloys

To date, the largest magnetostriiction is found in a sample of Fe$_{0.81}$Ga$_{0.19}$ rapidly quenched into water from 800°C. Figure 3 illustrates the dependence of magnetostriiction, $(3/2)\lambda_{100}$, on Ga concentration. For samples with $x = 0.17$, the $\alpha$-Fe phase is near equilibrium at room temperature, the magnetostriiction is about 300 ppm, and the effect of cooling rate on the sample is minimal. For $x = 0.24$, the magnetostriiction is about 270 ppm, and again the effect of cooling rate on the magnetostriiction is very small. However, between these values of $x$, a large cooling-rate dependent magnetostriiction peak appears. For $x = 0.19$, and $x = 0.21$, rapid quenching from 800°C greatly improves the magnetostriiction over furnace-cooled alloys. For $x = 0.19$, the improvement is $\sim40\%$. The room temperature magnetostriiction of quenched (disordered) Fe$_{1-x}$Ga$_x$ $(x \sim 0.19)$ exceeds that of all magnetostriictive 3d transition metals, such as Co, permendur, and Alfenol. This is fascinating since Ga is non-magnetic, i.e. the entire magnetostriiction arises from Fe in the diluted $\alpha$-Fe structure. Because of the sharp non-linear increase in magnetostriiction above that of Fe in the disordered $\alpha$-Fe–Ga alloys it is believed that the magnetostriiction is not due to conventional magnetoelastic effects but due to the onset of short range order and the presence of asymmetric clusters of Ga atoms along [100] directions in the $\alpha$-Fe structure. 3)

For very low values of $x$ there are few clusters. For large values of $x$ near 0.25, it becomes difficult if not impossible to form the disordered bcc structure since the alloys greatly prefer the ordered DO$_3$ or B2 states. Very significantly the measurements of $\lambda_{111}$ for Fe$_{1-x}$Ga$_x$ and Fe$_{1-x}$Al$_x$ show little or no change with $x$, in contrast to the great change in $\lambda_{100}$. 3, 5)

To determine the intrinsic value of the magnetostriiction in the Fe–Ga alloys, measurements of $\lambda_{100}$ were made from room temperature to near absolute zero. (Note that the temperature dependence of $\lambda_{100}$ of pure Fe is unusual, possessing two magnetostriiction peaks, a small one between $\sim173^\circ$C and $27^\circ$C and a larger one near the Curie temperature. 9) The magnetostriiction of Fe$_{1-x}$Ga$_x$, $0 < x < 0.20$, was found to be simple and decreases only a few percent with temperature from $-268^\circ$C to $27^\circ$C. 3, 5) The satisfying result of a similar small decrease in the magnetostriiction over the same temperature range is illustrated in Fig. 4. Here the magnetostriictions, $(3/2)\lambda_{100}$, of bcc Fe and rapidly cooled Fe$_{0.81}$Ga$_{0.19}$ are compared. Note the expanded scales. The large value of $(3/2)\lambda_{100}$ is intrinsic and not the result of an anomalous temperature dependence. Angular dependences of the strains of Fe$_{0.81}$Ga$_{0.19}$ at $-269^\circ$C and $22^\circ$C are illustrated in Fig. 5. The curves are excellent fits to $\lambda_{100} \sim 3/2 \cos^2 \theta$ dependence where $\lambda_{100} = (3/2)\lambda_{100}$. It should be pointed out that the satisfying agreement between the magnetization and magnetostriiction temperature dependences was not observed for Fe$_{0.76}$Ga$_{0.24}$. For this composition, while the magnetization still exhibits a small increase with decreasing temperature from $27^\circ$C to $-269^\circ$C, the magnetostriiction loses nearly half of its value over the same range. See Fig. 6. Both furnace-cooled and rapidly quenched samples were tested and revealed the same temperature dependence. This anomalous behavior, which
yields a peak in magnetostriction above room temperature, is not understood. The normal temperature dependence for the magnetostriction was again observed for the larger Ga concentration of $x = 35\%$.\(^{10}\)

Because of the large magnetization ($\sim 1.7\ T$) of these alloys, the magnetic fields required to achieve the large magnetostriction, even under large compressive stresses are small. Figures 7 and 8 illustrate the dependence of the magnetostriction vs. magnetic field for various compressive stresses up to $\sim 95\ MPa$ for furnace cooled Fe\(_{0.83}\)Ga\(_{0.17}\) and quenched Fe\(_{0.83}\)Ga\(_{0.19}\) alloys, respectively. The 17% alloy saturates at a slightly lower magnetic field and has a lower saturation magnetostriiction. The magnetostriction increases by 18% as the $\alpha$-Fe structure is extended from 17% to 19% Ga. The saturation magnetization for the 19% sample is slightly smaller than the 1.75 T value for the 17% sample. However in all cases, fields less than 32 kA/m are required to effectively achieve saturation magnetostriction at stresses up to $\sim 100\ MPa$. The effect of rapid quenching in obtaining the large strains in Fe\(_{0.83}\)Ga\(_{0.19}\) is shown in Fig. 9. In this figure are compared magnetostriction curves vs field for stresses of 20 MPa and 50 MPa before and after quenching. A remarkable 30% increase in magnetostriction is observed for $H \geq 32\ kA/m$.

5. Magnetostriction of Fe–Ga–Ni and Fe–Ga–Mo Alloys

In this section we explore the effect of small amounts of Ni on the magnetostriction of Fe\(_{1-x}\)Ga\(_x\) ($x = 0.11$ and $x = 0.16$) alloys. We have shown earlier that while
\( \lambda_{100} \) increases dramatically with substitutions of Ga into bcc Fe, the small negative value of \( \lambda_{111} \) of bcc Fe remains almost unchanged.\(^{1,2}\) Thus the magnetostriction of the highly magnetostrictive Fe–Ga alloys is very anisotropic; \( \lambda_{100}/\lambda_{111} \approx -10.\) Bozorth (using A. Schulze’s data), has inferred that small percentages of Ni when added to Fe substantially reduces \( \lambda_{111} \) and decreases the absolute magnetostrictive anisotropy.\(^{11}\) In Fig. 10, we compare the magnetostrictions along the [100] direction for Fe\(_{0.86}\)Ga\(_{0.14}\)Ni\(_{0.03}\) and Fe\(_{0.814}\)Ga\(_{0.16}\)Ni\(_{0.026}\) furnace-cooled alloys. For the Fe\(_{0.86}\)Ga\(_{0.14}\)Ni\(_{0.03}\) sample, the decrease in the magnetostriction inferred from the Ni-free sample is severe (\( \sim 40\% \)). For the Fe\(_{0.814}\)Ga\(_{0.16}\)Ni\(_{0.026}\) sample, the decrease is not as large (\( \sim 15\% \)). Our measurements show an increase of \( \lambda_{111} \) from \(-16\) ppm for the Ni-free sample of Fe\(_{0.87}\)Ga\(_{0.13}\) to \( \sim 0 \) for Fe\(_{0.811}\)Ga\(_{0.152}\)Ni\(_{0.0127}\).\(^{12}\) Thus, while the magnitude of \( \lambda_{111} \) is reduced by the addition of Ni, \( \lambda_{100} \) is also reduced. The substitution of small amounts of Ni for Fe also reduces the strains available under various compressive stresses. Figure 11 illustrates the field dependence of the magnetostriction Fe\(_{0.86}\)Ga\(_{0.14}\)Ni\(_{0.03}\) under various stresses up to 122 MPa. This figure can be compared to Fig. 7 for the Ni free Fe–Ga alloy.

We have also examined the effect of small Mo substitutions for Fe in the Fe–Ga alloys. Hall has shown that \( \lambda_{100} \) of bcc Fe increases moderately with the addition of 2–4\% Mo.\(^{13}\) Figure 12 compares the magnetostrictions (3/2)\( \lambda_{100} \) and (3/2)\( \lambda_{111} \) for Fe\(_{0.84}\)Ga\(_{0.15}\)Mo\(_{0.03}\) and Fe\(_{0.85}\)Ga\(_{0.10}\)Mo\(_{0.05}\), respectively. Here, while \( \lambda_{100} \) is again slightly reduced, (3/2)\( \lambda_{111} \) increases in negativity to \(-36\) ppm. This increase in the negativity of \( \lambda_{111} \) is unprecedented. Fe–Ga–Mo has a large negative anisotropy of approximately \(-5\) (Note, however, because of the difficulty of synthesizing identical compositions of the [100] and [111] samples, the alloy compositions for the [100] and [111] strains are slightly different.). The values of magnetostriction and magnetization under compressive stresses for Fe\(_{84}\)Ga\(_{11}\)Mo\(_{0.03}\) are illustrated in Fig. 13. The saturation magnetizations are above 1.7 T in all alloys. Compare Figs. 7, 11, and 13.
Magnetostrictive Properties of Galfenol Alloys Under Compressive Stress

Fig. 11 Room temperature magnetic field dependences of (a) magnetostriction along the [100] direction and (b) magnetization of Fe$_{0.83}$Ga$_{0.135}$Ni$_{0.035}$ for various compressive stresses.

Fig. 12 Angular dependence of the room temperature magnetostriction (a) along the [100] direction for Fe$_{0.84}$Ga$_{0.13}$Mo$_{0.03}$ and (b) along the [111] direction for Fe$_{0.85}$Ga$_{0.10}$Mo$_{0.05}$ for $H = 1200$ kA/m. Note: $\lambda_{100} > 0$; $\lambda_{111} < 0$.

6. Summary

Fe$_{1-x}$Ga$_x$, in its simple bcc structure, exhibits magnetostrictions, $(3/2)\lambda_{100}$’s, as high as 395 ppm at room temperature, much larger than common Fe and all other known 3d transition metal alloys. Rapid quenching increases the solubility of Ga in bcc Fe and thus the magnetostriction increases with increasing $x$ for $0.17 < x \leq 0.19$. Our observed maximum magnetostriction occurs in samples of $\sim 19\%$ Ga in Fe that were rapidly quenched into water from 800$^\circ$C. For $x \leq 0.19$, we observed a normal (small) decrease in magnetostriction with temperature from $-269^\circ$C to 22$^\circ$C. On the other hand, for both quenched and furnace-cooled samples of $x = 0.24$ (near Fe$_{2}$Ga), we find an anomalous increase in magnetostriction over the same temperature range. Thus, a peak in the magnetostriction occurs above room temperature. This is not consistent with the characteristic normal decrease in magnetization with temperature. $\lambda_{111}$ remains negative at room temperature in all binary and ternary alloys reported to date. The addition of small amounts of Mo to the Fe–Ga alloy increases the magnitude of $\lambda_{111}$ while the addition of Ni decreases the magnitude of $\lambda_{111}$ to near zero.

7. Acknowledgments

This work was supported by the U.S. Office of Naval Research, the Carderock Division of the Naval Surface Warfare Center’s In-house Laboratory Independent Research Program sponsored by the Office of Naval Research administered under Program Element 0601152N, and the Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. W-7405-ENG-82.

REFERENCES


