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# Magnetostriction of iron-germanium single crystals

D. Wu

*Iowa State University*

Qingfeng Xing

*Iowa State University, qxing@ameslab.gov*

R. William McCallum

*Iowa State University, mccallum@ameslab.gov*

Thomas A. Lograsso

*Iowa State University, lograsso@ameslab.gov*

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# Magnetostriction of iron-germanium single crystals

## Abstract

The addition of nonmagnetic Ga into body-centered cubic Fe enhances the magnetostriction constant  $\lambda_{100}$  over tenfold. Literature reports for substitution of Ge at low concentrations suggest that the addition of Ge also enhances the magnetostriction. In this work, the magnetostriction and microstructure of Fe–Ge were investigated to correlate magnetostriction with microstructure. The magnetostriction of  $\text{Fe}_{100-x}\text{Ge}_x$  single crystals with  $x$  between 0.05 and 0.18 varies with Ge concentration and correlates with phase changes. The value of  $(3/2)\lambda_{100}$  increases with Ge additions in the A2 single phase region (up to  $x \sim 10$ ), reaching a maximum of 94 ppm at the solubility limit of the disordered A2 phase. Further increases in Ge in the A2+D0<sub>3</sub> two-phase region ( $12 < x < 16$ ) result in a decrease in magnetostriction which changes from positive to negative. For Ge contents with  $x > 16$ , magnetostriction remains negative with an absolute value of strain of 129 ppm at 18 at. % Ge. This behavior is similar to that observed for Fe–Si alloys.

## Keywords

crystal microstructure, germanium alloys, iron alloys, magnetostriction, Iron-Germanium, Single Crystal, Magnetostriction

## Disciplines

Condensed Matter Physics | Metallurgy

## Comments

The following article appeared in *Journal of Applied Physics* 103 (2008): 07B307 and may be found at <http://dx.doi.org/10.1063/1.2829393>.



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## Magnetostriction of iron-germanium single crystals

D. Wu,<sup>a)</sup> Q. Xing, R. W. McCallum, and T. A. Lograsso  
*Materials and Engineering Physics Program, Ames Laboratory, Iowa State University,  
 Ames, Iowa 50011-3020, USA*

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The addition of nonmagnetic Ga into body-centered cubic Fe enhances the magnetostriction constant  $\lambda_{100}$  over tenfold. Literature reports for substitution of Ge at low concentrations suggest that the addition of Ge also enhances the magnetostriction. In this work, the magnetostriction and microstructure of Fe–Ge were investigated to correlate magnetostriction with microstructure. The magnetostriction of  $\text{Fe}_{100-x}\text{Ge}_x$  single crystals with  $x$  between 0.05 and 0.18 varies with Ge concentration and correlates with phase changes. The value of  $(3/2)\lambda_{100}$  increases with Ge additions in the A2 single phase region (up to  $x \sim 10$ ), reaching a maximum of 94 ppm at the solubility limit of the disordered A2 phase. Further increases in Ge in the A2+D0<sub>3</sub> two-phase region ( $12 < x < 16$ ) result in a decrease in magnetostriction which changes from positive to negative. For Ge contents with  $x > 16$ , magnetostriction remains negative with an absolute value of strain of 129 ppm at 18 at. % Ge. This behavior is similar to that observed for Fe–Si alloys. © 2008 American Institute of Physics. Iron-Germanium Single Crystal Magnetostriction [DOI: 10.1063/1.2829393]

### INTRODUCTION

As one of the ductile and low-cost Fe-based magnetostrictive alloys, Fe–Ga alloy has been studied extensively during the past few years.<sup>1–5</sup> It has been found that the addition of nonmagnetic Ga into body-centered cubic Fe significantly enhances the magnetostriction to a maximum of 398 ppm at Ga content near 19 at. % and 353 ppm near 27 at. % Ga.<sup>1</sup> The role that Ga plays in Fe–Ga alloys still remains open, even though extensive efforts<sup>3,6–9</sup> have been made to explain the phenomenon. In order to gain insight into this enhanced magnetostriction, it is worth exploring other elemental additions which can also improve the magnetostriction of iron. Results by Hall<sup>10</sup> for Ge concentrations lower than 6 at. % showed that the addition of Ge also enhances the magnetostriction of iron. However, the magnetostriction was not investigated at higher concentrations where peak values are found in Fe–Ga. Since the maximum strains are observed near the solubility limit for Ga in bcc  $\alpha$ -Fe, the region near solubility limit of Ge should also be of particular interest. In this paper, we report on the single crystalline magnetostriction of  $\text{Fe}_{100-x}\text{Ge}_x$  and its dependence on Ge concentration for  $x$  between 0 and 18. It was found that the changes of the magnetostriction,  $(3/2)\lambda_{100}$ , with Ge additions are correlated with the structural changes in Fe–Ge alloys.

### EXPERIMENT

Appropriate amounts of iron and germanium were arc melted several times under an argon atmosphere to form buttons. The buttons were remelted and then dropped into a copper cast mold chilled by water to ensure compositional homogeneity throughout the ingot. Fe–Ge single crystals

were grown from the as-cast ingot by Bridgman method in alumina crucibles. Details of the Bridgman growth are described elsewhere.<sup>1</sup> Disks with a diameter of 6.35 mm and a thickness of 2.5 mm were sectioned from the ingot and were oriented with surface plane parallel to Fe (100) plane. Magnetization measurements in the plane of the disk showed that the magnetization along the major crystallographic axis in the plane of the disk saturated at fields well below 20 kOe with the magnetization at 20 kOe having a value of 0.995, the value at 50 kOe. A strain gauge was glued on one side of the disk, parallel to the [100] direction. Magnetostriction was measured in a 20 kOe saturating magnetic field applied in the (100) disk plane by rotating the sample around the disk axis. Details of the strain measurements are given in Ref. 3.

The microstructure was examined by transmission electron microscopy (TEM) in order to correlate the magnetostriction with structure. TEM foils were electropolished and in some cases, then ion milled for 15 min. A Tecnai F20XT TEM and a Philips CM30 TEM were employed at a nominal operation voltage of 200 kV. Sample composition was determined by energy-dispersion spectroscopy (EDS) available on a scanning electron microscope (SEM) using standards for calibration.

### RESULTS AND DISCUSSION

The magnetostriction constant,  $(3/2)\lambda_{100}$ , was extracted by fitting the strain gauge data with a simple angular expression,  $\lambda_{100} \cos^2(\theta + \alpha) + \lambda_{100} \cos^4(\theta + \alpha) + c$ , where  $\alpha$  is the phase angle and  $c$  is a constant which has contributions from both the magnetostriction and the balance point of the strain gauge bridge. Figure 1 illustrates the change in differential strain data, the angle dependent portion of the experimental strain gauge data, for Fe–10.6 at. % Ge and Fe–17.7 at. % Ge single crystals as a function of the angle  $\theta$  between [100] direction and the direction of the applied magnetic field. If  $\alpha$

<sup>a)</sup>Electronic mail: wudm@ameslab.gov.

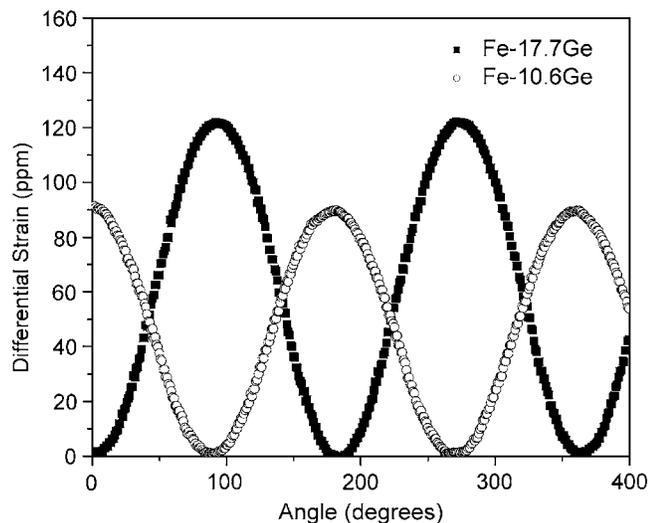


FIG. 1. Measured differential strain of Fe–10.6 at. % Ge and Fe–17.7 at. % Ge single crystals along the [100] direction vs the angle between the [100] direction and applied magnetic field direction, showing the differences in magnitude and the signs of the magnetoelastic constant values between the two alloys.

is zero, the maximum strain along [100] occurs parallel to the axis and the magnetostriction is considered to be positive. On the other hand, if  $\alpha = \pi/2$ , the maximum strain occurs for the field perpendicular to [100] and the magnetostriction is negative. In the case of the Fe–Ge samples, the contribution of the higher order term was generally less than 3 ppm. The  $\pi/2$  phase shift between the two data sets clearly shows a change in sign of the magnetostriction.

The value of  $(3/2)\lambda_{100}$  as a function of Ge concentration is presented in Fig. 2. It increases slowly with Ge additions up to  $\sim 10$  at. % Ge with a value of 94 ppm, and then decreases rapidly with further Ge additions up to  $\sim 18$  at. % Ge. Interestingly, the magnetostriction changes from positive to negative at  $\sim 14$  at. % Ge. For Ge contents  $> 16$  at. %, the magnetostriction continues to decrease reaching  $-129$  ppm at Fe–17.7 at. % Ge.

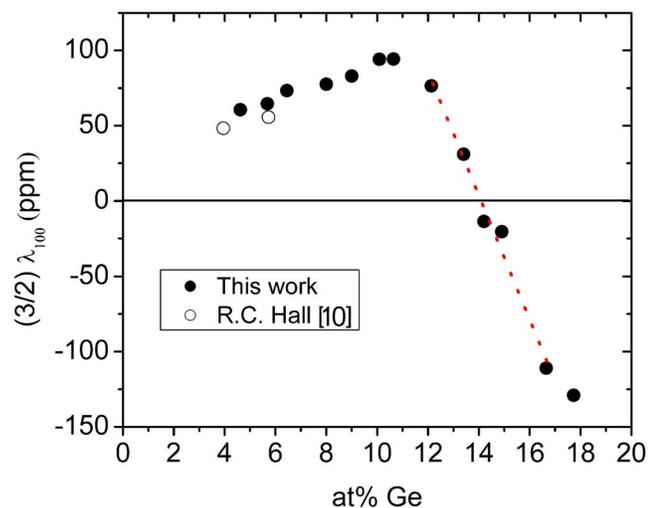


FIG. 2. (Color online) Magnetostriction constant,  $(3/2)\lambda_{100}$ , of Fe–Ge single crystals as a function of Ge composition in the slow-cooled state. The data between 12 and 17 at. % Ge is linearly fitted, as shown by the inserted dotted line. Data from Hall's work are shown as open circles for comparison.

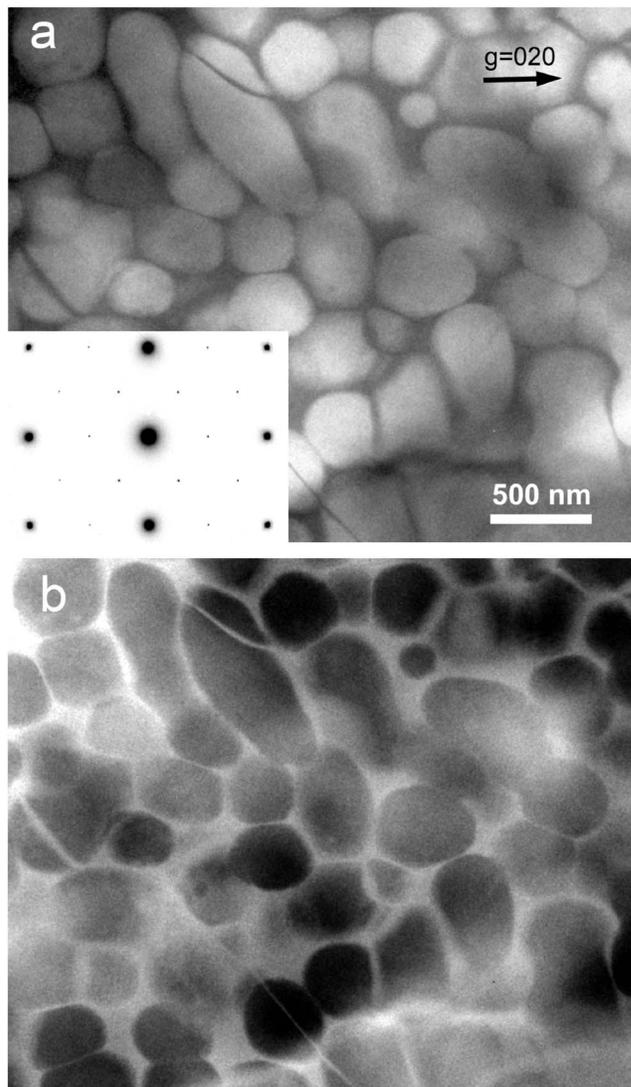


FIG. 3. TEM results of Fe–14.2 at. % Ge used to determine the presence of A2/D<sub>03</sub> mixture: (a) dark-field image under two-beam condition with a (020) superlattice reflection strongly excited, showing mixture of A2 phase (dark) and D<sub>03</sub> phase (bright), (b) contrast-inversed image of (a) clearly showing that D<sub>03</sub> phase (dark) dispersed in the A2 matrix (bright). The [011] zone-axis diffraction pattern indicating the presence of D<sub>03</sub> phase by the superlattice reflections (weak spots) is inserted in (a).

Structural investigations reveal that the changes in magnetostriction correlate with the phase changes in the Fe–Ge system. TEM characterization shows a single phase of A2 structure up to 10 at. % Ge, a mixture of two phases between 12 and 16 at. % Ge, and a single phase of D<sub>03</sub> structure when Ge concentration is higher than 16 at. % in general agreement with the assessed phase diagram.<sup>11</sup> Structural identification was accomplished by selected-area and nanobeam diffraction and, in particular, inspection for existence of superlattice reflections corresponding to the chemically ordered phases. For Fe–10 at. % Ge, no superlattice reflections was observed on the [011] zone-axis diffraction pattern. This reveals that the alloy is single phase A2 (disordered  $\alpha$ -Fe). Between 12 and 16 at. % Ge, the [011] diffraction pattern shows the presence of D<sub>03</sub> phase, as superlattice reflections of D<sub>03</sub> phase appears [see Fig. 3(a)]. The dark-field imaging [see Figs. 3(a) and 3(b)] and nanobeam diffraction

further indicate that the  $D0_3$  phase is dispersed in the A2 matrix. Note that no B2 structure was observed, even though the Fe–Ge phase diagram<sup>11</sup> suggests the presence of B2 phase between 10 at. % Ge and 16 at. % Ge. Above 16 at. % Ge, superlattice reflections are clearly visible and no A2 phase was found in the dark-field images, which demonstrates that the alloy is a  $D0_3$  single phase.

Lack of B2 phase in the two-phase region does not conflict with the previous literature. In Ref. 12, used for the assessed phase diagram,<sup>11</sup> the authors could not confirm the existence of the B2 phase. The morphology of the two-phase mixture that they observed is similar to what we obtained. However, nanobeam diffraction was used in our work to confirm the presence of the A2 phase, and no superreflections were observed for the phase. A thorough investigation of the phase change with germanium concentration is underway.

## DISCUSSION

The general magnetostrictive behavior of Fe–Ge single crystals shows the same general trend with composition as found in Fe–Ga crystals. The present results clearly show that the magnetostriction of Fe–Ge alloys is structurally dependent. Alloy compositions falling within single phase regions show a monotonic increase in absolute magnetostriction with Ge addition for both A2 and  $D0_3$  phases. The A2 phase shows increasing positive values while  $D0_3$  shows increasing negative values. The present results clearly demonstrate that not only the magnitude but also the sign of magnetostriction are strongly dependent on local atomic environment.

Within the two-phase region ( $\sim 12$ – $16$  at. % Ge), the measured magnetostriction of A2+ $D0_3$  mixture is determined by the competition between the expansion/contraction of the two phases. This competition is dependent on both the relative volume fractions of  $D0_3$  phase as well as the relative elastic properties of the two phases. Assuming that the difference in elastic properties is negligible (as the composition difference between the phases is small), then one can use a simple rule of mixture to describe the two-phase behavior. As the  $D0_3$  phase shows negative magnetostriction, the increase of volume fraction of  $D0_3$  will naturally lead to a linear decrease in the measured  $(3/2)\lambda_{100}$ . Fitting the data to a least square line (dotted line) between 12 and 16 at. % Ge, as shown in Fig. 2, suggest behavior consistent with a rule of mixture relationship.

The magnetostrictive behavior of Fe–Ge alloys is similar to that reported for Fe–Si alloys<sup>13</sup> where the behavior was explained by effect of ordering. This similarity is not entirely unexpected, as Si and Ge lie adjacent to each other in the same column of the periodic table and have same electronic

configuration, which is believed to contribute to magnetostriction.<sup>6</sup> The fact that the magnetostriction has changed from positive to negative as Fe–Ge alloys as they undergo chemical ordering also reflects the importance of local atomic arrangement in determining the magnetoelastic interactions in Fe-based alloys. Such a sign reversal between disordered bcc Fe (A2) and ordered  $D0_3$  has been predicted by first principle calculations for Fe–Ga.<sup>6</sup>

## SUMMARY

The magnetostriction of Fe–Ge single crystals with various Ge concentrations in the range of 0–18 at. % was measured. In the composition range where the A2 phase is present ( $< 10$  at. % Ge), it was found that the magnetostriction was positive and increased in magnitude with increasing Ge concentration. In contrast, the  $D0_3$  phase at higher Ge concentration (16–18 at. % Ge) shows negative magnetostriction which also increased in magnitude with increasing Ge concentration. Within the two-phase A2+ $D0_3$  region (10–16 at. % Ge), the magnetostriction decreased linearly between the positive and negative values of the terminal phases suggesting that a simple rule of mixtures can be used to account for the transition in both sign and magnitude of magnetostriction.

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