

Extraordinary magnetoelasticity and lattice softening in bcc Fe-Ga alloys

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Extraordinary magnetostrictive behavior has been observed in Fe-Ga alloys with concentrations of Ga between 4% and 27%. λ_{100} exhibits two peaks as a function of Ga content. At room temperature, λ_{100} reaches a maximum of 265 ppm near 19% Ga and 235 ppm near 27% Ga. For compositions between 19% and 27%, λ_{100} drops sharply to a minimum near 24% Ga and exhibits an anomalous temperature dependence, decreasing by as much as a factor of 2 at low temperatures. This unusual magnetostrictive behavior is interpreted on the basis of a single maximum in the magnetoelastic coupling $|b_1|$ of Fe with increasing amounts of nonmagnetic Ga, combined with a strongly temperature dependent elastic shear modulus ($c_{11} - c_{12}$) which approaches zero near 27% Ga. λ_{111} is significantly smaller in magnitude than λ_{100} over this composition range, and has an abrupt change in sign from negative for low Ga concentrations to positive for a concentration of Ga near 21%. © 2003 American Institute of Physics. [DOI: 10.1063/1.1540130]

INTRODUCTION

It was recently pointed out that Ga, when substituted for Fe in the common α -Fe structure, increases the tetragonal magnetostriction λ_{100} over tenfold.¹ This is reminiscent of the earlier results by Hall, who observed that, similarly, the addition of Al to Fe enhanced λ_{100} fourfold over the same concentration range.² Both Ga and Al have large solubility ranges in Fe and have an inclination to retain local bcc-like symmetry, both in the disordered alloy and in the $B2$ and $D0_3$ ordered structures. It has been shown that rapid cooling from the large high temperature region of solubility of Ga into Fe retains the disordered bcc structure at room temperature for samples with less than 20% Ga.³ For our samples treated in this way the magnetostriction increased still further.⁴ Cullen *et al.* proposed a model for the increases in magnetostriction for Fe-Ga and Fe-Al alloys as arising from the pairing of Ga atoms along $\langle 100 \rangle$ axes.⁵

In this article, we report magnetostriction and elastic constant measurements on $\text{Fe}_{100-x}\text{Ga}_x$ ($4 < x < 27$) prepared by furnace cooling ($\sim 10^\circ\text{C}/\text{min}$) and by rapid quenching into water at room temperature. Our results show a double peak in the magnetostriction (λ_{100}) versus Ga concentration

at both cryogenic and room temperatures, with unorthodox temperature dependences of the magnetostrictions in the region between the peaks. The unusual double peak dependence of magnetostriction on Ga concentration is interpreted on the basis of (1) a magnetoelastic energy $|b_1|$ that increases rapidly for small concentrations up to $\sim 19\%$ Ga, plus (2) a near-linear softening of the shear elastic constant extending at least to 27% Ga.

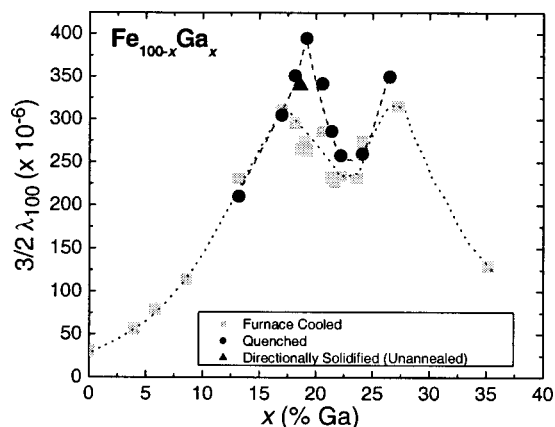


FIG. 1. $(3/2)\lambda_{100}$ as a function of Ga concentration for $\text{Fe}_{100-x}\text{Ga}_x$.

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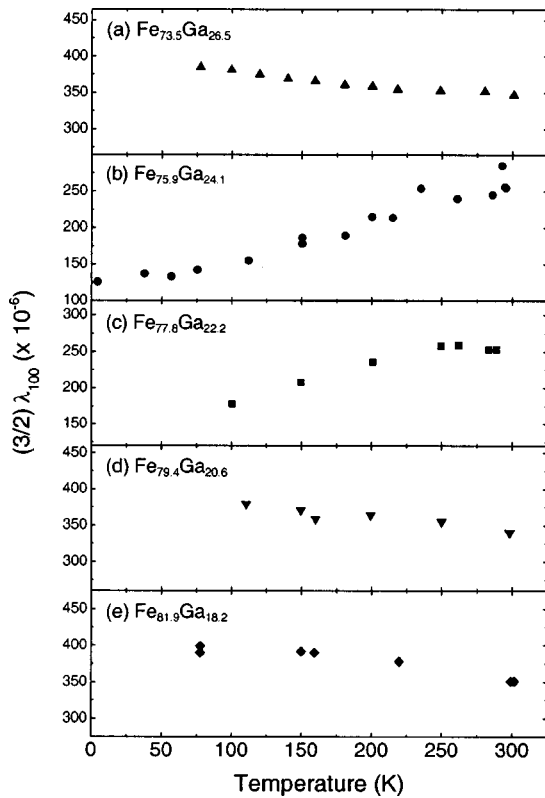


FIG. 2. Temperature dependence of $(3/2)\lambda_{100}$ for various Fe-Ga alloys.

SAMPLE PREPARATION

Single crystal alloys of Fe-Ga have been prepared by Bridgman growth of arc-cast ingots of electrolytic Fe (99.999% pure) and Ga (99.999% pure) in alumina crucibles. The ingots were stabilized in the crucible for 1 h at 1650 °C and lowered at a rate of ~2 min/h. Heat treatment at 1000 °C between 72 and 168 h followed the crystal growth. Oriented single crystal disks (~0.3 cm×0.6 cm diam) and parallelepipeds (0.1 cm×0.2 cm×0.3 cm) were cut from the boule by electronic spark erosion techniques. Furnace cooled samples were cooled at a rate of ~10°/min from 1000 °C; quenched samples were heated to 800 or 1000 °C in evacuated quartz tubes and held for 1 h before being dropped into water at room temperature. X-ray diffraction confirmed that all of

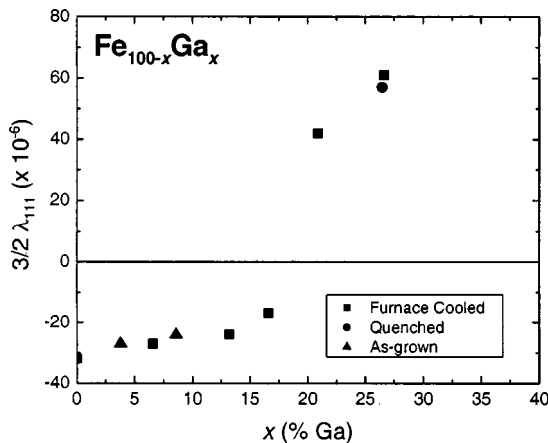


FIG. 3. $(3/2)\lambda_{111}$ as a function of Ga concentration for $Fe_{100-x}Ga_x$.

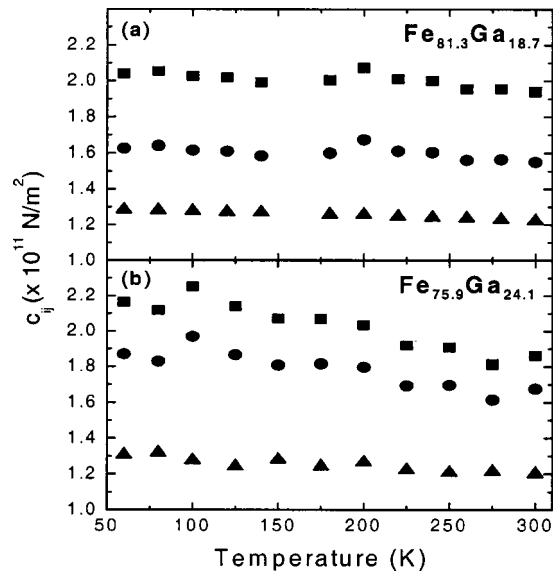


FIG. 4. Elastic constants c_{11} (■), c_{12} (●), and c_{44} (▲), as a function of temperature for (a) $Fe_{81.3}Ga_{18.7}$ and (b) $Fe_{75.9}Ga_{24.1}$.

the samples have bcc-like structure, but the local ordering of Ga and Fe on the bcc lattice sites was not definitively determined.

MAGNETOSTRICTION MEASUREMENTS

Magnetostriction constants $(3/2)\lambda_{100}$ and $(3/2)\lambda_{111}$ were measured by standard strain gauge techniques. To evaluate $(3/2)\lambda_{100}$, small 0.6-cm-diam samples with (001) faces were affixed with strain gauges oriented along [100] crystalline directions and rotated 360° in the presence of magnetic fields ranging from 5 to 25 kOe. To lowest order, peak-to-peak strain values at saturating fields equal $(3/2)\lambda_{100}$. To determine $(3/2)\lambda_{111}$, (110) disks were affixed with strain gauges oriented along [111] directions. Measurements were made as a function of temperature from 4 to 300 K.

Values of $(3/2)\lambda_{100}$ for $Fe_{100-x}Ga_x$ ($4 < x < 35$) at room temperature are shown in Fig. 1. Two widely separated peaks are found for both slow cooled and rapidly quenched samples. Between these peaks $(3/2)\lambda_{100}$ has an unusual temperature dependence. Figure 2 illustrates temperature dependencies for quenched samples from $x = 18.2$ to 26.5. Normal

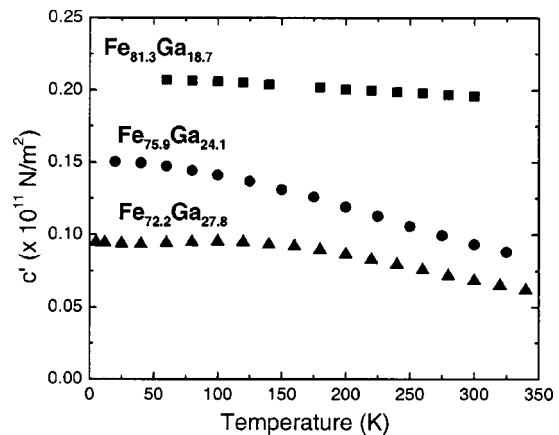


FIG. 5. c' as a function of temperature for $Fe_{81.3}Ga_{18.7}$ (■), $Fe_{75.9}Ga_{24.1}$ (●), and $Fe_{72.2}Ga_{27.8}$ (▲).

TABLE I. Room temperature tetragonal magnetoelastic constants for $\text{Fe}_{100-x}\text{Ga}_x$.

	$(c_{11}-c_{12})/2$ (GPa)	$(3/2)\lambda_{100}$ ($\times 10^{-6}$)	b_1 (MJ/m ³)
Fe (Ref. 9)	48	30	-2.9
5.8% Ga	40 (Ref. 7)	79	-6.3
13.2% Ga	28 (Ref. 7)	210	-11.8
17% Ga	21	311	-13.1
18.7% Ga	19.7	395	-15.6
24.1% Ga	9.4	270	-5.1
27.2% Ga	6.8	350	-4.8

magnetostrictive behavior, i.e., magnetostriction decreasing modestly with increasing temperature (decreasing magnetization), is found for the $x=18.2$, 20.6, and 26.5 alloys. On the other hand, the magnetostriction *increases strongly* with increasing temperature for $x=22.2$ and 24.1 alloys.

Room temperature values for $(3/2)\lambda_{111}$ are displayed in Fig. 3. Note the change in sign. Entirely different slopes in the magnetostriction versus Ga concentration are found for small x ($x < 20$) than for large x ($x > 20$).

ELASTIC CONSTANT MEASUREMENTS

Resonant ultrasound spectroscopy (RUS) techniques were used to determine the elastic constants of Fe-Ga over the same range of Ga concentration. Single crystal samples in the shape of small parallelepipeds, with $\{100\}$ faces and unequal principal axes, were mounted at their corners between two transducers, and the resonant frequencies recorded. Using normal mode frequency analysis, density, and sample dimensions, c_{11} , c_{12} , and c_{44} for the samples were calculated. Elastic constant measurements were made over the temperature range from 4 K to room temperature.

From Fig. 4, the temperature dependences of the elastic constants for $x=18.7$ and 24.1 can be contrasted. While c_{44} values are quite similar, c_{11} and c_{12} have different magnitudes and temperature dependences. A profound difference is found in the values of the tetragonal shear elastic constant $c' \equiv \frac{1}{2}(c_{11}-c_{12})$ (see Fig. 5). Here the c 's are compared for $x=18.7$, 24.1, and 27.8. Three features are noted. First, c' is nearly an order of magnitude smaller than the rhombohedral shear elastic constant c_{44} .⁶ Second, c' becomes progressively softer as Ga is increased to 27.8% over the whole temperature range.⁷ Third, the slopes of the modulus versus temperature curves are entirely different: a normal small decrease in c' is found for $x=18.7$, an extraordinary large decrease in c' is found for the sample near the minimum in the magnetostriction peak ($x=24.1$), and a more complex temperature dependence is found for $x=27.8$.

DISCUSSION

From a knowledge of magnetostriction constants and elastic constants it is possible to compute the values of the conventional magnetoelastic energy constants b_1 and b_2 . From Kittel⁸ $b_1 = -(3/2)\lambda_{100}(c_{11}-c_{12})$ and $b_2 = -3\lambda_{111}c_{44}$. Table I displays values of c' , $(3/2)\lambda_{100}$, and b_1 for Fe-Ga alloys at room temperature. *Note that this calculation yields only one peak in the values of b_1 versus con-*

TABLE II. Room temperature rhombohedral magnetoelastic constants for $\text{Fe}_{100-x}\text{Ga}_x$.

	c_{44} (GPa)	$(3/2)\lambda_{111}$ ($\times 10^{-6}$)	b_2 (MJ/m ³)
Fe (Ref. 9)	116	-32	7.4
8.6% Ga	119	-27	6.4
13.2% Ga	~119	-24	5.7
20.88% Ga	~120	42	-10.1
28.63% Ga	~120	61	-14.6

centration. This peak corresponds to the magnetostriction peak at ~19% Ga. The second magnetostriction peak can be attributed to the softening of the $c_{11}-c_{12}$ elastic constant. Most of the anomalous temperature dependence of λ_{100} for the 24.1% sample appears to come from the temperature dependence of the elastic constants. The small temperature dependence for the 27.8% sample may result from a combination of an increase with temperature due to elastic softening plus a decrease with temperature due to a rapidly falling magnetization ($T_C \cong 600$ K).³

Table II displays values of c_{44} , $(3/2)\lambda_{111}$, and b_2 for Fe-Ga alloys over a similar composition range at room temperature. Note that, unlike b_1 , b_2 changes sign. The sign change in b_2 occurs at nearly the same composition as the peak in b_1 , indicating a ordering transition that affects both of them. When $b_1=b_2$, the material is magnetoelastically isotropic. Thus the Fe-Ga alloys system exhibits a large “-” magnetoelastic anisotropy (characteristic of Fe) for small x , but reverts to a large “+” anisotropy for large x (see Tables I and II). Since the calculated values for b_1 and b_2 at 77 K are comparable to those at room temperature, this behavior can be seen as a fundamental property of the Fe-Ga system.

ACKNOWLEDGMENTS

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⁶These anomalously soft values for c' affect the relative accuracies of the different elastic constants that are obtained from the RUS normal mode analysis. Values for c' are fitted with high accuracy due to the large number of low frequency (soft) tetragonal shear modes. But the individual elastic constants c_{11} , c_{12} , and c_{44} are determined from a proportionately smaller number of modes, leading to a larger experimental uncertainty in their values, and perhaps accounting for the scatter in Fig. 4.

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