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Magnetic field dependence of galfenol elastic properties

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Abstract
Elastic shear moduli measurements on $Fe_{100-x}Ga_x$ (x = 12–33) single crystals (via resonant ultrasound spectroscopy) with and without a magnetic field and within 4–300 K are reported. The pronounced softening of the tetragonal shear modulus c' is concluded to be, based on magnetoelastic coupling, the cause of the second peak in the tetragonal magnetostriction constant λ_{100} near x = 28. Exceedingly high ΔE effects (~ 25%), combined with the extreme softness in c' (c'<10 >GPa), suggest structural changes take place, yet, gradual in nature, as the moduli show a smooth dependence on Ga concentration, temperature, and magnetic field. Shear anisotropy (c_{44}/c') as high as 14.7 was observed for $Fe_{71.2}Ga_{28.8}$.

Keywords
iron alloys, gallium alloys, ferromagnetic materials, shear modulus, softening, magnetoelastic effects, magnetostriction

Disciplines
Condensed Matter Physics | Metallurgy

Comments

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Magnetic field dependence of galfenol elastic properties

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Elastic shear moduli measurements on $Fe_{100-x}Ga_x$ ($x=12-33$) single crystals (via resonant ultrasound spectroscopy) with and without a magnetic field and within 4–300 K are reported. The pronounced softening of the tetragonal shear modulus c' is concluded to be, based on magnetoelastic coupling, the cause of the second peak in the tetragonal magnetostriction constant λ_{100} near $x=28$. Exceedingly high ΔE effects ($\sim 25\%$), combined with the extreme softness in c' ($c' < 10$ GPa), suggest structural changes take place, yet, gradual in nature, as the moduli show a smooth dependence on Ga concentration, temperature, and magnetic field. Shear anisotropy (c_{44}/c') as high as 14.7 was observed for $Fe_{71.2}Ga_{28.8}$. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1855711]

Mechanically strong and malleable, with a high relative permeability and reduced cost of fabrication, while reaching a magnetostriction as high as one order of magnitude above that of pure Fe, $Fe_{100-x}Ga_x$ (galfenol) has become the material of interest in magnetomechanics. As one of the elements with high solubility in α-Fe,[1] Ga generates the highest magnetostriction in this group of alloys: $\frac{3}{2}\lambda_{100}$ reaches over 400 ppm (Fig. 1).[2] Interestingly, the magnitude of λ_{111} magnetostriction is only weakly affected by the addition of Ga. This strong magnetostriction anisotropy together with the peculiar doublepeak in the λ_{100} vs Ga concentration curve have been the motivation for the study of elastic properties of galfenol.

Room temperature elastic moduli for $x=12\%$, 14%, and 20% Ga were previously reported by Wuttig *et al.*[3] Temperature dependent moduli for $x=18.7\%$, 24.1%, and 27.8% Ga have been previously measured.[2] In both cases, no magnetic field was applied.

A model[3] has been proposed to explain the increase in tetragonal magnetostriction in Fe-Ga alloys as arising from Ga atoms pairing along [100] directions in a premartensitic short-range ordering process. The model advances the idea that embryos of a martensitic phase grow around the Ga pairs, as the Ga concentration increases, in an approach to a cubic-to-tetragonal transformation at $x=26\%$, where the c' elastic modulus is assumed to extrapolate to zero. λ_{100} is predicted to increase continuously with x.

This paper reports extensive measurements of the shear elastic moduli of galfenol with x ranging between 12% and 33%, with and without magnetic field, and as a function of temperature.

The structural complexity of galfenol, likely the source of its unusual magnetoelastic properties, is apparent from its complex phase diagram.[4] As more Ga atoms are introduced in the alloy, chemical ordered phases, such as DO_3, $B2$, DO_{19}, or $L1_2$ develop and continue to grow over the composition range. Above 19% Ga, two phase mixtures may exist depending on the thermal treatment. Both quenched and furnace-cooled samples were used in an attempt to attain pure or mixed phases, or different degrees of ordering. The samples were annealed at 1000 °C followed by either 10 °C/min slow cooling or water quenching. Details of sample preparation are given elsewhere.[2] X-ray diffraction confirms that all investigated samples are single crystals of a cubic structure.

Measurements of the elastic moduli within 4–301 K and 0–15 kOe have been performed using resonant ultrasound

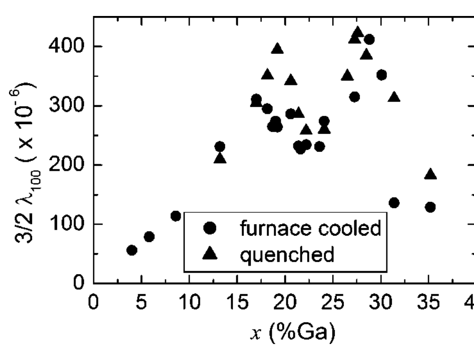

FIG. 1. Magnetostriction constant $\frac{3}{2}\lambda_{100}$ vs Ga concentration x for $Fe_{100-x}Ga_x$ at room temperature.

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spectroscopy[5] (RUS) on [100]-oriented rectangular parallelepipeds. In RUS, the complete elastic moduli tensor of an object with a well-defined geometry is determined through an iterative algorithm from its dimensions, mass, and a set of normal modes of vibration (first 30 resonances, on average). The resonances are detected with a pair of piezoelectric transducers acting as source and receiver, lightly pressed against the sample. The classical corner-to-corner[5] method could not be used here since an unbalanced torque due to the magnetic field removes the sample from its mount. We have developed an alternative RUS probe based on an original idea emerged from Migliori's group at Los Alamos National Laboratories[6] which allowed us to measure the resonance frequencies with a magnetic field applied. In this probe, the sample is held *flat* between opposite faces by the transducer pair, under minimum pressure. Since neither the transducers nor the samples can be finished to microscopically planar surfaces, a maximum of three contact points, *randomly* distributed, exists between the transducer and its adjacent sample face. This imperfection is the key factor that allows excitation and detection of all normal modes, while the sample is mechanically stable in the presence of the magnetic field. An eventual missed resonance, rarely occurring, is found during a successive remount.

Two of the three independent elastic moduli for a cubic system, the shear c' and c_{44} [note: $c'=\frac{1}{2}(c_{11}-c_{12})$], are likely to play a major role in the magnetoelasticity of galfenol, due to magnetoelastic coupling. Results of c_{44} and c' dependence on (a) Ga concentration x in the alloy, (b) temperature, and (c) magnetic field are presented in the following section. In (a) and (b) the measurements were done under a field of 15 kOe, more than double the saturation value, oriented along a [100] crystalline axis. For each sample, the measurements were performed in this order: at 15 kOe, the resonances were measured at temperatures decreasing from 301 K to 4 K. The sample was then warmed to 301 K, where the resonance frequencies were measured at 14 intermediate field values between 15 kOe and 0. At zero field, the temperature was lowered again from 301 K to 4 K, in order to obtain the corresponding zero-field elastic moduli values.

The results are summarized below.

(a) Values of c_{44} and c' as a function of Ga concentration for furnace-cooled and quenched samples are listed in Table I for two temperatures. Here, we concentrate on the room temperature results as temperature dependence is discussed in part (b). Bearing in mind the x dependence of λ_{100} and λ_{111}, a "tandem" reaction of the couples (λ_{100}, c') and (λ_{111}, c_{44}) is observed. c_{44} reacts modestly to the Ga addition, while c' has a very strong response. c' becomes extremely soft with increasing Ga concentration, as in an approach to a phase transition. Nevertheless, it starts increasing after reaching a minimum of 7.9 GPa at 28.8% Ga, where λ_{100} raises to a second peak. c' is a smooth function of x, maintaining a nonzero value throughout the entire range investigated. The very low value of c', together with the flat response of c_{44}, leads to the extremely large shear anisotropy of 14.7 ([110] highly auxetic) for $Fe_{71.2}Ga_{28.8}$.

(b) c' temperature dependences for various Ga concentrations at 15 kOe are plotted in Fig. 2. Apart from the 33% samples, the cooling process does not affect this response significantly. A very strong temperature dependence of c' is observed for all samples with $x > 19\%$. It is also important to mention that c_{44} (not shown), compared with c', has a much weaker temperature dependence for all samples.

(c) The following expression, with $H_s = 15$ kOe as the saturating field, is used to report the magnetic field influence on the moduli:

$$\Delta c = [c(H_s) - c(H=0)]/c(H=0), \tag{1}$$

where $c = c'$ or c_{44}. Room temperature values of Δc for both shear moduli are listed in Table II. Typical Δc values for ferromagnetic materials are under 0.2%. For Fe-Ga, we see

TABLE I. Shear moduli c_{44} and c' and shear anisotropy (c_{44}/c') in saturating field (15 kOe), at 300 K and 4 K, as a function of Ga concentration and heat treatment (FC, furnace cooled; Q, quenched).

	c_{44}(GPa)		c'(GPa)		$A=c_{44}/c'$
x Ga%	(301 K)	(4 K)	(301 K)	(4 K)	(301 K)
0[a]	116	121.9	48	52.5	2.4
12_{FC}	126.8	131.25	32.5	34.9[b]	3.9
12_Q	124.7	130.0	32.1	34.4	3.9
19_{FC}	123.9	131.7	16.6	20.1	7.5
19_Q	121.9	127.4	19.9	21.2[b]	6.1
24.4_{FC}	127.5	141[c]	9.5	14.4	13.5
25_Q	124.6	137[c]	10.0	14.6	12.5
26.3_Q	125.2	135.3	8.9	12.9	14.1
27.3_Q	123.7	138.1[c]	8.7	12.7	14.2
27.8_{FC}	122.7	127.2[c]	8.6	11.5	14.3
28.8_{FC}	116.6	127.1[c]	7.9	10.2	14.7
33.3_{FC}	95.6	104.0[c]	22.2	24.5	4.3
33.3_Q	103.8	112.8	10.5	12.6	9.9

[a]Pure Fe data from Ref. 8.
[b]9 K value.
[c]40 K value.

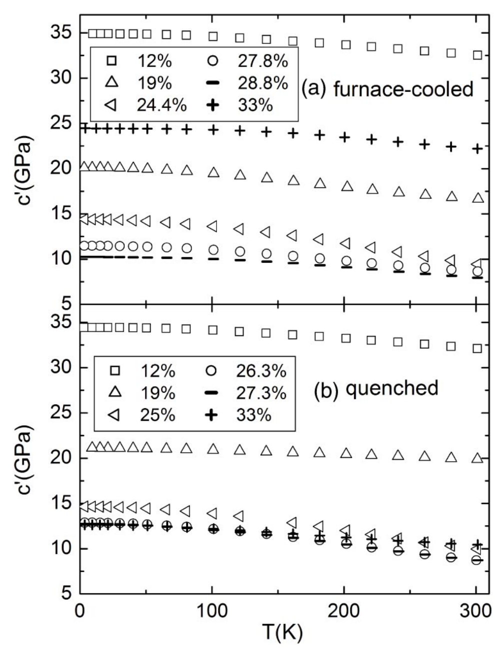

FIG. 2. Temperature dependence of the tetragonal shear modulus c' for furnace-cooled (a) and quenched (b) samples at various Ga concentrations. Measurements taken in saturating field (15 kOe).

TABLE II. Room temperature values for Δc_{44} and $\Delta c'$.

x(Ga%)	Δc_{44}(%) (301 K)	$\Delta c'$(%) (301 K)
12_{FC}	0.5	0.25
12_{Q}	1.0	0.4
19_{FC}	0.6	6.8
19_{Q}	0.4	2.2
24.4_{FC}	10.7	12.3
25_{Q}	1.5	11
26.3_{Q}	1.2	6.3
27.3_{Q}	5.1	6.9
27.8_{FC}	13.7	25.3
28.8_{FC}	5.1	24.8
33.3_{FC}	0.6	0.9

significantly higher numbers, reaching more than 25%, as c' approaches its minimum at $x=28\%$ Ga. Once more, in accord with the trends seen in (a) and (b), c' responds to the applied magnetic field in a more significant way than c_{44} (not shown). Note that a large $\Delta c'$ also implies very low zero-field modulus (minimum c' value is 6.3 GPa at 28.8% furnance cooled).

$\Delta c'$ temperature dependence for all samples with small or large x, furnace-cooled or quenched, is monotonic and weak [e.g., Figs. 3(a) and 3(d)], similar to conventional ferromagnetic materials. Within the particular range of (26–29)% Ga, $\Delta c'$ temperature dependence for the furnace-cooled samples becomes unusual [Figs. 3(b) and 3(c)] due to an abnormal temperature dependence of c' at zero field. No temperature hysteresis was observed. c_{44} does not exhibit this behavior.

The coupling between magnetostriction and elastic constants is incorporated in the magnetoelastic energy, which, for a cubic system, to first order, is given by

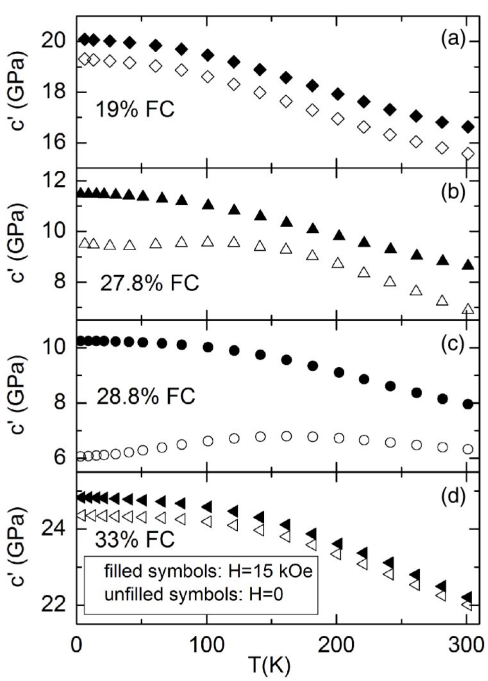

FIG. 3. c' temperature dependence with and without field for slow-cooled (FC) alloys, outside (a,d) and inside (b,c) the minimum-c' range. The difference between the field and nofield values indicates the $\Delta c'$ effect.

$$E_{\mathrm{me}} = b_1(\alpha_x^2\varepsilon_{xx} + \alpha_y^2\varepsilon_{yy} + \alpha_z^2\varepsilon_{zz}) + b_2(\alpha_x\alpha_y\varepsilon_{xy} + \alpha_y\alpha_z\varepsilon_{yz} + \alpha_z\alpha_x\varepsilon_{zx}), \quad \text{with } b_1 = -3(\lambda_{100}\times c'), \quad b_2 = -3(\lambda_{111}\times c_{44}), \tag{2}$$

where α_i's are the direction cosines between magnetization and the crystal axis, ε_{ij} are the conventional Cartesian strain components, and b_1 and b_2 are the magnetoelastic energy constants, defined as the product between corresponding tetragonal and rhombohedral magnetostriction and elastic constants. A clear interdependence within the pairs (λ_{100}, c') and (λ_{111}, c_{44}) has been observed. The mechanisms coupling λ_{100} and c' at the first and second peaks are different, most probably due to a difference in the structure. The first peak in λ_{100}, at $x=19\%$, has been attributed to an increase in the magnetoelastic energy constant b_1, with the onset of short range Ga ordering. The pronounced softening of c' at $x=28.8\%$, a strong characteristics of the system, being maintained for all magnetic configurations and over the entire temperature range, is sufficient to explain the second peak in λ_{100} even if b_1 remains flat in that region.

The very unusual $\Delta c'$ observed for the furnace-cooled samples within the region of the second peak is not understood. An interpretation can be inferred, however, if an analogy is considered. The sites of two of the Fe atoms in galfenol's DO_3-like structure, very probable to occur in the furnace-cooled samples at x close to 29%, are similar with those of Ni and Mn in the Ni–Mn–Ga system which undergoes transitions between different martensitic states.[7] In the case of galfenol, the magnetic field is able to inhibit the growth of the eventual martensitic variants present in the alloy. Small regions of martensitic phase clusters were suggested in the previously proposed model for galfenol.[3] The model also suggested that a cubic-to-tetragonal transformation occurs at $x=26\%$. Based on the fact that the elastic moduli are smooth functions of the variables investigated and c' does not extrapolate to zero, we believe that this transformation is approached but it does not fully take place. Also, the ordering process evolves gradually.

Development of a model to explain the observed magnetoelastic phenomena awaits detailed characterization of the ordered alloy phases present in these samples.

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