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The effect of partial substitution of Ge for Ga on the elastic and magnetoelastic properties of Fe–Ga alloys

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Abstract

Both components of the tetragonal magnetoelastic constant b_1: the saturation magnetostriction, $\lambda^{\gamma,2} = (3/2)\lambda_{100}$, and the magnetic-field saturated shear elasticity, $c' = (c_{11}-c_{12})/2$, were investigated over a wide temperature range for the magnetostrictive$Fe_{1-x-y}Ga_xGe_y$ alloys, ($x+y \cong 0.125$, 0.185, and 0.245; $x/y \cong 1$ and 3). The magnetostriction was measured from 77 to 425 K using standard strain gage techniques. Both shear elastic constants (c' and c_{44}) were measured from 5 to 300 K using resonant ultrasound spectroscopy. Six alloy compositions were prepared to cover three important regions: (I) the disordered solute α-Fe region, (II) a richer solute region containing both disordered and ordered phases, and (III) a rich solute region containing ordered multiphases. Our observations reveal that, when the data is presented versus the total electron/atom (e/a) ratio, the above regions for both the ternary and binary alloys are in almost perfect alignment. Following this analysis, we find that the magnetoelastic coupling, b_1, peaks for both the binary and the ternary alloys at $e/a \sim 1.35$. The values of c' as well as of $\lambda^{\gamma,2}$ in region I of the ternary alloys, when plotted versus e/a, fall appropriately between the binary limits.

Keywords

elastic constants, ferromagnetic materials, gallium alloys, germanium alloys, iron alloys, magnetoelastic effects, magnetostriction, ultrasonic measurement

Disciplines

Condensed Matter Physics | Metallurgy

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The effect of partial substitution of Ge for Ga on the elastic and magnetoelastic properties of Fe–Ga alloys

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Both components of the tetragonal magnetoelastic constant b_1: the saturation magnetostriction, $\lambda^{\gamma,2}=(3/2)\lambda_{100}$, and the magnetic-field saturated shear elasticity, $c'=(c_{11}-c_{12})/2$, were investigated over a wide temperature range for the magnetostrictive $Fe_{1-x-y}Ga_xGe_y$ alloys, ($x+y\cong 0.125$, 0.185, and 0.245; $x/y\cong 1$ and 3). The magnetostriction was measured from 77 to 425 K using standard strain gage techniques. Both shear elastic constants (c' and c_{44}) were measured from 5 to 300 K using resonant ultrasound spectroscopy. Six alloy compositions were prepared to cover three important regions: (I) the disordered solute α-Fe region, (II) a richer solute region containing both disordered and ordered phases, and (III) a rich solute region containing ordered multiphases. Our observations reveal that, when the data is presented versus the total electron/atom (e/a) ratio, the above regions for both the ternary and binary alloys are in almost perfect alignment. Following this analysis, we find that the magnetoelastic coupling, b_1, peaks for both the binary and the ternary alloys at $e/a\sim 1.35$. The values of c' as well as of $\lambda^{\gamma,2}$ in region I of the ternary alloys, when plotted versus e/a, fall appropriately between the binary limits. © *2010 American Institute of Physics*. [doi:10.1063/1.3368108]

The profound interest in Galfenol (Fe–Ga) as an active material led to a vigorous area of research in Fe-based alloys. Fe–Ge, a binary alloy that emerged from that effort,[1] helped elucidate some of the unsolved questions of Fe–Ga. It is natural to consider that an alloy with both Ga and Ge will further extend the understanding of magnetostrictive Fe-based alloys. The phase diagrams of Fe–Ga and Fe–Ge illustrate distinct phase distributions for each alloy,[2] with three major regions of interest. In order of increasing solute concentration, they are as follows. The first region is characterized by a disordered arrangement of the solute atoms within the α-Fe bcc crystalline structure. The second region is a two-phase region characterized by the formation of an ordered phase within the disordered matrix. The third region is fully ordered and may be characterized by the coexistence of multiple phases. Fe–Ge has a stable phase distribution, which is reproducible and independent of thermal treatment in all three regions. Fe–Ga has a complex distribution of phases that is highly dependent on thermal treatment at the transition between regions one and two. Despite the marked difference between these two binary alloys, the work presented here shows that the magnetoelastic coupling factor b_1 of Fe–Ga–Ge ternary alloys follows the weighted average of those of Fe–Ga and Fe–Ge. The tetragonal magnetoelastic coupling factor was calculated from magnetostriction and elastic constants measured for six compositions of Fe–Ga–Ge as a function of temperature. The measurements and calculations are presented in this paper.

Single crystal ingots of $Fe_{1-x-y}Ga_xGe_y$ were prepared using the Bridgman method.[3] The aim during the synthesis process was to obtain samples with two different values of x/y for a given value of $x+y$. The following sample identification scheme was introduced: I, II, and III to represent $x+y$ values of ~12.5, ~18.5, and ~24.5 at. %, while (a) and (b) to represent an x/y value of ~1 and ~3, respectively. The compositions were determined to within 0.33 at. % by wavelength-dispersive x-ray spectroscopy and are listed in Table I.

For a direct comparison of the physical properties of the investigated alloys, a generalized variable, one that accounts for the different electronic structures of the solutes, was necessary. A factor known to be a measure of phase stability[4] and commonly used when comparing physical properties of different alloys is the total number of valence electrons per total number of atoms in a given volume, e/a. For an alloy of the type $Fe_{1-x-y}Ga_xGe_y$, $e/a=n_{Ga}x+n_{Ge}y+n_{Fe}(1-x-y)$, where n represents the number of valence electrons. Using $n_{Fe}=1$ (suggested by the average number of *sp*-electrons from first principle electronic structure calculations[5]), $n_{Ga}=3$, and $n_{Ge}=4$, the e/a values for the investigated samples were calculated and are listed in Table I. The range of e/a covers the transition from disordered to ordered, crossing the solubility limit of the bcc α-Fe phase ($e/a\sim 1.35$) for the added elements. The three sample types belong in distinct regions: below the solubility limit (I), in the mixed disordered-ordered phase region (II), and in the high-solute, fully ordered region (III).

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TABLE I. Sample composition, e/a value, and type, for the magnetostriction (upper) and elasticity (lower) experiments.

	$1-x-y$ (at. % Fe)	x (at. % Ga)	y (at. % Ge)	$x+y$ (at. %)	x/y	e/a	Type
Magneto-striction samples	88.1	6.2	5.7	11.9	1.1	1.30	Ia
	86.9	9.9	3.2	13.1	3.1	1.29	Ib
	81.8	9.5	8.7	18.2	1.1	1.45	IIa
	80.8	14.5	4.7	19.2	3.1	1.43	IIb
	75	13.1	11.9	25.0	1.1	1.62	IIIa
	76.2	18.2	5.6	23.8	3.1	1.53	IIIb
RUS samples	88.3	6.1	5.6	11.7	1.1	1.29	Ia
	87.2	9.7	3.1	12.8	3.1	1.29	Ib
	82.1	9.5	8.4	17.9	1.1	1.44	IIa
	81.2	14.3	4.5	18.8	3.2	1.42	IIb_1
	80.8	14.6	4.6	19.2	3.2	1.43	IIb_2
	75.9	12.7	11.4	24.1	1.1	1.60	IIIa
	76.6	18.1	5.3	23.4	3.4	1.52	IIIb

The saturation value of the tetragonal magnetostriction $(3/2)\lambda_{100}$ was measured for the six single crystal disks (Table I—upper) in the slow-cooled (SC) state as a function of temperature. The standard strain gage technique[3] was used. The samples were ~6 mm in diameter, ~3 mm in thickness, with faces oriented parallel to a (100) plane of the iron lattice. A magnetic field of 15 kOe, well above saturation, was used for temperatures between 78 and 300 K and a field of 4.3 kOe was used for temperatures between 300 and 423 K. In the high-temperature setup, the maximum allowed field of 4.3 kOe was sufficient to saturate the sample, and the results obtained from both setups at 300 K are in agreement. Resonant ultrasound spectroscopy was used to determine the two independent shear moduli of the alloy, $c'=(c_{11}-c_{12})/2$ and c_{44}, for the seven single crystal parallelepipeds (Table I—lower) from 5 to 300 K. The faces of the parallelepipeds (millimeters in size) were oriented parallel to (100) planes of the iron lattice. The elasticity measurements were performed in a saturating magnetic field of 20 kOe oriented along the longest side of the parallelepiped. The crystals were first investigated in their SC state. Subsequently they were reheated at 1273 K for 4 h and quenched in ice-water. For the samples in their quenched state, the elasticity measurements were repeated versus temperature while the magnetostriction measurements were repeated at room temperature only. The IIIa disk sample fractured during quenching, which made it unusable.

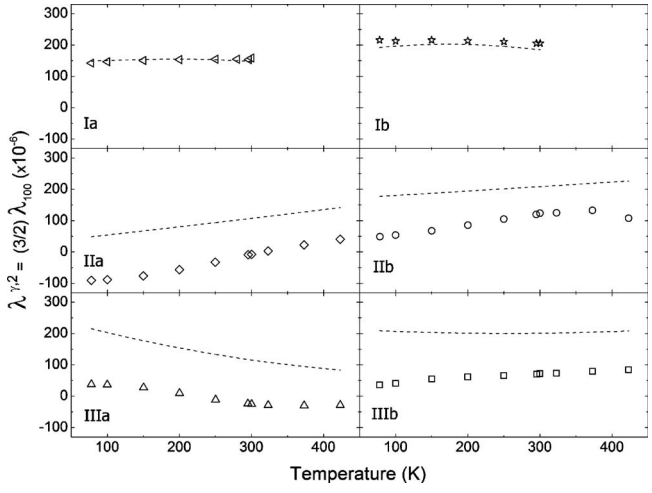

FIG. 1. Tetragonal magnetostriction $(3/2)\lambda_{100}$ of $Fe_{1-x-y}Ga_xGe_y$ (SC) vs temperature (data points) and the corresponding weighted average (dotted line) from binary Fe–Ga (Ref. 3) and Fe–Ge (Ref. 1) data. The same scale is used for all graphs.

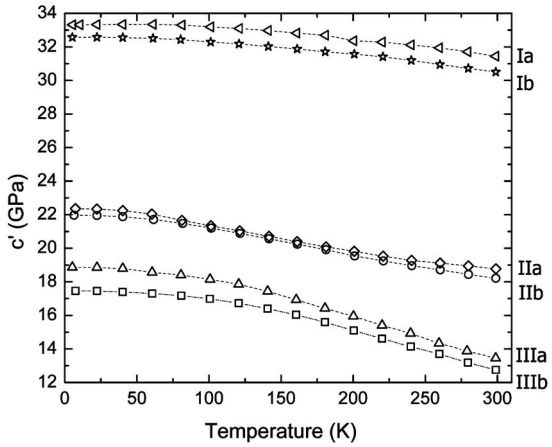

FIG. 2. Tetragonal shear modulus, c', of the $Fe_{1-x-y}Ga_xGe_y$ single crystals (SC state) vs temperature.

The saturation values of the tetragonal magnetostriction, $(3/2)\lambda_{100}=\lambda^{\gamma,2}$, for the Fe–Ga–Ge crystals in SC state are shown in Fig. 1 as a function of temperature. The results are compared with the weighted average ($x/y=1$ or 3) of the existing SC data for the binary Fe–Ga (Ref. 3) and Fe–Ge (Ref. 1) alloys, taken at corresponding e/a values. Qualitatively, the weighted average is a good representation of the measured ternary data, in terms of the slope $(d\lambda^{\gamma,2}/dT)$, for all six samples. Quantitatively, a good agreement was found only for the type I samples. These are samples where the added element (Ga and/or Ge) dilutes within the α-Fe bcc structure at random, with no preferred lattice sites. The averaging is therefore physically meaningful.

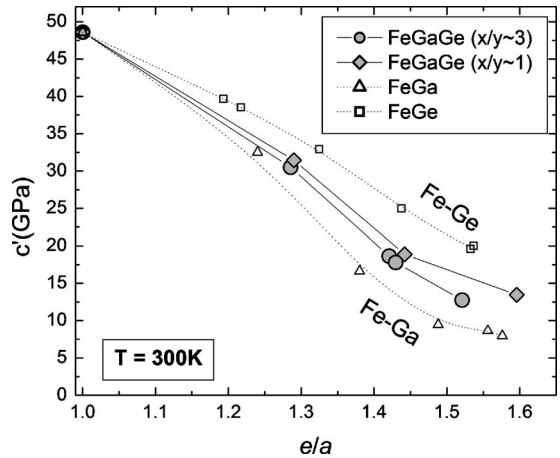

FIG. 3. Tetragonal shear modulus, c', of $Fe_{1-x-y}Ga_xGe_y$ compared with c' of the binary Fe–Ga (Ref. 6) and Fe–Ge (Ref. 1) alloys vs electron-to-atom ratio, e/a, at 300 K (all data are from SC single crystals).

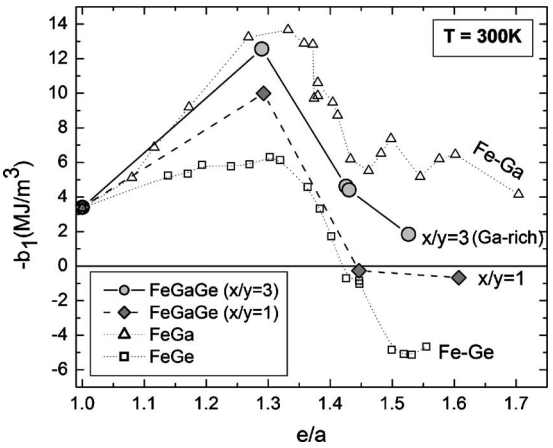

FIG. 4. Magnetoelastic coupling $-b_1=3\lambda_{100}c'$ of $Fe_{1-x-y}Ga_xGe_y$ contrasted with those of the binary alloys Fe–Ga (Ref. 1) and Fe–Ge (Ref. 1) as a function of electron-to-atom ratio, e/a, at 300 K (all data are from SC single crystals).

For types II and III, the weighted average of the binary data exceeds the measured ternary values by ~130 and ~170 ppm, respectively. These samples have solute concentrations beyond the solubility limit where ordered phases exist. The ternary crystals settle into a structure with a magnetostriction that is closer to that of the ordered Fe–Ge, even when Ga-rich. This suggests that Ge favors chemical ordering and its presence in the ternary alloy changes the phase distribution more rapidly than a simple weighted average would predict. Therefore, the high value of the weighted average for the type II and III alloys results from an over emphasis of the Fe–Ga magnetostriction. A quantitative understanding of these high-solute concentration ternary alloys will require careful measurements of their phase distributions by powder x-ray diffraction, which is made difficult by the similarity in the x-ray form factors of Fe, Ga, and Ge.

The tetragonal shear constant c' (saturation-field value) for Fe–Ga–Ge in the SC state is shown in Fig. 2 as a function of temperature. An average of IIb_1 and IIb_2 (within 5% difference) was used for IIb. The temperature dependence of c' for the ternary alloys follows the common trends observed for Fe–Ga (Ref. 6) and Fe–Ge (Ref. 1), i.e., $c'(T)$ is weak at low solute and strong at high-solute concentrations. A notable feature is that, if the solute's electronic contribution is considered, the softening of $c'(x)$ depends significantly on the *type* of solute (Ga or Ge) at high-solute concentrations. This can be seen in Fig. 3, where c' of Fe–Ga–Ge is plotted together with c' of Fe–Ga (Ref. 6) and Fe–Ge (Ref. 1) versus e/a at room temperature. This feature is maintained at all temperatures.

Compared to c', the rhombohedral shear modulus, c_{44}, shows a much weaker dependence on both the amount of solute and temperature. This behavior is similar to that observed for the Fe–Ga (Ref. 3) and Fe–Ge (Ref. 7) binary alloys. For the e/a range studied, c_{44} of Fe–Ga–Ge lies between 123 and 128 GPa at 300 K, and between 128 and 136 GPa at 5 K. Quenching did not affect significantly the elastic constants of the SC Fe–Ga–Ge crystals (differences of less than 4% were observed for c' of samples in the two states).

Using the magnetostriction and elastic constants values, the tetragonal magnetoelastic coupling factor was calculated using $-b_1=3\lambda_{100}c'$. The results for the SC crystals are shown in Fig. 4, together with $-b_1$ for the SC binary alloys.[1] Before the onset of ordering ($e/a\sim1.35$), the magnetoelastic coupling factor increases with the amount of solute dissolved into the α-Fe structure and its magnitude depends on the electronic structure of the solute(s). The value of $-b_1$ measured for the ternary alloy follows the compositionally weighted average of $-b_1$ for the binary alloys. Beyond the solubility limit, the formation of ordered phases leads to a decrease in $-b_1$. Despite the lower than the weighted average values of λ_{100} found for the type II and III samples, $-b_1(e/a)$ still follows a weighted average of $-b_1$ of the binary alloys in those regions (an exception, not yet explained, is found for sample IIa). What leads to this behavior is the very low shear constant c' of Fe–Ga at $e/a>1.35$. At these concentrations, c' of Fe–Ga is ~2 times lower than that of Fe–Ge, and the effect of the high λ_{100} of Fe–Ga is compensated by this low c' value when $-b_1$ is calculated.

The values for $-b_1$ at 300 K for the four quenched samples for which the magnetostriction could be measured were close to those of the samples in their SC state and are listed in Table II.

In summary, we find that the magnitude of the tetragonal magnetoelastic coupling of Fe–Ga–Ge can be estimated from those of the binary alloys utilizing an electronic concentration (e/a ratio) basis.

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TABLE II. Magnetoelastic coupling $-b_1=3\lambda_{100}c'$ (MJ/m^3) at 300 K for samples in their slow-cooled (SC) and quenched (Q) states.

	Sample type			
	Ia	Ib	IIa	IIIb
$-b_1$ (SC)	10.00	12.57	−0.26	1.84
$-b_1$ (Q)	9.33	11.86	0.35	1.64

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