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Magnetostriction, elasticity, and D0₃ phase stability in Fe–Ga and Fe–Ga–Ge alloys

Abstract

The contrast between the saturation tetragonal magnetostriction, $\lambda^2 = (3/2)\lambda_{100}$, of Fe_{1-x}Ga_x and Fe_{1-y}Ge_y, at compositions where both alloys exhibit D0₃ cubic symmetry (*second peak* region), was investigated. This region corresponds to $x = 28$ at. % Ga and $y = 18$ at. % Ge or, in terms of $e/a = 2x + 3y + 1$, to an e/a value of ~ 1.55 for each of the alloys. Single crystal, slow-cooled, ternary Fe_{1-x-y}Ga_xGe_y alloys with $e/a \sim 1.55$ and gradually increasing y/x were investigated experimentally (magnetostriction, elasticity, powder XRD) and theoretically (density functional calculations). It was found that a small amount of Ge ($y = 1.3$) replacing Ga in the Fe–Ga alloy has a profound effect on the measured λ^2 . As y increases, the drop in λ^2 is considerable, reaching negative values at $y/x = 0.47$. The two shear elastic constants $c' = (c_{11} - c_{12})/2$ and c_{44} measured for four compositions with $0.06 \leq y/x \leq 0.45$ at 7 K range from 16 to 21 GPa and from 133 to 138 GPa, respectively. Large temperature dependence was observed for c' but not for c_{44} , a trend seen in other high-solute Fe alloys. The XRD analysis shows that the metastable D0₃ structure, observed previously in slow-cooled Fe–Ga at $e/a = 1.55$, is replaced with two phases, fcc L1₂ and hexagonal D0₁₉, at just 1.6 at. % Ge. The two are the *stable* phases of the assessed Fe–Ga phase diagram at $x \sim 28$. Notably, at $y = 7.8$, only the D0₃ phase (the equilibrium phase of Fe–Ge at $e/a = 1.54$) was found in the ternary alloy. The theory also shows that the D0₃ instability is removed for compositions with $y \geq 3.9$, when D0₃ becomes the structure's ground-state phase. Thus, the high, positive λ^2 value for Fe–Ga at $x = 28$ could be the result of the high sensitivity of its metastable D0₃ structure.

Keywords

density functional theory, elastic constants, elasticity, gallium alloys, germanium alloys, iron alloys, magnetostriction, phase diagrams, X-ray diffraction

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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Magnetostriction, elasticity, and D0₃ phase stability in Fe–Ga and Fe–Ga–Ge alloys

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The contrast between the saturation tetragonal magnetostriction, $\lambda^{\gamma,2} = (3/2)\lambda_{100}$, of Fe_{1-x}Ga_x and Fe_{1-y}Ge_y, at compositions where both alloys exhibit D0₃ cubic symmetry (*second peak* region), was investigated. This region corresponds to $x = 28$ at. % Ga and $y = 18$ at. % Ge or, in terms of $e/a = 2x + 3y + 1$, to an e/a value of ~ 1.55 for each of the alloys. Single crystal, slow-cooled, ternary Fe_{1-x-y}Ga_xGe_y alloys with $e/a \sim 1.55$ and gradually increasing y/x were investigated experimentally (magnetostriction, elasticity, powder XRD) and theoretically (density functional calculations). It was found that a small amount of Ge ($y = 1.3$) replacing Ga in the Fe–Ga alloy has a profound effect on the measured $\lambda^{\gamma,2}$. As y increases, the drop in $\lambda^{\gamma,2}$ is considerable, reaching negative values at $y/x = 0.47$. The two shear elastic constants $c' = (c_{11} - c_{12})/2$ and c_{44} measured for four compositions with $0.06 \leq y/x \leq 0.45$ at 7 K range from 16 to 21 GPa and from 133 to 138 GPa, respectively. Large temperature dependence was observed for c' but not for c_{44} , a trend seen in other high-solute Fe alloys. The XRD analysis shows that the metastable D0₃ structure, observed previously in slow-cooled Fe–Ga at $e/a = 1.55$, is replaced with two phases, fcc L1₂ and hexagonal D0₁₉, at just 1.6 at. % Ge. The two are the *stable* phases of the assessed Fe–Ga phase diagram at $x \sim 28$. Notably, at $y = 7.8$, only the D0₃ phase (the equilibrium phase of Fe–Ge at $e/a = 1.54$) was found in the ternary alloy. The theory also shows that the D0₃ instability is removed for compositions with $y \geq 3.9$, when D0₃ becomes the structure's ground-state phase. Thus, the high, positive $\lambda^{\gamma,2}$ value for Fe–Ga at $x = 28$ could be the result of the high sensitivity of its metastable D0₃ structure. © 2011 American Institute of Physics. [doi:10.1063/1.3535444]

I. INTRODUCTION

It was recently found¹ that the magnetoelastic properties of dissimilar Fe-based alloys, i.e., with solutes residing in different groups of the Periodic Table, could be correlated when e/a is used as the common variable. The parameter e/a , generally used as a measure of phase stability, represents the total number of valence electrons per total number of atoms in a given volume. For example, for a single crystal of Fe_{1-x-y}Ga_xGe_y, e/a is calculated to be $2x + 3y + 1$ by taking into account the average valence of Fe, Ga, and Ge.¹ Note that the binary alloys, Fe–Ga and Fe–Ge, are limiting cases of this ternary example with either $y \rightarrow 0$ or $x \rightarrow 0$, respectively. At $e/a \sim 1.55$, although it was found that both Fe–Ga² and Fe–Ge³ exhibit D0₃ cubic symmetry in their slow-cooled form, an unexplained contrast was observed between the alloys' tetragonal magnetostriction (saturation value), $\lambda^{\gamma,2} = (3/2)\lambda_{100}$. The results show that the room temperature $\lambda^{\gamma,2}$ of Fe_{1-x}Ga_x has a large and *positive* peak² ($\lambda^{\gamma,2} > 400 \times 10^{-6}$) at $x \sim 28$ at. % ($e/a = 1.56$), whereas $\lambda^{\gamma,2}$

of Fe_{1-y}Ge_y has a *negative* peak³ ($\lambda^{\gamma,2} < -125 \times 10^{-6}$) at $y \sim 18$ at. % ($e/a = 1.54$). This region has been commonly referred to as the *second-peak* region.

In this study we used ternary Fe_{1-x-y}Ga_xGe_y alloys with $e/a \sim 1.55$ and $0 < y/x \leq 0.5$. A gradual increase of y/x at a fixed e/a ensured a detailed examination of the transition between the structure responsible for the large and positive $\lambda^{\gamma,2}$ in Fe–Ga and that of Fe–Ge leading to a negative $\lambda^{\gamma,2}$. Note that the D0₃ structure that was experimentally determined for both binary alloys is either overpopulated (in Fe–Ga) or underpopulated (in Fe–Ge) with respect to the stoichiometric composition of 25 at. % for the structure. The large difference in the number of Fe atoms associated with each of the binary alloys (72 and 82 at. %, respectively) is expected to significantly change the band structure and the magnetoelastic properties associated with each structure.

The following concurrent analyses were performed on the selected compositions of Fe–Ga–Ge alloys: (i) saturation magnetostriction measurements as a function of temperature ($77 \text{ K} < T < 300 \text{ K}$), (ii) elastic constants measurements as a function of temperature ($7 \text{ K} < T < 300 \text{ K}$) with resonant ultrasound spectroscopy, (iii) powder x-ray diffraction for structure

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analysis, and (iv) density functional calculations at 0 K for stability and elastic constant evaluation for the investigated structures.

II. EXPERIMENT

Single crystal ingots of $\text{Fe}_{1-x-y}\text{Ga}_x\text{Ge}_y$ were prepared at Ames Laboratory using the Bridgman method² with two parameters under control, the electron per atom ratio, e/a , and the y/x ratio. Following growth, all samples were slow-cooled (SC) at 10 K/min following postgrowth annealing at 900 °C. The compositions of the investigated alloys were measured to within 0.33 at. % by wavelength-dispersive x-ray spectroscopy and are listed in Table I, separated by the analysis to which they were subjected. Segregation during crystal growth made it difficult to maintain e/a close to the desired value of 1.55 in the synthesized samples, while such a problem does not exist for the modeled structures, also shown in Table I.

Tetragonal magnetostriction measurements were made with the standard rotating-gage technique³ for four alloy compositions between 77 and 300 K. The gage-instrumented disk samples were rotated in a magnetic field of 20 kOe, well above saturation. 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 four alloy compositions between 7 and 300 K. A magnetic field of 20 kOe, well above saturation, was applied along the longest side of the resonating rectangular parallelepiped during the measurements. Powder x-ray diffraction measurements were conducted at room temperature in a Panalytical X'Pert diffractometer with Cu K_α radiation at 45 kV and 40 mA. The scans were performed for 2θ ranging between 20° and 90°, with a 0.02° scan step. Silicon powder was used as an internal standard.

III. THEORETICAL CALCULATIONS

Density functional calculations using the Vienna *ab initio* simulation package^{4,5} (VASP) were performed to find the elastic constants of Fe–Ga–Ge alloys in a D0_3 configuration with compositions similar to those of the measured samples (Table I). The projector-augmented-wave⁶ (PAW) pseudopo-

TABLE I. Composition, e/a , and y/x values, for samples subjected to magnetostriction measurements (MS), resonant ultrasound spectroscopy (RUS), powder x-ray diffraction (XRD), and density functional calculations (DFC).

Analysis	$1-x-y$ (at. % Fe)	x (at. % Ga)	y (at. % Ge)	e/a	y/x
MS	76.2	22.5	1.3	1.49	0.06
	75.3	21.2	3.5	1.53	0.17
	80.5	13.7	5.8	1.45	0.42
	77.7	15.2	7.1	1.52	0.47
RUS	75.9	22.7	1.4	1.50	0.06
	75.3	21.2	3.5	1.53	0.17
	80.5	13.7	5.8	1.45	0.42
	78.6	14.8	6.6	1.49	0.45
XRD	73.4	25.0	1.6	1.55	0.06
	74.2	21.9	3.9	1.56	0.18
	76.6	15.6	7.8	1.55	0.50
	73.4	25.0	1.6	1.55	0.06
DFC	74.2	21.9	3.9	1.56	0.18
	76.6	15.6	7.8	1.55	0.50
	74.2	21.9	3.9	1.56	0.18
	76.6	15.6	7.8	1.55	0.50

tentials technique was used to represent the ionic cores, and the generalized-gradient approximation proposed by Perdew and Wang (GGA-PW91)⁷ was adopted in the formulation of the exchange-correlation functions. Since the arrangements of Ga and Ge atoms are unknown in the synthesized samples, a number (four to five) of cubic supercell models with 128 atoms were tested at each composition, with no Ga–Ga, Ge–Ge, or Ga–Ge first neighbors, and maintaining the D0_3 configuration. The lattice sizes and atomic positions of these structures were optimized through energy minimization procedures guided by atomic forces. The properties reported at each composition are a statistical average of the different possible structures according to Boltzmann statistics and the structure's total energy. An energy cutoff of 400 eV for the plane-wave basis was used. The Brillouin zone was sampled using a $(3 \times 3 \times 1)$ Monkhorst-Pack k -point mesh grid. The elastic constants were calculated by evaluating and fitting the total energy versus strain ($|\epsilon| < 0.02$) with third-order polynomials. The results for the different configurations at a given composition reveal that the arrangement of Ga and Ge atoms in the 128-atom cells affects the elastic constants mostly for structures where the Ge concentration is high (e.g., c' and c_{44} range between 6 and 23 GPa and between 118 and 129 GPa, respectively, for the $y = 7.8$).

IV. RESULTS AND DISCUSSION

The saturation values of the tetragonal magnetostriction, $(3/2)\lambda_{100} = \lambda^{\gamma,2}$, for the investigated Fe–Ga–Ge crystals are shown in Fig. 1(a) as a function of temperature. The results fall between those of the limiting binary alloys at the closest available e/a , $\text{Fe}_{72.7}\text{Ga}_{27.3}$ (Ref. 8) ($y = 0$ and $e/a = 1.55$) and $\text{Fe}_{82.3}\text{Ge}_{17.7}$ (Ref. 9) ($x = 0$ and $e/a = 1.53$). A positive $d\lambda^{\gamma,2}/dT$ is observed for all four samples. The key result here is the effect of a minute amount of Ge on the magnetostriction of Fe–Ga. At only 1.3 at. % Ge [label A in Fig. 1(a)] the magnetostriction of the ternary sample drops by 35% from that of Fe–Ga at 300 K, and as much as 80% at 77 K. The values of $\lambda^{\gamma,2}$ continue to drop vigorously as y increases. For

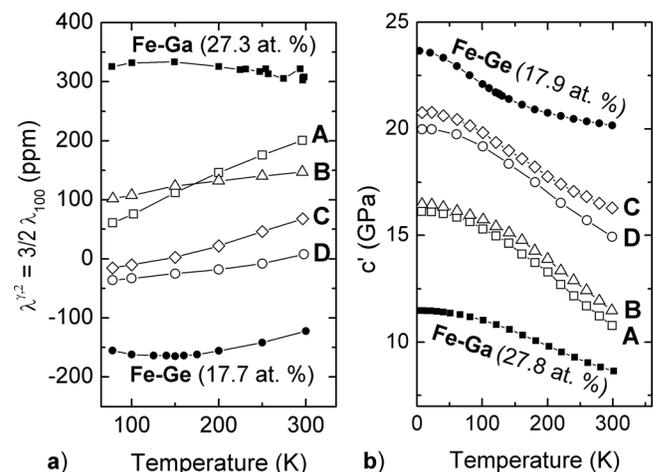


FIG. 1. Tetragonal magnetostriction (a) and shear modulus (b) of the investigated $\text{Fe}_{1-x-y}\text{Ga}_x\text{Ge}_y$ single crystals; data for the binary Fe–Ga (Refs. 8 and 10) and Fe–Ge (Ref. 9) alloys is shown for comparison. Labels A, B, C, and D, are used in order for the samples listed in Table I, with A corresponding to the lowest Ge composition sample.

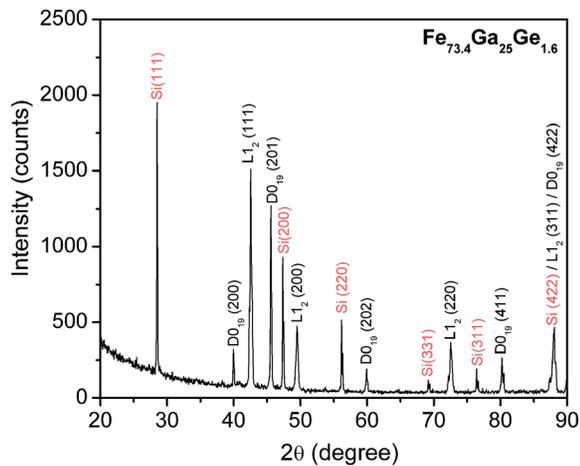


FIG. 2. (Color online) Powder x-ray spectrum of the $y = 1.6$ at. % Ge ternary sample showing a mixture of $L1_2$ and $D0_{19}$ phases.

the 7.1 at. % Ge sample [label D in Fig. 1(a)], which contains twice as much Ga as Ge, $\lambda^{7,2}$ exhibits mostly negative values that are closer to those of Fe–Ge than those of Fe–Ga.

The tetragonal shear constant, c' , for the investigated Fe–Ga–Ge crystals is shown in Fig. 1(b) as a function of temperature. Again, the results fall in between those of the limiting binary alloys, $Fe_{72.2}Ga_{27.8}$ (Ref. 10) ($y = 0$ and $e/a = 1.56$) and $Fe_{82.1}Ge_{17.9}$ (Ref. 9) ($x = 0$ and $e/a = 1.54$). Typical for high-solute Fe alloys,¹ a large temperature dependence, linked with lattice anharmonicity, is observed for c' , whereas the c_{44} shear modulus (not shown) is practically constant. The drop of c' from 7 to 300 K is between 33% [for the 1.4 at. % Ge sample, labeled A in Fig. 1(b)] and 21% [for the 5.8 at. % Ge sample, labeled C in Fig. 1(b)]. Variations are expected in the values of the measured constants as a function of growth-history/heat-treatment and/or e/a . Note that the C-labeled ternary sample has a particularly low e/a , of 1.45.

The powder XRD analysis reveals that already at $y = 1.6$ at. % Ge the structure of the ternary alloy differs significantly from that of the metastable $D0_3$ structure² of SC Fe–Ga. Two phases were found in the 1.6 at. % sample (spectrum shown in Fig. 2), the fcc $L1_2$ and the hexagonal $D0_{19}$. Coincidentally, these are the stable phases present in the assessed phase diagram¹ of Fe–Ga at $x \sim 28$ at. %. The lower-temperature ($T < 600$ K) phase, $L1_2$, is expected since the samples were slow-cooled. Also, at this composition, the density functional theory calculations indicate that the $D0_{19}$ and $L1_2$ phases have energies that are lower (more stable) than that of the $D0_3$ phase by 0.13 and 0.19 eV per Fe_3Ga unit, respectively. A similar result was obtained for the 3.9 at. % Ge sample, with stronger lines for the $D0_{19}$ phase. However, a significantly different result was obtained for the 7.8 at. % Ge sample, where single-phase $D0_3$ was found. This is, coincidentally, the equilibrium phase of the binary Fe–Ge alloy at $y \sim 18$ at. % found to be stable in both experimental and theoretical investigations.^{9,11} These results indicate that the metastable formation of $D0_3$ in the binary Fe–Ga system is quickly suppressed with minor additions of Ge. On further addition of Ge, the stability is reversed and we speculate, based on the theoretical calculations summarized below, that a stable $D0_3$ structure forms in $Fe_{1-x-y}Ga_xGe_y$ alloys with $y \geq 7.8$.

TABLE II. Theoretical and experimental (lowest temperature) shear elastic constants, c' and c_{44} , at the investigated compositions.

Composition	Theory ($T = 0$ K)		Experiment ($T = 7$ K)	
	c' (GPa)	c_{44} (GPa)	Composition	c' (GPa) c_{44} (GPa)
$Fe_{73.4}Ga_{25.0}Ge_{1.6}$	-1.58	115.3	$Fe_{75.9}Ga_{22.7}Ge_{1.4}$	16.12 137.8
$Fe_{74.2}Ga_{21.9}Ge_{3.9}$	3.61	129.0	$Fe_{75.3}Ga_{21.2}Ge_{3.5}$	16.47 134.7
—	—	—	$Fe_{80.5}Ga_{13.7}Ge_{5.8}$	20.77 135.7
$Fe_{76.6}Ga_{15.6}Ge_{7.8}$	12.2	123.2	$Fe_{78.6}Ga_{14.8}Ge_{6.6}$	20.00 133.3

The elastic constants obtained from density functional calculations are listed in Table II alongside the experimental data at the lowest temperature. Recall that all calculations are based on the ideal $D0_3$ structure of Fe_3Ga , where Ga atoms are gradually replaced by Ge atoms and the equilibrium lattice spacing is optimized through energy minimization. The 1.6 at. % Ge configuration is found to be unstable ($c' < 0$) in the assumed $D0_3$ ground state. The next higher composition, with 3.9 at. % Ge, is stable but with a very low c' . The 7.8 at. % Ge composition has a c' value that is closest to the measured one; this is the first ternary composition (in order of increasing y) in which the $D0_3$ phase was found by XRD analysis.

In summary, the theory and the XRD results show that the $D0_3$ instability is removed at $y/x \geq 0.5$, at which compositions the structure's ground-state phase becomes $D0_3$. The result is consistent with previous studies in the $y/x \rightarrow \infty$ limit, i.e. on Fe–Ge^{2,9} alloys. Further theoretical studies indicate that the instability of $D0_3$ in the low-Ge (including $y = 0$) structures can be removed by applying a hydrostatic-type strain of 0.3% to the optimized structure¹² or by including small pockets of A2 phase throughout the structure. Thus, the positive and peculiarly high $\lambda^{7,2}$ value of Fe–Ga at $x \sim 28$ at. % could be the result of the high sensitivity of its metastable $D0_3$ structure.

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