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Magnetoelastic coupling in $\text{Fe}_{100-x}\text{Ge}_x$ single crystals with $4 < x < 18$

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In this paper we examine the elastic (c' and c_{44}) and magnetostrictive (λ_{100} and λ_{111}) behaviors of $\text{Fe}_{100-x}\text{Ge}_x$ for $4 < x < 18$, quantities used further to find the fundamental magnetoelastic coupling constants b_1 and b_2 at room temperature. The x dependence of b_1 and b_2 for $\text{Fe}_{100-x}\text{Ge}_x$ is contrasted to those of $\text{Fe}_{100-x}\text{Ga}_x$ and $\text{Fe}_{100-x}\text{Al}_x$. While the rhombohedral shear elastic constant c_{44} is almost insensitive to the type and amount of solute, the tetragonal shear constant c' shows a pronounced and rapid softening with increasing x for all three alloys but with different decreasing slopes. Similarly, while the rhombohedral magnetostriction λ_{111} behavior is analogous for all three alloy systems, showing a sign change from negative to positive at the onset of chemical order, the tetragonal magnetostriction λ_{100} behavior differs. For the Ga and Al alloys, λ_{100} maintains positive values over the entire x range, both curves showing large peak values, whereas λ_{100} of $\text{Fe}_{100-x}\text{Ge}_x$ exhibits a moderate positive peak followed by a negative dip, both of comparable magnitude. Finally the tetragonal coupling constant $-b_1$ of Fe–Ge shows a marked, sharp decrease as chemical order occurs at $x \sim 12$ at. % Ge. The decline continues until the ordered $D0_3$ phase is fully established at $x \sim 18$ at. % Ge. The peak value of $|b_1|$ for Fe–Ge is approximately half of those for Fe–Ga and Fe–Al. This smaller value of $|b_1|$, obtained for the higher electron concentration Ge alloy, is consistent with predictions based on band structure calculations. The rhombohedral coupling constant $-b_2$ shows a consistent sign change at the occurrence of chemical ordering in both Fe–Ga and Fe–Ge. © 2009 American Institute of Physics. [DOI: 10.1063/1.3061864]

I. INTRODUCTION

The $\text{Fe}_{100-x}\text{X}_x$ ($\text{X}=\text{Ga}, \text{Al}, \text{Ge}, \text{Si}, \text{Zn}, \text{Be}$) alloys are a new class of materials designed for energy conversion applications due to their significant magnetostrictive properties. Despite the concentrated efforts directed toward Fe–Ga, the origin of its large and double-peaked magnetostriction¹ is not well understood. One source of the difficulties lies in the complexity of phases present in the alloy at Ga concentrations higher than 18 at. %.² In contrast, the Fe–Ge alloy has the advantage of a clean and reproducible phase distribution, as indicated by its phase diagram.² The magnetostriction of furnace-cooled single crystals of Fe–Ge has been recently investigated³ up to compositions of 18 at. % Ge. It was found that the increase in the tetragonal magnetostriction λ_{100} is directly linked to the structural changes that take place in the alloy. The structure was investigated with transmission electron microscopy (TEM) and nanobeam diffraction. In this paper we report (i) new measurements of magnetostriction for Fe–Ge single crystals in the quenched state for $4 < x < 18$, (ii) the first measurements of elastic constants of Fe–Ge covering the range $6 < x < 18$, and (iii) calculations of the magnetoelastic coupling constants b_1 and b_2 , the fundamental quantities governing the piezomagnetic interactions in a ferromagnetic material. All measurements were per-

formed at a temperature of 300 K. Further, the coupling constants for three alloy systems, $\text{X}=\text{Ga}, \text{Ge}$, and Al , are compared. In the (Ga, Al) pair, the elements have a similar electronic configuration and produce a similar increase in $|b_1|$. For the (Ga, Ge) pair, the additional electron in Ge results in lowering the coupling constant $|b_1|$ significantly. The coupling constant b_2 shows a firm sign change at the occurrence of chemical ordering for both Fe–Ga and Fe–Ge. For Fe–Al b_2 has not yet been evaluated.

II. EXPERIMENT

Single crystal ingots of $\text{Fe}_{100-x}\text{Ge}_x$ ($x < 18$) were prepared using the Bridgman method.³ The disks used in Ref. 3 (6.35 mm in diameter and 2.5 mm in thickness) with the flat surfaces oriented along a (100) plane of the iron lattice were retreated thermally to obtain the quenched state at the same composition. Standard strain-gauge techniques^{1,3} were used to measure the saturation value of the tetragonal magnetostriction $(3/2)\lambda_{100}$ of the quenched samples. In addition, samples of three other compositions were tested in both the slow-cooled (SC) and the quenched (Q) states. Similar disks but oriented along a (110) plane of the iron lattice were used to measure the saturation value of the rhombohedral magnetostriction $(3/2)\lambda_{111}$ at five compositions between 4 and 18 at. % Ge. Additional single crystal samples ($6 < x < 18$) were prepared for measurements of elastic constants. The samples were cut and polished in the shape of parallelepi-

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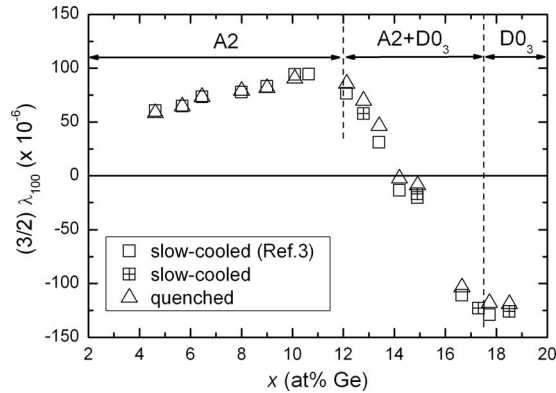


FIG. 1. Tetragonal magnetostriction $(3/2)\lambda_{100}$ for $\text{Fe}_{100-x}\text{Ge}_x$ single crystals in SC (squares) and Q (triangles) states at room temperature. The empty squares are a reproduction from the work of Wu *et al.* (Ref. 3).

peds (millimeter size) with faces oriented along (100) planes of the iron lattice. Resonant ultrasound spectroscopy⁴ was used on these samples to determine the two independent shear moduli (also known as elastic constants) of the alloy, $c' = (c_{11} - c_{12})/2$ and c_{44} , as a function of composition. Magnetostriction and elastic constants are the two variables upon which the magnetoelastic coupling constants b_1 and b_2 depend. The elastic modulus measurements were done in a saturating magnetic field of 15 kOe, oriented along the longest [100] axis of the parallelepiped. The composition of all samples was determined within 0.33 at. % by wavelength-dispersion spectroscopy.

III. RESULTS AND DISCUSSION

Shown in Fig. 1 and Table I are room temperature values of $(3/2)\lambda_{100}$ and $(3/2)\lambda_{111}$, respectively, as a function of Ge concentration and thermal treatment of the crystals. The correlation between composition and phase found by Wu *et al.*³ is marked on the figure. Their TEM analysis showed very clearly a pure A2 phase (disordered α -Fe) up to 10 at. % Ge, a single $D0_3$ phase above 17.5 at. % Ge, and a balanced mixture of the two phases between 12 and 16 at. % Ge. No B2 phase was found. Similar to other $\text{Fe}_{100-x}\text{X}_x$ systems studied before,^{5,6} λ_{100} of Fe–Ge increases monotonically up to the solubility limit ($x \sim 10$) before a sharp decrease caused by the ordering of the Ge atoms (the appearance of the $D0_3$ phase). Here, it is observed that, unlike in other alloys of the group, the quenching of the Fe–Ge crystals does not affect the magnetostriction value significantly at any composition. This behavior is consistent with a distinctive feature of the Fe–Ge phase diagram,² where phase boundaries are vertical (i.e., remain fixed at the same at. % Ge indifferent of tem-

TABLE I. Rhombohedral magnetostriction $(3/2)\lambda_{111}$ for $\text{Fe}_{100-x}\text{Ge}_x$ single crystals in SC and Q states at room temperature for x covering the transition from A2 to $D0_3$ phases.

		x (at. % Ge)					
		4.9	8.37	10.5	12.41	14.27	17.22
$(3/2)\lambda_{111}(\times 10^{-6})$	SC	−22.6	−15.4	−8.7	2.6	8.5	12.6
	Q	−21.2	−14.5	−10.1	2.6	8.3	13.8

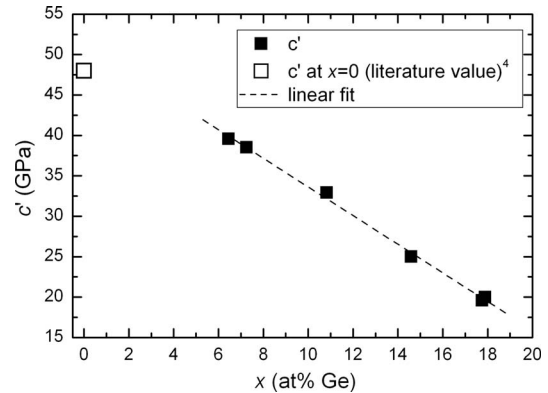


FIG. 2. Tetragonal shear modulus c' for $\text{Fe}_{100-x}\text{Ge}_x$ single crystals in SC state at room temperature. Measurements were taken in a saturating magnetic field of 15 kOe. The value at $x=0$ is extracted from literature (Ref. 4).

perature). Further, the $D0_3$ phase is stable at high temperatures where precipitation occurs readily; once nucleation is initiated, the proportion of A2 and $D0_3$ phases remains fixed indifferent of cooling rate. Comparing the Ge and Ga systems,² the finite slope of the A2/ $D0_3$ phase boundary in Fe–Ga suggests that cooling rate can greatly affect the relative amount of A2 retained in the crystal. Quenching Fe–Ga from high temperatures has been shown to inhibit the formation of the ordered $D0_3$ phase, allowing the A2 single phase to be maintained at compositions up to ~ 21 at. % Ga. Delaying the ordering (i.e., the formation of the $D0_3$ phase) in Fe–Ga resulted in a further increase in the $(3/2)\lambda_{100}$ peak value from 340 ppm (SC) to 398 ppm (Q).

In contrast to λ_{100} , $|\lambda_{111}|$ is only modestly affected by the amount of Ge present in the alloy. The sign of λ_{111} , however, changes as chemical ordering occurs at $x \sim 12$. The same behavior has been observed in Fe–Ga.¹

Shown in Fig. 2 and Table II are room temperature values of the two shear moduli, c' and c_{44} , respectively, as a function of Ge concentration. The presence of the Ge atoms in the host Fe lattice is felt quickly by the tetragonal shear modulus c' , which shows a pronounced and rapid softening. The rhombohedral shear modulus c_{44} does not change its value significantly with x , mimicking the behavior of λ_{111} . These trends, for both c' and c_{44} , were also observed in the Fe–Ga system.⁴ Given the insensitivity of the magnetostriction of Fe–Ge to the cooling rate (Fig. 1 and Table I) and considering previous results on Fe–Ga (Ref. 4) which did not show a significant change in the elastic constant values for different cooling rates, the elastic constants of Fe–Ge measured and presented here are only for samples in the SC state.

Using the magnetostriction and elastic constant values, the tetragonal and rhombohedral magnetoelastic coupling constants were calculated using the relationships $-b_1 = 3\lambda_{100}c'$ and $-b_2 = 3\lambda_{111}c_{44}$. Since λ and c were measured at

TABLE II. Rhombohedral shear modulus c_{44} for $\text{Fe}_{100-x}\text{Ge}_x$ single crystals in SC state at room temperature. Measurements were taken in a saturating magnetic field of 15 kOe.

x (at. % Ge)	0 ^a	6.45	7.25	10.83	14.59	17.75	17.85
c_{44} (GPa)	116	120.3	122.9	124.4	124.2	126.7	124.8

^aPure Fe data from literature (Ref. 4).

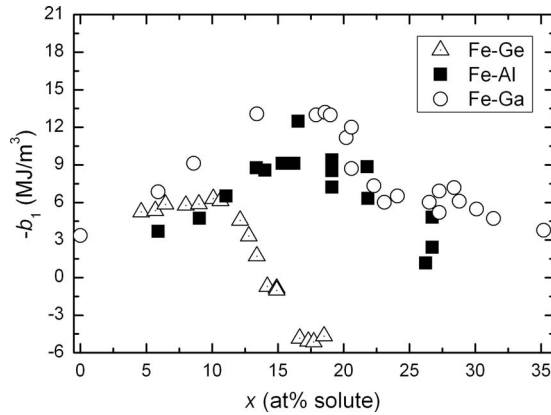


FIG. 3. Comparison of room temperature magnetoelastic coupling constant $-b_1 = 3\lambda_{100}c'$ vs x for $\text{Fe}_{100-x}\text{X}_x$ ($\text{X} = \text{Ga}, \text{Al}, \text{Ge}$).

different x values, extrapolations were necessary. For the calculation of $-b_1$, c' was fitted (dashed curve in Fig. 2) with a linear function. $-b_1$ was then evaluated at the same x values where the tetragonal magnetostriction (in the SC state) was measured. The results are shown in Fig. 3, together with $-b_1$ of two other alloys, $\text{Fe}_{100-x}\text{Ga}_x$ and $\text{Fe}_{100-x}\text{Al}_x$. The Al data are a direct reproduction from Ref. 7. The Ga data were processed using λ and c values from Ref. 4 (c' versus x was fitted with a linear function as well; details of this calculation, including Fe–Ga $-b_1$ temperature dependence, will be given in a future report).

The comparison between the three Fe-based alloys is made in direct relation to the relative location within the Periodic Table of the guest elements. Al and Ga, with similar electronic configuration, produce a significant and comparable increase in the coupling factor $-b_1$. The extra electron present in Ge has a powerful effect on $-b_1$, lowering its magnitude considerably. The coupling constant $-b_1$ for Fe–Ge does not vary much in absolute value ($\sim 6 \text{ MJ/m}^3$) but changes sign at the transition from the A2 to the D0_3 phase. For compositions where at least 50% of the volume is in the D0_3 phase ($x \sim 14 \text{ at. \% Ge}$), $-b_1$ is negative. This is a feature that is unique to Fe–Ge within the three systems compared. The pronounced softening of the tetragonal shear modulus c' is a common feature for Fe–Al, Fe–Ga, and Fe–Ge.

Focusing on Fig. 3 again, it is observed that the second peak in $-b_1$ versus x for Fe–Ga (at $x \sim 29$), although not as predominant as in λ_{100} versus x , does not appear at all in the other two systems. This interesting peculiarity of Fe–Ga cannot be assessed from the comparison with Fe–Al or Fe–Ge.

The results for $-b_2$ are listed in Table III. A linear fit was used to obtain c_{44} at the x values where λ_{111} was measured. The SC magnetostriction data were used for the $-b_2$ calculations as well. Note that the behavior of $-b_2(x)$ mirrors that of $\lambda_{111}(x)$, as c_{44} has a much weaker dependence on x . Because of the low $|b_1|$ values, the two coupling factors for Fe–Ge are

TABLE III. Magnetoelastic coupling constant $-b_2 = 3\lambda_{111}c_{44}$ for $\text{Fe}_{100-x}\text{Ge}_x$ (SC) at room temperature.

x (at. % Ge)	4.9	8.37	10.5	12.41	14.27	17.22
$-b_2$ (MJ/m^3)	−8.13	−5.62	−3.19	0.96	3.18	4.76

essentially comparable in absolute value. In contrast, $-b_2$ of Fe–Ga varies from a nearly constant of -6.7 MJ/m^3 at $6 < x < 20$ (A2 phase) to 11 MJ/m^3 at $x > 20$ (D0_3 phase and possibly others),⁶ with an overall maximum of $|b_1/b_2|$ of ~ 2 .

IV. SUMMARY

The effect of chemical ordering just above the solubility limit is indeed detrimental to the tetragonal magnetostriction λ_{100} , as seen in both the SC and Q states of Fe–Ge. It is concluded that, if rapid quenching can preserve the disordered A2 phase further, assuming the phase diagram allows it, an increase in magnetostriction and $|b_1|$ can be obtained (as observed in the Fe–Ga case). An indicator that chemical order in the alloy was initiated is the sign change in λ_{111} , and consequently in $-b_2$.

The comparison between three iron-rich alloys $\text{Fe}_{100-x}\text{X}_x$ ($\text{X} = \text{Ga}, \text{Al}, \text{Ge}$) shows that the strong magnetoelastic coupling factor $-b_1$ (that corresponds to a tetragonal distortion) in the Fe–Ga and Fe–Al crystals is drastically weakened when the solute X bears an extra electron, such as in the case of the Fe–Ge crystal. This observation is consistent with band filling calculations⁸ and suggests Zn additions as a possible candidate for obtaining elevated magnetoelastic effects.

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