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Combinatorial Investigation of Magnetostriction in Fe-Fa and Fe-Ga-Al

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Combinatorial investigation of magnetostriction in Fe–Ga and Fe–Ga–Al

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A high-throughput high-sensitivity optical technique for measuring magnetostriction of thin-film composition-spread samples has been developed. It determines the magnetostriction by measuring the induced deflection of micromachined cantilever unimorph samples. Magnetostriction measurements have been performed on as-deposited Fe–Ga and Fe–Ga–Al thin-film composition spreads. The thin-film Fe–Ga spreads display a similar compositional variation of magnetostriction as bulk. A previously undiscovered peak in magnetostriction at low Ga content was also observed and attributed to a maximum in the magnetocrystalline anisotropy. Magnetostrictive mapping of the Fe–Ga–Al ternary system reveals the possibility of substituting up to 8 at. % Al in Fe₇₀Ga₃₀ without significant degradation of magnetostriction. © 2008 American Institute of Physics. [DOI: 10.1063/1.2980034]

The discovery of large magnetostriction in Fe_{100-x}Ga_x (15 < x < 31) in 2000 has stimulated attempts to discover related materials with high magnetostriction.^{1–3} In particular, the embrittlement of Fe–Ga, caused by the large amount of included Ga, has led to significant efforts to at least partially replace the Ga content with a suitable ternary element. The goal of the ternary alloying is to increase the overall workability of the material, in particular its ability to be hot rolled without brittle fracture, while preserving or perhaps enhancing the magnetostriction.^{4–7}

The majority of the alloying work has been performed with traditional bulk samples, where limited compositional sampling and run-to-run variations can cause promising compositions to be overlooked. To avoid this and to sample the large and complicated composition-property phase space inherent in this system, it is of interest to apply the combinatorial strategy. In this approach, hundreds of samples with varying compositions are deposited in a single deposition, processed simultaneously, and then rapidly characterized for their figure of merit.⁸

In this letter we describe a high-throughput high-sensitivity optical technique for measuring magnetostriction in composition spreads. By monitoring the deflection of cantilever unimorph composition spreads, the magnetostrictive properties across ternary phase diagrams can be rapidly mapped. Previously, cantilever arrays have been used to perform high-throughput mapping of shape memory alloys and residual strains in deposited thin films.^{9–11} In the present, experiment, this technique is combined with rapid x-ray diffraction analysis and transmission electron microscopy (TEM) to provide insight into the role of microstructure in magnetostrictive materials. Here, composition spreads of Fe–Ga and Fe–Ga–Al have been deposited and character-

ized. In binary Fe–Ga, bulk trends were reproduced in the high-Ga regime. Also, a previously unknown region of enhanced magnetostriction was observed. Measurements on Fe–Ga–Al reveal a large region of magnetostriction connecting the known Fe–Ga and Fe–Al magnetostrictive composition regions. In addition, compositions based on Fe₇₀Ga₃₀ containing as much as 8 at. % Al have been found which do not result in the degradation of magnetostriction as had been observed previously for similar compositions.^{6,7}

All samples were deposited in an ultrahigh vacuum sputtering chamber with three magnetron sputtering guns oriented in a nonconfocal geometry, the details of which can be found elsewhere.¹⁰ The base pressure before deposition was lower than 4×10^{-11} Pa, and depositions were carried out in high purity Ar (99.9995%) at pressure of .6 Pa. The substrates were 3 in. (100) Si wafers with 3 μ m of thermally oxidized SiO₂ which had been patterned into an array of ~ 100 cantilevers, using the standard Si bulk micromachining technique. Each cantilever was 8.5 mm in length \times 2 mm wide and approximately 65 μ m thick. The samples were deposited at room temperature, with a target-sample distance of 12.5 cm. To deposit binary spreads of Fe–Ga a pure Fe target and an intermetallic Fe₂Ga₃ target were cosputtered at 75 W and 40 W, respectively. Ternary Fe–Ga–Al spreads were deposited using the same powers for the Fe and Fe₂Ga₃ targets as the binary Fe–Ga samples but cosputtered with an Al target sputtered at 50 W. We have used the quenching of high-temperature phases that is inherent to sputtering depositions onto room temperature substrates for phase formation, and no postdeposition annealing was carried out. The film thickness after deposition was measured via cross-sectional scanning electron microscopy and determined to be about 0.5 μ m.

To measure the magnetostriction of the unimorph cantilever composition-spread samples, the deflection of a laser

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reflected off each cantilever was monitored as a function of applied magnetic field. During measurements the wafer was placed on an *X-Y-Z* stage, which was located between the pole faces of an electromagnet with a field range of ± 2 T. The electromagnet was placed on a rotatable table, which allowed the magnetic field and the cantilevers to be rotated independently. This allowed the cantilevers to be oriented so that during measurement the magnetic field was applied along the long axis of the cantilever. A 635 nm power-stabilized diode laser (20 mW) was used as the light source. A condensing lens with a 0.5 m focal length was used to focus the spot size down to a diameter of less than 0.3 mm, allowing individual measurement of each cantilever. The reflected light was collected with a position sensitive detector (PSD). A light chopper operating at 400 Hz was used to modulate the light intensity, allowing for lock-in detection of the PSD output, yielding sensitivity to the change in cantilever angle of $90 \mu\text{rad}$. Magnetic torque, caused by a misalignment of the plane containing the cantilever's magnetic moment and the magnetic field, was minimized prior to each cantilever measurement.

The raw deflection of the cantilever is converted to an effective magnetostrictive constant λ_{eff} through the model placed, forth by du Tremolet de Lachiesserie and Peuzin,¹²

$$\lambda_{\text{eff}} = \frac{2(D_{\parallel} - D_{\perp})E_s t_s^2 (1 + \nu_f)}{9E_f L^2 t_f (1 + \nu_s)}, \quad (1)$$

where L is the length, E_f and E_s are Young's moduli of the film and substrate, respectively, t_f and t_s are their respective thicknesses, and ν_s and ν_f are their respective Poisson ratios. D_{\parallel} and D_{\perp} represent the measured displacement of the tip of the cantilever out of the plane of the cantilever when the field is applied in the plane of the cantilever, parallel and perpendicular to its long axis, respectively.

All of the constants for the Si substrate are known. For Fe–Ga, however, it is difficult to extrapolate bulk values of $E_f/(1 + \nu_f)$ to their thin-film values.¹² In previous studies of magnetostriction in various Fe containing material systems, both amorphous and crystalline, this ratio was set to 50 GPa.^{13,14} Here, we follow this convention to facilitate comparisons with other work. In addition, in order to minimize the time required for characterization, magnetic fields were only applied along the length of the cantilever (D_{\parallel}), and it is assumed that D_{\perp} is zero. This means that the magnitude of the values reported here represent a lower bound for the magnetostriction, as volume conservation dictates that the two deflections are opposite in sign.

It should be noted that the calculation of λ_{eff} with Eq. (1) is carried out without regard to the crystalline orientation of the sample with respect to the magnetic field, and thus the cubic magnetostriction constants λ_{100} and λ_{111} are not explicitly contained in the formula. If the crystalline orientation of the sample is determined, the relationship of λ_{eff} to the cubic magnetostriction constants can be explicitly derived.

Fe–Ga is a well-known magnetostrictive materials system, which has been shown to have λ_{100} values as high as 270 ppm (or a change in length of .027%). Fe–Ga exhibits a nonmonotonic dependence of magnetostriction on Ga concentration: as the amount of Ga is increased from pure Fe two clear maxima develop in the magnetostriction. The first maximum occurs around $\text{Fe}_{81}\text{Ga}_{19}$ and is thought to be associated with establishing a local order.⁵ The second maximum

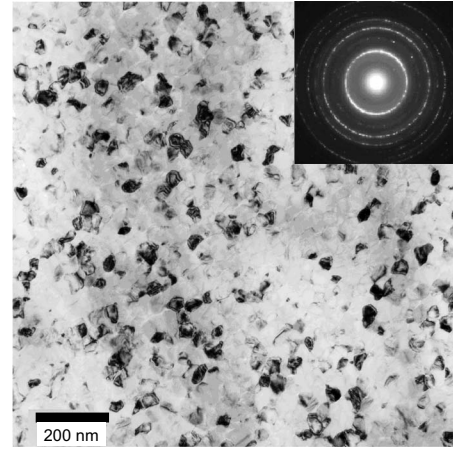


FIG. 1. Plan-view TEM image of an as-deposited $\text{Fe}_{67}\text{Ga}_{33}$ thin film showing nanocrystalline morphology with an average grain size of 25 nm. The inset is a selected area diffraction pattern.

occurs around $\text{Fe}_{70}\text{Ga}_{30}$ and is caused by a drop in the elastic constant C' , which enhances the magnetostriction.^{1,15}

X-ray diffraction and TEM of the as-deposited composition-spread samples showed our films to be randomly oriented polycrystalline throughout the investigated composition range. From these measurements, calculation of λ_{100} is possible through the standard polycrystalline formula.¹⁶ In the range where λ_{100} is much larger than λ_{111} , λ_{111} can be ignored and λ_{eff} is dominated by λ_{100} . An approximate value for λ_{100} can then be calculated from λ_{eff} through the relation $\lambda_{\text{eff}} = \frac{2}{5}\lambda_{100}$. In the bulk literature magnetostriction is generally reported as $\frac{3}{2}\lambda_{100}$, and we will use this convention throughout this letter. Plan-view TEM studies (Fig. 1) showed the samples to be nanocrystalline with an average grain size of 25 nm across the entire spread. The ring pattern of the selected area diffraction (Fig. 1 inset) shows the sample to be polycrystalline with all possible orientations appearing. More detailed microstructural studies of Fe–Ga thin films will appear elsewhere.

Figure 2 presents the magnetostriction, plotted as $\frac{3}{2}\lambda_{100}$, measured from a thin-film cantilever bimorph spread sample of Fe–Ga. To compare the trend, the data are plotted with bulk data from Clark *et al.*¹⁷ From the figure it is clear that the bulk trend of magnetostriction as a function of Ga concentration is captured. The two peaks in magnetostriction appear at $x=21$ and 32 with a maximum value for $\frac{3}{2}\lambda_{100}$ of ≈ 190 ppm. The magnetostriction values deduced from our thin-film samples are roughly half of the values reported for bulk. There are a number of reasons for this including residual strain from deposition and the fact that we are reporting a lower bound, as discussed above. The discrepancy in the positions of the peaks could be due to the compositional variation across each cantilever. From wave dispersive spectroscopy measurements, this compositional variation was found to typically be 1.5 at. %. In addition, it is known that the position of the first peak in magnetostriction is very sensitive to the thermal treatment history of the samples.¹⁷ It is possible that the high rate of quenching of the thin-film samples from the plasma suppresses the onset of ordering to higher Ga contents.

A close look at the low Ga tail of the graph reveals a minor peak in magnetostriction at about $x=4.5$. This third peak occurs in the composition region between $3 < x < 9$ and

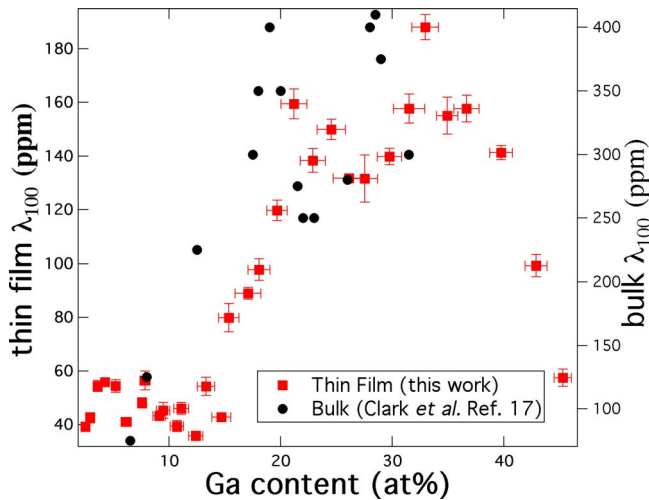


FIG. 2. (Color online) $\frac{3}{2}\lambda_{100}$ measured from Fe–Ga thin-film composition spread plotted against values from previous bulk studies (Ref. 17) as a function of Ga content. The compositional trend and values agree well with bulk values. A third minor maximum is observed at a Ga concentration of 4.5 at. %. The error bars represent the standard deviation of the magnetostriction estimated from the experimental noise after smoothing and were taken at each measurement point.

displays a maximum $\frac{3}{2}\lambda_{100}$ of ≈ 60 ppm at $x=4.5$. Bulk samples in the region of this maximum, made by arc-melting, were tested for their magnetostriction and exhibited an identical trend to the thin-film samples. From previous bulk studies of this region it is known that there is a local maximum of magnetocrystalline anisotropy and the net magnetic moment of individual Fe atoms at this composition.¹⁸ Therefore the peak in magnetostriction at 5 at. % Ga is most likely related to the peak in the magnetocrystalline anisotropy.

Figure 3 plots the variation of $\frac{3}{2}\lambda_{100}$ in ternary Fe–Ga–Al. A composition region displaying large magnetostriction extending from Fe_{100-x}Ga_x ($0 < x < 30$) to Fe_{100-x}Al_x ($0 < x < 25$) is observed. In a previous study where Al was alloyed with Fe–Ga, it was found that near the compound Fe₆₃Ga_{27-x}Al_x, λ_{100} decreases by 36% with the addition of 3 at. % Al.^{6,7} We find that as long as one stays closely within

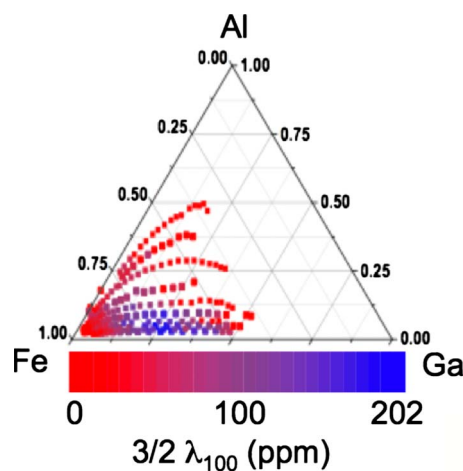


FIG. 3. (Color online) $\frac{3}{2}\lambda_{100}$ measured from a Fe–Ga–Al composition spread. A region of large magnetostriction is seen stretching from Fe–Ga to Fe–Al.

the narrow range of ternary composition region connecting Fe_{100-x}Ga_x ($0 < x < 30$) and Fe_{100-x}Al_x ($0 < x < 25$), magnetostriction is maintained as Al is substituted for Ga. In fact, our data plotted in Fig. 3 show that it is possible to substitute as much as ≈ 7 at. % Al to Fe₇₀Ga₃₀ and only decrease λ_{100} by $\approx 18\%$.

In conclusion a high-throughput measurement technique for measuring magnetostriction in thin-film bimorph cantilever composition-spread samples has been developed. The technique measures the deflection of the cantilevers, which can then be used to calculate the λ_{100} values for each composition. The technique was tested on the well-known binary magnetostrictive material Fe–Ga as well as on ternary Fe–Ga–Al. For Fe–Ga, the bulk trend of magnetostriction as a function of Ga content was reproduced. A third maximum in magnetostriction was observed, with a value of ≈ 60 ppm at 4.5 at. % Ga, which had not been reported previously in bulk studies and is most likely related to the peak in magnetocrystalline anisotropy and magnetization per Fe atom. In Fe–Ga–Al, an approach for alloying Al in to Fe₇₀Ga₃₀ while preserving the magnitude of magnetostriction has been demonstrated.

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