Magnetostrictive effect in single crystal $Fe_{1-x}Ga_x$ thin films

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The magnetic properties of single crystal $Fe_{1-x}Ga_x$ thin films deposited on ZnSe/GaAs(001) and MgO(001) substrates by molecular beam epitaxy were investigated by vibrating sample magnetometry and angle dependent ferromagnetic resonance. Depositions on the ZnSe buffer layer feature a strong uniaxial anisotropy that scales with the thin film magnetostriction of the samples, while depositions on MgO(001) substrates result in a purely cubic anisotropy whose cubic anisotropy constant, K_1, switches sign at a lower Ga concentration than is seen in bulk. © *2010 American Institute of Physics*. [doi:10.1063/1.3367971]

I. INTRODUCTION

Pinning a highly magnetostrictive material to a substrate and applying a magnetic field will impart an anisotropic stress, controllable by the magnetic field, that may modify the magnetic properties of the film including the magnetic anisotropy and magnetization dynamics. Candidates for these pinned films are often found in materials that have had success in bulk applications. Alloys of bulk $Fe_{1-x}Ga_x$ have generated recent interest because of their unique magnetostrictive and high tensile loading properties.[1,2] Of particular interest is the large and anisotropic behavior of the magnetostriction as a function of Ga concentration, where $(3/2)\lambda_{100}$ features a double peak with values exceeding 400 ppm.[1,2] The first peak near $x=0.19$ is due to a maximum in the magnetoelastic coupling constant, B_1, while the second peak around $x=0.27$ is related to the softening of the elastic shear constant, $(c_{11}-c_{12})/2$, as a result of the crystal undergoing a bcc to bct transition.[2–4] Furthermore, it is possible to induce large magnetostriction in these materials with a small applied magnetic field.[5] In thin film form, the epitaxial pinning of this highly magnetostrictive material may generate large variations in the magnetic anisotropy with Ga concentration that can be compared to the bulk behavior.

II. EXPERIMENTAL

Single crystal $Fe_{1-x}Ga_x$ thin films (~17 nm) of various Ga concentrations were prepared on GaAs(001), with a 75 nm ZnSe buffer layer, and MgO(001) substrates by molecular beam epitaxy as previously described.[6,7] All samples were capped with 4 nm Al to prevent the formation of any oxides. The thicknesses and concentrations of the films were determined by Rutherford backscattering. The magnetic properties of the samples were investigated using vibrating sample magnetometry and angle dependent ferromagnetic resonance (FMR).

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III. RESULTS AND DISCUSSION

As shown by the magnetic hysteresis loops in Fig. 1, $Fe_{1-x}Ga_x$ thin films grown on MgO(001) and ZnSe/GaAs(001) substrates have very different responses to an applied magnetic field. The $Fe_{0.85}Ga_{0.15}$ samples deposited on MgO have a purely cubic anisotropy [Fig. 1(a)], whereas

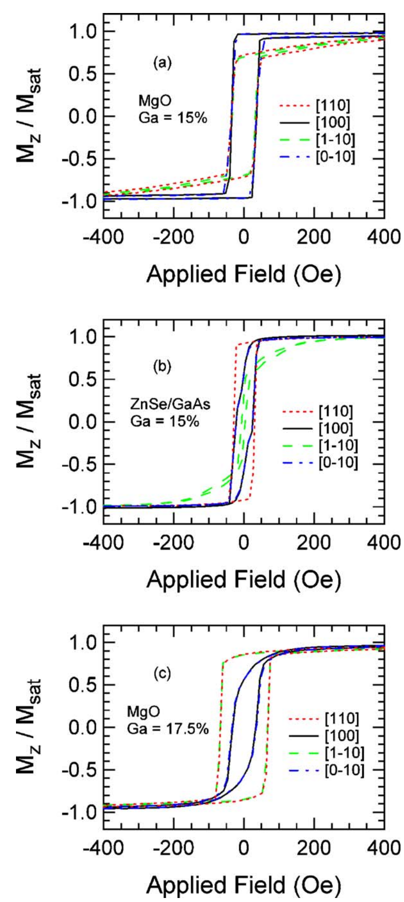

FIG. 1. (Color online) Depositions of single crystal $Fe_{1-x}Ga_x$ thin films on ZnSe/GaAs(001) substrates (b) have a strong uniaxial anisotropy, while depositions on MgO(001) substrates [(a) and (c)] are purely cubic. The switching of the easy and hard axes between $Fe_{0.85}Ga_{0.15}$ (a) and $Fe_{0.825}Ga_{0.175}$ (c) indicates a change in sign of the cubic anisotropy constant, K_1.

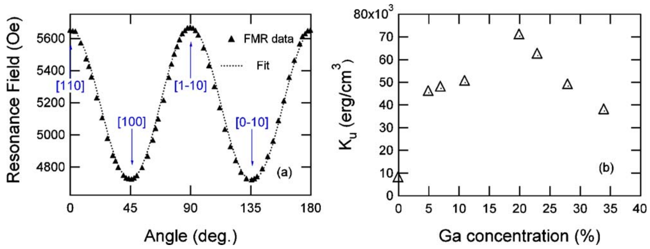

FIG. 2. (Color online) The reduced experimental Q-band FMR data for a 17 nm pure iron film deposited on the ZnSe buffer layer (triangles) (a), and the effective uniaxial anisotropy of $Fe_{1-x}Ga_x$ samples deposited on ZnSe/GaAs(001) as a function of Ga concentration, which features a single peak near 20% Ga (b) ($K_u^{eff}=0$ for depositions on MgO).

samples deposited on the directionally bonded ZnSe buffer layer feature a strong uniaxial anisotropy [Fig. 1(b)].

The cubic and uniaxial anisotropies in our $Fe_{1-x}Ga_x$ samples deposited on ZnSe/GaAs(001) have been quantified by angle dependent FMR using the following form for the free energy density of a ferromagnetic thin film:

$$E = -\boldsymbol{M}\cdot\boldsymbol{H} + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + 2\pi(\boldsymbol{M}\cdot\hat{\boldsymbol{n}})^2 + K_u^{eff}\sin^2(\alpha-\alpha_u) - K_n(\boldsymbol{M}\cdot\hat{\boldsymbol{n}}/M)^2. \quad (1)$$

The five terms are the Zeeman energy; the cubic magnetocrystalline anisotropy, with α_1, α_2, and α_3 being the typical direction cosines; the demagnetization energy for a film; the in-plane uniaxial magnetic anisotropy, where K_u^{eff} includes any effective contributions from the substrate, capping layer, magnetoelastic coupling, etc., and α being the angle of the magnetization in the film plane and α_u the angle of the easy axis of the in-plane uniaxial anisotropy (in this case along [110]); and lastly the large perpendicular anisotropy that forces the magnetization in the film plane. The magnetic anisotropies were computed using the above expression for the free energy and the equations for the FMR condition,[8] and the experimental data acquired at 34.6 GHz (Q-band) and fit for a pure iron film are shown in Fig. 2(a) using $K_u^{eff}=8\times10^3$ ergs/cm^3 and $K_1=5.2\times10^5$ ergs/cm^3. The strength of the uniaxial anisotropy as a function of Ga concentration is shown in Fig. 2(b) and exhibits a single peak near $x=0.20$.

The presence of a uniaxial magnetic anisotropy for Fe thin films deposited on zinc-blende surfaces has been well established,[9–11] and has been attributed to magnetostriction and an anisotropic strain relaxation that has its origins in the directional bonding from the zinc-blende surface.[11,12] The anisotropic strain relaxation creates an elastic shear strain, and through magnetoelastic coupling introduces an energy term that has a uniaxial angle dependence,[12] which is accounted for in our K_u^{eff} term. Epitaxial depositions on substrates lacking an anisotropic strain relaxation [e.g., MgO(001)] will lack a magnetoelastically coupled uniaxial magnetic anisotropy. However, for substrates compliant to an elastic shear strain, the effect of an enhanced magnetostriction is to enhance the anisotropy of the strain relaxation. Therefore, one would expect the strength of the uniaxial term to scale with the magnetostriction.

The trend of the FMR extracted uniaxial anisotropy as a function of Ga concentration, including the single peak near $Fe_{0.80}Ga_{0.20}$, is reminiscent of the magnetostriction of bulk $Fe_{1-x}Ga_x$ alloys, but an expected second peak in the uniaxial anisotropy is not observed in thin films. The absence of a second peak can be explained as follows. A first principles calculation of magnetostriction is determined by minimizing the free energy of a material with respect to the elastic constants. In bulk form, all of the elastic constants are free parameters and the minimization results in the usual expression for the magnetostriction: $(3/2)\lambda_{100}^{bulk}=-B_1/(c_{11}-c_{12})$.[13,14] However, there is only one free parameter for any thin film epitaxially pinned to a substrate, and the resulting magnetostriction is: $(3/2)\lambda_{100}^{film}=-B_1/c_{11}$.[15] Bulk magnetoelastic and elastic constants are listed in Table I for several Ga concentrations (data taken from Ref. 16), along with the corresponding thin film magnetostriction, assuming the bulk values remain valid. It is interesting to note that the strength of the uniaxial anisotropy seen in the $Fe_{1-x}Ga_x$ thin films deposited on ZnSe/GaAs(001) substrates scales with the predicted thin film magnetostriction, most importantly that they both feature a single peak near a Ga concentration of 19%.

Comparing Figs. 1(a) and 1(c) reveals that the easy axis of magnetization has switched from ⟨100⟩ for $Fe_{0.85}Ga_{0.15}$ to ⟨110⟩ for $Fe_{0.825}Ga_{0.175}$ samples deposited on MgO substrates. This signals that the cubic anisotropy constant, K_1, has become negative by 17.5% Ga, in contrast to bulk $Fe_{1-x}Ga_x$ alloys where K_1 switches sign near 20% Ga.[17] Also, the shape of the hysteresis loops for the $Fe_{0.825}Ga_{0.175}$ sample and the low magnetic remanence of only 86% along the easy axis suggest that K_1, though negative at this Ga concentration, likely has a very small value.

It is well established that for cubic crystals the effect of magnetostriction is to modify the cubic anisotropy, where K_1 can be written as $(K+\Delta K)$ with $\Delta K=\{(c_{11}-c_{12})[(3/2)\lambda_{100}]^2 - 2c_{44}[(3/2)\lambda_{111}]^2\}$ being the contribution from magnetostriction.[13,14] For FeGa alloys this modification is to

TABLE I. The magnetoelastic energy constants, elastic constants, and bulk and thin film magnetostriction values of $Fe_{1-x}Ga_x$ alloys for several Ga concentrations (λ in ppm). Note the much reduced magnetostriction in thin film form, as well as the single peak [(data taken from (Ref. 16)].

Ga (%)	B_1 (MJ/m^3)	c_{11} (GPa)	c_{12} (GPa)	$(3/2)\lambda^{bulk}$	$(3/2)\lambda^{film}$
0	−2.88	237	141	30	12
18.7	−15.56	195.7	156.3	395	80
24.1	−5.02	186.1	167.5	270	27
27.2	−4.76	220.8	207.2	350	22

enhance K_1, but the much reduced thin film values of $(3/2)\lambda_{100}^{\text{film}}$ shown in Table I result in a negligible enhancement. However, the magnetostriction for bulk $Fe_{0.83}Ga_{0.17}$ alloys results in an enhancement of about 4×10^4 ergs/cm^3,[16] and can explain why K_1 remains positive for bulk $Fe_{0.825}Ga_{0.175}$ alloys, whereas in thin films K_1 has become negative by 17.5% Ga.

IV. SUMMARY

Depositions of $Fe_{1-x}Ga_x$ single crystal thin films grown on MgO(001) and ZnSe/GaAs(001) substrates have very different magnetic properties. The samples deposited on MgO have a purely cubic anisotropy, and it is found that the cubic anisotropy constant, K_1, switches sign at a lower Ga concentration than is seen in bulk $Fe_{1-x}Ga_x$. The enhancement in the bulk cubic anisotropy is attributed to the much larger bulk magnetostriction. Depositions on the ZnSe buffer layer feature a strong uniaxial anisotropy that scales with the thin film magnetostriction of the samples, and is attributed to an interplay between the magnetostriction and an anisotropic strain relaxation.

This work demonstrates that the effect of depositing magnetostrictive materials as thin films can be heavily influenced by the substrate. Depositions on substrates that allow anisotropic strain relaxation, such as GaAs and InAs,[18] may feature a strong uniaxial anisotropy, while depositions on nondirectionally bonded substrates can have a strongly modified cubic anisotropy. It is also important to highlight that the difference between the bulk and thin film magnetostriction values due to the bonding of the material to a substrate predicts that some highly magnetostrictive materials in the bulk, such as $Fe_{0.81}Ga_{0.19}$, may have significantly reduced values as thin film single crystals. Conversely, there possibly exist materials whose bulk magnetostriction is insignificant but have dramatically enhanced magnetostriction in the form of single crystal thin films.

ACKNOWLEDGMENTS

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[1] A. E. Clark, J. B. Restorff, M. Wun-Fogle, T. A. Lograsso, and D. L. Schlagel, IEEE Trans. Magn. **36**, 3238 (2000).
[2] A. E. Clark, K. B. Hathaway, M. Wun-Fogle, J. B. Restorff, T. A. Lograsso, V. M. Keppens, G. Petculescu, and R. A. Taylor, J. Appl. Phys. **93**, 8621 (2003).
[3] A. E. Clark, M. Wun-Fogle, and J. B. Restorff, IEEE Trans. Magn. **37**, 2678 (2001).
[4] M. Wuttig, L. Dai, and J. Cullen, Appl. Phys. Lett. **80**, 1135 (2002).
[5] R. A. Kellogg, A. B. Flatau, A. E. Clark, M. Wun-Fogle, and T. Lograsso, J. Intell. Mater. Syst. Struct. **16**, 471 (2005).
[6] A. McClure, S. Albert, T. Jaeger, H. Li, P. Rugheimer, J. A. Schaefer, and Y. U. Idzerda, J. Appl. Phys. **105**, 07A938 (2009).
[7] A. McClure, E. Arenholz, and Y. U. Idzerda, "Ferrimagnetic ordering of single crystal $Fe_{1-x}Ga_x$ thin films," J. Vac. Sci. Technol. (submitted).
[8] H. Suhl, Phys. Rev. **97**, 555 (1955).
[9] G. A. Prinz, G. T. Rado, and J. J. Krebs, J. Appl. Phys. **53**, 2087 (1982).
[10] J. J. Krebs, B. T. Jonker, and G. A. Prinz, J. Appl. Phys. **61**, 2596 (1987).
[11] J. J. Krebs, B. T. Jonker, and G. A. Prinz, J. Appl. Phys. **61**, 3744 (1987).
[12] O. Thomas, Q. Shen, P. Schieffer, N. Tournerie, and B. Lepine, Phys. Rev. Lett. **90**, 017205 (2003).
[13] C. Kittel, Rev. Mod. Phys. **21**, 541 (1949).
[14] R. C. O'Handley, *Modern Magnetic Materials Principles and Applications* (Wiley, New York, 2000).
[15] D. Sander, Rep. Prog. Phys. **62**, 809 (1999).
[16] A. E. Clark, *Structural Fe-based Alloys with High Magnetostriction* (Galfenol Workshop, University of Maryland, 2004).
[17] S. Rafique, J. Cullen, M. Wuttig, and J. Cui, J. Appl. Phys. **95**, 6939 (2004).
[18] Y. B. Xu, D. J. Freeland, M. Tselepi, and J. A. C. Bland, Phys. Rev. B **62**, 1167 (2000).