Overlayer-induced magnetic uniaxial anisotropy in nanoscale epitaxial Fe

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We used Brillouin light scattering to probe and quantify the effect Al overlayers have on the magnetic properties of epitaxial Fe layers grown on GaAs(001). In addition, we correlate the magnetic properties with structural properties obtained using reflection high-energy electron diffraction. We unexpectedly find that an epitaxial Al overlayer induces a significant volume component to the uniaxial magnetic anisotropy energy in 1.1–3.0 nm Fe layers of magnitude $(2.5\pm0.2)\times10^5$ erg/cm^3. Our data indicate that the origin of this volume component resides in the presence of an anisotropic strain and relaxation induced in the Fe layer. However, for thinner Fe layers, the overlayer suppresses the uniaxial magnetic anisotropy of Fe layers. We also find that the Al overlayer has no effect on the cubic magnetic anisotropy energy and effective magnetization as our measured values of these constants are consistent and in good agreement with previous reports of Fe layers with Au and Cu overlayers and those without an overlayer.

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INTRODUCTION

Ferromagnetic layers on semiconductors are of considerable interest for studying spin transport phenomena[1] as well as for spintronics applications, where spin-injection contacts require the deposition of ferromagnetic materials on semiconductors to generate spin-polarized currents inside the semiconductor. A promising material system for both basic and applied studies is Fe on GaAs since high-quality epitaxial Fe layers and interfaces are relatively easy to fabricate on GaAs,[2–4] allowing the magnetocrystalline anisotropy to be exploited. However, as the dimensions of these materials decrease to the nanometer scale, dramatic changes in the physical properties occur. Because of this, a fundamental understanding of how materials behave when confined to such dimensions is needed. This knowledge is also essential in order to engineer the magnetic anisostropy, structure, and electronic properties of magnetic nanostructures for new device applications. While considerable work has been done on the interface between Fe and GaAs,[3–13] very little is understood about the influence of overlayers, which are essential for confining the nanostructured Fe layers.[12] In this paper, we directly quantify the influence an overlayer has on the magnetic and structural properties of nanoscaled Fe on GaAs(001). Surprisingly, we find that an ultrathin epitaxial Al overlayer has a profound effect on the magnetic anisotropy and strain within the Fe layer.

EXPERIMENT

Surfaces of 50 mm commercial Si-doped GaAs(001) wafers were cleaned for 80 min under 1 keV Ar^+ bombardment with a final temperature of approximately 600 °C in our four-chamber Perkin-Elmer molecular beam epitaxy system. This system has a base pressure in the low 10^{-11} Torr range, and Auger electron spectroscopy of this sputtered GaAs surface confirms the prepared GaAs is free of any measurable contaminant. The reflection high-energy electron diffraction (RHEED) images in Figs. 1(a) and 1(b) show intense and streaked diffraction indicating well-ordered and smooth (4×6) GaAs(001) reconstructed surfaces.

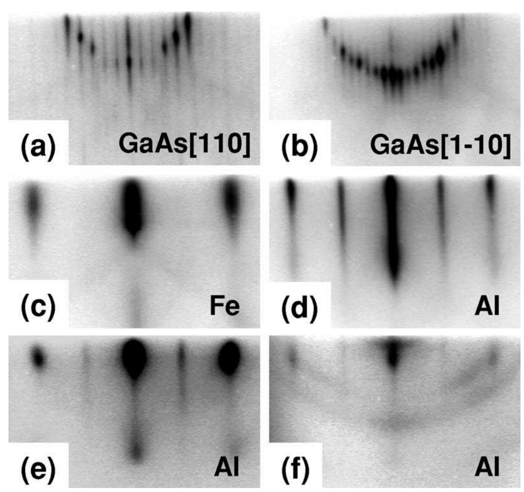

FIG. 1. 10 keV RHEED images along the GaAs ⟨110⟩ directions of (a),(b) the initial (4×6) GaAs(001) surface; (c) a 0.9 nm Fe layer; (d) a typical Al overlayer grown on a continuous and fully crystallized (>0.5 nm) Fe layer; and (e),(f) Al overlayers grown on 0.5 and 0.3 nm Fe layers, respectively, showing the mixed epitaxial and polycrystalline diffraction.

We deposited 0.2–3.0 nm Fe layers at 0.40 nm/min using a Knudsen cell while rotating the 20–40 °C substrates for increased uniformity. The RHEED image for a typical Fe layer above 0.7 nm in thickness is shown in Fig. 1(c). The body centered cubic (bcc) Fe layer is epitaxial on GaAs(001) due to the 1:2 ratio of lattice constants respectively with only a 1.4% mismatch. Immediately after growth of Fe, we deposited a 5 nm Al overlayer using an electron beam evaporation source. This Al overlayer is epitaxial when grown on Fe layers greater than 0.5 nm in thickness. Figure 1(d) shows the typical RHEED pattern we observe for an epitaxial face centered cubic (fcc) Al overlayer. The fcc Al lattice is rotated about the surface normal by 45°, in which configuration, the lattice mismatch between Al and Fe is only ≈0.1%. However, the Al RHEED pattern shows mixed polycrystalline and epitaxial diffraction when grown on Fe layers below 0.5 nm as shown in Figs. 1(e) and 1(f). This is consistent with the Al overlayer being epitaxial only when the underlying Fe is thick enough to completely coalesce and order.

We studied magnetic properties of our buried Fe layers with *ex situ* Brillouin light scattering (BLS), using a diode-pumped neodymiun-doped yttrium aluminum garnet (Nd:YAG) laser with a wavelength of 532 nm and an output power of 200 mW to probe thermally excited long-wavelength spin-wave modes. The details of our system are described elsewhere.[14]

SPIN-WAVE FITTING ROUTINE

A single analytical expression describing the surface spin-wave mode, or Damon-Eshbach mode, can be used for ultrathin ferromagnetic layers whose thickness is less than the exchange-correlation length (approximately 3.5 nm for Fe).[15,16] However, this equation was shown experimentally by Madami *et al.*[12] to be valid for Fe layers up to 10 nm. This expression is given in Eq. (1):

$$\left(\frac{\omega}{\gamma}\right)^2 = \left[H\cos(\phi-\phi_H) + H_\alpha + \frac{2A}{M_s}q_\parallel^2 + 4\pi M_s D\left(1-\frac{q_\parallel d}{2}\right)\right] \times \left[H\cos(\phi-\phi_H) + H_\beta + \frac{2A}{M_s}q_\parallel^2 + 2\pi M_s D q_\parallel d \sin^2\left(\phi-\phi_H+\frac{\pi}{2}\right)\right]. \tag{1}$$

Here, ω is the spin-wave frequency, γ=18.47 GHz/kOe is the gyromagnetic ratio (using a g factor of 2.1), H is the external magnetic field, ϕ is the in-plane (azimuthal) angle of the magnetization, ϕ_H is the direction of the applied in-plane H field, $H_{\alpha,\beta}$ are the anisotropy fields, A is the exchange stiffness constant, M_s is the saturation magnetization, $q_\parallel$ is the in-plane wave vector, $D=1-0.2338/n$ is the demagnetization factor,[17] n is the number of monolayers of Fe, and d is the thickness of the Fe layer.

The symmetry of the anisotropy for Fe layers grown on GaAs(001) is well documented[6,7,9–13] and was also confirmed in this study to consist of both in-plane uniaxial and in-plane cubic components. The easy axes of the cubic and uniaxial anisotropies lie along the ⟨100⟩ and [110] directions, respectively, and the hard axes lie along the ⟨110⟩ and $[1\bar{1}0]$ directions, respectively. Polar-magneto-optical Kerr effect measurements yielded no detectable spontaneous perpendicular component to the magnetization for all thicknesses of Fe

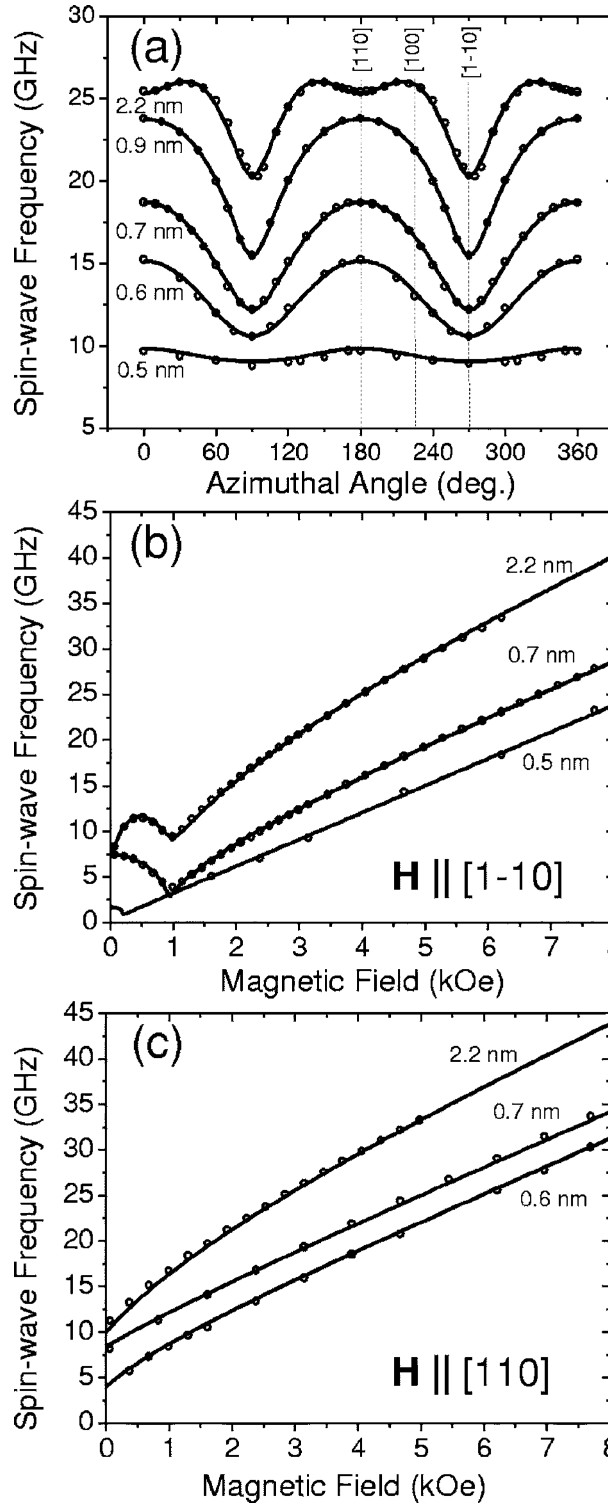

FIG. 2. BLS data (○) of the spin-wave frequency along with our fitted data (solid line) for (a) angular dependence at 3 kOe, and magnetic field dependence along the (b) $[1\bar{1}0]$ and (c) [110] directions.

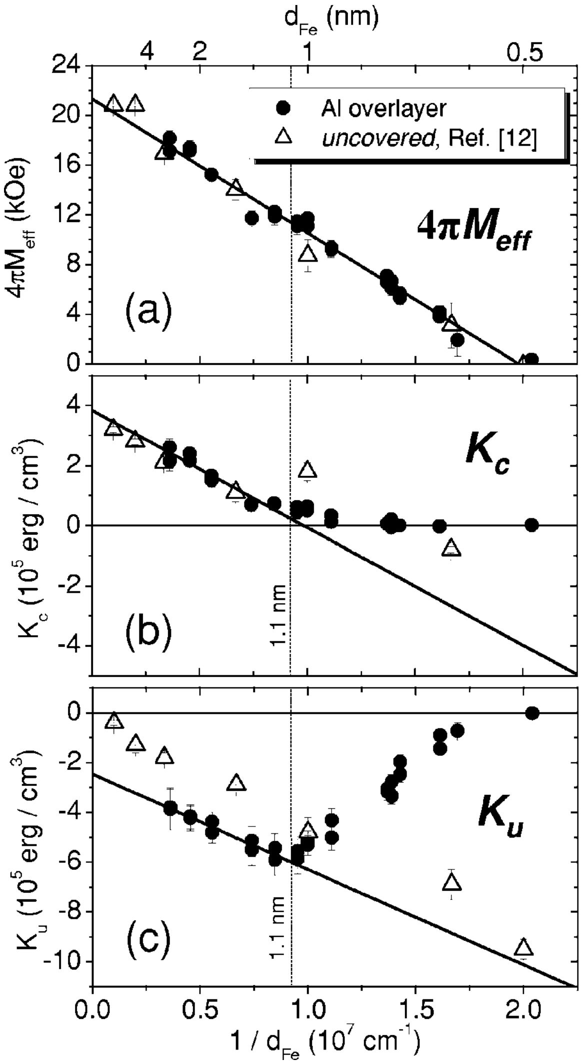

FIG. 3. Plots of our measured values of (a) M_{eff}, (b) K_c, and (c) K_u versus inverse thickness. Regression lines were fitted through the K_c and K_u data above 1.1 nm. For comparison, uncovered Fe data taken from Ref. 12 are included in the plot as the open points (△).

investigated in this study. From these symmetries, the total magnetic anisotropy energy (E) can be constructed with an additional out-of-plane anisotropy term as follows:

$$\begin{aligned} E &= E_{cubic} + E_{uni} + E_{out} \\ &= K_c[\cos^2\theta\sin^2\theta + \sin^4\theta\cos^2\phi\sin^2\phi] \\ &\quad + K_u\sin^2\theta\cos^2(\phi-\phi_0) + K_{out}\sin^2\theta \end{aligned} \tag{2}$$

where K_c is the cubic anisotropy constant, K_u is the uniaxial anisotropy constant, K_{out} is the out-of-plane anisotropy constant, θ=90° is the polar angle (confined to be in plane), ϕ is the in-plane (azimuthal) angle, and ϕ_o=45° is the direction of the uniaxial easy axis when K_u is negative. The anisotropy fields ($H_{\alpha,\beta}$) are calculated from Eq. (2) as follows:[15,16]

$$\begin{aligned} H_\alpha &= \frac{\partial^2}{M_s\partial\theta^2}E \\ &= \frac{K_c}{M_s}(2-\sin^2 2\phi) - \frac{2K_u}{M_s}\cos^2(\phi-\phi_o) + \frac{2K_{out}}{M_s}, \end{aligned}$$

$$\begin{aligned} H_\beta &= \frac{\partial^2}{M_s\partial\phi^2}E = \frac{2K_c}{M_s}(\cos^4\phi + \sin^4\phi - 6\sin^2\phi\cos^2\phi) \\ &\quad - \frac{2K_u}{M_s}[1-2\sin^2(\phi-\phi_o)]. \end{aligned} \tag{3}$$

The direction of the spontaneous magnetization is found by minimizing the total free energy of the system taking into account the above anisotropy energy and the external field term $M_sH\cos(\phi_H-\phi)$. We created a least-squares fitting program to simulate and fit our BLS data, from which we extracted quantitative values of magnetic constants. Figure 2 shows our BLS data along with simulated data obtained from a simultaneous fit of the spin-wave field dependence and angular scans for various Fe layer thicknesses. The experimental error bars for these BLS data are smaller than the data points in the plot and, as can be seen, there is excellent agreement between our BLS data and the simulated data.

RESULTS AND DISCUSSION

Unique fits of both M_s and K_{out} were not possible, allowing us to obtain only a value of an effective magnetization (M_{eff}) for an individual sample. Figure 3(a) shows a plot of M_{eff} versus the inverse Fe layer thickness ($1/d_{\mathrm{Fe}}$), exhibiting excellent linearity across all values of Fe layer thickness (d_{Fe}). In addition, the y intercept of the regression line in Fig. 3(a) yields a value of 21.30±0.36 kOe, within the error of the bulk Fe magnetization of 21.0 kOe, indicating that K_{out} is purely a surface term and has no volume component. By applying Eq. (4) to the regression line,[18] we obtain a value of 0.93±0.03 erg/cm^2 for the surface component of K_{out}:

$$4\pi M_{eff} = 4\pi D M_s - \frac{2K_{out}^{surf}}{M_s}\frac{1}{d_{\mathrm{Fe}}}. \tag{4}$$

The effective uniaxial (K_u) and cubic (K_c) anisotropy constants are plotted as functions of $1/d_{\mathrm{Fe}}$ in Figs. 3(b) and 3(c), respectively. The volume and surface components to each anisotropy term are related to the effective anisotropy through Eq. (5). The factor of 2 in both Eqs. (4) and (5) results from the presence of two interfaces Fe-GaAs and Fe-Al. The magnetic properties of these two interfaces are not the same; however, since we are not able to separate the individual properties of the two interfaces, we report the combined average:

$$K_{c,u} = K_{c,u}^{vol} + \frac{2}{d_{\mathrm{Fe}}}K_{c,u}^{surf}. \tag{5}$$

Here, the superscripts refer to the volume and surface components and subscripts refer to either the cubic or uniaxial term. This equation is used to separate the volume and surface components of $K_{c,u}$ from the data in Figs. 3(b) and 3(c).

TABLE I. Comparison of measured values of the surface and volume components in K_c, K_u, and K_{out} from this work and other recent published reports using BLS, FMR, and MOKE.

Anisotropy	Method	Overlayer material	Volume component (10^{15} erg/cm^3)	Surface component (10^{-2} erg/cm^2)	Reference
Uniaxial	BLS	Al	-2.5 ± 0.2	-1.9 ± 0.2	This work
(K_u)	BLS	Uncovered	-0.05 ± 0.1	-2.2 ± 0.1	12
	BLS	Au	0	-10 ± 1.0	10
	FMR	Cu		-3.2 ± 1.2	18
	FMR	Au	0	-12 ± 2	7
	MOKE	Au		-1.3 ± 0.01	9
Cubic	BLS	Al	3.8 ± 0.3	-2.0 ± 0.2	This work
(K_c)	BLS	Uncovered	3.4 ± 0.3	-1.2 ± 0.4	12
	BLS	Cu	3.7 ± 0.3	-3.2 ± 0.5	10
	FMR	Cu	4.6 ± 0.3	-5.1 ± 0.5	18
	FMR	Au	4.3 ± 0.2	-4.6 ± 0.2	7
	MOKE	Au	3.8 ± 0.2	-3.2 ± 0.2	9
Out of plane	BLS	Al	0	93 ± 3	This work
(K_{out})	BLS	Uncovered	0	40 ± 10	12
	MOKE	Cu	12 ± 7	90 ± 10	10
	FMR	Cu	0	172 ± 10	18

However, below 1.1 nm, we observe both K_u and K_c to be suppressed, deviating from a linear trend. This suppression of the anisotropy in the low-d_{Fe} region was previously observed with Cu (Ref. 12) and Au overlayers (Ref. 18) and was attributed to electronic and structural effects, respectively. Next, we present the evidence that it is a combination of both these effects.

As mentioned above, RHEED images of the Al overlayer grown on Fe layers below 0.5 nm consist of mixed epitaxial and polycrystalline diffraction, indicating the structure and crystallinity of the Fe layer is not completely established, consistent with the magnetocrystalline anisotropy also not being completely established. In addition, Madami *et al.*[12] performed a detailed *in situ* BLS study of *uncovered* Fe layers grown on (4×6) GaAs(001). Their data are also plotted in Fig. 3 as the open points. As can be seen, there is excellent agreement of M_{eff} and K_c with our data above 0.5 nm, indicating the suppression of K_c is independent of the presence of the Al overlayer. Also, K_c is the magnetic anisotropy of bulk Fe (bulk value$=4.5\times10^5$ erg/cm^3) and thus is highly correlated with its crystal structure. Finally, it was reported that the Fe interface with (4×6) GaAs(001) has a bulklike spin moment and is ferromagnetic.[3,4] Therefore, we conclude that the lack of a completely established structure is the cause of the suppression of K_c in the lower thickness region.

However, suppression of K_u in *uncovered* Fe layers is not observed. This indicates the presence of the Al overlayer, not any lack of structure in the Fe layer, is responsible for this phenomenon. In addition, Madami *et al.* found that a 1 monolayer Cu overlayer qualitatively suppresses the anisotropy for 0.6–1.5 nm Fe layers, strongly indicating that the surface bonding and electronic effects cause the suppression of K_u.

We fit linear regression lines through the data in Figs. 3(b) and 3(c) above 1.1 nm to separate the volume and surface contributions since in this region the data is clearly linear. Table I lists the values we obtained for the surface and volume components to K_u, K_c, and K_{out} and compares them to values experimentally determined in recent studies performed by other groups using BLS, ferromagnetic resonance (FMR) and magneto-optical Kerr effect (MOKE) techniques.[19]

As can be seen, the values for all the anisotropy constants listed in Table I are generally consistent among all groups and methods used, with one notable exception: we measure a significant nonzero value for the volume component of K_u. This result is surprising, since the uniaxial anisotropy in Fe on GaAs(001) is known to be interfacial in origin.[7,9,10] Since this volume component to K_u is not present in the uncovered Fe layer data shown in Fig. 3(c), as those data have a zero y intercept, we conclude that the presence of the Al overlayer is responsible.

In order to explain this phenomenon, we first note that an anisotropically strained magnetic layer will create a uniaxial anisotropy which results in the following magnetoelastic energy term[20] which must be added to Eq. (2):

$$E_{me} = K_{me}\sin^2(\alpha) = \left(\frac{2}{3}\lambda_s\sigma\right)\sin^2(\alpha) \tag{6}$$

where K_{me} is the magnetoeleastic anisotropy term, α is the angle between the stressed axis and the magnetization, λ_s is the saturation magnetostriction constant, and σ is the stress. Comparing Eq. (6) to the uniaxial anisotropy energy term in Eq. (2), we find that the terms are identical in form assuming [110] as the strained axis. Under the reasonable assumption that our ultrathin Fe layers are strained uniformly across the entire thickness of the layer, we conclude that K_{me} has to be a volume term. Since the intrinsic uniaxial anisotropy in Fe layer on GaAs is known to be interfacial in origin, we conclude that the volume component of K_u that we observe is K_{me}. This implies the Fe layer is strained anisotropically in plane along ⟨110⟩ directions within this region.

Recently, the structure and morphology of Fe layers on As-rich (2×4) GaAs(001) with 3 nm Al overlayers were studied in detail by x-ray diffraction.[11] The authors find that the Fe layer is fully strained to that of the GaAs substrate up to approximately 2 nm. Above this thickness the Fe layer begins to relax anisotropically along the in-plane ⟨110⟩ directions causing an in-plane uniaxial strained layer. However, the relaxation in the Fe layer would have to occur earlier in our samples, at 1.1 nm, to be consistent with the generation of a magnetoeleastic component of K_u. Although unclear as to why we observe an earlier relaxation, a relaxation in the Fe layer could be aided by our thicker epitaxial Al overlayer, which only has a lattice mismatch of 0.1% with Fe. Since this lattice mismatch is extremely small relative to the 1.4% lattice mismatch between Fe and GaAs, we propose that the epitaxial Al layer is increasing the *effective* thickness of Fe, and thus causing an earlier relaxation in Fe. The above discussion is supported by two reports of highly strained Fe layers grown on InAs (lattice mismatch $>5\%$) in which only a volume component to K_u was observed in the Fe layer,[10] and in a second report, in which a strong uniaxial anisotropy was only observed in the Fe thickness region where a strong anisotropic strain was measured.[21] In addition, Morley *et al.* showed that they could induce a uniaxial anisotropy in Fe on GaAs by mechanically straining the Fe layer.[22] Finally, an alternative explanation could reside in the magnetoelastic effects induced and observed in Fe-Al alloys.[23] However, it is reasonable to assume that any alloying between Fe and Al under our conditions would be strictly confined to the interface and therefore not affect the bulk or volume properties.

In conclusion, we find that an epitaxial Al overlayer unexpectedly induces a volume component to the uniaxial anisotropy for Fe layers between 1.1 and 3.0 nm. Our findings indicate that this volume term results from anisotropic strain formation within the Fe layer that creates a magnetoeleastic component to the uniaxial anisotropy. In addition, we find that both the cubic and uniaxial magnetic anisotropy are suppressed in Fe layers below 1.1 nm. Although our results show the structure of the initial Fe layer to be responsible for the suppression of the cubic anisotropy, the suppression of the uniaxial magnetic anisotropy is caused by the presence of the Al overlayer.

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