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Interface bonding versus strain-induced magnetic anisotropy in epitaxial Fe/semiconductor structures

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We report measurements of perpendicular magnetic anisotropies of epitaxial Fe/InAs(100) 4×2 and Fe/GaAs(100) 4×6 films using the polar magneto-optic Kerr effect (MOKE) and of the in-plane magnetic anisotropy, magnetization and *g*-factor using Brillouin light scattering (BLS). The magnetization is very close to the bulk value in all samples, demonstrating a lack of As intermixing. Both Fe/GaAs and Fe/InAs show large out-of-plane uniaxial surface anisotropy. Both show large in-plane fourfold surface anisotropies which oppose the bulk fourfold anisotropy. Fe/GaAs shows the well known in-plane uniaxial surface anisotropy which is attributed here to interface bonding. However, Fe/InAs shows almost no in-plane uniaxial surface anisotropy, despite the large strain of 5.4% compared to -1.2% for Fe/GaAs. The small in-plane uniaxial volume anisotropy which is observed for Fe/InAs is ascribed, due to the thickness dependence, to a magneto-elastic effect.

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I. INTRODUCTION

Fe films epitaxially grown on semiconductors are an interesting model system with which to probe the nature of the interface between a magnetic and nonmagnetic material. They also have potential technological applications as nonvolatile magnetic memory, microwave, and spin injection devices.[1] The possibilities for creating devices which combine magnetic and semi-conductor materials is increased by the fact that Fe forms an ohmic contact with InAs.[2]

It has been known for some time that bcc Fe grows epitaxially on GaAs (100) (a zinc blende structure)[3] with a misfit strain of $e_{[0\bar{1}\bar{1}]}$ (parallel to the $[0\bar{1}\bar{1}]$ axis) and $e_{[011]}$ (parallel to the $[011]$ axis) equal to -1.2% (compression) at the interface. The misfit strain for Fe grown on GaAs relaxes to zero along the $[0\bar{1}\bar{1}]$ direction at a thickness, *t*, of ~ 120 ML[4] (note the different direction convention adopted by Filipe *et al.*). To our knowledge no measurement has been made along the $[011]$ direction but given the long relaxation length and the fact that no strain anisotropy has been reported in this very well studied system it is likely that $\Delta e = e_{[011]} - e_{[0\bar{1}\bar{1}]}$ in Fe/GaAs is $\ll 1\%$.

On the other hand the epitaxial growth of Fe on InAs(100) has been recently discovered.[5] This system has the same crystallographic structure as Fe/GaAs (so the surface dangling bonds of the In atoms point along $[0\bar{1}\bar{1}]$ as in GaAs (100)) but it is more highly strained and in the opposite direction. At the interface the Fe lattice (bulk lattice constant 0.2866 nm) is expanded with an in-plane strain of $e_{[0\bar{1}\bar{1}]} = e_{[011]} = 5.4\%$. Fe grown on InAs has been shown to relax parallel to the $[0\bar{1}\bar{1}]$ direction faster (relaxation occurs over 5–10 ML) than perpendicular to it (relaxation over 5–25 ML).[6] In the range 5–25 ML the strain anisotropy $\Delta e \sim +1\%$. Below $t \sim 5$ M*L* the film is *isotropically* strained.

Magnetic characterization of the Fe/GaAs system[7] shows the existence of a well known uniaxial magnetic anisotropy (with an easy axis along $[0\bar{1}\bar{1}]$ for $t \gtrsim 5$ ML) which currently defies explanation. It dominates over the four-fold bulk anisotropy for $t \lesssim 30$ ML. It has been attributed either to anisotropic strain relaxation or to anisotropic interface bonding.[7–9]

MOKE experiments[8] show a qualitative difference in the anisotropy of Fe/InAs and Fe/GaAs. The in-plane anisotropy of Fe/InAs is found to be predominantly fourfold (i.e., dominated by the bulk anisotropy), but to have an overall easy axis along $[011]$ at a thickness $t = 8$ ML. However the magnetic anisotropies of Fe/InAs have not previously been quantitatively measured so the cause of this uniaxial anisotropy has been unclear.

If the lattice constant of the Fe decays back to the bulk value anisotropically with thickness a magneto-elastic anisotropy can be expected. The magnitude and sign of this anisotropy depend on the magneto-elastic constant B_1. A first order Taylor expansion of the magneto-elastic energy in the strain[10] predicts a uniaxial anisotropy of $K_{\mathrm{Uv}} = B_1 \Delta e$. The bulk value of B_1 for Fe is $\sim -3\times10^6\ \mathrm{J\,m^{-3}}$ (all anisotropies and magnetic quantities are expressed in SI units, the Sommerfeld convention is used throughout) but it has been shown that for thin Fe/W films[11] B_1 is substantially reduced and changes sign at $t \sim 20$ nm. To our knowledge B_1 has not been directly measured for Fe/GaAs or Fe/InAs thin films. However we assume B_1 to vary slowly over the thickness ranges studied in this work. Such a slow variation has been confirmed experimentally for Fe/W films, which are more highly strained than either Fe/GaAs or Fe/InAs.[11] This assumption is supported in the Fe/InAs system by the absence of any observable thickness variation in the in-plane uniaxial anisotropy. The only other possible explanation is that the volume and surface contributions cancel perfectly over the whole range, which is unlikely.

The magneto-elastic effect persists to the relaxation length of the lattice constant. In contrast, magnetic anisotropy caused by anisotropic interface bonding is strictly a surface term. It is thought to occur due to the uniaxial nature of Fe/Ga (Fe/In if applicable) bonds at the interface between the two materials. Thus it is possible to identify which of the two effects is dominant by measuring the strength of the anisotropies as a function of film thickness.

BLS is a powerful technique for measuring the magnetic anisotropies of thin magnetic films. The spin wave frequencies are measured by the change in frequency of Raman scattered photons.[12] Spin waves are deviations of the magnetization from the equilibrium direction. The frequencies are determined by the curvature of the potential well (with deviation of the magnetization) for the mean direction of the magnetization. The curvature of this potential well depends on the anisotropy energy experienced by the spins and so spin wave frequencies are higher when the magnetization is oriented along a low energy (easy axis) direction. BLS can therefore discover the magnitude and nature of the anisotropies. An additional benefit is that the samples are probed while magnetically saturated, which reduces the effect of impurities or disorder on the results.

The purpose of this work is to measure the Fe/InAs anisotropies for samples which have previously been structurally characterized and to identify the relative contributions of the two mechanisms in both Fe/GaAs and Fe/InAs by comparing the thickness dependence of the strain and magnetic anisotropies in the two systems.

II. EXPERIMENTAL METHOD

Stepped bcc Fe samples were grown by molecular beam epitaxy with thicknesses in the range 4.1–33 ML (Fe/GaAs) and 8–40 ML (Fe/InAs), they were capped with ~20 ML of Au. The growth took place at room temperature and at a pressure of $\sim 7\times10^{-10}$ mbar. Further details are as previously described.[13]

LEED images of the GaAs substrate taken immediately prior to growth show the surface to have a 4×6 reconstruction.[14] It is not known whether this is a "true" 4×6 or a "pseudo" 4×6 made up of patches of 4×2 and 1×6.[15] LEED and STM images of the InAs substrate[8] show a 4×2 In rich reconstruction.

Polar MOKE (incident angle of ~0.5°) and BLS experiments both took place *ex situ*. BLS experiments were performed in the Voigt geometry: **H** in-plane parallel to z and perpendicular to the incident light (with the x axis out-of-plane). The incident angle of the laser was 45°. The 514.5 nm line of an Ar^+ ion laser was used with an optical power at the sample surface of $\lesssim 100$ mW. The scattered light was directed into a Sandercock 3+3 pass interferometer as described previously.[16]

III. RESULTS AND DISCUSSION

The magnetic anisotropy energy density for a biaxial crystal may be written,

$$U_{\mathrm{an}}=K_{\mathrm{P}}u_{[100]}^2+K_{\mathrm{U}}u_{[011]}^2+K_1(u_{[100]}^2u_{[011]}^2+u_{[011]}^2u_{[0\bar{1}\bar{1}]}^2+u_{[100]}^2u_{[0\bar{1}\bar{1}]}^2), \tag{1}$$

where $\mathbf{u}=\mathbf{M}/M_s$ (M_s is the saturation magnetization) and the subscripts of the components of $\mathbf{u}$ denote the crystalline directions. $[100]\|x$ with $[011]$ and $[0\bar{1}\bar{1}]$ in the y–z plane. The angle between $[011]$ and z is defined to be η. Equation (1) is used in the analysis of the BLS results as detailed by Hicken *et al.*[16]

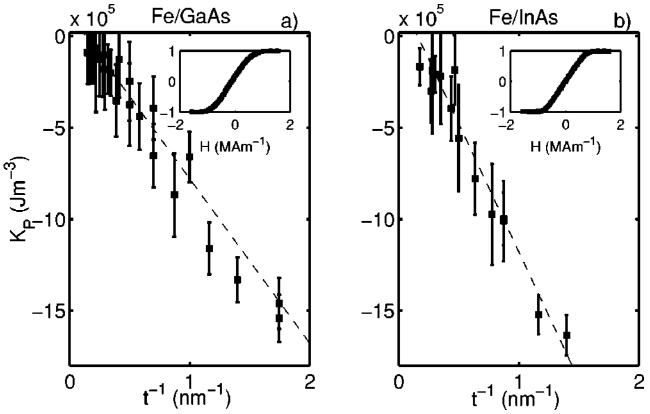

FIG. 1. Effective out-of-plane anisotropy [defined in Eq. (3)] of (a) Fe/GaAs and (b) Fe/InAs measured by polar MOKE. The lines are weighted best fits.

A possible fourth order term proportional to $u_{[100]}^4$ is not used in the analysis of the MOKE results (as justified below) or the BLS results (due to the small size of the spin wave fluctuations[16]).

K_{U} and K_{P} are in- and out-of-plane effective uniaxial anisotropies given by

$$K_{\mathrm{U}}=K_{\mathrm{Uv}}+\frac{K_{\mathrm{Us}}}{t}, \tag{2}$$

$$K_{\mathrm{P}}=K_{\mathrm{Pv}}+\frac{K_{\mathrm{Ps}}}{t}, \tag{3}$$

and for the four-fold anisotropy,

$$K_1=K_{1\mathrm{v}}+\frac{K_{1\mathrm{s}}}{t}. \tag{4}$$

Each is a combination of a volume and surface term (note that the surface term is the sum of the top and bottom surfaces). For a uniaxial crystal (with identical in-plane directions) $K_{\mathrm{Uv}}=0$.

The effective out-of-plane anisotropy, K_{P}, may be measured by polar MOKE,

$$K_{\mathrm{P}}=\frac{\mu_0 M_s}{2}(H_s-N_{\mathrm{P}}M_s), \tag{5}$$

where H_s is the out-of-plane saturation field and N_{P} is the out-of-plane demagnetizing factor.

The saturation field is taken from the maxima of the magnitude of the the second derivative of the MOKE signal intensity, i.e., $(\partial^2 M_x)/(\partial H_x^2)$.[17] Comparison with H_s derived from the gradient of the MOKE signal at $H\sim0$ shows the effect of the fourth order terms to be comparable to or smaller than the random error. Typical loop shapes are shown in the inserts of Fig. 1. The curvature of the MOKE loops is due to the large in-plane anisotropy constant (and is most apparent in the Fe/GaAs).

TABLE I. Table of anisotropy constants for epitaxial Fe/GaAs and Fe/InAs calculated using the measured values of M_s [see Fig. 4(a)&4(b)]. The $K_{1\mathrm{v}}$ terms include the bulk cubic anisotropy. The lower half of the table shows values taken from the literature. Values of the constants have not been adjusted to remove the effect of the overlayer.

Constant	Au/Fe/GaAs	Au/Fe/InAs
$K_{\mathrm{Us}}(\mathrm{J\,m^{-2}})$	$(10.0\pm1.0)\times10^{-5}$	0
$K_{\mathrm{Uv}}(\mathrm{J\,m^{-3}})$	–	$-(0.25\pm0.1)\times10^{4}$
$K_{\mathrm{Ps}}(\mathrm{J\,m^{-2}})$	$-(0.9\pm0.1)\times10^{-3}$	$-(1.4\pm0.1)\times10^{-3}$
$K_{\mathrm{Pv}}(\mathrm{J\,m^{-3}})$	$(1.2\pm0.7)\times10^{5}$	$(2.2\pm1.3)\times10^{5}$
$K_{1\mathrm{s}}(\mathrm{J\,m^{-2}})$	$-(3.2\pm0.5)\times10^{-5}$	$-(7.9\pm0.6)\times10^{-5}$
$K_{1\mathrm{v}}(\mathrm{J\,m^{-3}})$	$(3.7\pm0.3)\times10^{4}$	$(6.5\pm0.3)\times10^{4}$

Constant	Cu/Fe/GaAs (Ref. 23)	Au/Fe/GaAs
$K_{\mathrm{Us}}(\mathrm{J\,m^{-2}})$	$(3.2\pm1.2)\times10^{-5}$	[22]$(12.0\pm2)\times10^{-5}$
$K_{\mathrm{Uv}}(\mathrm{J\,m^{-3}})$	–	–
$K_{\mathrm{Ps}}(\mathrm{J\,m^{-2}})$	$-(1.7\pm0.1)\times10^{-3}$	[24]$-(1.1\pm0.2)\times10^{-3}$
$K_{\mathrm{Pv}}(\mathrm{J\,m^{-3}})$	–	–
$K_{1\mathrm{s}}(\mathrm{J\,m^{-2}})$	$-(5.1\pm0.5)\times10^{-5}$	[24]$-(2.1\pm0.2)\times10^{-5}$
$K_{1\mathrm{v}}(\mathrm{J\,m^{-3}})$	$(4.6\pm0.3)\times10^{4}$	[22]$(4.3\pm0.2)\times10^{4}$

The out-of-plane demagnetizing factor, $N_{\mathrm{P}}\gtrsim0.97$, differs from unity due to the surface roughness[18] and the atomic nature of thin films[19] (though the effect of this on the measurement of K_{P} is small). The roughness amplitudes and lateral length scales were taken from STM data for Fe/InAs[8] and Fe/GaAs (unpublished data).

By measuring K_{P} at a range of thicknesses the values of K_{Pv} and K_{Ps} may be found. K_{P} is plotted as a function of inverse thickness in Fig. 1 for Fe/GaAs and Fe/InAs. A weighted least square fit gives $K_{\mathrm{Ps}}=(-0.9\pm0.1)\times10^{-3}\ \mathrm{J\,m^{-2}}$ and $K_{\mathrm{Pv}}=(1.2\pm0.7)\times10^{5}\ \mathrm{J\,m^{-3}}$ for Fe/GaAs. The value of K_{Ps} was found to be in excellent agreement with the one reported by Urban *et al.*[25] The anisotropy constants of Fe/GaAs and Fe/InAs are summarized in Table I.

For Fe/InAs we must consider a possible effect from the variation of the out-of-plane strain (via magneto-elastic effects) over the thickness measured. However, the data of Fig. 1 appear linear, implying that the change in the out-of-plane volume uniaxial constant is not significant (it is presumably greatest for the thinnest films, but for these films the surface term dominates). A weighted least squares fit for the Fe/InAs data for which $t<25$ ML gives $K_{\mathrm{Ps}}=(-1.4\pm0.1)\times10^{-3}\ \mathrm{J\,m^{-2}}$ and $K_{\mathrm{Pv}}=(2.2\pm1.3)\times10^{5}\ \mathrm{J\,m^{-3}}$.

It is curious that K_{Pv} has the same sign in both Fe/GaAs and Fe/InAs. As the in-plane strain (and hence the tetragonal distortion) is of opposite sign in the two systems a difference in sign of K_{Pv} would be expected. It is possible that the magneto-elastic constant is different in each system, there is an out of plane lattice matching condition similar to that in Fe/Ag,[20] or that the effect is caused by the relaxation over the thickness range measured. However, without knowledge of the magneto-elastic constants for the two systems it is not possible to distinguish between these possibilities.

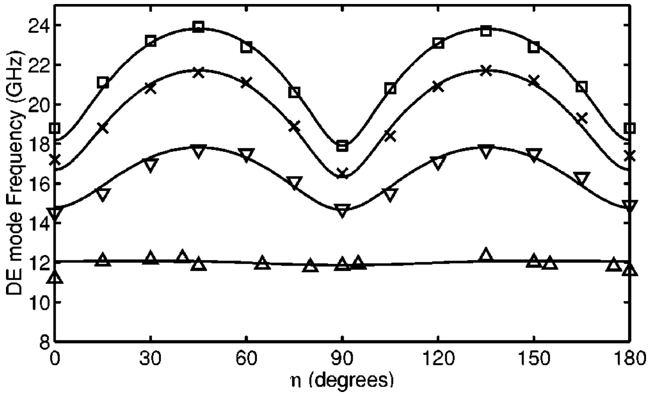

FIG. 2. Angular dependence of DE spin wave frequency in Fe/InAs with angle, η, between **H** field and [011] ($|\mathbf{H}|=1.6\times10^{5}\ \mathrm{Am^{-1}}$). □:40 ML, ×:25 ML, ▽:15 ML, and △: 8 ML. The lines are iterative best fits. The errors are given by the symbol size.

BLS measures the frequency of the surface spin wave mode (called the DE mode after Damon and Eshbach[21]) for the ultrathin films investigated in this work. The spin wave frequencies were fitted iteratively using the continuum model of Hicken *et al.*[16] with the anisotropy energy of Eq. (1) (though the surface terms were used as boundary conditions rather than effective anisotropies). Though the in-plane demagnetizing factor $(1-N_{\mathrm{P}})/2$ is only a few percent its effect on the DE mode frequency is significant at low external fields when $M_s\gg|\mathbf{H}|$ and it must be included in the fitting process. The BLS results for the two systems are discussed separately below, and then compared.

For each thickness of Fe/InAs the frequency of the DE spin wave mode was measured as the external **H** field varied in magnitude with fixed angle to the [011] axis, η, and as η was varied (at fixed field magnitude). The variation of DE mode frequency with external field allows the magnetization and *g*-factor to be obtained, and the directional variation reveals the anisotropy energy (for the measured magnetization at a given thickness). The frequency as a function of η is plotted in Fig. 2. The films have four-fold magnetic anisotropy, with little uniaxial anisotropy. The reduction of the frequency with decreasing thickness is caused by the negative out-of-plane surface anisotropy (which flattens the "energy paraboloid" that the spins oscillate in). The magnitude of the reduction in frequency is approximately five times greater than the reduction that would be expected in the absence of any surface anisotropy.[21]

The values of the anisotropy constants derived for Fe/InAs by fitting the BLS data are shown in Fig. 3(b). The four-fold anisotropy decreases inversely proportional to thickness, justifying the choice of Eq. (4). The data of Fig. 3(b) have been fitted without constraining the effective anisotropy to pass through the bulk value (zero for the uniaxial anisotropy and $\sim4.5\times10^{4}\ \mathrm{J\,m^{-3}}$ for the four-fold anisotropy). By this analysis quite a large four-fold volume term of $K_{1\mathrm{v}}=(6.5\pm0.3)\times10^{4}\ \mathrm{J\,m^{-3}}$ is found for Fe/InAs, approximately $2.0\times10^{4}\ \mathrm{J\,m^{-3}}$ greater than the bulk value. This enhancement may be explained as follows. The thin films measured here are strained over a significant portion of their thickness. The strain has both two- and four-fold compo-

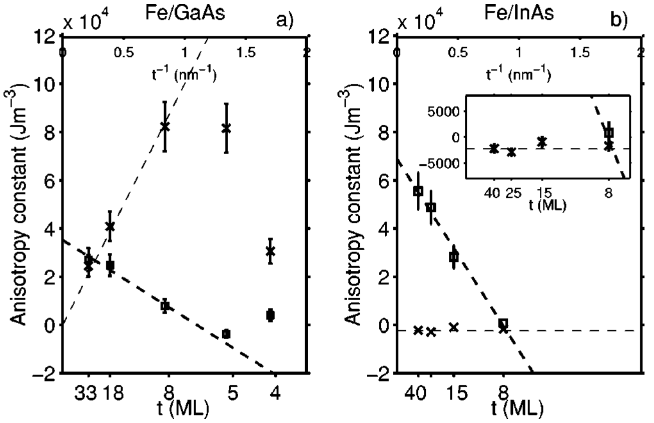

FIG. 3. Variation of anisotropy constants with inverse thickness (scale shown on the top of each figure) for (a) Fe/GaAs and (b) Fe/InAs. For reference, the actual thicknesses are shown at the bottom. □: K_1 and ×: K_U. Where no error bars are shown the errors are given by the symbol size. The inset of (b) is a magnified view of the region containing the K_U data.

nents, the latter contribute to K_{1v}. This may be expected to lead to a magneto-elastic contribution to the four-fold volume anisotropy. Note that the relevant magneto-elastic constant is not B_1 but the coefficient of the fourth order term in the expansion of magneto-elastic energy with strain, which again has not been measured, to our knowledge, for thin Fe/GaAs or Fe/InAs films.

The observation of square longitudinal MOKE loops along the [011] axis[6,8] of a 8 ML Fe/InAs film can now be explained: although the uniaxial anisotropy K_U is not large (compared to the bulk value of K_1) it is at approximately this thickness that the four-fold anisotropy K_1 is zero, allowing the uniaxial anisotropy to produce a easy axis at this thickness.

For the Fe/GaAs samples the DE mode frequency was measured as function of field along the [001], [011], and [010] directions (data not shown). The thickest ($t=33$ ML) film is predominantly fourfold while thinner ones are mostly uniaxial. Figure 3(a) shows that the variation in the uniaxial anisotropy is proportional to $1/t$ in a thickness range for which the strain is almost constant ($8 \leqslant t \leqslant 33$ ML).[4] The anisotropy constants of the thinnest films can be seen to deviate from the trend; this was also observed by Brockmann *et al.*[22]

Fe/GaAs also shows a volume contribution to the four-fold anisotropy, i.e., the intercept in Fig. 3(a) (also see K_{1v} values in Table I) is not at the bulk value of K_1, presumably due to the four-fold strain. The deviation is smaller and of the opposite sign to that in Fe/InAs, as might be expected for a magneto-elastic effect given the relative signs and magnitudes of the four-fold strain in the two systems.

The K_1 and K_U values extracted from the fitting procedure for Fe/GaAs are similar to those measured by other authors for similar systems,[22,23,24] see Table I.

The values of g and M_s are plotted in Fig. 4. Both Fe/GaAs and Fe/InAs can be seen to have a magnetization close to that of bulk Fe ($M_s = 1.71 \times 10^6$ Am^{-1}, $g = 2.09$) for all thicknesses, showing that there is little or no As intermixing with the Fe, even in the monolayers closest to the interface.

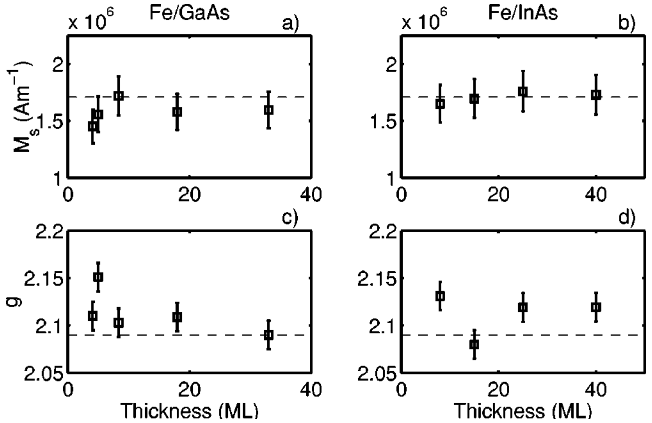

FIG. 4. (a)&(b) Magnetization and, (c)&(d) g-factor of (a)&(c) Fe/GaAs and b)&d) Fe/InAs. The dashed lines represent the bulk values ($M_s = 1.71 \times 10^6$ Am^{-1}, $g = 2.09$).

The Fe/GaAs and Fe/InAs systems behave in a similar manner in most respects, with the notable exception of their K_U dependence on thickness. The strong $1/t$ dependence of K_U in Fe/GaAs indicates a dominant interface contribution while the lack of thickness dependence of K_U in Fe/InAs suggests a volume effect.

Using $K_{Uv} = B_1 \Delta e$ with a strain anisotropy of 1% and the measured value of K_{Uv} (see Table I) the magneto-elastic constant B_1 for Fe/InAs may be estimated to be $\sim +2.5 \times 10^5$ J m^{-3}, considerably smaller than (and of opposite sign to) the bulk value. As the strain anisotropy has not been measured for Fe/GaAs a similar estimation cannot be made for that system. However, it is very difficult to explain the thickness dependence of K_U in the Fe/GaAs as a strain anisotropy induced effect given the magnitude of K_U in Fe/GaAs and the constraint on the strain anisotropy. A direct measurement of the magneto-elastic constants for the two systems would provide an independent check on this and the other results obtained in this paper.

IV. CONCLUSIONS

Fe/GaAs and Fe/InAs have been assessed using BLS and quantitative magnetic parameters extracted. The two systems were grown under similar conditions and shown by LEED and STM[8,14] to possess high quality crystal structures. The two systems show qualitatively similar dependencies of K_1 and K_P on t and similar M_s and g values. Fe/GaAs, for which there is no evidence of anisotropic strain, shows a large in-plane uniaxial anisotropy, K_U. Fe/InAs, which is anisotropically strained,[8] has a negligible value of K_U. The striking difference in the magnitude and t dependence of the in-plane uniaxial anisotropy in the two systems suggests that it has a different origin in each. In Fe/InAs the lack of a strong t dependence shows that strain causes a small magneto-elastic effect and we conclude from our analysis that the interface bonding does not give rise to a uniaxial anisotropy in contrast to the case of Fe/GaAs. In contradistinction, the same analysis leads to the conclusion that in Fe/GaAs the $1/t$ dependence of the uniaxial anisotropy is due to the presence of a large interface bonding term in the absence of a significant magneto-elastic term. The stunning difference in the uniaxial anisotropy behavior of the two systems we attribute to the difference between the Fe/In and Fe/Ga chemical interface bonds, i.e., interface bonding rather than strain is fundamental to the origin of the uniaxial anisotropy.

ACKNOWLEDGMENTS

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