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# Magnetostriction and magnetocrystalline anisotropy constants of ultrathin epitaxial Fe films on GaAs, with Au overlayers

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## Abstract

In this paper, we present the first measurements on the variation of the saturation magnetostriction constant with film thickness of ultrathin epitaxial Fe films on GaAs(100) substrates. Furthermore, we explore whether there is a link between magnetostriction and the uniaxial anisotropy in these Fe films. The Fe film thickness ranged from seven monolayers (ML) (having only uniaxial anisotropy) to 50 ML (almost pure cubic anisotropy). The anisotropy constants were determined from the normalized magnetization loops, using a magneto-optic Kerr effect (MOKE) fitting technique that convolutes a magnetic energy density model with the dependence of the MOKE signal on the angle between the pass plane of the analyser and the plane of incidence of the laser light on the sample. Each film was uniformly strained along the [011] direction, while the magnetization was measured along the [0 $\bar{1}$ 1] direction using a MOKE magnetometer. From the change in anisotropy field as a function of strain (Villari effect), the magnetostriction constant in the [011] direction was calculated. It is demonstrated that the saturation magnetostriction constant in the [011] direction is significantly different to the equivalent value in bulk Fe, and increases in magnitude as the thickness of the Fe film decreases. It will also be shown that the uniaxial anisotropy constant has a linear dependence on the magnetostriction constant for each film.

## 1. Introduction

Ultrathin epitaxial Fe films on GaAs(100) substrates have been the subject of a large amount of study as they may be a model system for certain spintronic applications [1]. For film thicknesses less than 100 monolayers (ML), a uniaxial anisotropy is present in the film as well as the (bulk-like) cubic anisotropy [2–5]. For Fe thicknesses less than 10 ML, only the uniaxial anisotropy is observed [6]. The origin of this uniaxial anisotropy is uncertain [7, 8], but it has been attributed to the properties of the Fe–GaAs interface [4, 9] or to an interface anisotropy [10]. It

is reasonable to assume that the epitaxial misfit strain and morphological changes at surfaces and interfaces may couple with the magnetostriction to give magnetoelastic anisotropy. This paper investigates the magnetoelastic properties of the films, by measuring the saturation magnetostriction constant. This may be the origin of the uniaxial component. Furthermore, the magnetostriction must be characterized as there is potential for extrinsically generated magnetoelastic anisotropy which could impinge on device applications.

For single-crystal Fe/GaAs films, the in-plane magnetization process is assumed to proceed by pure and coherent rotation of the moments. Allowing for the crystallography of the GaAs wafer, and the epitaxial growth relationship for Fe on GaAs(100), the in-plane magnetic energy density ( $F$ ) of the film is given by

$$F = \frac{1}{4} K_1(t) \sin^2 2(\varphi - a) + K_u(t) \sin^2 \left( \varphi - a + \frac{\pi}{4} \right) - HM \cos \varphi \quad (1)$$

where  $K_1(t)$  is the first-order cubic magnetocrystalline anisotropy constant,  $K_u(t)$  is the uniaxial anisotropy constant,  $a$  is the angle between the magnetic field and the [001] direction in the film and  $\varphi$  is the angle between the magnetic field ( $H$ ) and the in-plane magnetization ( $M$ ). Both anisotropy constants are allowed to be functions of the Fe layer thickness,  $t$ . The second-order magnetocrystalline anisotropy constant does not contribute to the free energy for the particular film crystallography considered here. The direction of the magnetization in the film is found by solving  $\frac{dF}{d\varphi} = 0$ , for known anisotropy constants and field directions. Thus the normalized magnetization at a given field is  $\frac{M}{M_{\text{sat}}} = \cos \varphi_{\text{min}}$ , where  $\varphi_{\text{min}}$  is the equilibrium angle between the given applied field and the magnetization.

The anisotropy constants were determined using the magneto-optic Kerr effect (MOKE) fitting method. For a MOKE magnetometer, the output intensity of the photodetector depends on the angle ( $\theta_a$ ) between the pass plane of the analyser and the plane of incidence of the laser [11]. The intensity is determined by considering the resultant electric field,  $E_r$ , of the laser after it has been reflected off the sample, which is given by [11]

$$E_r = E_0 [(m_t^2 r_{\text{pp}}^t + m_l^2 r_{\text{pp}}^l) \cos \theta_p \cos \theta_a + m_l^2 r_{\text{ps}}^l \sin(\theta_p - \theta_a) + r_{\text{ss}}^l \sin \theta_p \sin \theta_a] \quad (2)$$

where  $m_t$  and  $m_l$  are the transverse and longitudinal magnetizations with respect to the plane of incidence of the laser, and  $E_0$  the incident electric field amplitude. The angles  $\theta_p$  and  $\theta_a$  are the angles between the plane of incidence and the transmission axis of the polarizer and analyser respectively. The terms  $r_{\text{pp}}^l$ ,  $r_{\text{pp}}^t$ ,  $r_{\text{ps}}^l$  and  $r_{\text{ss}}^l$  are the Fresnel reflection coefficients for light reflected from a magnetic film [12–14]. Thus the intensity of the laser measured on the photodetector is the square of the resultant electric field (equation (2)), and is given by

$$\begin{aligned} \frac{I}{I_0} = & [|m_l^2 r_{\text{pp}}^l + m_t^2 r_{\text{pp}}^t|^2 \cos^2 \theta_p \cos^2 \theta_a + |m_l^2 r_{\text{ps}}^l|^2 \sin^2(\theta_p - \theta_a) + |r_{\text{ss}}^l|^2 \sin^2 \theta_p \sin^2 \theta_a \\ & + [(m_l^2 r_{\text{pp}}^l + m_t^2 r_{\text{pp}}^t) m_l^2 r_{\text{ps}}^l + \text{c.c.}] \cos \theta_p \cos \theta_a \sin(\theta_p - \theta_a) \\ & + [(m_l^2 r_{\text{pp}}^l + m_t^2 r_{\text{pp}}^t) r_{\text{ss}}^{l*} + \text{c.c.}] \cos \theta_p \cos \theta_a \sin \theta_p \sin \theta_a \\ & + [r_{\text{ss}}^l m_l^2 r_{\text{ps}}^{l*} + \text{c.c.}] \sin \theta_p \sin \theta_a \sin(\theta_p - \theta_a)]. \end{aligned} \quad (3)$$

For the experiments carried out the longitudinal magnetization was measured; thus the polarizer angle was parallel to the plane of incidence, i.e.  $\theta_p = 0^\circ$ , and the analyser angle was set just off extinction,  $\theta_a = 88^\circ$ . Hence substituting this polarizer angle into equation (3) gives

$$\begin{aligned} \frac{I}{I_0} = & |m_t^2 r_{\text{pp}}^t + m_l^2 r_{\text{pp}}^l|^2 \cos^2 \theta_a + |m_l^2 r_{\text{ps}}^l|^2 \sin^2(\theta_a) \\ & - [(m_t^2 r_{\text{pp}}^t + m_l^2 r_{\text{pp}}^l) m_l^2 r_{\text{ps}}^{l*} + \text{c.c.}] \cos \theta_a \sin \theta_a. \end{aligned} \quad (4)$$

The transverse and longitudinal magnetizations are taken to be  $m_t = m_s \sin(\varphi + \frac{\pi}{2})$  and  $m_l = m_s \cos(\varphi + \frac{\pi}{2})$ , i.e. the longitudinal magnetization is parallel to the plane of incidence of the laser. Hence simplifying equation (4), with the above expressions, and collecting the constants together, the normalized intensity at the detector ( $I/I_0$ ) is [15, 16]

$$\frac{I}{I_0} = A \cos^2 \theta_a + (B \cos^2 \theta_a) \cos \varphi + (C \sin \theta_a \cos \theta_a) \sin \varphi + (D \sin^2 \theta_a) \sin^2 \varphi \quad (5)$$

where  $A$ ,  $B$ ,  $C$  and  $D$  are constants which depend on the refractive index of Fe ( $n$ ), the magneto-optic constant ( $Q$ ), and the angle of incidence of the laser beam on the film. These constants are derived elsewhere [15, 16]. For  $\theta_a$  close to  $90^\circ$ , all four terms in equation (5) are the same order of magnitude. Hence the measured magnetization loop is non-symmetric. For each film, the anisotropy constants were determined by convoluting the magnetic energy density (equation (1)) with the output of the photodetector (equation (5)), which was then fitted to the measured normalized magnetization data for the hard axis,  $a = \pm \frac{\pi}{4}$  [16].

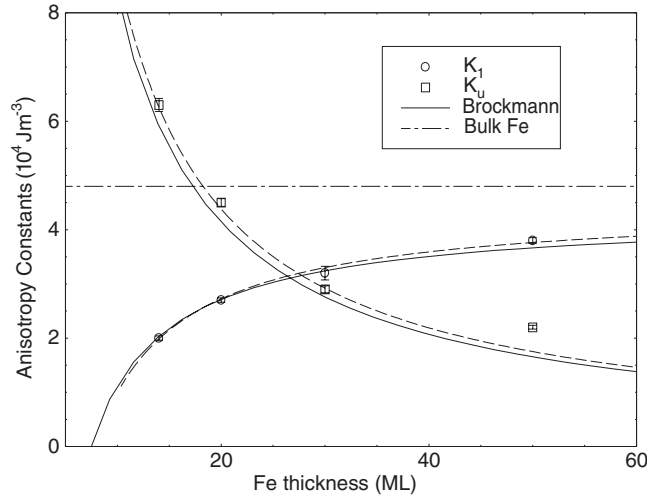
## 2. Experimental details

The epitaxial Fe films on GaAs(100) substrates with Au overlayers were fabricated using molecular beam epitaxy (MBE) [17]. The GaAs(100) substrates were purchased from Wafer Technology Ltd, with the major flat parallel to the  $[0\bar{1}1]$  crystallographic direction. Prior to each film's deposition, they were etched using  $\text{H}_2\text{SO}_4$  (sulfuric acid): $\text{H}_2\text{O}_2$  (hydrogen peroxide): $\text{H}_2\text{O}$  (de-ionized water) at a ratio of 4:1:1, followed by de-ionized water rinsing and dehydrating using isopropyl alcohol (IPA). Once in the MBE system, the substrates were cleaned using an ion sputter at  $200^\circ\text{C}$  for 20 min. They were then annealed at  $550^\circ\text{C}$  for 45 min, and then allowed to cool. The surface flatness and reconstruction of the GaAs(100) substrates were determined by reflection high energy electron density (RHEED). The Fe films were then grown at  $50^\circ\text{C}$  and  $1 \times 10^{-10}$  mbar. The growth rate was kept constant, by ensuring the emission current between the filament and the source material was constant. For the Fe film, the flatness and the uniformity along the  $[011]$  direction was checked using RHEED. The patterns showed epitaxy on GaAs(100) with the relationship  $\text{Fe}(100)\langle 001 \rangle \parallel \text{GaAs}(100)\langle 001 \rangle$ . The evaporation procedure was then repeated for the 7 ML thick Au overlayer. The thickness of the Fe films ranged from 7 to 50 ML.

The magnetization (presented as normalized to saturation) was measured on a MOKE magnetometer. The films were strained along the  $[011]$  direction, using a specially designed bending tool, over four different bend radii ( $R = 220\text{--}280$  mm). The magnetizations were measured along the  $[0\bar{1}1]$  direction (Villari effect). The absolute magnetizations were measured on a vibrating sample magnetometer (VSM).

## 3. Results and discussion

The anisotropy constants were determined using the MOKE fitting method outlined in section 1. For the 30 ML Fe film, the anisotropy constants determined by this method were  $K_u = 29\,000 \pm 2\,300 \text{ J m}^{-3}$  and  $K_1 = 32\,000 \pm 2\,500 \text{ J m}^{-3}$ . From the literature [9] (solid curve in figure 1) the constants are  $K_u = 27\,800 \text{ J m}^{-3}$  and  $K_1 = 32\,400 \text{ J m}^{-3}$ ; thus the data from the fitting are within error of the literature data. In figure 1, the anisotropy constants are plotted as a function of thickness. It is observed that the cubic anisotropy constants increased as the film thickness increased, tending towards values characteristic of bulk Fe, while the uniaxial anisotropy constant decreased as the film thickness increased. From the literature the



**Figure 1.** Anisotropy constants for the Fe/GaAs films, as a function of Fe thickness. The solid curves represent the anisotropy constants model presented by Brockmann [9]. The dashed curves represent the equations which describe the anisotropy constants as a function of thickness. The bulk Fe cubic anisotropy constant is also plotted.

thickness dependence of the anisotropy constants can be most simply represented by [4]

$$K = K_v + \frac{K_s}{t} \quad (6)$$

where  $K_v$  is the volume component of the anisotropy,  $K_s$  is the surface or interface component of the anisotropy and  $t$  is the Fe film thickness. Equation (6) was applied to determine the thickness dependence of the anisotropy constants in figure 1. Thus the cubic components were  $K_{1v} = 44500 \pm 600 \text{ J m}^{-3}$  and  $K_{1s} = (-5.02 \pm 0.2) \times 10^{-5} \text{ J m}^{-2}$  and the uniaxial components were  $K_{uv} = 0 \text{ J m}^{-3}$  and  $K_{us} = (1.27 \pm 0.08) \times 10^{-4} \text{ J m}^{-2}$  (dashed curves in figure 1). Comparing to data in the literature [9] (solid curve in figure 1), there is a good agreement between the data from this MOKE fitting method constants and the literature. This gives confidence in the later analysis.

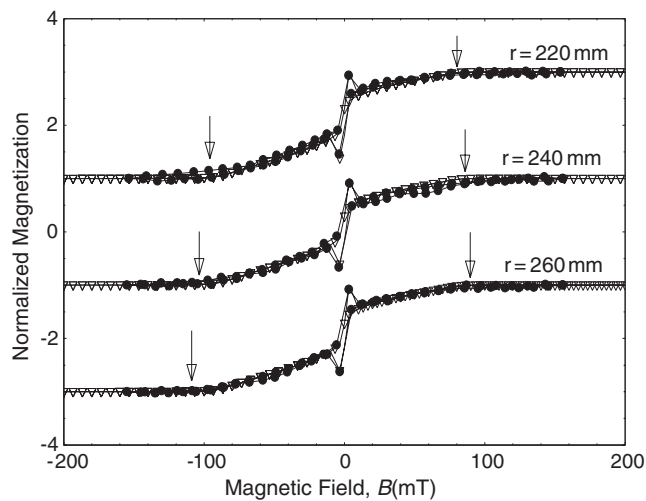
The saturation magnetostriction constant ( $\lambda_s$ ) for the [011] direction was determined by uniformly straining each film over a range of bend radii (figure 2). From figure 2, it is seen that the anisotropy fields decreased as the strain on the film was increased. This means the magnetostriction constant was negative along the field direction. The experimental magnetostriction constants were determined by plotting the anisotropy fields as a function of the inverse bend radius, and using the equation [18]

$$\lambda_s = \frac{dH_k}{d\frac{1}{R}} \frac{2\mu_0 M_s (1 - \nu^2)}{3\tau Y} \quad (7)$$

where  $H_k$  is the anisotropy field,  $R$  is the bend radius,  $\nu$  is the Poisson ratio,  $\tau$  is the thickness of the substrate and  $Y$  is the Young's modulus of the substrate.

The magnetostriction constants were also determined by adding a uniaxial strain anisotropy term ( $K_\sigma = \frac{3}{2}\lambda_s\sigma$ ), to the magnetic energy density equation (equation (1)) and using the MOKE fitting method:

$$F = \frac{1}{4}K_1(t) \sin^2 2(\varphi - a) + K_u(t) \sin^2\left(\varphi - a + \frac{\pi}{4}\right) + K_\sigma \sin^2(\varphi - b) - HM \cos \varphi \quad (8)$$

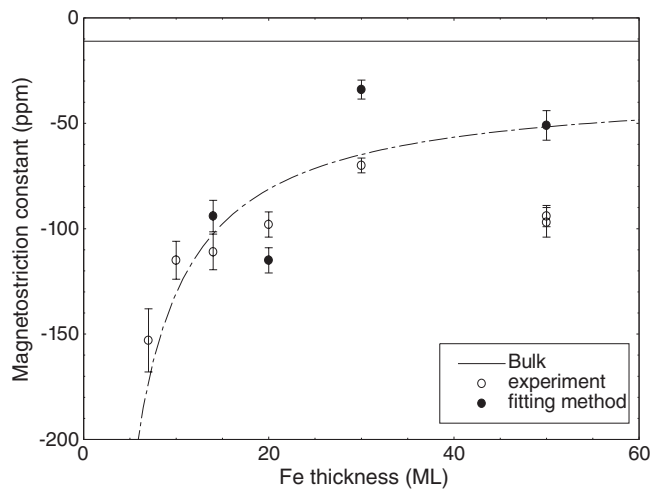


**Figure 2.** Normalized magnetization for the 30 ML Fe/GaAs films as a function of applied magnetic field and bend radius,  $r$ . The solid circles represent the measured data and the black triangles represent the MOKE fitting method data. The arrows mark the anisotropy field for each loop.

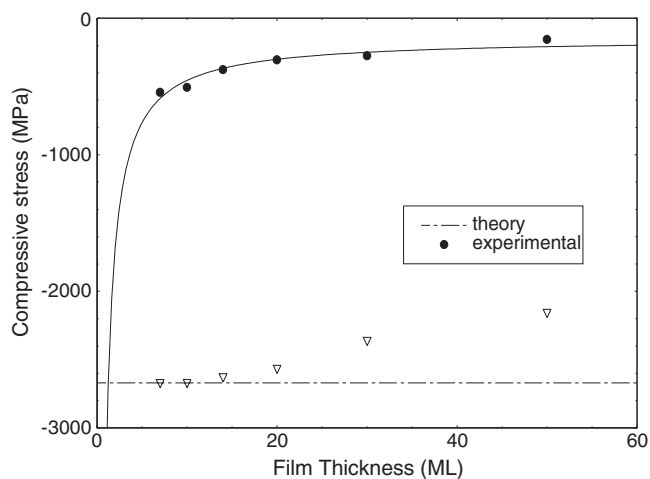
where  $b$  is the angle between the applied field and the applied stress ( $\sigma$ ). For each bend radius, the anisotropy constants determined for the unstrained film were used (figure 1), so the only unknown variable in equation (8) was  $K_\sigma$ . This assumed that the original uniaxial anisotropy constant did not have any strain dependence. A second set of magnetostriction constants were determined by assuming that  $K_u = 0$  in equation (8), so only  $K_\sigma$  was determined. This was allowed as in equation (8), the two uniaxial terms can be combined to give  $(K_u + K_\sigma) \sin^2(\varphi - \frac{\pi}{2})$ , as the films were strained along the [011] axis, with the field along the  $[0\bar{1}1]$  axis. This is effectively giving  $K_u$  a strain dependence, which is ascribing it to a magnetoelastic effect. For both data sets, the magnetostriction constants were determined by plotting the strain anisotropy constant,  $K_\sigma$ , against the inverse bend radius, and assuming the uniform stress relation  $\sigma = \frac{\tau Y}{2R(1-\nu^2)}$  for the ultrathin film on the substrate. From figure 3, it is observed that there is good agreement between the two sets of magnetostriction constants determined from the MOKE fitting method (equation (8)), which means that assuming the uniaxial anisotropy term had strain dependence was valid.

For all six films measured, the magnetostriction constants for the [011] direction are plotted in figure 3. It is seen that for all films the magnetostriction constant was negative, and had a more negative value than bulk Fe in the [011] direction. In general, the magnetostriction constant increased to more negative values as the thickness of the Fe decreased, with an inverse thickness dependence (dashed curve in figure 3). From previous research on the magnetostriction constant of ultrathin magnetic films, it was found that the behaviour of the film depended on the substrate [19], the roughness of the substrate surface [20] and the fabrication method [21]. The magnetostriction constant can increase or decrease as the film thickness decreases, which is taken to be an interface effect described by Néel's phenomenological model [22]. Hence for the Fe/GaAs films, the change in the magnetostriction as a function of thickness can indeed be attributed to an interface effect.

As the uniaxial anisotropy and magnetostriction constant both vary with thickness, it is possible that the uniaxial anisotropy was caused by strain due to the lattice mismatch between the Fe atoms and the GaAs lattice. If this was the case then  $K_u$  is a stress

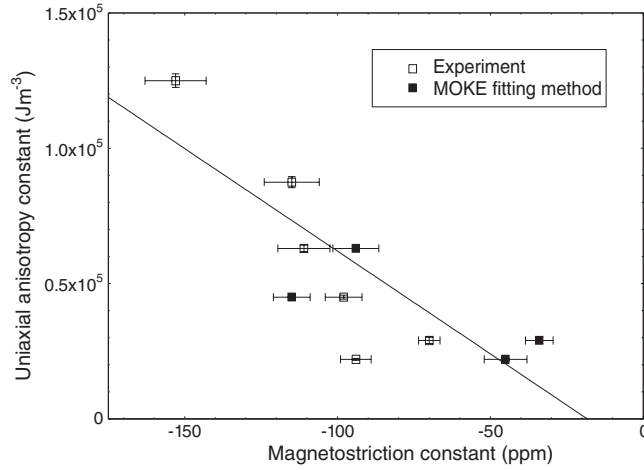


**Figure 3.** Magnetostriction constant for Fe/GaAs along the [011] direction, as a function of film thickness. The open shapes represent the experimental data, the solid circles represent the MOKE fitting method data with  $K_u$  and  $K_\sigma$  in equation (7), and the solid squares represent the MOKE fitting method data with  $K_u = 0$  in equation (7). The dashed curve is proportional to  $1/t$ .



**Figure 4.** Compressive stress due to the lattice mismatch of Fe on GaAs substrate as a function of Fe film thickness. The dashed line represents the lattice stress calculated from the elastic constants for strain  $\varepsilon = -1.3\%$ , and the triangles represent the stress calculated using Thomas's lattice strain as a function of thickness [10]. The solid shapes represent the stress calculated from the uniaxial anisotropy constant and magnetostriction constant, and the solid curve is proportional to  $1/\text{film thickness}$ .

anisotropy constant given by  $\frac{3}{2}\lambda_s\sigma$ . Thus for each film, the stress ( $\sigma$ ) which could have caused the uniaxial anisotropy was determined from the experimental anisotropy constants and the magnetostriction constants (figure 4). From figure 4, it is observed that the stress was compressive and has inverse dependence on thickness (solid curve). The effective stress



**Figure 5.** Uniaxial anisotropy constant as a function of the magnetostriction constant for each Fe/GaAs film. The open shapes represent the magnetostriction constants determined from experimental data, and the closed shapes represent the magnetostriction constants determined from the MOKE fitting method. The solid line is a guide for the eye.

arising from the lattice mismatch can be calculated from

$$\sigma = \left( c_{11} - \frac{c_{12}^2}{c_{11}} \right) \varepsilon_1 + \left( c_{12} - \frac{c_{12}^2}{c_{11}} \right) \varepsilon_2 \quad (9)$$

where  $c_{11}$  and  $c_{12}$  are the elastic constants of Fe, and  $\varepsilon_i$  is the lattice mismatch. The values of the elastic constants for Fe are found in the literature [8] (we have to take bulk values, but this may be invalid for an ultrathin film). For a GaAs(100) substrate in the  $1 \times 1$  reconstruction, the lattice mismatch with Fe is  $\varepsilon_1 = \varepsilon_2 = -1.3\%$ . This gave a constant stress which was an order of magnitude larger than the stress calculated from  $K_u$  (dashed curve). From figure 4, the Fe thickness which has the calculated lattice stress is 1.2 ML. One reason for this difference in the stress estimated from the anisotropy and the lattice mismatch is that as the film thickness increases, the stress due to the lattice mismatch decreases, i.e. the lattice relaxes, and hence the uniaxial anisotropy decreases. This change in lattice strain in Fe/GaAs films was confirmed by Thomas [10]. Using the data presented by Thomas, the change in the *average* compressive stress with film thickness due to the relaxation of the lattice was determined (triangles in figure 4). These stresses are still roughly five times larger than the stresses calculated from the uniaxial anisotropy constants. Thomas used an Al overlayer, which may have changed the magnetic properties of the film, compared to an Au overlayer [23]. It is not possible to determine the magnetostriction constants of Fe/GaAs films for thicknesses less than 5 ML, as the films are not magnetic at room temperature. Thus it is impossible to determine how the uniaxial anisotropy constant changes near the interface. Thomas [10] suggested that the uniaxial anisotropy for films thinner than 15 ML was due to the interface anisotropy, while for thicker films the magnetic anisotropy is the result of a competition between magnetoelastic coupling and interface anisotropy. Our results show that the uniaxial anisotropy is related to the strain, but probably not due to the lattice mismatch, which agrees with Thomas. Plotting the unstrained uniaxial anisotropy constants against the magnetostriction constant gives an approximately linear relationship (figure 5). This upholds the conclusion that the uniaxial anisotropy is related to the magnetoelastic energy in the film.



#### 4. Conclusions

The anisotropy constants of the Fe/GaAs films in this paper were determined by using the MOKE fitting method. The constants calculated were within error of those found in the literature or Brillouin light scattering experiments. The saturation magnetostriction constant along the [011] direction of Fe/GaAs films increased to more negative values as the film thickness decreased. It was also determined that the uniaxial anisotropy is related to the strain in the film, although whether its origin is the lattice mismatch between the Fe film and the GaAs lattice is still uncertain.

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