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Anisotropies and magnetostriction constants of epitaxial Co films on GaAs(100) substrates

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Abstract

Metastable body centred cubic (bcc) Co has been grown as an epitaxial thin film on GaAs, using molecular beam epitaxy. In this paper, epitaxial Co films were grown on GaAs(100) substrates with 1×1 surface construction. The film thicknesses cover the bcc–hcp (hexagonal close packed) Co transition. The magnitudes and symmetries of the anisotropy for these films were estimated from normalized magnetization data measured using a magneto-optic Kerr effect magnetometer. It was determined for these films that there was an early onset of the hcp phase and of the uniaxial anisotropy component. The magnetostriction constants were determined by straining the films over a series of bend radii and determining the change in the anisotropy field (the Villari effect). The large uniaxial anisotropy observed in one case may be ascribed to a large magnetostriction constant for that film.

1. Introduction

Bulk magnetic Co has a hexagonal close packed (hcp) crystal structure. Its anisotropy is uniaxial with the easy direction along the c -axis, normal to the close packed plane. In 1985 Prinz fabricated, using molecular beam epitaxy (MBE), a thin film ($t = 35.7$ nm) of Co on a GaAs(110) wafer, which had the metastable body centred cubic (bcc) structure [1]. The bcc structure of the Co film was deduced from reflection high energy electron diffraction (RHEED) analysis. Over recent years further research on this metastable phase of Co on GaAs has been reported with a wide range of results and conclusions [2–7]. This paper gives a review of the major results for these films, and attempts to systematize the findings. Further structural and magnetic data on metastable Co films are presented, for Co films grown on the 1×1 surface construction of GaAs(100).

From the literature, the standard technique for fabricating the bcc Co/GaAs films has been MBE. This technique controls the energy of the atoms, allowing epitaxial films to be

grown. A range of characterization techniques has been used to determine the properties of the films, including transmission electron microscopy (TEM) [3, 7], Brillouin light scattering (BLS) [5], polarized neutron reflection (PNR) [2] and magneto-optic Kerr effect (MOKE) magnetometry [2, 3, 7].

The first bcc Co film [1] was grown on the (110) plane of a GaAs wafer, while most of the other Co films were grown on the (001) plane of GaAs [2, 3, 5, 7]. This difference in GaAs orientation has led to differences in the Co crystal structure grown for a given Co film thickness. For the Co/GaAs(110) films, the Co bcc structure has been grown to thicknesses of 35.7 nm [1, 4]. Early work on the Co/GaAs(001) films [2] suggested the bcc phase grew up to 5 nm, with the bcc phase being distorted at and above 4 nm. For films thicker than 5 nm, a polycrystalline hcp phase grew. This was confirmed by TEM measurements by Gu *et al* [3] who determined that at 5 nm two hcp structures grew, with their *c*-axes perpendicular to each other. The growth process, for the Co/GaAs(001) films was published by Wu *et al* [7], who clarified that the Co initially grows in the bcc structure up to 1.5 nm thick, with the epitaxial relationship $(001)[001]\text{Co} \parallel (001)[001]\text{GaAs}$ [7]. After 1.5 nm, the structure starts to contain two hcp structures, which grow with the relation $(1\bar{2}10)[0001]\text{Co} \parallel (001)[\bar{1}10]\text{GaAs}$ and $(1\bar{2}10)[0001]\text{Co} \parallel (001)[110]\text{GaAs}$, i.e. the *c*-axis of the hcp structure is either along the [110] direction or the $[\bar{1}10]$ direction in the plane of the film [5]. The only work which disagrees with this is that of Subramanian *et al* [6], which claims a bcc Co/GaAs(001) film of thickness 21.6 nm. Unfortunately this discrepancy cannot be clarified as no structural evidence, such as TEM or RHEED, was presented by Subramanian *et al*. In general we conclude that bcc Co can be grown up to 1.5 nm thick on GaAs(001), compared to 35.7 nm on GaAs(110). For the Co/GaAs(001) films, the surface reconstruction of the GaAs was either 4×6 [3, 5] or 4×2 [7].

These changes in the Co crystal structure lead to changes in the anisotropy in the films. Madami *et al* [5] suggested that for films thinner than 1.6 nm, the in-plane anisotropy was cubic, which came from the bcc structure. At about 1.6 nm, the bcc cell distorted and the hcp structures started to grow. This distortion of the structure changed the anisotropy to uniaxial. Once the hcp structure had started to grow, the anisotropy was fourfold or cubic. This was due to the [0001]-axis of the hcp phase lying in the surface plane either along [110] or $[\bar{1}10]$ for the bcc phase with the two directions being equal in energy. As the films got thicker (between 5 and 7 nm), the [0001]-axis preferentially grew along the GaAs $[\bar{1}10]$ direction, which led to dominant uniaxial anisotropy, mixed with the cubic anisotropy. From figure 1, it is observed that for all Co films with thicknesses less than 7 nm the dominant in-plane anisotropy was cubic, apart from for the 1.6 nm Co film studied by Madami *et al* [5]. Similarly for most of the Co films thicker than 7 nm, uniaxial anisotropy was observed, with the easy axis along the $[\bar{1}10]$ direction. Thus the changes in the anisotropy as a function of thickness for the Co/GaAs(001) films are consistent within the literature, and can be supported by the structural studies.

The anisotropy constants (uniaxial and cubic) determined for the Co/GaAs(001) films in the literature are plotted as a function of film thickness (figure 1). From figure 1(a) it is observed that the cubic anisotropy constants, K_1 , are described by the relationship [8]

$$K_1 = K_{1v} + \frac{K_{1s}}{t} \quad (1)$$

where K_{1v} is the cubic volume anisotropy component and K_{1s} is the cubic surface or interface anisotropy component [9, 10]. For these Co/GaAs films, the cubic anisotropy constant values are $K_{1v} = 24\,100 \pm 850 \text{ J m}^{-3}$ and $K_{1s} = (2.31 \pm 0.08) \times 10^{-5} \text{ J m}^{-2}$ (solid black line in figure 1(a)). The 1.6 nm point was neglected as this was the thickness where the bcc cell distorted [5]. As equation (1) describes the relationship between the cubic anisotropy constants and the film thickness, this means the bcc contribution to the cubic anisotropy follows standard thickness dependence. From figure 1(b), it is observed that the uniaxial anisotropy constants,

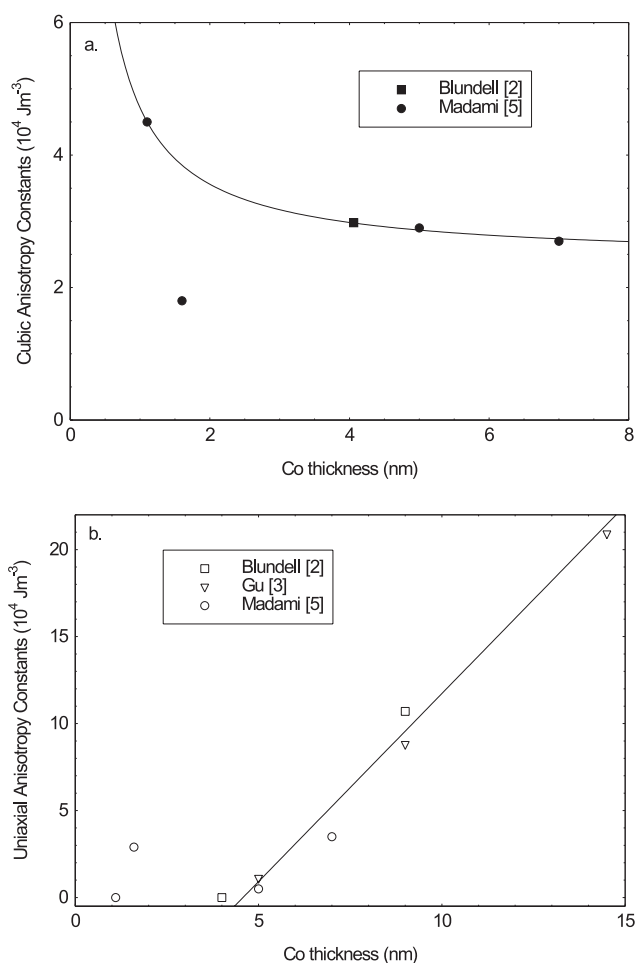


Figure 1. (a) Cubic anisotropy constants for epitaxial Co on GaAs(001) substrate as a function of film thickness ($t < 8$ nm). The solid line is a fit through the cubic anisotropy constants. (b) Uniaxial anisotropy constants for epitaxial Co on GaAs(001) as a function of film thickness ($t < 15$ nm). The solid line is a linear fit through the $t > 2$ nm anisotropy constants. The data are taken from the literature.

K_u , as a function of film thickness do not obey equation (1). For the Co films thicker than 4 nm, the relationship between the uniaxial anisotropy constants and the film thickness is linear (solid black line). The uniaxial anisotropy constants are increasing towards the bulk Co uniaxial anisotropy constant of $41 \times 10^4 \text{ J m}^{-3}$ [11]. This ties in with the [0001]-axis preferentially growing along the GaAs $[\bar{1}10]$ direction, as the Co thickness increases. For thicknesses less than 4 nm, no uniaxial anisotropy is observed in the films (figure 1(b)), apart from at 1.6 nm.

There is imperfect epitaxy between the hcp Co and GaAs for any GaAs orientation. For the Co $(\bar{1}2\bar{1}0)$ plane on GaAs(001), the lattice mismatch for the [210] direction along the [110] direction is 7.9% and the [001] direction along the $[\bar{1}10]$ direction is 1.8%. This could lead to an increase or change in the anisotropy of the Co films due to the internal strain [12]. Bulk Co has non-zero saturation magnetostriction constants in the (001) plane, $\lambda_s = -93$ ppm, but along the [001] direction, $\lambda_s = 0$ ppm. This combination may be assumed to give rise to a

magnetoelastic contribution to the magnetic anisotropy. In order to evaluate this contribution an independent measure of the saturation magnetostriction constants of the thin films is required. Following on our work on Fe on GaAs(100) [13, 14] we have determined the magnetostriction constants as a function of film thickness.

2. Experimental set-up

The epitaxial Co films on GaAs(100) substrates with a Cr overlayer were fabricated using molecular beam epitaxy (MBE) [15]. Prior to each film's deposition, the substrates were etched using H₂SO₄ (sulfuric acid):H₂O₂ (hydrogen peroxide):H₂O (de-ionized water) at a ratio of 4:1:1, and this was followed by de-ionized water rinsing and de-hydrating using isopropyl alcohol (IPA). Once in the MBE system, the substrates were cleaned using an ion sputter at 200 °C for 20 min. They were then annealed at 550 °C for 45 min, and then allowed to cool. The surface flatness and construction of the GaAs(100) substrates were determined by reflection high energy electron diffraction (RHEED). The RHEED images showed that the surface construction of the GaAs was 1 × 1. The Co films were then grown at 50 °C and 1 × 10⁻¹⁰ mbar. The thicknesses of the four Co films fabricated ranged from 0.7 to 3.5 nm. For the Co film, the flatness and uniformity along the [011] direction and the crystal structure were checked using RHEED (figure 2). The evaporation procedure was then repeated for the 2 nm thick Cr overlayer. RHEED images of the Cr overlayers were taken after deposition. For all the films, the RHEED images showed two lines of dots, which means that the Cr grew with a cubic lattice structure on the Co.

The magnetization (presented as normalized to saturation) was measured using a MOKE magnetometer. To determine the magnetostriction constants, the films were strained along the [0 $\bar{1}$ 1] direction, over four different bend radii ($R = 220$ – 280 mm), using a bending tool. The anisotropy fields were measured along the [011] direction (the Villari effect). This technique has been used successfully in the study of Fe/GaAs films [13, 14]. The experimental magnetostriction constants were determined by plotting the anisotropy fields as a function of the inverse bend radius, and using the equation [16]

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

where H_k is the anisotropy field, R is the bend radius, ν is the Poisson ratio, τ is the thickness of the substrate and Y is the Young's modulus of the substrate.

3. Results and discussion

3.1. In-plane anisotropy

At the end of the growth of the Co onto the GaAs(100) substrate, before the Cr capping layer was grown, a RHEED image was taken (figure 2). From these RHEED images, the crystal structures of these films can be determined [7]. For the 0.7 nm film (figure 2(a)), two lines of dots are observed, which relate to a bcc structure; this is consistent with the results in the literature [7]. For the 1.4 nm film (figure 2(b)), an additional set of dots between the bcc lines are observed; this means that the structure contains hcp structures, as well as a bcc structure. From the literature [5, 7], the hcp structures started to grow at thicknesses above 1.6 nm, and thus we have an early onset of the hcp growth in our films. This could be due to the GaAs surface construction; our films were grown on the 1 × 1 surface construction, while most of the films in the literature were grown on either the 4 × 2 [2, 7] or 4 × 6 [3, 5] surface reconstructions.

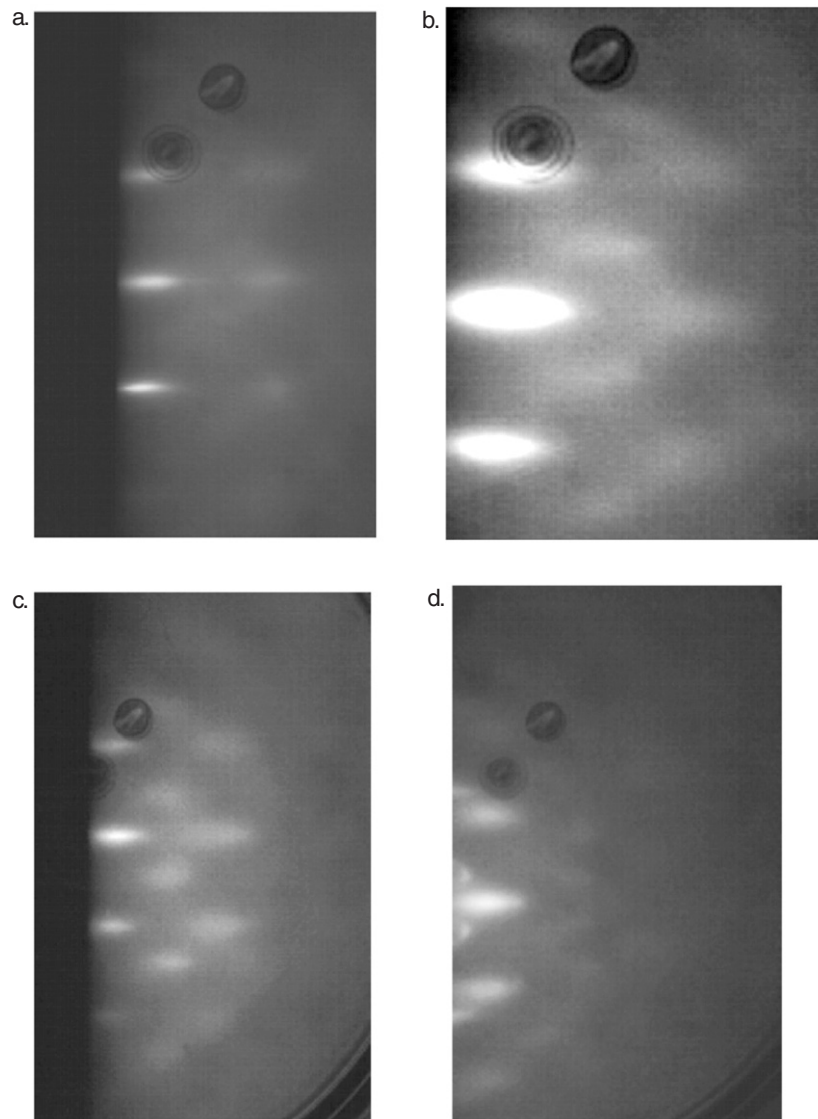


Figure 2. RHEED images of the Co/GaAs(100) films for Co thicknesses of (a) 0.7 nm, (b) 1.4 nm, (c) 2.1 nm and (d) 3.5 nm.

For the 2.1 and 3.5 nm films (figures 2(c) and (d)), the crystal structures also have a mixing of the hcp and bcc structures, which is consistent with the literature [3, 7]. We are unable to say if there was further change to the co-structure from the Cr cap.

The 0.7 nm thick Co film showed no magnetization from measurements made using the MOKE magnetometer. This is probably due to the Co growing in clusters (Volmer–Webber mode) rather than as continuous films (van der Merwe mode) up to a certain thickness. For the 1.4 nm film (figure 3(a)), the in-plane anisotropy observed was cubic (or fourfold), which is consistent with the previous data (figure 1(a)). The magnetization loops measured for the field along the hard axes ($[01\bar{1}]$ and $[0\bar{1}1]$) have no coercivity and the remanent magnetization

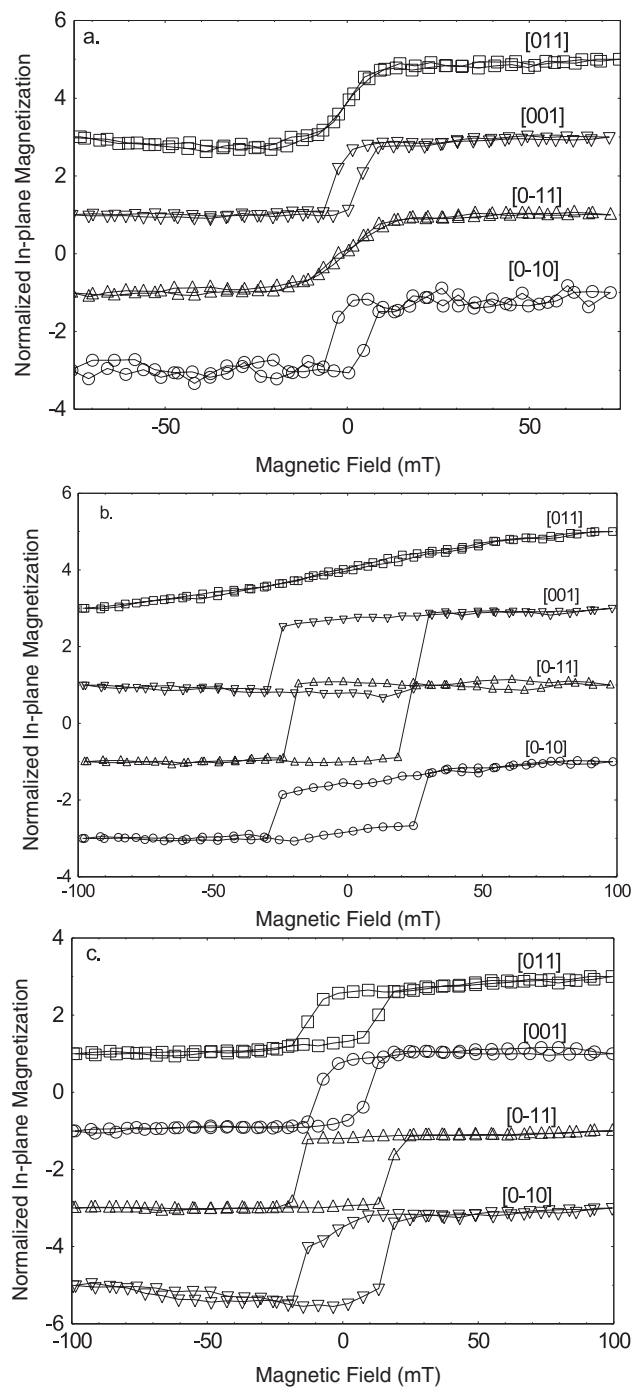


Figure 3. Normalized in-plane magnetization as a function of magnetic field, for the (a) 1.4 nm Co/GaAs film, (b) 2.1 nm Co/GaAs film and (c) 3.5 nm Co/GaAs film.

is approximately zero (figure 3(a)); thus they do not comply with the coherent rotation magnetization model. For the 2.1 nm Co film, the in-plane anisotropy observed was uniaxial

(figure 3(b)), with the hard axis along the [011] direction of the GaAs substrate. This suggests that the c -axis of the hcp Co is only along the $[0\bar{1}\bar{1}]$ GaAs direction. For the 3.5 nm Co film, the in-plane anisotropy is weakly uniaxial (figure 3(c)). The uniaxial hard axis magnetization loop ([011] GaAs direction) has large coercivity and remanent magnetization (figure 3(c)) in comparison to the 2.1 nm Co film hard axis magnetization loop. One possible reason for the magnetization loops measured for the 3.5 nm Co/GaAs film being almost isotropic is the mixed crystal structure (hcp and bcc).

From the literature [3, 5, 7] and figure 1(a), it would be expected that for all three film thicknesses, cubic anisotropy would be dominant, as the films were thinner than 7 nm. Only the 1.4 nm film had cubic anisotropy; the other two films (2.1 and 3.5 nm) had uniaxial anisotropy. As both of these films were thicker than 1.6 nm and had hcp structures present in the film (figures 2(c) and (d)), the uniaxial anisotropy observed was not due to the distortion of the bcc cell [5]. Another possible reason for the 2.1 and 3.5 nm films having uniaxial anisotropy is the surface construction of the GaAs(100) substrate. For our films, the surface construction was 1×1 ; thus there are no Ga–Ga dimers or trenches on the surface [17], while some of the other Co films were grown on the 4×6 [3, 5] surface reconstruction of GaAs, which is Ga rich and has trenches along the $[0\bar{1}\bar{1}]$ direction [17]. This difference in GaAs surface may have led to the differences observed in the in-plane anisotropies and the crystal structure. For our films, the RHEED images show that the 1.4 nm Co film had hcp structure (figure 2(b)). Thus the hcp structure grew at smaller thicknesses compared to Wu's data [7], where the hcp structure was observed in films thicker than 2 nm. This suggests that the [0001]-axis grew equally along the GaAs [011] and $[0\bar{1}\bar{1}]$ directions, as the in-plane anisotropy was cubic (figure 3(a)). For the 2.1 and 3.5 nm films, the hcp structure was observed in the RHEED images (figures 2(c) and (d)), and the measured anisotropy was uniaxial; this suggests that the Co grew with the [0001]-axis along the GaAs $[0\bar{1}\bar{1}]$ direction. This again occurred at Co thickness less than those in the literature (figures 1(a) and (b)). This suggests that the bcc Co films grown on the 1×1 GaAs surface construction were less stable than those grown on the 4×6 GaAs surface reconstruction. Thus an early onset of the hcp structure and uniaxial anisotropy was observed.

The overlayer on epitaxial films can also change the magnetocrystalline anisotropy [18]. For our films, Cr was used to cap the films and stop the Co oxidizing. In the literature, Wu *et al* used Cu as the overlayer and determined that there was no difference between the capped and uncapped magnetization hysteresis loops of the Co/GaAs films [7]. Gu *et al* also used Cr as the overlayer [3]. For their Co/GaAs film, cubic anisotropy was observed at 5 nm, and at 9 nm the uniaxial anisotropy was dominant, which is consistent with the rest of the literature [2, 5]. Also from epitaxial Co/Cr(001) superlattice measurements, the interdiffusion of the Cr into the Co at the interface is less than 0.2 nm [19]. Thus the Cr overlayer could have affected the anisotropy of our films, but it would seem more likely that the GaAs surface reconstruction was the dominant factor.

3.2. Magnetostriction constants

For all the Co/GaAs(100) films, the magnetostriction constants were determined using equation (2). For each Co film, the magnetostriction constant was determined for the direction [210], which lay along the GaAs[011] direction. The bulk magnetostriction constants were also determined by assuming that the [0001]-axis grew along the $[0\bar{1}\bar{1}]$ direction, then using the magnetostriction equations for a hexagonal crystal [20]. For bulk Co, the magnetostriction constant along the [001] direction is 0 ppm and that along the [210] direction is -50 ppm [21]. From figure 4, it is observed that the magnetostriction constants of all three films were negative. The 1.4 nm film's magnetostriction constant ($\lambda_s = -82 \pm 30$ ppm) was similar to the bulk

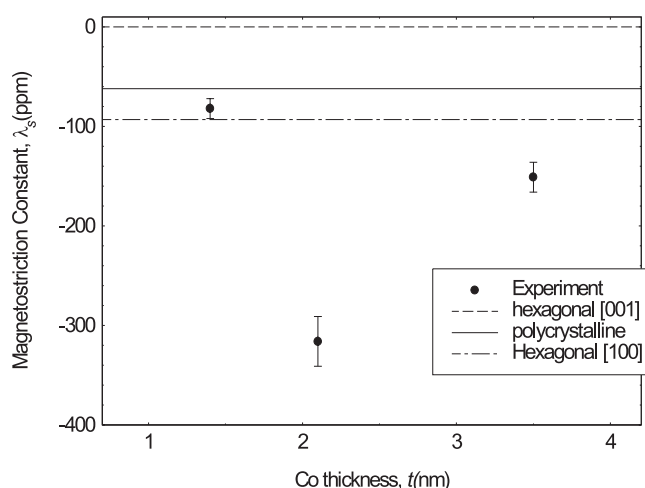


Figure 4. Magnetostriction constant for the Co/GaAs(100) films as a function of film thickness. The solid black line represents the polycrystalline Co magnetostriction constants, and the dashed lines represent the bulk hcp Co magnetostriction constants.

values, while the 2.1 and 3.5 nm Co films' magnetostriction constants ($\lambda_s = -316 \pm 85$ and -151 ± 45 ppm respectively) were more negative than the bulk values. The large magnetostriction constants for the 2.1 and 3.5 nm Co films could be due to the large lattice mismatch between the hcp Co film and the GaAs substrate. The 2.1 nm Co film had the most negative magnetostriction constant, and the strongest uniaxial anisotropy. This suggests that the large magnetostriction constant of the film could have increased the uniaxial anisotropy of the film in comparison to the 3.5 nm Co film [12].

4. Conclusions

For epitaxial Co grown on GaAs(100), with a 1×1 surface construction, the metastable bcc phase was observed in the RHEED images at 0.7 nm, but the film was not magnetic. For 1.4 nm Co/GaAs(100), a mixed bcc and hcp structure was observed. The film had cubic anisotropy, due to the c -axis of the hcp structure being equally along the $[011]$ and $[01\bar{1}]$ directions. For the 2.1 and 3.5 nm Co/GaAs(100) films the anisotropy was uniaxial, suggesting that the hcp structure observed in the RHEED images was growing with the c -axis along the $[01\bar{1}]$ direction of the GaAs. The anisotropy data suggest that the surface construction of the GaAs films is an important factor in how the Co films are fabricated. For the Co/GaAs(001) films reported in the literature, the surface reconstruction was 4×6 , and the uniaxial anisotropy became dominant at 7 nm, while for the 1×1 surface construction, the uniaxial anisotropy was dominant by 2.1 nm. The magnetostriction constants were measured for the 1.4, 2.1 and 3.5 nm Co films, and were determined to be negative and larger than the bulk Co values. Also the Co/GaAs films with the largest magnetostriction constants (2.1 and 3.5 nm) had uniaxial anisotropy.

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