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Anisotropy of an ultrathin Cu(111)/Co(111)/Cu(111) wedge

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Abstract. An anisotropy study is reported on an ultrathin fcc Cu(111)/Co(111)/Cu(111) wedge grown on a sapphire substrate by molecular beam epitaxy. Room temperature polar and longitudinal magneto-optical Kerr effect (MOKE) measurements were analysed by two different methods to determine the volume and surface contributions to the anisotropy. The values obtained by the 'area method' by calculating the difference in area between the in-plane and perpendicular magnetization curves and also by simulation of the polar MOKE curves were not in agreement. The 'area method' was found to be unreliable for analysing MOKE data. Values of the anisotropy obtained by simulation agreed with the expected bulk value for fcc Co of 0.0 MJ m^{-3} and the surface anisotropy was found to lie in the range 0.4–0.5 mJ m^{-2}.

1. Introduction

Magnetic surface anisotropy in epitaxial magnetic ultrathin films and multilayers remains a subject of major technological importance and fundamental scientific interest [1]. An understanding of why the magnetization favours a given direction in a material is important for the engineering of magnetic structures with the desired properties. It is well known that a surface or interface anisotropy which is more than an order of magnitude larger than the bulk anisotropy may occur in cubic transition metal thin films [2]. In cases where the anisotropy field is greater than the demagnetizing field, perpendicular magnetization may be established which is of importance for potential applications.

Ultrathin Co structures have been studied extensively since the discovery of the perpendicular magnetic anisotropy in sputter-deposited Co/Pd multilayers [3]. The magnetic anisotropy of the Co/Cu(111) system has been studied by several groups using a variety of different techniques such as conventional magnetometry [4–6], torque magnetometry [7, 8], Brillouin light scattering [5], magneto-optical Kerr effect (MOKE) [9, 10] and ferromagnetic resonance (FMR) [11]. However, the values of the anisotropy constants quoted in the literature vary considerably although in all studies the total effective anisotropy is found to be negative indicating that in-plane magnetization is favoured except at very low Co thicknesses where a transition to perpendicular magnetization is sometimes seen [4, 5, 8].

Most reported work has concentrated on studies of multilayers [4, 5, 7] or samples consisting of Co(111) grown on a single layer of Cu(111) [6, 8, 10, 11]. Only one previous study has been reported on a Cu(111)/Co(111)/Cu(111) trilayer in which the Cu(111) thickness was varied [10]. In this paper, we report an anisotropy study of an ultrathin Cu/Co/Cu(111) wedge grown on sapphire with varying Co thickness.

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Experimentally the determination of the magnitude of the surface or interface anisotropy is difficult due to contributions from other terms (such as magnetocrystalline, shape and magnetoelastic anisotropy) which must be separated from the total anisotropy. For the analysis of thin films and multilayers the effective magnetic anisotropy, K_{eff}, of a magnetic layer of thickness, t, may be written as [12]

$$K_{eff} = K_v + 2K_s/t \tag{1}$$

where K_v is the bulk or volume contribution per unit volume (including the shape anisotropy) and K_s is the surface or interface contribution per unit area. The factor of two is included to account for the fact that each magnetic layer is assumed to be bounded by two identical interfaces.

This analysis has been widely used in experimental studies [4–6, 11] since values for K_v and K_s can be determined by plotting a graph of $K_{eff}t$ against t. K_v can then be obtained from the slope, the sign of which gives the sign of K_v and therefore gives an indication of whether perpendicular or in-plane magnetization is favoured. The intercept at $t = 0$ gives $2K_s$. A critical thickness $t_\perp$ can also be obtained when $t = -2K_s/K_v$ (i.e. $K_{eff} = 0$), indicating the thickness below which the sample is perpendicularly magnetized because the surface contribution outweighs the volume anisotropy.

However de Jonge *et al* [1] pointed out that this expression must be utilized with caution. Firstly the above description is based on the assumption that the local anisotropy at the interface is experienced by the system as a whole and that the system behaves as one magnetic entity such that the individual magnetic moments are aligned. This is true only when the anisotropy is much smaller than the intralayer exchange, or in other words, when the layer thickness is less than the 'exchange length'; in Co-based films the exchange length is around 30 Å. Secondly the validity of separating the anisotropy into volume and surface terms is uncertain when considering layer thicknesses of a few monolayers. Chappert and Bruno [13] also pointed out that it is not always the case that K_v is independent of film thickness particularly as stress can take the form of a $1/t$ dependence on K_v which would mistakenly be assumed to be due to a surface contribution in the above analysis. Kohlhepp *et al* [8] argued that in order to isolate the true Néel surface anisotropy, systems should be used where the strain anisotropies can be neglected or are independent of t. They suggested that Co(111) on Cu(111) is expected to be a good candidate for such a system since the lattice mismatch is small and strain contributions are only expected to be significant for Co thicknesses well in excess of 30 Å.

Recent work by Farle *et al* [11] has shown that this might not be the case in the Co/Cu(111) system. Conversely they found that strain played a dominant role, obtaining a value of K_v for hcp Co/Cu(111) films of less than 5.5 monolayers which could almost entirely be derived from the expected magnetoelastic anisotropy values for either hcp Co and trigonal distorted fcc and did show a $1/t$ dependence in K_v. However, they found that the magnetoelastic contribution was very small in thickness regimes where a relaxed cubic fcc structure dominated.

The use of the MOKE to measure magnetization has several advantages over conventional magnetometry in the study of the anisotropy of ultrathin films since the magnetization is only monitored in the region illuminated by a focused laser spot. It therefore provides a highly localized measure of the magnetization and allows a positional scan along a wedge shaped magnetic layer. The magnetic properties can be investigated as a function of layer thickness in one single sample thus being free of the problems associated with structural variations in samples grown at different times.

In this paper we report room temperature polar and longitudinal MOKE measurements of an ultrathin Cu(111)/Co(111)/Cu(111) wedge grown on a sapphire substrate by molecular beam epitaxy (MBE). We obtain the surface and volume contributions to the magnetic anisotropy

by two different analyses and discuss the validity of each method. Finally these results are discussed in terms of previously published values for the Co(111)/Cu(111) system.

2. Experimental details

A Cu/Co/Cu wedge was grown in a VG80M MBE facility with a base pressure of 10^{-11} mbar on a polished single crystal sapphire substrate oriented normal to the $[11\bar{2}0]$ direction. To achieve a high degree of uniformity across the sample the substrate was rotated at 1 Hz throughout the growth process except during deposition of the wedged Co layer. A 30 Å Nb (110) buffer layer was first deposited on the substrate of dimensions 35 mm long × 10 mm wide followed by a 30 Å Cu (111) buffer layer. A nominally 0–35 Å wedged layer of Co was then deposited across the length of the substrate using a stepper motor driven shutter [14]. A further 30 Å Cu layer was grown to produce a symmetric Cu(111)/Co(111)/Cu(111) structure and finally a protective 15 Å layer of Au was deposited to prevent damage and oxidation of the sample. This produced a wedged sample of total nominal thickness of 70–110 Å with a nominal Co thickness gradient of 1.1 Å mm^{-1}.

Since the anisotropy of thin Co layers is known to vary substantially with thickness it was necessary to check the nominal growth thicknesses by an independent *ex situ* method. A wedged sample does not lend itself easily to low angle x-ray measurements as the beam footprint is around 1 cm at these angles so the range of thicknesses being sampled would be too large for the measurement to be meaningful. After the MOKE measurements were completed on the wedge, three 2 mm pieces were cut from the wedge at different nominal thicknesses for the x-ray measurements. The size of the piece chosen was a trade-off between sufficient x-ray count rate and nominal thickness range across the sample. For a 2 mm piece, the thickness would be expected to vary by 2 Å across the piece. However since x-rays provide averaged information the averaged thickness of Co was assumed to correspond to the centre of the sample.

Low angle x-ray measurements were performed using a Siemens two-circle diffractometer using Cu Kα radiation. The total thicknesses were calculated from the position of the Kiessig fringes. The thickness gradient was found to be constant at 1.24 Å mm^{-1} which agrees well with the nominal value of 1.1 Å and is well within the bounds of experimental errors in both the nominal growth rate and Kiessig fringe measurements.

Room temperature polar and longitudinal MOKE measurements were made as a function of Co thickness by measuring hysteresis loops at various positions along the wedge. The laser spot was focused to a 100 μm diameter spot size using lenses. The magnetic anisotropy constants, K_v and K_s, were then determined from these measurements by two different methods: the 'area method' and by simulation of the perpendicular magnetization. The 'area method' consists of calculating the area between the parallel (longitudinal MOKE) and perpendicular (polar MOKE) curves which gives a measure of the magnetic anisotropy energy, K_{eff}. Further details are given in section 3.2. The values of K_v and K_s were then obtained by plotting a graph of $K_{eff}t_{Co}$ against t_{Co} as explained earlier in this paper. The method of simulation is described fully in section 3.3.

3. Results

3.1. MOKE measurements

Longitudinal and polar MOKE measurements sampled at various positions along the wedge are shown in figures 1 and 2, respectively. Inspection of these graphs indicates that for Co

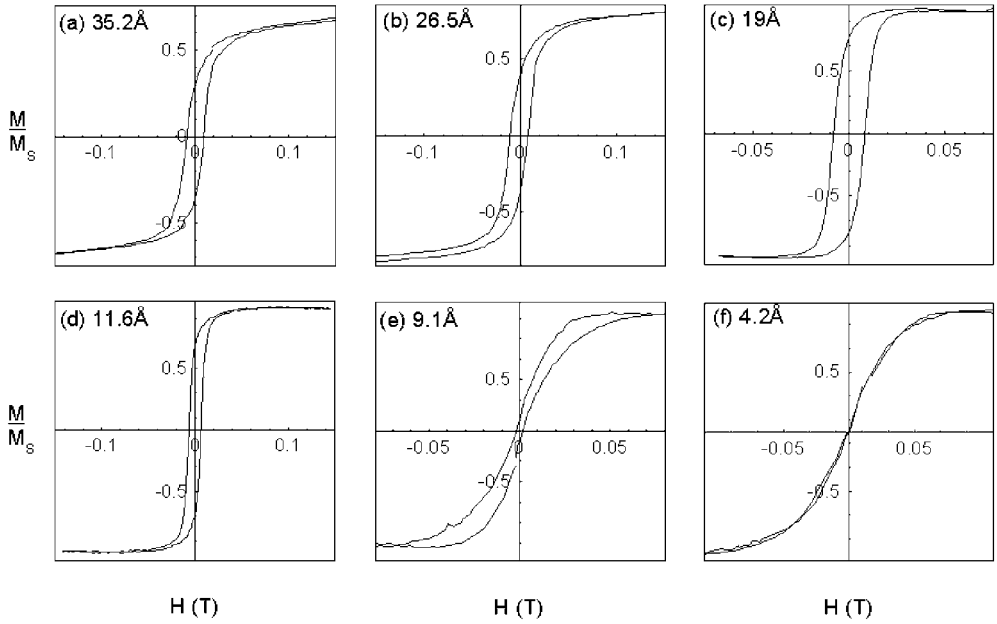

Figure 1. Longitudinal MOKE curves at different positions along the wedge corresponding to: (a) 35.2 Å; (b) 26.5 Å; (c) 19 Å; (d) 11.6 Å; (e) 9.1 Å and (f) 4.2 Å of Co.

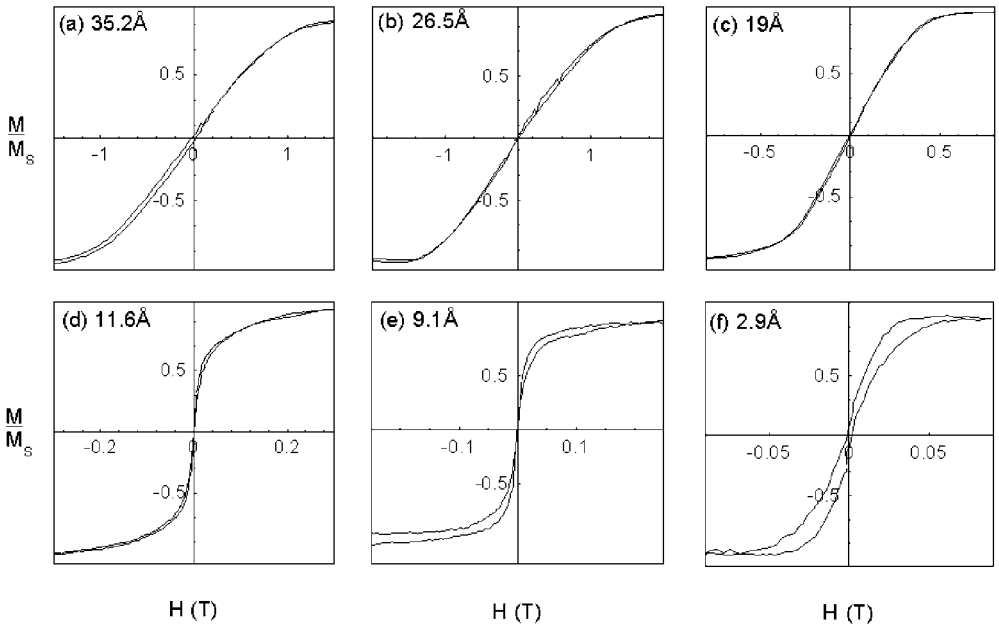

Figure 2. Polar MOKE curves at different positions along the wedge corresponding to: (a) 35.2 Å; (b) 26.5 Å; (c) 19 Å; (d) 11.6 Å; (e) 9.1 Å and (f) 2.9 Å of Co.

thicknesses of 11.6 Å and above the magnetization lies in the plane of the sample. The longitudinal and polar MOKE curves for the 9.1 Å Co thickness (figures 1(e) and 2(e)) suggest that the magnetization is beginning to lie out of the plane of the sample, although

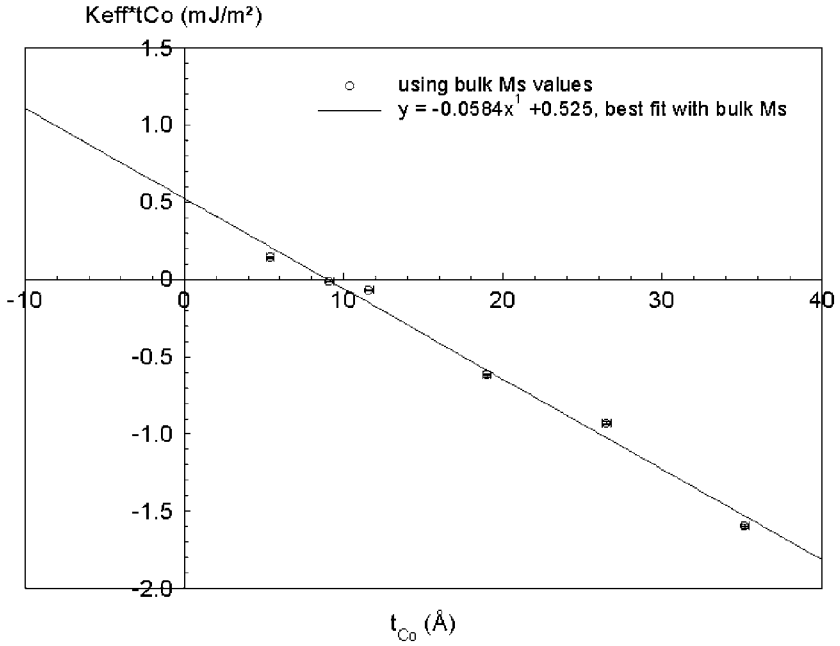

Figure 3. Plot of $K_{eff}t_{Co}$ versus t_{Co} for the ultrathin Co wedge. The equation for the straight-line fit to the data is shown in the legend.

significant hysteresis is still present in the longitudinal measurement. In contrast no hysteresis is discernible in figure 1(f) at the 4.2 Å Co position suggesting that the sample is perpendicularly magnetized. Further evidence is provided by the hysteresis observed in the polar MOKE curve at the 2.9 Å Co position (figure 2(f)).

3.2. Anisotropy constants obtained using the 'area method'

In order to extract a measure of the surface and bulk anisotropy constants, K_s and K_v, the polar and longitudinal MOKE curves for each Co thickness were plotted together and the area between the curves was computed to determine the magnetic anisotropy energy, K_{eff}. In samples showing hysteresis the two halves of the loop were averaged to remove the hysteresis as suggested by de Jonge *et al* [1]. In all cases the field available was sufficient to saturate each sample so no extrapolation was necessary.

Initially the saturation magnetization, M_s, was assumed to have the bulk value for Co of 1.422 MJ T^{-1} as is customary when using the area method to analyse MOKE measurements since MOKE cannot provide a direct measure of the value for the magnetization [1]. The validity of assuming bulk values for M_s is discussed later. Figure 3 shows the expected linear dependence as predicted by equation (1). A straight line least squares fit (full line) to the data gives values of $K_v = -0.60 \pm 0.03$ MJ m^{-3} and $K_s = +0.26 \pm 0.04$ mJ m^{-2}. Since K_v is negative it implies that in-plane magnetization is favoured in these samples which is also borne out by inspection of the magnetization loops already examined (refer to figures 1 and 2), except at very low Co thicknesses. The point of interception of the line with the x-axis gives the critical thickness of Co at which perpendicular magnetization is favoured. From figure 3 this is seen to be 9 Å and this is in agreement with visual inspection of the polar and longitudinal MOKE loops in figures 1 and 2.

The main contribution to K_v is the shape anisotropy, or demagnetizing term, which is $\frac{1}{2}\mu_0 M_s^2 = -1.27$ MJ m^{-3} for Co [1]. Subtraction of this term gives a value of $+0.67 \pm 0.03$ MJ m^{-3} for K_v. Magnetocrystalline (MC) and magnetoelastic (ME) anisotropy may also be present although the ME contribution is expected to be very small in Co(111) due to the small lattice mismatch between Co and Cu [8]. The MC anisotropy is expected to be negligible for fcc Co and +0.53 MJ m^{-3} for hcp Co [1] although Lee *et al* [15] found that the MC anisotropy is not negligible for fcc Co.

It is therefore difficult to explain the fitted value of K_v which lies outside the range of both the expected hcp and fcc values for the MC anisotropy. It is usual to account for values of K_v outside these ranges by invoking a ME explanation [1] but even if we assumed the Co to be hcp in orientation, the ME anisotropy would still be higher than expected for the case of Co(111) on Cu(111) [11]. This discrepancy could not be resolved using a thickness dependent M_s as suggested by Givord *et al* [17]. When the values for K_{eff} were recalculated using $M_s(t)$ values the fitted values for K_v and K_s reduced only slightly and, considering the experimental error, we find that it predicts the same values as for the bulk M_s calculation.

Although no high-angle x-rays were performed on this particular sample all other similar Co/Cu structures grown in the same MBE chamber have been found to be of the fcc orientation up to thicknesses of around 30 Å, beyond which a small hcp peak is observed in the x-ray data. In an NMR study, Thomson *et al* [16] also found that similar Co(111) layers sandwiched by Cu, grown by MBE, retained an fcc structure up to thicknesses of 60 Å, beyond which a hcp structure was favoured. We therefore expect all Co thicknesses studied here to be fcc in structure except the 35.2 Å sample. However, as pointed out by Farle *et al* [11], in the case of the low thicknesses of Co where determination of phase by x-rays is impractical, it is difficult to be sure of the exact phase present in the films since the fcc(111) and hcp(0001) structures differ only by the layer sequence (ABCA versus ABA). In their *in situ* study of a single layer of Co(111) grown on a Cu(111) single crystal, they observed evidence of changing phases from trigonal fcc or hcp stacking to a relaxed fcc structure due to a marked change in the slope of the K_{eff} against $1/t$ graph. However this behaviour has only been reported for uncapped Co/Cu(111) films and since this behaviour has not been reproduced in either this study or previous work on Cu capped films, it seems likely that the capped films studied here do not show this marked structural transition.

There is a further possibility that pinholes would play some role in the anisotropy in these ultrathin films since it is well known that it is difficult to obtain good layer-by-layer growth of MBE-grown Co/Cu(111) since it is susceptible to fcc/hcp stacking faults [18]. This can cause the formation of a twinned fcc structure in the subsequent Cu capping layer which and allows pinholes to develop between the twins [19]. Although we have no measurements of the number and type of pinholes in these samples, we would expect this to change as the Co thickness increased. Since no marked change is seen in the slope of figure 3, i.e. no thickness dependence of K_v or K_s is seen, it seems unlikely that pinholes play a dominant role in determining the anisotropy of these films although further work would be needed to investigate this in detail.

3.3. Simulations of the magnetization

The free energy E, of a thin film can be written as

$$E = (-K_i + \tfrac{1}{2}\mu_0 M_s^2)\cos^2\theta - \mu_0 M_s H \cos(\theta - \phi) \tag{2}$$

where K_i contains all the first-order anisotropy energy contributions excluding shape anisotropy, which is given by the $\frac{1}{2}\mu_0 M_s^2$ term for a saturated film. The interaction between the applied field, H, and the resulting magnetization is described by the last term where θ

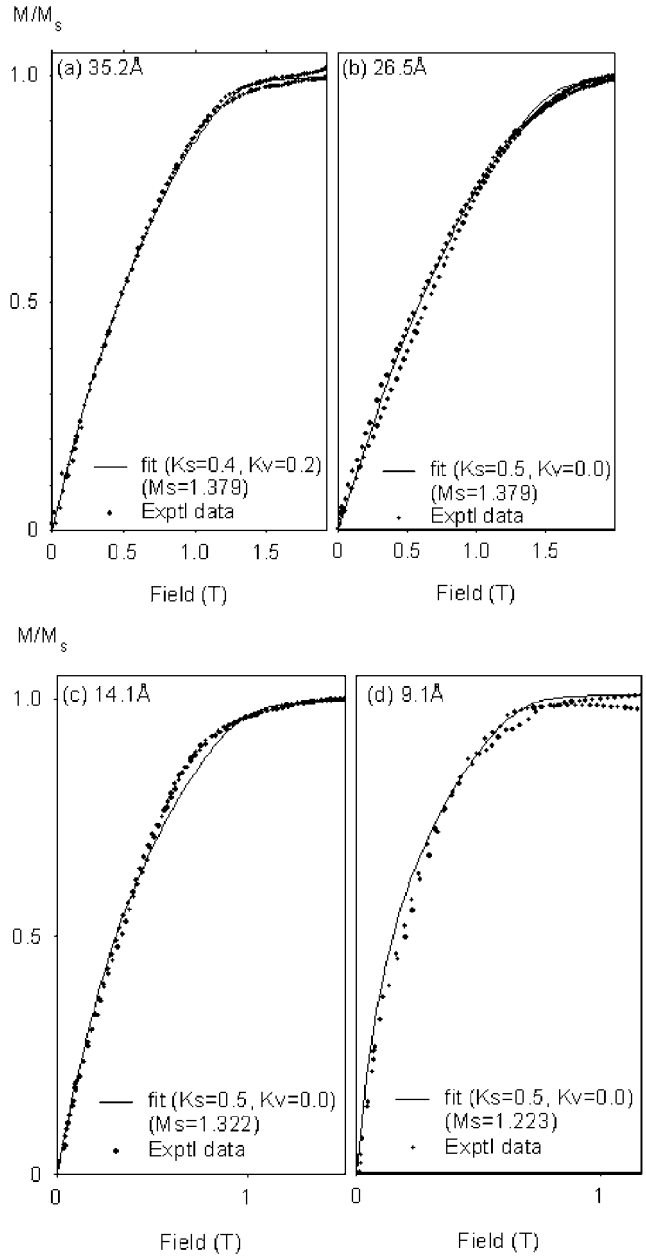

Figure 4. Best-fit simulations of the wedge at: (a) 35.2 Å; (b) 26.5 Å; (c) 14.1 Å and (d) 9.1 Å of Co. The fits are shown as solid lines along with the experimental data points. The reduced M_s values and the values of K_v and K_s used in both sets of simulations are shown in the legend text.

and ϕ are the angles between the film normal and, respectively, the magnetization and field. If this equation is minimized for the energy as a function of applied field we obtain the field dependence of the equilibrium angle $\theta_{eq}(H)$ and the field component of the magnetization $M = M_s \cos(\theta_{eq} - \phi)$ and thus magnetization curves can be obtained. By adjusting the

parameters in the calculated perpendicular, magnetization curves can then be compared to normalized experimental polar magnetization curves. Values for the anisotropy constants can thus be obtained.

A second-order MC volume contribution was also included in the simulation but it was found to have a very small effect. The simulations in figure 4 are for one field sweep from positive to negative saturation. Preliminary simulations showed that an offset angle of 2° was required to obtain curves with the similar rounding of the approach to saturation found in all the experimental polar MOKE curves. This suggests that either there had been a slight misalignment of the sample with respect to the field during the measurements or that the Co layers lay at a slight angle with respect to the top surface of the sample which is possible in wedged layers. Ives *et al* [10] reported that a similar value of offset angle was necessary to obtain a close fit to polar MOKE data of Cu/Co/Cu wedged trilayers.

In addition, close inspection of the polar MOKE data in figure 2 reveals that the curves are not symmetric with respect to the positive and negative field. This may be due to the fact that the relationship between the magnetization and the intensity recorded by the photodiode is not perfectly linear resulting in a small second-order term being present [10]. Hence both the positive and negative field regions of the experimental data were fitted and where the best-fit values differ, a range of values are quoted. It should also be noted that introduction of an offset angle produces a slight asymmetry in the simulated curves which was seen to follow the behaviour of the experimental data reasonably well. Simulations are not reported for the 5.4 Å and 2.9 Å samples since their polar MOKE curves displayed marked hysteresis indicating that K_{eff} is positive, i.e. perpendicular magnetization is favoured. In this case the films tend to consist of up and down domains and the magnetostatic contribution of $\frac{1}{2}\mu_0 M_s^2$ given in equation (2) is not valid [1].

Initially simulated perpendicular magnetization, curves were calculated using the values of $K_v = 0.7$ MJ m^{-3}, $K_s = 0.26$ mJ m^{-2} obtained from the area method assuming both a bulk value of M_s and also the appropriate reduced $M_s(t)$ value. These simulations did not fit the experimental polar MOKE data; for all Co thicknesses the simulated curves saturated in fields well below the corresponding experimental data. Since the value for the MC anisotropy is expected to be negligible in fcc Co, a further set of simulations were produced by setting $K_v = 0.0$ and allowing K_s to vary until a satisfactory fit was obtained. When a bulk M_s value was assumed, best-fit values for K_s ranged from 1.4 mJ m^{-2} for the 35.2 Å sample to 0.5 mJ m^{-2} for the 9.1 Å sample although no systematic variation in K_s was observed as a function of Co thickness.

A more systematic variation in K_s was found when the fits were re-computed with varying values of M_s, taken from the values quoted by Givord *et al* [17]. Best-fit simulations (see figure 4) gave K_s to be 1.2 for the 35.2 Å sample and constant at 0.4–0.5 for all other thicknesses simulated. It was mentioned previously that hcp regions have been found to appear in x-ray measurements of Co samples above 30 Å in thickness and so a MC contribution from these regions might be expected. Further simulations were performed for the 35.2 Å sample by setting K_s to 0.45 and allowing K_v to vary. Inspection of figure 4(a) shows that a value of K_v of 0.2 fits the data well implying that some hcp regions are present since K_v lies between the expected values for fcc (0.0) and hcp (0.53).

4. Discussion

We have obtained the anisotropy constants for the ultrathin Co wedge by two different methods of analysing the magnetization. The area method, by far the most commonly used in the published literature, produced values of K_v of +0.67 MJ m^{-3} (or −0.6 if the shape anisotropy

is included) and K_s of 0.3 mJ m^{-2}. Previous work on sputtered [5] and MBE-grown [7, 8] Co/Cu(111) multilayers and MBE-grown trilayers [10] quoted as having a fcc structure report values of K_v which lie close to the bulk value of -1.27 MJ m^{-3} for the shape anisotropy suggesting that MC and ME contributions to the anisotropy are small as expected. Den Broeder *et al* [4] reported a value close to that expected for bulk hcp Co for evaporated films although no evidence was provided on the structure of the samples. Overall the values for K_s quoted in the literature are wide ranging in value although they are generally positive ranging from +0.53 to -0.02, indicating that a perpendicular magnetic interface anisotropy is present in Co/Cu. The value of K_s is reasonable when compared to the values previously found [4–8, 10] whereas K_v is not. All previously quoted values have been shown to lie within the hcp and fcc range expected for bulk Co.

It should also be noted that the values of K_{eff} are not necessarily expected to be the same for Cu-covered Co/Cu(111) films compared to uncovered Co/Cu(111) layers studied in a vacuum. Recently Farle *et al* [11] reported a factor of two to four reduction in K_{eff} in Co(111) grown on single crystal Cu(111) when measured in UHV conditions, which may be due to the difference in the interface anisotropy terms since Cu capped samples contain both Co/Cu and Cu/Co interfaces compared to the vacuum/Co and Co/Cu interfaces for uncapped *in situ* studies. Farle *et al* [11] also found evidence that capping the Co(111) layer with Cu influenced the arrangement of the Co planes below. Since it is therefore difficult to make a careful comparison between films which are capped and uncapped, we have limited our discussion above to reports on capped films, which include those on trilayers and multilayers.

It is possible that a systematic error was present during the calculation of the area between parallel and perpendicular MOKE curves which could not be included in the error analysis. It was mentioned previously that the MOKE data on these films are often asymmetric and so it is possible that the area calculated is erroneous. Another source of error became evident from the simulations which required a 2° offset angle in order to fit the polar MOKE curves. This was explained to have the effect of rounding the approach to saturation and would thereby change the area calculated considerably. Since the in-plane magnetization curves were not simulated we are therefore unsure whether the rounding effect was occurring here and, if it was, whether it would increase the error further or cancel it out. The use of the area method in this case is therefore considered to be unreliable. Recently the values reported by Hillebrand *et al* [9] for a sputtered layer of Co(111) on Cu(111) using MOKE and the area method were almost double those obtained when they analysed Brillouin light scattering data from the same samples.

In addition, simulations of our data using the values obtained by the area method did not fit the data at any of the Co thicknesses. Good fits to the data were produced by setting $K_v = 0.0$ and varying K_s. When a bulk M_s was assumed, K_s was found to vary in a non-systematic manner. However by using appropriate reduced values for M_s it was concluded that K_s lay in the range 0.4–0.5 mJ m^{-2} for all thicknesses above 5.4 Å. The 35.2 Å sample could be fitted successfully if a K_v of 0.2 MJ m^{-3} was introduced to account for the appearance of hcp regions in the sample. K_s is consistent with the value reported by Ives *et al* [10] of 0.51 mJ m^{-2} for a single Co layer grown by MBE on GaAs. It is also close to the 0.53 value quoted by Lamelas *et al* [6] for fcc multilayers grown by MBE. In fact Ives *et al* [10] also performed measurements on a Co/Cu trilayer grown by our MBE system, reporting a value of 0.39 which is again consistent with our results. Moreover, they assumed a negligible volume contribution of -0.02, close to the zero value we used. It should be noted, however, that they assumed a bulk value of M_s but this is consistent with our findings since the thickness of Co used was 15 Å which would correspond to a 95% bulk M_s value in our study. Overall the most reliable values of K_s and K_v found from our simulations lie well within the range previously quoted in the literature.

The thickness, $t_\perp$, at which the onset of perpendicular magnetization is observed is expected to occur at a higher thickness in hcp compared to fcc oriented films due to the increase in MC anisotropy for hcp systems. Den Broeder *et al* [4] and Kohlepp *et al* [8] found that perpendicular magnetic anisotropy was limited to below 3 Å of Co although Lee *et al* [5] observed a critical thickness of 10 Å in Co/Cu multilayers grown by MBE. We observed the transition at 5.4 Å of Co in this wedged Cu/Co/Cu trilayer sample, by inspection of the MOKE curves (figures 1 and 2). This is in agreement with the value of $t_\perp$ of 6 ± 1 Å calculated from the fitted values.

5. Conclusions

We have described a detailed analysis of the anisotropy constants calculated from MOKE measurements on an ultrathin Co wedged layer sandwiched by Cu(111). We have shown how the widely used 'area method' to calculate the bulk and surface contributions to the magnetic anisotropy does not agree with the values obtained by simulation of the polar MOKE data. Our simulations have shown that the most realistic values of K_s (0.4–0.5 mJ m^{-2}) and K_v (0.0 MJm^{-3}) values were obtained by using a value for the saturation magnetization that varied with Co thickness. K_v was seen to increase to 0.2 MJ m^{-2} for the 35 Å sample suggesting that some hcp regions were present. In summary, the ultrathin Co wedge was found to have a negative total anisotropy indicating that in-plane magnetization is favoured for most thicknesses of Co. A transition from in-plane to perpendicular magnetization was observed in the MOKE data for Co thicknesses of 6 ± 1 Å and below.

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