Magnetisation reorientation in ultra-thin Fe films on Cu(100) upon deposition of Co

Jonathan Hunter Dunn,[a]* Dimitri Arvanitis,[b] Roger Carr[a] and Nils Mårtensson[b]

[a]Stanford Synchrotron Radiation Laboratory, Stanford Linear Accelerator Center, Stanford University, Stanford, CA 94309, USA, and [b]Department of Physics, University of Uppsala, Box 530, S-751 21 Uppsala, Sweden. E-mail: hdunn@slac.stanford.edu

Ultra thin films of Fe deposited on the (100) surface of a Cu single crystal exhibit a net perpendicular magnetic anisotropy. The addition of very low coverages of Co results in the easy magnetic direction reorienting into the film plane. This behavior is in contrast to the addition of similar amounts of Fe, whereupon the ferromagnetic response vanishes. This result is discussed in terms of the anisotropy energies derived from the spectroscopic data.

Keywords: Ultrathin films; **Magnetic anisotropy energy**; **XMCD.**

1. Introduction

The continued interest in the magnetic properties of ultra thin films is due to the novel structural and magnetic behavior they exhibit. The demands of the magnetic data storage industry provide substantial motivation for the investigation of such systems with a view to exploiting the magnetic phenomena they manifest in high capacity, compact storage devices. In particular, ultra thin transition metal films combine macroscopic remnant magnetism with the possibility of stabilising alternative crystallographic structures when prepared on single crystal substrates.

Fe grown on the (100) surface of Cu is a prime example of such a system. In the ultra thin limit, below 4 atomic layers, the structure is face centered tetragonal (fct) (Müller *et al.* 1995) due to the crystallographic mismatch between the two elements. This is accompanied with a net perpendicular magnetic anisotropy and an increased magnetic moment (Hunter Dunn *et al.*, 1996): the so called 'high spin' state, as predicted by theory (Söderlind *et al.*, 1992). With increasing thickness, there follows a structural transformation in which the strain is relaxed. The system then adopts the bcc crystallographic structure and the easy magnetic direction is oriented in the film plane. A transition thickness just in excess of 4 atomic layers (AL), for films grown below 150 K, (Müller *et al.*, 1995) and 10 AL, for films grown at room temperature, is reported (Schmailzl *et al.*, 1994). For well ordered growth a substrate temperature below 150 K during evaporation minimises intermixing at the interface. Shortly annealing to 300 K orders the layers (Müller *et al.*, 1995 and Giergiel *et al.*, 1995). Repeated short annealing cycles were found to improve LEED patterns but tended to degrade the magnetic properties associated with the high spin phase (Platow *et al.*, 1998). Similarly, annealing to temperatures above 350 K had an irreversible impact on the magnetism (Shen *et al.*, 1995). These facts indicate the metastable nature of the system.

2. Experiment

For this work we used X-ray Magnetic Circular Dichroism (XMCD) over the Fe $L_{3,2}$ edges to probe the magnetic response from three Fe/Cu(100) preparations before, and after, deposition of half an atomic layer of Co. In each case an ultra thin film of Fe was prepared and characterised for its magnetic properties before adding the sub monolayer coverages of Co. The thickness of the films was accurately calibrated using their 'edge jump' ratio (Hunter Dunn *et al.*, 1996). The present set of samples were 2.7, 3.0 and 3.9 AL. All these samples yielded similar results. Film surface order was monitored by means of LEED which showed patterns that compared favorably with those in the literature for this thickness range (Müller *et al.*, 1995). In our previous work (Hunter Dunn *et al.* 1996), using $L_{3,2}$ edge absorption spectra, sum rules (Carra *et al.*, 1993) were applied to determine magnetic moments for both the ultra thin fct, and thicker bcc films. For the bcc films we found a spin moment of 2.26 μ_B, in excellent agreement with theory. The high spin phase of Fe showed a significant increase in the magnetic response, giving moments in excess of 3.00 μ_B per atom.

The present set of experiments were performed at beamline 5.2 of the Stanford Synchrotron Radiation Laboratory, California. The energy resolution of the photon beam was better than 1.5 eV at the Fe $L_{3,2}$ edges. To ensure the highest possible quality all samples were prepared and measured *in situ*, in a ultra high vacuum system custom built for magnetic measurements. This includes a pair of coils for magnetising the samples and a circular channelplate detector on the axis of the chamber and sample rotation. The coils and channelplate assembly may be rotated through 360° facilitating any combination of applied field and sample relative to the incident x-ray light without altering the detection geometry. The samples were measured in remamnence. In addition to the channel plate, the sample drain current was measured simultaneously. Both detection channels agreed extremely well when appropriately normalised. The films were prepared by evaporation, using electron bombardment heating of purified bulk Fe and Co. Apart from the brief annealing after deposition, the Cu(100) substrate was maintained below 150 K throughout the experiments. The chamber pressure during evaporation was better than 1×10^{-9} mbar. During the measurements contamination from adsorbed impurities remained less than 0.2 % of a monolayer.

3. Results

The result of the XMCD measurements are shown in Figures 1 and 2. Before the addition of the Co, the ultra thin Fe film exhibits a perpendicular magnetisation direction relative to the film plane. (Figure 1.) The magnitude of the magnetic contrast, evident in the L edge resonance features, is substantial, reflecting the high spin phase of the sample. The addition of 0.5 AL of Co causes the easy magnetisation direction to rotate into the film plane. This behavior is in contrast to the addition of more Fe, whereupon the ferromagnetic response vanishes completely before re-emerging in-plane for films in excess of 4 AL. The size of the magnetic contrast is also reduced. This is primarily due to a reduction in the magnetic moment per atom. See Table 1 below. The addition of more Co, to a total of 2.5 AL Co, rendered the Fe non-ferromagnetic while the Co exhibited a small in-plane ferromagnetic response. While it is not known why ultra-thin Fe films exhibit no ferromagnetic response in 4 AL thickness range, theoretical work (M. Uhl *et al.*, 1994) suggests a spin spiral configuratioon in which the magnetic moment in each

succesive layer is rotated by 90° relative to the preceeding layer resulting in a average moment of zero. Spectra were taken over both the Fe and Co edges from which it is clear that the magnetisation of two elements couples ferromagnetically. (Figure 2.) No sizable change in the average height of the absorption peaks was found, suggesting that there is minimal charge transfer between the Fe and Co *d* states.

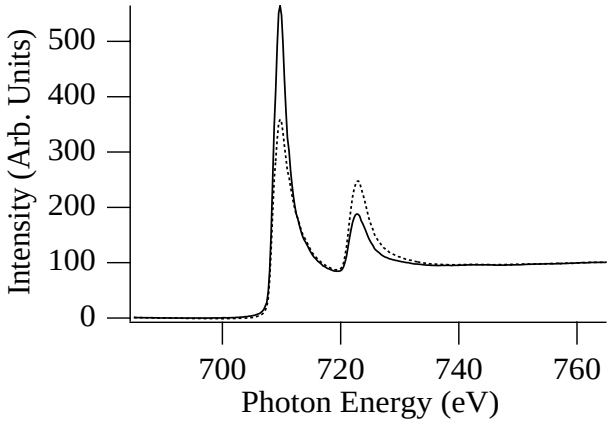

Figure 1
Fe $L_{3,2}$ edge XMCD spectra from a 3.0 AL ultra thin Fe film stabilised on the (100) surface of Cu. The difference in the *L* edge resonance intensity is a maximum when the x-rays are at normal incidence indicating that the easy magnetisation direction is perpendicular to the plane of the film.

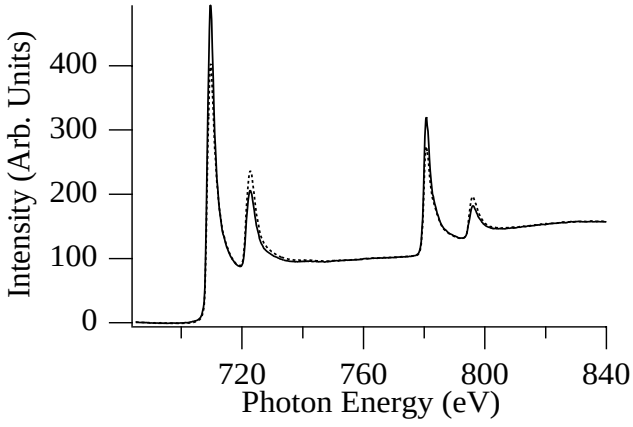

Figure 2
Grazing incidence XMCD spectra from the sample portrayed in Figure 1 after the addition of 0.5 AL of Co. The magnetic contrast in the $L_{3,2}$ edge resonance features is now a maximum when measuring at grazing x-ray incidence indicating that the easy magnetisation direction rotated into the film plane. In addition, the size of the XMCD response is reduced.

4. Discussion

Table 1 below summarises the results of applying sum rules (Carra *et al.*, 1993) to the x-ray absorption spectra shown in Figures 1 and 2. From these values the Magneto Crystalline Anisotropy (MCA) energy, E_{MCA}, is derived by applying,

$$E_{\mathrm{MCA}} \approx \frac{\xi}{20}(\Delta M_L), \tag{1}$$

where ξ is the spin orbit coupling constant, $\approx 68\mu$eV, (Bruno *et al.*, 1989) and $\Delta M_L = (M_L^{\perp} - M_L^{\parallel})$. $M_L^{\perp}$ and $M_L^{\parallel}$ are the orbital moments in μ_B perpendicular to and in the plane of the film respectively. Due to the fact that this approach, based on perturbation theory, is known to fail in the case of Fe where the exchange splitting is similar in magnitude to the 3*d* band width, a corrective factor of 4 was suggested (Bruno *et al.*, 1989). Despite this, it was found to overestimate the MCA energy in the case of ultra-thin Fe films by a further factor of 5 (Hunter Dunn *et al.* 1996). The denominator in Equation 1 contains these factors previously necessary for quantitative agreement between application of Equations 1 and 2. As discussed below, for the present case, the error, and thus the corresponding corrective factor, is smaller than those found before (Hunter Dunn *et al.*, 1996). For the dipolar contribution to the total anisotropy energy per atom, $E_{Dipolar}$, we take,

$$E_{\mathrm{Dipolar}} = \frac{M_S^2}{2}. \tag{2}$$

While valid for bulk analysis, it is known that the continuous approximation (Equation 2) breaks down along the surface normal for the ultra thin films, underestimating this contribution (Farle *et al.*, 1988). However, it is still a reasonable approximation and holds for a qualitative discussion of the results presented here.

It is clear from Table 1 that certain qualitative information is readily available. We will follow the convention that negative energies favor an in-plane magnetisation. The first row of the table contains previous experimental results (Hunter Dunn *et al.*, 1996) where application of the sum rules to data from a bcc Fe film gave results in excellent agreement with theory. We have taken this sample as a standard from which the in-plane orbital moment is used in the application of Equation 1. In this case the MCA energy that would give a perpendicular component is virtually zero. (The fact that it is non-zero arises from the distortion of the bcc film on the Cu surface.) This result is expected since the magnetisation direction in such samples *is* in the film plane.

With the orbital moment of the 3 AL Fe film we can now apply Equation 1 (including the corrective factor) which yields a MCA energy of 272 μeV/atom. This is just larger than the dipolar contribution of -242 μeV/atom correctly predicting a perpendicular magnetisation direction. It is quite possible that the distorted crystallographic structure and less than perfect surface of the experimental sample introduce lateral finite size effects that would similarly influence the moments and anisotropy terms, stabilising the perpendicular magnetisation further. Following our sign convention, the addition of the Co changes the sign of the experimental value of the MCA energy. Again, using the value of the orbital moment derived previously for a bcc Fe film, (Hunter Dunn *et al.*, 1996) we again apply Equation 1 but this time use the difference of the two in-plane orbital moments and find an MCA energy of -136 μeV/atom. Together with the negative dipolar contribution this is sufficient to rotate the easy magnetisation direction into the film plane as is observed experimentally.

Table 1
The experimental moments, obtained from the spectra shown in Figures 1 and 2 using sum rules, and the derived MCA energy and dipolar contribution to the total anisotropy energy. [a] From ref Hunter Dunn *et al.*, 1996. [b] From ref Hunter Dunn *et al.*, 1997.

Fe/Cu(100)	E_{MCA} (μeV/atom)	ΔM_L (μ_B)	M_L (μ_B)	M_S (μ_B)	$E_{Dipolar}$ (μeV/atom)
20 AL (bcc)[a]	< 10[b]		0.16(2)	2.26(2)	-137
3 AL Fe	272	0.08	0.24(2)	3.0(3)	-242
3 AL Fe + 0.5 AL Co	-136	0.04	0.20(2)	2.08(2)	-116

5. Conclusion

In conclusion, we have presented experimental results for the Fe/Cu(100) system before and after the addition of low coverages of Co. Commensurate with previous work, we find a perpendicular magnetisation direction for the high spin Fe/Cu(100) system. Post Co deposition we observe that the remnant magnetisation direction reorients into the film plane and is accompanied with a reduction in magnetic moment. Anisotropy energies derived from magnetic moments extracted from the spectroscopic data using sum rules provide a qualitative picture of the observed reorientation.

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