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1989 J. Phys.: Condens. Matter 1 4407

(http://iopscience.iop.org/0953-8984/1/27/013)

J. Phys.: Condens. Matter 1 (1989) 4407–4413. Printed in the UK

Investigation of the magnetic properties of sandwiched epitaxial Fe and Co films using the magneto-optic Kerr effect

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Received 20 January 1989

Abstract. Using the surface magneto-optical Kerr effect, we have performed magnetic hysteresis measurements at 300 K on Ag–8-monolayer BCC Fe–Ag(001) and Cu–7-monolayer FCC Co–Cu(001) sandwich structures. Both films magnetically saturate for fields in the region of 100–200 Oe applied in the plane of the film. These findings confirm the validity of previously obtained polarised neutron reflection data, from which the absolute value of the magnetic moment per atom had been estimated. In the case of the Fe structure we find evidence of planar shape magnetic anisotropy, whereas for the Co structure we observe an in-plane magneto-crystalline anisotropy and very strong planar shape anisotropy. For each sample, our results are consistent with the existence of both long-range ferromagnetic order and planar interfaces.

1. Introduction

The magnetic properties of ultrathin epitaxial films are a subject of strong and growing scientific (Fu *et al* 1985) and technical (Connell 1986) interest. Unusual magnetic properties arise not only because of the two-dimensional nature of the film, but also as a result of the modified atomic coordination and electronic structure at the interface. In this paper we describe a study of Fe and Co films incorporated into sandwich structures. By using the same non-magnetic metal for both substrate and overlayer, it is possible to create equivalent magnetic–normal-metal interfaces. This geometry is of interest since extensive spin-polarised band structure calculations have been made for such structures, which investigate the role of the interface in determining the magnetic properties of the film (Fu and Freeman 1987). In experimental investigations of such systems several considerations arise. Firstly, the non-magnetic overlayer chemically seals the film, permitting studies outside the preparation chamber. Secondly, enhanced sensitivity is achieved with polarised neutron reflection (PNR) measurements, from which the absolute value of the magnetic moment per atom can be obtained (Bland *et al* 1987b). Thirdly, the optimum overlayer thickness for PNR measurements is 10–20 nm, which corresponds to the value of the optical skin depth in most metals. Consequently, the sensitivity of the Surface Magneto-Optical Kerr Effect (SMOKE) measurements is reduced by approxi-

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0953-8984/89/274407 + 07 $02.50

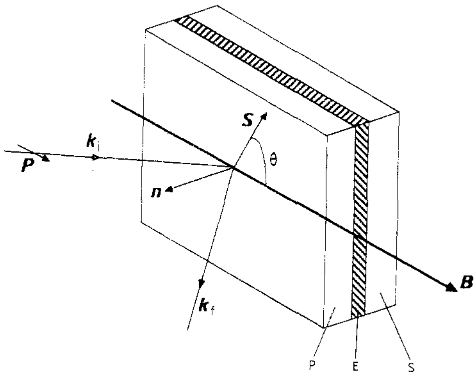

Figure 1. Geometry for observing the magneto-optic Kerr effect, described in § 2. Key: P, protective overlayer; E, epitaxial film; S, substrate; ***P***, polarisation; ***S***, symmetry axis; $\boldsymbol{k}_i$, incident wavevector; $\boldsymbol{k}_f$, reflected wavevector.

mately one order of magnitude. However, our recently developed SMOKE apparatus is sufficiently sensitive that hysteresis loops can still be observed from these buried films.

We chose to study epitaxial Ag–BCC Fe–Ag(001) and Cu–FCC Co–Cu(001) sandwich structures. The Fe–Ag(001) system is ideally suited for epitaxial growth as the lattice mismatch between the primitive surface cells of BCC Fe and Ag(001) is only 0.8%, with the Fe laterally expanded (Koon *et al* 1987). The Co–Cu(001) system exhibits a higher lattice mismatch of 2.9%, but good epitaxial growth can still be obtained for ultrathin films (Clarke *et al* 1987). The motivation for the hysteresis measurements reported here was to demonstrate the sensitivity of our SMOKE apparatus and to complement the neutron reflection measurements already carried out on these samples (Bland *et al* 1987a, 1989b).

2. The surface magneto-optical Kerr effect

The magneto-optic Kerr effect relates the change in the polarisation state of incident light reflected from a ferromagnetic surface to the magnetisation of the material.

When the incident wave vector $\boldsymbol{k}$ has a resolved component along the direction of the magnetisation $\boldsymbol{M}$ within the sample, inequivalent interactions occur for right and left circularly polarised light. The origin of the effect is due to the spin-orbit interaction within the medium and this gives rise to off-diagonal terms in the dielectric tensor which governs the reflectivity (Argyres 1955). Thus, in the general case, when a plane polarised beam is reflected or transmitted by a magnetic medium, it become elliptically polarised with the principal axis of polarisation rotated away from the plane of the incident ploarisation. In ferromagnetic materials the effect is particularly large due to the high magnetic susceptibility. In the case of transmission, this is referred to as the Faraday effect and a field-dependent rotation occurs. In reflection geometry the phenomenon is known as the magneto-optical Kerr effect, with both the refractive index and the reflectivity being dependent on the handedness of the polarisation, and therefore the light is elliptically polarised in addition to being rotated.

SMOKE measurements are usually carried out in one of the three geometries (see figure 1), namely; longitudinal (***B*** parallel to the plane of incidence and perpendicular to ***n***), transverse (***B*** perpendicular to both the plane of incidence and ***n***) or polar (***B*** parallel to both the plane of incidence and ***n***).

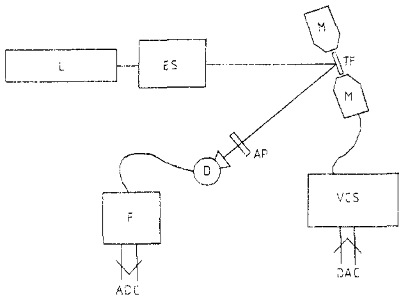

Figure 2. The complete layout of the SMOKE apparatus. Key: L, 20 mW He–Ne laser; ES, external intensity stabiliser; M, magnet (±270 Oe); TF, thin film sample; D, detector; VCS, voltage controlled current supply; AP, analysing polariser; F, 50 Hz notch, 100 Hz notch and low-pass filters; DAC, digital-to-analogue converter (8-bit); ADC, analogue-to-digital converter (8-bit, 10 μs conversion time).

For all of these geometries, the magnetisation in the sample is parallel to the applied magnetic field. In the longitudinal geometry there is no rotation at normal incidence and the magnitude of the rotation increases with the angle of incidence. By contrast, in the polar geometry a rotation can be obtained at all angles of incidence. This can be understood by noting that the magnitude of the Kerr rotation is determined by the projection of $\boldsymbol{k}$ along $\boldsymbol{M}$. In the longitudinal geometry $\boldsymbol{k} \cdot \boldsymbol{M}$ vanishes at normal incidence, whereas in the polar geometry $\boldsymbol{k} \cdot \boldsymbol{M}$ is always non-zero.

The light only penetrates the metal a distance approximately equal to the skin depth; thus even for bulk samples, it is only the material within a few tens of nanometers of the surface that contributes to the observed rotation. In the case of ferromagnetic transition metals this rotation may be as large as one degree. For films thin on the length scale of the skin depth, the total rotation is approximately proportional to the film thickness and it is therefore experimentally feasible to detect the rotation induced by a single monolayer (Moog and Bader 1985).

3. Experimental procedure

The apparatus used for obtaining the SMOKE data was essentially the same as that described by Bland *et al* (1989a). The output from a He–Ne laser is intensity stabilised using a Pockels cell placed between parallel polarisers, which provides a variable degree of attenuation so that the light incident on a reference photodiode is of a constant intensity. The p-polarised light is reflected from the thin epitaxial film sample (see figure 1) and after passing through an analysing polariser, its intensity is recorded by a second photodiode (see figure 2). Any small change in the polarisation state of the light will result in a change in the recorded intensity. A micro-computer is used to ramp the magnetic field repeatedly, to record and then average the resulting hysteresis loops.

It might be thought that maximum sensitivity would be obtained with the analysing polariser set at extinction, but this is not the case. As discussed by Bland *et al* (1989a), the slight ellipticity of the reflected polarisation state, the scattered light from the surface and the presence of intensity-independent noise sources mean that the optimum signal-to-noise ratio is obtained when the analysing polariser is set a small angle away from extinction. The optimum position of the analyser depends on the relative contribution of the various noise sources and how they vary as a function of detected intensity (typically 1–10 degrees away from extinction).

Modifications to the existing system included the incorporation of a more powerful laser source (20 mW He–Ne, Spectra Physics 106-1) which reduces the relative importance of the noise sources that are independent of the detected intensity. Improved software, high speed analogue-to-digital converters and a voltage-controlled current source based on a high-power audio amplifier enable sweeping of the magnetic field at rates up to 20 Hz.

For each cycle of the magnetic field, 100 data points are recorded and so for a sweep frequency f_0, the information relating to the magnetic properties of the film is contained within a frequency window from f_0 to $50f_0$. The noise at lower frequencies is not important since absolute intensities are not required, and the noise at higher frequency is strongly attenuated by the low-pass filter preceding the analogue-to-digital (A–D) converter. In addition, two high-Q notch filters at 50 and 100 Hz remove noise related to the AC mains. However, most of the noise within the frequency window f_0–$50f_0$ cannot be removed by simple filtering (as this would obviously remove the signal, too); instead one relies on averaging many hysteresis loops in order to obtain an adequate signal-to-noise ratio. The noise spectrum is not flat in frequency and the ability to scan the field at a wide range of frequencies enables one to find an optimum value of f_0, which in our case was found to be approximately 4 Hz.

The absolute change in the intensity as a function of magnetic field is very small (less than 1 part in 10^3) and as a result great care must be taken to ensure a stable optical path so that the detector collection efficiency remain constant. To this end, the samples were glued to a rigidly mounted non-magnetic support which is physically separate from the magnetic assembly.

4. SMOKE results for sandwiched Fe and Co films

The two sandwiched epitaxial film structures were prepared under UHV conditions by evaporation from high purity metal sources. Atomic cleanliness and surface structure were monitored during growth by Auger spectroscopy and low energy electron diffraction; the thickness of the protective overlayer was determined by using polarised neutron reflection (Bland *et al* 1987a, Johnson *et al* 1988).

Figure 3 shows hysteresis loops obtained at room temperature for the 8-monolayer BCC Fe film and figure 4 for the 7-monolayer FCC Co film, corresponding to various orientations of the $\boldsymbol{B}$-field with respect to the crystal axes of the epitaxial films. An incident angle of approximately 10° was used in the longitudinal geometry and 60° in the polar geometry. The $\boldsymbol{B}$-field was repeatedly swept between ± 270 Oe at 4 Hz. A total of 2000 loops were collected for each orientation and averaged loops were normalised so that comparisons in the form of the loops could be readily made.

In the case of the Fe film, similar behaviour was observed for all field orientations in the longitudinal geometry, but magnetic saturation could not be obtained for the polar geometry. This is consistent with the existence of planar shape magnetic anisotropy, forcing the easy magnetisation direction to lie in the plane of the film. The longitudinal hysteresis loops exhibit rapid switching, with a coercive field in the region of 130 Oe. This result has important implications for the PNR studies carried by Bland *et al* (1989b), since the low value of the coercive field confirms that the film was magnetically saturated (the applied field was 800 Oe) during the experiment and therefore the deduction of a reduced magnetic moment per atom was valid.

For the Co film, different behaviour is obtained compared with the iron film both in the polar and longitudinal geometries. In the longitudinal geometry, the form of the hysteresis loop is found to depend on the orientation of the $\boldsymbol{B}$-field with respect to the

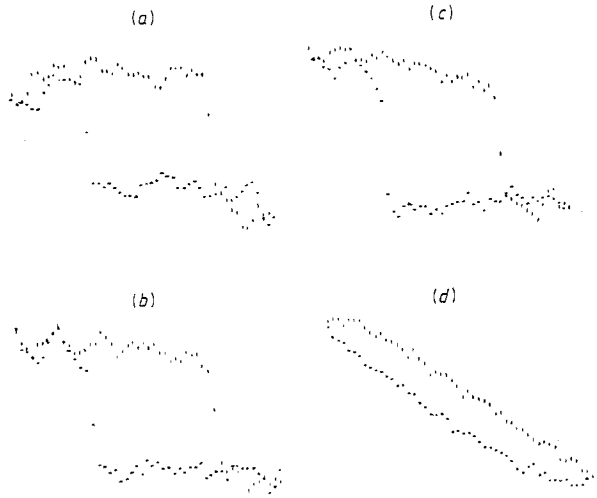

Figure 3. Hysteresis loops obtained from an epitaxial iron film at 300 K for various orientations of the B-field with respect to the crystal axes of the film. 15 nm Ag–8-monolayer BCC Fe–Ag(100); magnetic field swept between ±270 Oe at 4 Hz; data collection time approximately 500 s, (*a*) $\theta = 0°$, $B \perp n$; (*b*) $\theta = 45°$, $B \perp n$; (*c*) $\theta = 90°$, $B \perp n$; (*d*) $\theta = 0°$, $B \parallel n$.

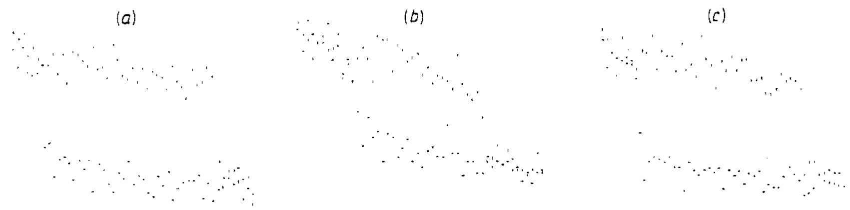

Figure 4. Hysteresis loops obtained from an epitaxial cobalt film at 300 K for various orientations of the B-field with respect to the crystal axes of the film. 10 nm Cu–7-monolayer FCC Co–Cu(001); magnetic field swept between ±27° Oe at 4 Hz; data collection time approximately 500 s. $B \perp n$ in all cases. (*a*) $\theta = 0°$; (*b*) $\theta = 45°$; (*c*) $\theta = 90°$.

crystal axis, indicating the presence of a magneto-crystalline anisotropy. The same quadrangular hysteresis loops are obtained when the field is applied along the [100] direction and perpendicular to it, with a coercive field in the region of 200 Oe. With the field aligned along the [110] direction, a lower coercive field of approximately 130 Oe is observed. Therefore, as in the case of the Fe film, the Co film was also magnetically saturated during the PNR investigations, which indicated a value for the magnetic moment per atom close to that of the bulk material (Bland *et al* 1987a). No magneto-optical signal could be detected in the polar geometry, which implies an extremely hard magnetisation direction normal to the plane of the film. Such behaviour is consistent with the existence of a positive anisotropy constant (Pierce 1987), but complete hysteresis loops need to be obtained for the polar geometry to quantify the anisotropy.

5. Discussion

The hysteresis loops presented in this paper demonstrate the high intrinsic sensitivity of SMOKE techniques as a tool for examining the magnetic behaviour of ultrathin epitaxial films. The reflected signal recovered from an 8-monolayer film buried beneath an overlayer whose thickness is equal to the skin depth for the probe light is comparable in size to the signal obtained from a free monolayer. However, the capability to investigate buried films is an important advantage, since SMOKE measurements can then be carried out on samples already characterised by PNR.

It is interesting to compare the hysteresis behaviour that we observe for the sandwiched Fe film with the findings of Stampanoni *et al* (1987), who studied the magnetic properties of free BCC Fe films during epitaxial growth on Ag(001). In their work (at room temperature) the remanent magnetisation was found to lie in the plane of the film for all thicknesses in the range 1–10 monolayers. We also found that the direction of easy magnetisation lay in the plane of the film and this suggests that the double interface does not introduce strong perpendicular anisotropy.

The strong anisotropy observed for the Co structure may be expected to influence the temperature dependence of the magnetisation. According to Levy and Motchane (1971), a gap in the spin-wave spectrum weakens the linear dependence of the magnetisation upon temperature predicted by 2D spin-wave theory. A weak temperature dependence of the magnetisation is observed for ultrathin sandwiched FCC Co films in the range 4–300 K (Pescia *et al* 1987, Bland *et al* 1987a).

An important feature of the longitudinal hysteresis loops obtained for both films is the almost bistable value of the magnetisation. The fact that the magnetisation at zero field is equal to the saturation value indicates the presence of long range ferromagnetic order (i.e. ordering over distances comparable with the laser probe beam diameter) and that the film behaves as though it were a single domain at zero field. Similar behaviour has been observed for a free FCC Co monolayer by Beier *et al* (1988) and spin-polarised band structure calculations have predicted such ferromagnetic ordering for both FCC Fe–Ag(001) (Fu *et al* 1985) and FCC Co–Cu(001) (Li *et al* 1988) films. For both Fe and Co sandwich structures, the in-plane coercive fields are substantially larger than those observed for free epitaxial Fe films (Moog and Bader 1985) and for a free FCC Co monolayer (Beier *et al* 1988), Elmers and Gradmann (1988) have demonstrated that a non-magnetic coating overlayer strongly increases the in-plane magnetic anisotropy of ultrathin Fe(110) films. It is possible that the overlayer modifies the domain structure or that the dominant mechanism for magnetisation reversal differs for the sandwiched films. To verify these hypotheses, direct observations of the local magnetic domain structure are required, such as have been recently performed using scanning electron microscopy with polarisation analysis (Pierce 1987).

Finally, it should be noted that the large difference in the saturation field obtained for the polar and longitudinal geometries gives a clear indication of the quality of the epitaxial growth. In an ideal thin film the well defined planar interfaces give rise to an easy magnetisation direction which lies in the plane of the film, since for in-plane magnetisation the magnetic dipole–dipole interaction is minimised. By contrast, in a polycrystalline film with granular microstructure comparable values for the saturation field arise due to the lower demagnetisation factor associated with the higher local symmetry (Stoner and Wohlfarth 1948). Therefore, we can infer a high quality of epitaxial growth with sharp interfaces for both samples

6. Conclusions

In this paper we have demonstrated that, despite the reduced signal associated with SMOKE measurements on films buried by metallic overlayers, the high sensitivity of our apparatus enables the observation of the magnetic hysteresis behaviour of ultrathin sandwiched films. This is a particular advantage, since such structures are required for polarised neutron reflection (PNR) investigations.

Our measurements on Ag–8-monolayer BCC Fe–Ag(100) and Cu–7-monolayer FCC Co–Cu(001) confirm the validity of previously obtained PNR data, from which the absolute value of the magnetic moment per atom had been estimated (Bland *et al* 1987a, 1989b).

In the case of the Fe structure we find evidence of planar shape magnetic anisotropy, whereas for the Co structure we observe an in-plane magneto-crystalline anisotropy and a very hard magnetisation axis normal to the interface. Magnetic saturation occurs in both samples for fields applied in the plane of the film with strengths of 130 Oe for the Fe structure and 130–200 Oe for the Co structure, depending on the in-plane orientation. The increased in-plane coercive fields are associated with the presence of the non-magnetic overlayer. In both films, the form of the hysteresis loops is consistent with the presence of long-range ferromagnetic order, as predicted theoretically (Fu *et al* 1985, Li *et al* 1988) and confirms the existence of planar interfaces, indicative of epitaxial growth.

Acknowledgments

We would like to thank Dr J M Huntley and Dr J E Field for the loan of the 20 mW He-Ne laser, N Bett, D Greer and B Marrah for technical support and Dr P Johnson for valuable discussions. JACB gratefully acknowledges the financial support of the Science and Engineering Research Council.

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