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Adsorbate influence on the coercive field of ultrathin Co/Cu{110}

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ABSTRACT

We report on experimental coercive field measurements for ultrathin films of Co on Cu{110}, revealing a complex dependence upon the surface coverage of oxygen and hydrogen adatoms. With reference to first-principles calculations, we rationalise this behaviour in terms of initial reaction with surface contaminants and/or adsorption at defects, followed by (in the case of oxygen) subsequent surface ordering and eventual formation of surface oxide.

Spintronic devices manipulate the quantum spin state of the electron for the purpose of conveying information, exploiting the fact that it is much faster to flip the spin of a polarised current than it is to start or stop that current [1,2]. The giant magnetoresistance (GMR) effect [3–7] is a controlling factor in these devices, which are typically complicated many-layered structures incorporating a multitude of different magnetic and non-magnetic materials [8]. Amongst the most interesting candidate materials for spintronic applications are those involving alternating Co/Cu layers. These structures have been the subject of multiple investigations since the discovery that they are GMR active [9], with the strength of the effect critically dependent on interfacial properties. The varying effects of adding whole or partial capping layers of a variety of metallic atoms have been extensively studied [10–12], however there is little published work that examines the effect on thin film samples of common non-metallic surface impurities, such as those generated through O_2 or H_2 adsorption. These species are naturally of great interest, as even a low coverage of such adsorbates has been shown to alter dramatically the magnetic properties of native ferromagnetic surfaces [13–20]. One particularly interesting phenomenon is the reorientation of surface magnetic anisotropy observed upon adsorption of CO on Co/Cu{110} [21]. In this work we present the first results illustrating a complex relationship between the coercive field of the same thin film system and the coverage of surface impurities arising from adsorption of gas-phase oxygen and hydrogen. Our new results reveal a distinct effect, which we believe relates to the magnitude of the coercive field, rather than to an interchange of easy and hard axes.

The experiments were performed in an ultra-high vacuum (UHV) chamber with a base pressure in the low 10^{-10} mbar range, rising to 8×10^{-10} mbar during Co deposition. The single crystal Cu{110} substrate was prepared via cycles of 2 kV Ar^+ sputtering and subsequent annealing to 800 K, until low energy electron diffraction (LEED) measurements indicated a well-ordered surface. Ultrathin magnetic Co films with thicknesses of 10 ML were deposited by vapours sublimated from a Co rod and then condensed on a clean Cu{110} surface. All growth preparations and measurements were carried out at room temperature. The overlayer is epitaxial, with Co adopting the *fcc* crystal structure rather than its native *hcp* geometry but not strictly layer-by-layer; elongated 'island' structures, oriented in the $\langle 001 \rangle$ direction, are observed in scanning tunneling microscope (STM) images [21]. Nevertheless, with an average Co thickness of 10 ML, it is anticipated that essentially no bare Cu remains exposed at the surface. In situ magneto-optic Kerr effect (MOKE) measurements were performed in the longitudinal geometry, with a maximum field of 2 kG. Angle-dependent MOKE measurements of a 10 ML Co film grown on the clean Cu{110} surface have previously shown that a dominant uniaxial anisotropy is present (consistent with the substrate's position as the highest-symmetry stepped *fcc* surface [22,23]), resulting in easy $\langle 001 \rangle$ and hard $\langle 1\bar{1}0 \rangle$ directions [21].

The Co-covered surface was then dosed with varying quantities of oxygen and hydrogen. Fig. 1 shows the effects of adsorption on the coercivity of 10 ML Co grown on the clean Cu surface. The

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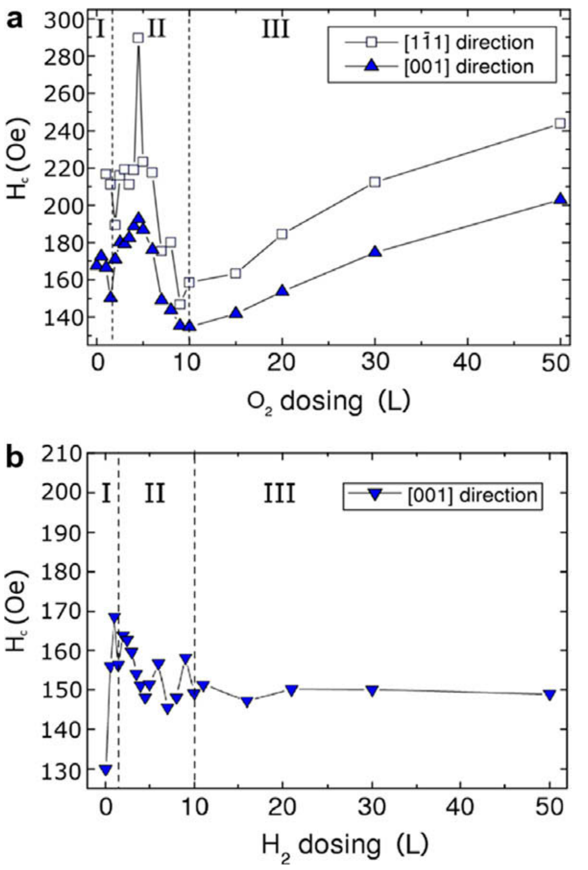

Fig. 1. Evolution of the coercive field as a function of (a) O_2 and (b) H_2 dosage on the clean Co/Cu{1 1 0} surface.

results presented here were obtained in the $\langle 1\bar{1}1\rangle$ and/or $\langle 001\rangle$ azimuths, with the same behaviour essentially observed in both. If the change in the coercive field was purely a consequence of rotation of the easy axis (as is known to occur upon CO adsorption [21]), then little variation would be seen in the $\langle 1\bar{1}1\rangle$ azimuth. Hope et al. [21] have also investigated the effect of oxygen and (indirectly) hydrogen adsorption on this system, and proposed that neither of these species switch the easy axis of magnetisation. It appears, therefore, that the phenomena observed here are *general* modifications of the coercive field magnitude, rather than a reorientation of surface magnetic anisotropy. The Co films show an initial coercive field of around 130–170 G, with the exact value depending on the surface roughness (i.e. varying with the number of sputter/anneal cycles used in preparation of the crystal substrate). Evolution of the coercive field (H_c) upon gas dosing was found to be independent of the initial H_c value, with the curve maintaining a similar shape (albeit offset) for runs with differing initial H_c.

In the case of O_2 dosing, three distinct regimes are observed: initial (I), intermediate (II), and oxidation (III) stages. Initially (regime I), the coercive field shows oscillations upon small amounts of O_2 dosing (<1.5 L). A peak H_c value becomes apparent at an O_2 dosage of around 5 L (regime II) followed by a trough at around 10 L (onset of regime III).

The initial rapid changes in the coercive field (regime I in Fig. 1a) are assigned to probable adsorption in defect sites and/or displacement of residual adsorbed gases on the surface. As the coercivity reaches a peak in regime II, LEED spots reach their maximum sharpness. These spots fade with increasing oxygen dosing beyond ~5 L and background intensity gradually increases. In *some* experimental runs, weak half-order spots are observed at 5 L dosage, consistent with a (2 × 1) overlayer. We believe, therefore, that the peak in coercivity is associated with the attainment of maximum surface order, possibly at 0.5 ML coverage. Certainly it is believed that pre-adsorbed oxygen can act as a surfactant, promoting layer-by-layer growth of Co on Cu{1 1 0} [24,25]; it may be that the role of the oxygen in the present experiments is to smooth, somewhat, the relatively rough Co film. Beyond 5 L, further dosing of oxygen results in an increasingly disordered overlayer, consequently lowering the coercive field. At 10 L dosage (onset of regime III) all LEED spots disappear, suggesting incipient formation of a surface oxide. This not only increases the roughness of the surface, but also results in a thinning of the magnetic cobalt layer leading to an increase in the coercivity.

For H_2 dosing, three different regimes were again found, albeit of different character from the oxygen case: initial (I), intermediate (II), and saturation (III) stages. After an initial sharp increase in H_c (regime I in Fig. 1b), further dosing of H_2 leads to a gradual oscillatory decrease of the coercivity until ~10 L dosing (regime II), after which H_c remains relatively constant (regime III). The initial increase in H_c upon minimal H_2 dosage is proposed to be, as in the oxygen case, a consequence of adsorption preferentially occurring at defect sites. The oscillatory behaviour observed in regime II is a more complex issue. Clearly the coercive field has an intricate (and reproducible) dependence on the hydrogen coverage in this region, but we are unable to proffer a definitive explanation as to the cause of this effect. The LEED pattern does not reveal evidence for any ordered structure that might influence coercivity, but H adatoms are such weak scatterers that this fact alone does not entirely preclude the possibility. Due to the stability of the coercive field with hydrogen dosages above 10 L, we suspect that this exposure corresponds to saturation coverage (onset of regime III).

While the experimental evidence alone can offer us some explanation for the behaviour of the coercive field in these systems, a theoretical analysis of the influence of the adatoms on the surface is desirable and should aid further comprehension.

The first-principles density functional theory (DFT) calculations reported in this work were performed using the CASTEP code [26]. Ultrasoft pseudopotentials [27] and the Perdew–Wang form of the generalised gradient approximation [28] were used to describe the electron–ion interactions and to calculate the exchange–correlation energies respectively. The plane wave basis set was given a cutoff value of 340 eV and the electronic spin is relaxed in all calculations. Quantification of atomically-resolved spin magnetic moments in this letter are performed via a Bader type analysis of the system [29]. The cobalt pseudopotential includes non-linear core corrections (NLCC) [30] and the Broyden-Fletcher-Goldfarb-Shanno (BFGS) method was used for geometry optimisation.[2]

The adsorption of gas-phase species on a Co{1 1 0} surface was modelled using a supercell consisting of six layers of *fcc* cobalt in a (2 × 1) unit cell, with a vacuum region of approximately 11 Å. The lateral dimensions of the supercell were fixed by the copper lattice constant.[3] For each gas-phase adsorbate, five high-symmetry initial adsorption sites were considered (Fig. 2). Adsorption was allowed on only one side of the cobalt slab, with the bottom

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[2] Applicability of these parameters was determined through trial calculations performed for bulk *fcc* cobalt and copper; the Brillouin zone corresponding to the primitive unit cell was sampled with a 6 × 6 × 6 **k**-point mesh according to the Monkhorst-Pack scheme [31]. The lattice parameter and magnetic moment of bulk cobalt were found to be 3.547 Å and 1.68μ_B, respectively, in good agreement with previous theoretical [32,33] and experimental results [34]. The lattice parameter of copper was calculated to be 3.596 Å, also agreeing well with experimental [34] and theoretical [35] values.

[3] Our previous (unpublished) calculations for Co overlayers on Cu{1 1 0} show that beyond about 6 ML the magnetic properties at the vacuum interface are essentially indistinguishable from those of a native *fcc* surface whose lateral dimensions are held fixed at values consistent with the copper lattice constant.