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LETTER TO THE EDITOR

Effects of atomic-scale Cu structures on the magnetic anisotropy and magneto-optical response of ultrathin Co films

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Abstract. We present magneto-optical Kerr effect measurements of epitaxial Co/Cu(001) films studied *in situ* as submonolayer quantities of Cu are deposited under ultrahigh-vacuum conditions. An enhancement of the magneto-optical response coincides with changes in the remanence behaviour, which are shown to be due to a variation in the ratio of the uniaxial and cubic in-plane magnetic anisotropy strengths. The data are consistent with the preferential growth of Cu atoms at atomic steps due to the substrate: these Cu 'wires' reduce the step-induced uniaxial anisotropy and their electronic structure strongly affects the magneto-optical response.

The influence of interfaces and non-magnetic spacer layers on the magnetic properties of ultrathin films has become a key issue in magnetic multilayer research [1–5]. Increasingly it is recognized that atomic-scale defects, e.g. roughness or atomic steps, can strongly affect the observed magnetic properties. Experimentally, it is now well known that the presence of a continuous overlayer can affect the observed magnetic properties of a single magnetic film: reports include a variation in the Curie temperature T_C [6, 7], of the perpendicular interface anisotropy [4, 8–11] and of the coercive field H_c for in-plane Co films [12]. For in-plane systems, in contrast with perpendicular systems, the Néel model [13] predicts no surface anisotropy term, but preliminary experiments on Co films grown on Cu(001) [14] provided evidence for non-monotonic changes in H_c with concurrent changes in the Kerr loop amplitude as Cu is deposited. These findings suggest that changes in electronic structure affect the resulting magnetic properties, but the mechanism was unclear. Studies on vicinal surfaces [15–17] reveal that the step-induced magnetic anisotropy can change due to submonolayer coverages of non-magnetic materials. Recently we have reported observations of strongly varying magnetic properties in the Co/Cu(001) system due to a Cu overlayer, for a wide range of Co thicknesses [18]. This work suggested that both the step-induced uniaxial anisotropy and the magneto-optical signal are affected by the Cu overlayer although it was not clear precisely how this occurred. In this letter we quantify the large changes in the uniaxial in-plane magnetic anisotropy which occur as submonolayer quantities of Cu are deposited and compare the results obtained using substrates of varying surface roughness. In this way, we are able to explain the correlation between the observed enhancement of the magneto-optical response and the strong change in the step-induced magnetic anisotropy in terms of the formation of atomic-scale Cu 'wires' in the vicinity of the steps on the substrate.

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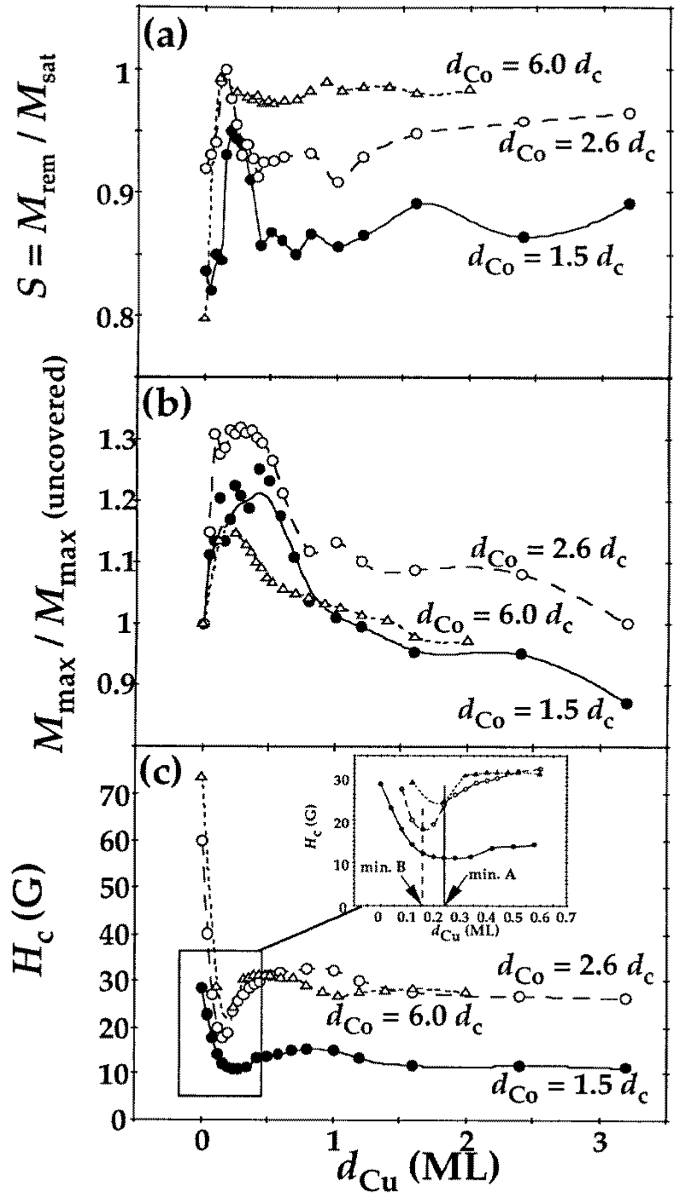

Figure 1. The behaviour of (a) the squareness S, (b) the loop amplitude M_{max}, and (c) the coercive field H_c, for the three films. The differing initial values of S are due to the particular substrate used, as described in the text; the inset in (c) shows the slight difference in peak coverage d_{peak} at which the minima are observed for the two substrates A and B. The lines are guides to the eye.

All of the experiments were carried out *in situ* in UHV with a base pressure of 1.0×10^{-10} mbar, and a pressure of lower than 5.0×10^{-10} mbar during deposition. Magnetic measurements were made using the transverse magneto-optical Kerr effect (MOKE), in which a change in the sample magnetization direction is detected as an intensity change in the reflected laser signal (a Helmholtz coil is used to apply a magnetic field along the [110] axis of the sample). The Cu single-crystal substrates used were cleaned by cycles of 1 kV Ar^+ sputtering and annealing to 700 K, until LEED and Auger techniques indicated a well ordered surface with no contamination. Three commercially supplied Cu crystals were used, to eliminate artefacts due to a particular substrate; a variety of Co film thicknesses were grown, in order to identify the causes of the observed behaviour. The Co film thickness is measured in terms of d_c, a highly reproducible reference thickness corresponding to the critical point for the onset of ferromagnetism at room temperature, and which is reported to be in the range 1.0–1.7 monolayers (ML) [8, 19, 20] (in agreement with our evaluation of 1.3 ML using the ratio of Auger peak heights; no differences could be measured between different substrate crystals). Co films and Cu overlayers were deposited by electron-beam evaporation, at typical speeds of 0.1 ML min^{-1} and 0.025 ML min^{-1} respectively.

M–H loops were recorded as Cu overlayers were grown on three Co films: thicknesses of $1.5d_c$, $2.6d_c$ and $6.0d_c$, on two substrates denoted A (the $1.5d_c$ and $6.0d_c$ films) and B (the $2.6d_c$ film). In each case the loops reveal changes in the loop amplitude M_{max}, loop squareness S, and coercive field H_c (M_{max} is the magnetic signal observed at the largest applied field strength, $S = M_{remanence}/M_{saturation}$). We are particularly interested here in the submonolayer coverage range; the behaviour shows further non-monotonic structure at higher Cu coverages, associated with the completed Cu/Co interface (including a reduction in the Curie temperature T_C), and is discussed elsewhere [18]. In figure 1, we display the behaviour of S, M_{max} and H_c as the Cu overlayer is grown. All of the Co thicknesses display the same qualitative behaviour: for around 0.2 ML Cu we see a pronounced peak in S accompanied by a sharp minimum in H_c and a concurrent but broader maximum in M_{max}. A slight difference in the position of the minimum in H_c is observed between the two substrates, displayed in the inset of figure 1(c); this will prove to be important in our interpretation. Further Cu deposition causes S to fall, then vary in a complex fashion within a limited range, and we see in all of the cases a secondary maximum in H_c in the range 0.5–0.9 ML Cu, and a decrease in M_{max} to a value close to or below that for the uncovered film. In general, less strong changes in S and H_c are seen on substrate B, and we find that the differing values of S obtained for uncovered Co films depend on the particular substrate used.

The variation of S is surprising since the [110] direction is expected to be a fourfold easy axis of the ideal Co film. The reduced remanence could arise either from domain formation or due to a rotation of the spins with respect to the [110] axis. In the following model, we assume that domains do not form at remanence (although the effect of domains cannot be fully excluded without an *in situ* domain-imaging technique such as spin-SEM). According to this description the non-unity remanence that we observe implies that the magnetization is rotated slightly away from the crystallographic [110] axis. Therefores an additional symmetry-breaking anisotropy term is present and we can use the measured remanence behaviour as a form of torque magnetometry to quantify its strength. Following Krams *et al* in describing the Co/Cu(1 1 13) system [8, 21], we express the magnetic anisotropy energy of the Co film as the sum of a fourfold in-plane anisotropy term K_1 and a uniaxial anisotropy term K_u. The following expression relates the value of S to the ratio of the anisotropy coefficients K_u and K_1:

$$\frac{K_u}{K_1} = -2S\sqrt{1 - S^2}. \qquad (1)$$

From this analysis we infer that as Cu atoms are deposited changes in K_u/K_1 are responsible for the variation in remanence that we observe. In figure 2 we show how

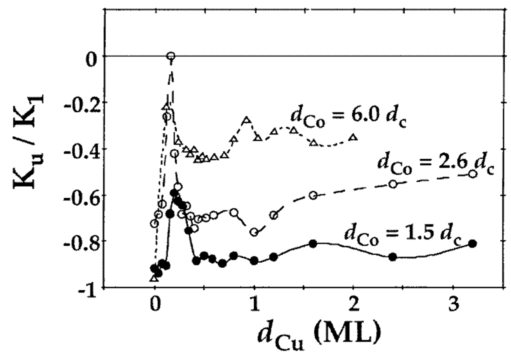

Figure 2. The behaviour of the ratio K_u/K_1, deduced from the data for S. The sharp minimum at $d_{Cu} = 0.2$ ML correlates with the squarest loops recorded in a given sequence, representing approximately fourfold behaviour in the samples. The lines are guides to the eye.

the ratio K_u/K_1, derived from S, changes as a function of Cu overlayer thickness for the Co films of figure 1. The ratio is a minimum when the squarest loop is recorded, as the system tends towards the ideal fourfold behaviour. This model may also account for the observed minimum in H_c, if the magnetization reversal in the Co/Cu(001) system proceeds via domain wall motion. By taking the value of S that we observe for the uncovered $1.5d_c$ film, and using the value of $K_1(d_{Co} = 1.5d_c) = -0.75 \times 10^6$ erg cm^{-3} reported by Krams *et al* [8], we estimate the value of the uniaxial anisotropy strength to be $K_u(\text{uncovered}) = 7 \times 10^5$ erg cm^{-3} for the $1.5d_c$ film. This corresponds to an anisotropy energy per unit area of surface of the film of $k_u \approx 1 \times 10^{-2}$ erg cm^{-2}.

Studies of Co films on vicinal surfaces [8, 15–17, 21, 22] demonstrate that a uniaxial anisotropy can arise from steps on the substrate. Weber *et al* [15, 16] have shown that the step-induced anisotropy changes as a Cu overlayer is grown. For significantly miscut Cu substrates, the anisotropy changes are large enough to reorient the easy axis of the Co film by 90°. For the Fe/W(110) system, Albrecht *et al* [23] report a step anisotropy energy of $k_{step} = 1.0 \times 10^{-13}$ J m^{-1} = 1.0×10^{-8} erg cm^{-1}. Assuming that the Co/Cu(001) and Fe/W(110) systems have comparable magnetic step anisotropy energies and attributing the value of k_u estimated above to the step anisotropy energy density, we deduce a terrace width of the order of 100 Å which compares well with the results of STM studies of Cu(001) substrates [24–26]. We thus attribute the uniaxial magnetic anisotropy in our samples to the presence of steps on the substrate [27]. We infer that after ≈0.2 ML Cu deposition, structures formed by the adsorbed Cu atoms result in this step-induced anisotropy being cancelled. We expect the coverage d_{peak} at which the anisotropy cancellation occurs to be proportional to the step density, within a restricted range. The value of 0.2 ML coverage is thus a property of the substrate used, but corresponds to sufficient Cu atoms in the vicinity of the steps to suppress the uniaxial character of the uncoated Co step atoms. This conclusion is supported by the fact that we see the pronounced features of the behaviour at the same Cu coverage for all of the Co thicknesses on a particular substrate. This implies that the Co films must maintain the same step morphology as the substrate, even up to thicknesses of $6.0d_c$ as is expected for pseudomorphic growth. For substrate B $d_{peak} = 0.16$ ML whereas for both the films grown on substrate A $d_{peak} = 0.24$ ML (see the inset of figure 1(c)). The larger initial value of S obtained for the films grown on substrate B (figure 1(a)) confirms that this substrate is smoother. Hence the peak coverage is indeed found to increase with roughness (step density) as expected. Optical and x-ray techniques were used to investigate the miscut of the substrates, which is less than 0.25° (the limiting resolution of the Laue technique).

We next consider the behaviour of M_{max}. Measurements of the magnetization of Co/Cu(001) during growth [28] show that, in order to obtain a 20% increase in the absolute magnetization upon deposition of 0.2 ML of Cu, the incoming Cu atoms would need to carry an unphysically large magnetic moment which is of the same order as that of a Co atom. However, the Cu atoms are likely to acquire a spin polarization as they are adsorbed, as is predicted theoretically [3, 29] and as reported for closely related Co/Cu systems [30–32]. The magneto-optical behaviour can be strongly affected by such an induced polarization in a non-magnetic overlayer, as reported for example for Ru/Co [33]. We therefore deduce that the enhancement of M_{max} must be magneto-optical in origin rather than a pure change in the magnetization itself. It is predicted theoretically [3, 34, 35] that hybridization of the Cu and Co electronic states can lead to a suppression of the magnetic moment of the Co atoms. It appears that this is occurring for Cu coverages above 0.2 ML, where M_{max} decreases more rapidly than can be explained by optical attenuation of the signal by the overlayer. This implies a competition between these two effects, and we conclude that the enhancement of the magneto-optical response dominates for the lowest Cu coverages, while at higher coverages the reduction in the Co moments dictates the behaviour—thus we see a peak in the value of M_{max} as the Cu thickness increases. We assume that since an additional mechanism is involved in this process, namely the hybridization of the Co and Cu electronic states, the peak in M_{max} is broader than the features of S and H_c which are sensitive principally to the step anisotropy.

A key feature of our data is that the peak in M_{max} is always concurrent with the sharp minimum in H_c and maximum in S. Our model is consistent with this behaviour if we assume that initially the Cu atoms adsorb preferentially at the steps, creating 'wires' on the surface of the sample, as has been seen directly by STM for other epitaxial metal overlayer systems [36, 37]. Initially the steps will dominate the behaviour, while after a small amount of deposition the steps will become 'diluted' with Cu atoms. The magneto-optical enhancement is a maximum at the same peak coverage d_{peak} at which the ratio K_u/K_1 is minimized, which implies that Cu 'wires', sufficient to suppress the uniaxial anisotropy of the steps, have an electronic structure that also causes a maximum in the magneto-optical response. Then as a complete monolayer coverage is approached, the distinct step-like nature of the sample returns as is evident from the increase in the uniaxial anisotropy strength, the electronic structure of the Cu layer approaches that of a complete layer, and hence the magneto-optical response decreases slightly. This explains why, for larger Cu coverages, S is less than 1 and M_{max} decreases.

In summary, we have shown that as a Cu overlayer is deposited in submonolayer steps on a Co/Cu(001) film, concurrent strong, non-monotonic changes in the magnetic anisotropy and in the magneto-optical response of the system occur at the same Cu coverage for all of the Co thicknesses up to 8 ML. We explain the behaviour in terms of a model of preferential growth of atomic-scale Cu 'wires' at steps on the surface of the Co film, which reduces the step-induced uniaxial anisotropy K_u, and simultaneously polarizes the Cu 'wires' giving rise to a strongly enhanced magneto-optical response. An estimate of the required step separation based on this model is in reasonable agreement with published STM studies of Cu(001) [24–26]. The maximum loop squareness occurs for $d_{Cu} = 0.15$–0.25 ML according to the substrate. Our results indicate that the critical thickness required to fully suppress the influence of the Co step atoms exceeds that corresponding to a single line of Cu atoms at the step edge. This suggests that the electronic properties of the step region continue to change with increasing Cu coverage. We have thus demonstrated the strong magneto-optical effects which can occur in partially polarized atomic-scale Cu wires, and believe that there is strong evidence to support our model of Cu 'wires' forming at the terrace edges. Our results illustrate the possibility of studying the magnetic and magneto-optical properties of atomic-scale structures and we hope will stimulate future theoretical studies of the electronic structure of such atomic-scale 'wires'.

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