Direct observation of the stabilization of ferromagnetic order by magnetic anisotropy

S. Hope, B.-Ch. Choi, P. J. Bode, and J. A. C. Bland
Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, United Kingdom
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We have engineered an experimental realization of an isotropic two-dimensional *XY* magnet by depositing submonolayer coverages of Cu onto a CO exposed 5 ML Co/Cu(110) film with zero cubic anisotropy component K_1 at room temperature. For a Cu coverage of 1.02 ML, the uniaxial anisotropy component vanishes also, and we observe a corresponding loss of ferromagnetic order at remanence. Further Cu deposition restores the uniaxial anisotropy and the magnetic order.

The theory of Mermin and Wagner[1] proved that long-range magnetic order cannot exist at any finite temperature in a two-dimensional (2D) magnet that is described purely by the Heisenberg Hamiltonian with isotropic, short-range interactions. The theory extends to the case of a 2D magnet with spins constrained to lie in the plane, i.e., an isotropic 2D XY magnet.[1–3] However, experimentally a quasi-2D magnetic system may be realized by growing a few atomic layers of a ferromagnet on top of a nonmagnetic substrate, and the existence of long-range magnetic order in such systems is well established. It is now widely accepted that 2D ferromagnetism in systems of infinite size is stabilized by magnetic anisotropies[4] (including the dipole shape anisotropy) and therefore to experimentally verify the theory of Mermin and Wagner one would need to engineer a 2D magnet in which the inherent anisotropies balance to give a net isotropic state. This of course is extremely difficult to achieve experimentally, and to date only systems which exhibit reorientation phase transitions (RPT's) have provided an opportunity to approximate this scenario.[5,6] One can imagine that at a critical thickness or temperature the perpendicular surface anisotropy and in-plane shape anisotropy might cancel (ignoring the higher order anisotropy terms), creating an almost perfectly isotropic film.

Pappas *et al.*[7] studied the reorientation phase transition as a function of temperature in the fcc Fe Cu(100) system using spin-polarized secondary-electron spectroscopy. They found that the remanent magnetization M_R vanished along all three perpendicular directions over an extended temperature range $\Delta T_S \cong 30\,\mathrm{K}$, where T_S is the switching temperature for the RPT. However, later scanning electron microscopy with polarization analysis (SEMPA) measurements by Allenspach *et al.*[4] on the same system ruled out the loss of ferromagnetic order as a reason for the vanishing of remanence observed by Pappas *et al.* Allenspach *et al.* discovered that the formation of reverse-oriented stripe domains were responsible for the loss of M_R reported earlier, and that this stripe domain pattern only appears as a consequence of the near cancellation of the surface and shape anisotropy contributions in this system.

Recent Brillouin light-scattering (BLS) measurements by Krams *et al.*[8] were thought to provide indirect evidence that ferromagnetic order is stabilized by in-plane magnetic anisotropies at room temperature in ultrathin Co/Cu(001). They found that the in-plane anisotropy constant $K_{\text{in-plane}}$ tends to zero at a Co thickness of $d_C^* \sim 1.7\,\mathrm{ML}$, which is in excellent agreement with the Co thickness ($d_C \sim 1.0–1.7\,\mathrm{ML}$) at which the onset of magnetic order is known to occur in this system.[9,10] However, later work by Schumann *et al.*[11] revealed that the onset of magnetic order in the Co/Cu(001) system can be accurately described by a percolation phase transition at a Co thickness of ~1.3 ML. Therefore, the loss of $K_{\text{in-plane}}$ observed by Krams *et al.* may simply reveal the fact that the Co atoms are not yet physically connected across the sample at a Co thickness of d_C^* and so would not exhibit a measurable anisotropy.

To directly investigate the role of anisotropy in stabilizing magnetic order, it is necessary to vary the anisotropy strength independently of parameters such as film thickness and temperature. The unique properties of the Co/Cu(110) system allow us to achieve this goal in principle, since in this system it is possible to extinguish both the in-plane cubic and uniaxial anisotropy contributions at a constant Co thickness (above the critical thickness for the onset of magnetic order) at room temperature. BLS measurements by Hillebrands *et al.*[12] have shown that the in-plane cubic anisotropy contribution K_1 is strongly suppressed by strain below $d_{\mathrm{Co}} \sim 50\,\text{Å}$ (40 ML), and tends to zero below a Co thickness of approximately 15 ML. Furthermore, we have previously reported[13] that the easy axis in this system is observed to switch through 90° from the [001] to the [1–10] direction, as the sign of the effective in-plane uniaxial anisotropy constant K_U^{eff} is reversed as a result of the adsorption of submonolayer coverages of residual CO gas from the UHV environment. K_U^{eff} includes the contribution of the shape anisotropy associated with the average strength of the dipole-dipole interactions. In addition to this uniaxial anisotropy, the dipole-dipole interactions can, in principle, lead to residual contributions to the total energy, which we will address later. The easy axis switch can be controllably and completely reversed by depositing submonolayer coverages of a Cu overlayer[14] on top of the CO/Co interface and therefore for a sufficient quantity of Cu, K_U^{eff} must pass through zero.

In this work we have deposited 5 ML of Co on to the Cu(110) surface and allowed the residual CO gas in the UHV chamber to switch the easy axis. For this Co thickness, K_1 is essentially zero according to the BLS measurements of Hillebrands *et al.*[12] and therefore only K_U remains. Incre-

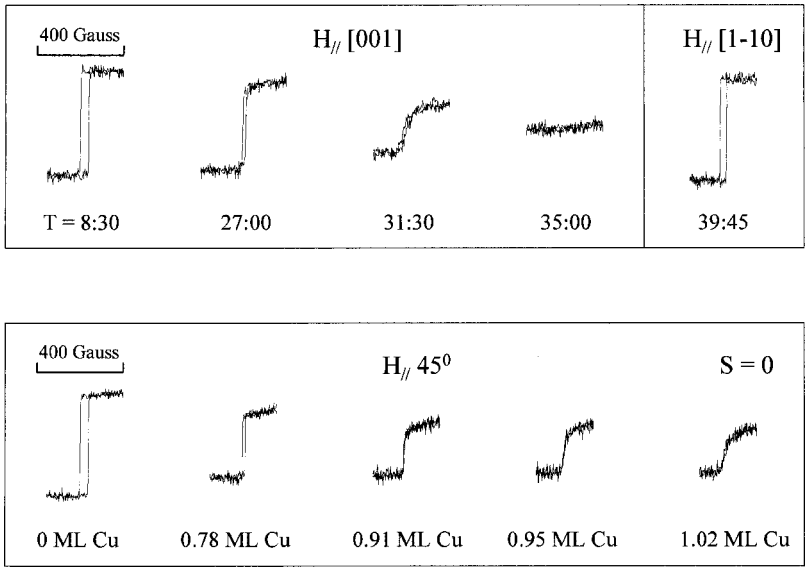

FIG. 1. (Top panel) The CO-induced easy axis switch from the [001] to the [1–10] direction on a 5 ML Co/Cu(110) film at room temperature. The time in minutes and seconds after the end of the Co deposition is given underneath each loop. After 35 min the [001] direction has become the hard axis. Rotating the sample so that the field is applied parallel to the [1–10] direction shows that this direction is now the new easy axis. (Bottom panel) The field is applied parallel to a direction midway between [001] and [1–10], i.e., 45°. Submonolayer increments of Cu overlayer are deposited on to the CO/Co interface. This has the effect of reversing the CO-induced easy-axis switch by reversing the sign of the effective in-plane uniaxial anisotropy K_U^{eff}. A Cu coverage of 1.02 ML has reduced the loop squareness S and hence remanence M_R to zero along this direction. All loop amplitudes are in the same arbitrary units and hence comparable.

mental submonolayer quantities of Cu were then deposited in an attempt to reduce K_U^{eff} to zero and therefore convert the system from a 2D Ising to an isotropic 2D XY magnet. All experiments were performed under UHV conditions with a base pressure of 3.0×10^{-10} mbar. The single-crystal Cu(110) substrate (miscut less than 0.5°) was prepared via cycles of 1 kV Ar^+ sputtering and annealing to 700 K, until Auger electron spectroscopy (AES) and low-energy electron-diffraction measurements indicated a clean well ordered surface. Co was evaporated from an Omicron commercial electron beam evaporator with an integral flux monitor, at a constant rate of 0.4 ML min^{-1}. Cu was evaporated extremely slowly from an e-beam evaporator at a rate of 0.015 ML min^{-1} which allowed very fine tuning of the effective in-plane uniaxial anisotropy K_U^{eff}. The thickness of the Co and Cu layers was estimated from the relative heights of the Co and Cu AES peaks. This procedure gives an error in the absolute thickness of ~25%. However, by keeping the evaporation rate constant, the relative thicknesses are known much more accurately. Magnetic measurements were performed using the magneto-optic Kerr effect (MOKE) in the transverse geometry.

The top panel of Fig. 1 shows the MOKE loops recorded after a deposition of 5 ML of Co on to the Cu(110) surface. The numbers underneath each loop indicate the time in minutes and seconds that have elapsed since the end of the Co deposition. Initially, the easy axis is along the [001] direction as indicated by the square loop with unity remanence when the field is applied parallel to the [001] direction. As residual CO gas is adsorbed on to the surface, the loop shape can be seen to evolve as the easy axis begins to switch through 90°. After 35 min the loop is indicative of a magnetic hard axis. Applying the field along the [1–10] direction reveals that this direction is now the new easy axis. The sample was then rotated so that the field was applied parallel to a direction midway between [001] and [1–10], i.e. 45°. The bottom panel of Fig. 1 shows a series of loops recorded with the field applied along this direction as incremental submonolayer coverages of Cu overlayer were deposited. A deposition of 1.02 ML of Cu has reduced the coercive field and loop squareness $S=M_{\text{rem}}/M_{\text{max}}$ (defined as the ratio of remanent to maximum Kerr signal) to zero. Rotating the sample so that the field could be applied along intermediate directions showed that the remanent magnetization M_R was zero for all in-plane directions (Fig. 2 top panel shows representative loops along three directions). Therefore, we conclude that we have engineered an isotropic 2D XY magnet since both K_U^{eff} and K_1 are now zero (we have assumed that higher-order anisotropy terms are zero or negligible). The bottom panel of Fig. 2 shows the result of depositing further Cu. The effective in-plane uniaxial anisotropy has changed sign and the easy axis has switched back to the [001] direction, restoring M_R and the coercive field along the 45° and [001] directions.

It is interesting to investigate the sharpness of the transition to zero remanence as a function of Cu overlayer thickness, as observed with the field applied along the 45° direction. Figure 3 shows a plot of the loop squareness S for the same sample as studied in Figs. 1 and 2. The plot shows that S is never at unity, which is to be expected since the 45° direction never becomes an easy axis direction during the easy axis switch for very thin Co/Cu(110) films.[14] Clearly M_R drops very sharply to zero at a Cu thickness of ~1 ML as K_U^{eff} tends to zero, then rises very sharply as K_U^{eff} changes sign. The solid line is a guide to the eye but we can estimate from the plot that M_R is zero over a Cu thickness range of less than 0.05 ML within the resolution of our measurements. The sharpness of the transition and the extremely narrow width over which $M_R=0$ suggests that as we reach the

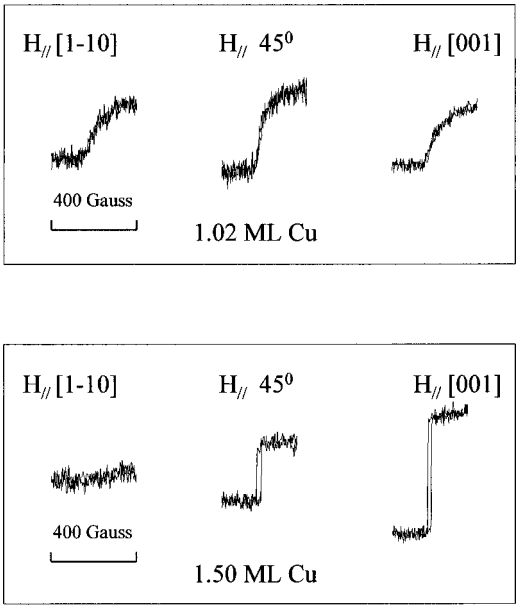

FIG. 2. (Top panel) An angle-dependent study shows that the remanence has fallen to zero along all in-plane directions for a Cu coverage of 1.02 ML, indicating that the sample is magnetically isotropic with an absence of spontaneous magnetic order in the zero applied field. This situation corresponds to the point where $K_1=0$ and $K_U^{\text{eff}}=0$. (Bottom panel) Further Cu coverage has completed the reversal of the easy axis switch and restored the remanence and coercive field along the [001] and 45° directions. The [001] direction is the easy axis again and K_U^{eff} has changed sign again. All loop amplitudes are in the same arbitrary units and hence comparable.

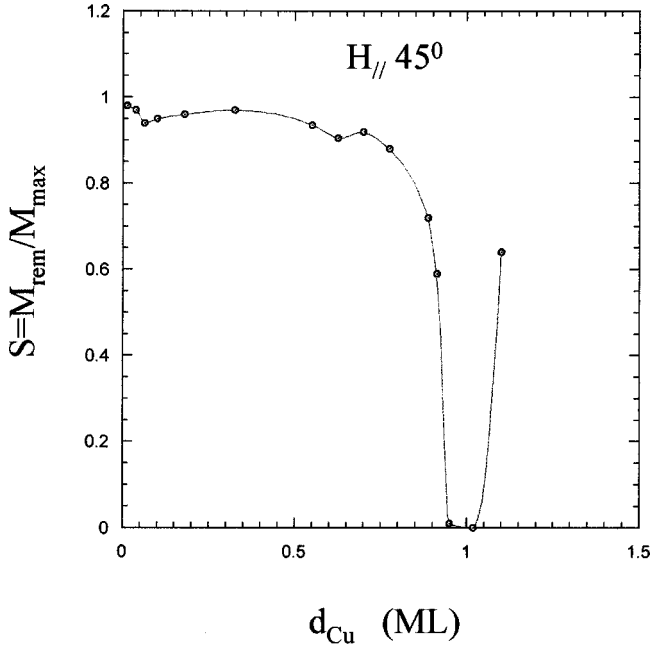

FIG. 3. A plot of the loop squareness S as a function of Cu overlayer thickness with the field applied parallel to the 45° direction. The transition to zero remanence occurs very sharply at a thickness of nearly 1 ML as K_U^{eff} tends to zero, then rises sharply as K_U^{eff} changes sign. The solid line is a guide to the eye but we can estimate that the remanence remains at zero for less than 0.05 ML of Cu, suggesting that as we reach the point of extinguishing the in-plane anisotropies the magnetism becomes exquisitely unstable to thermal fluctuations resulting in a loss of magnetic order in zero applied field. Further Cu deposition restores the uniaxial anisotropy and M_R.

point of extinguishing the effective in-plane anisotropy field, the magnetism becomes exquisitely unstable to thermal fluctuations resulting in a loss of ferromagnetic order, as expected theoretically.[1–3] This of course assumes that there is a direct correspondence between the *microscopic* and *macroscopic* quantities of spontaneous magnetization M_S, and remanent magnetization M_R, respectively. Allenspach has tested this correspondence[4] by combining SEMPA and MOKE measurements on the Co/Cu(001) system in the vicinity of T_C and found M_S and M_R to be equivalent. This correspondence can be expected to extend to all in-plane magnetized ultrathin systems.[4]

If we are to be certain that the loops displayed in Fig. 2 (top panel) represent an intermediate state of zero in-plane anisotropy and zero ferromagnetic order, the possibility of domain formation must be ruled out. If reverse-oriented domains formed as K_U^{eff} tended to zero and these domains were smaller than the MOKE laser spot size ($\sim$1 mm), then it is possible that the averaged Kerr signal would display zero remanence along all in-plane directions and a sizeable magnetic response M_H in an applied field, as observed. However, we have previously observed that for a 6.5 ML Co/Cu(110) film, the magnetization remains in a single-domain state as K_U^{eff} is driven to zero with increasing Cu overlayer thickness[14] up to $\sim$1 ML. Only within a very narrow Cu thickness range (0.9–1 ML) did M_R fall sharply as K_U^{eff} passed through zero and changed sign. The [001] direction was then immediately established as the new easy-axis direction in a single-domain state. Therefore, in the present case we need only consider the possibility of remanent domain formation in the critical region as K_U^{eff} is driven through zero. However, there is no energetic advantage for domain formation to occur since the domain-wall thickness is given by

$$\delta \propto \sqrt{\frac{A}{K}},$$

where A is the exchange stiffness and K is the anisotropy. Clearly, as K tends to zero the domain-wall thickness tends to infinity and hence domains cannot form. We attribute the slight asymmetry in the loops to instrumental asymmetries in the experimental setup which would not influence measurements of the remanence or coercive field.

We attribute the sizeable magnetic response M_H in small applied fields, (as observed in Fig. 2, top panel) to be the result of 2D spin blocks.[15] An inherent property of 2D magnets at temperatures slightly above T_C is that there exists large thermal fluctuations of spin blocks which can be easily oriented parallel to a small external field thereby inducing an appreciable magnetization. It must be emphasized that the Mermin Wagner theorem strictly only applies to infinite systems. As originally pointed out by Berezinski and Blank[16] and later by Bramwell,[17] finite-size effects caused by defects, or the dimensions of the sample for instance, will always result in a finite magnetization in a 2D XY magnet. The fact that our MOKE measurements reveal a complete absence of magnetization at remanence in the isotropic state suggests that finite-size effects and residual dipole-dipole interactions

are overwhelmed at room temperature. However, since we have engineered the anisotropy field in this system independently of other parameters such as temperature and magnetic thickness, we have directly demonstrated that ferromagnetic order is stabilized by magnetic anisotropy in 2D ultrathin films.

In conclusion, we have engineered a close approximation to an isotropic 2D XY magnet by depositing submonolayer coverages of Cu onto a CO exposed 5 ML Co/Cu(110) film. This thickness of Co is above the critical thickness for the onset of magnetic order in this system ($d_C = 4.6$ ML),[18] and therefore T_C is initially well above room temperature. The unique properties of the Co/Cu(110) system allow us to alter the in-plane anisotropy fields independently of the Co thickness and temperature. The theoretical description of a 2D XY magnet indicates that either magnetic anisotropy, dipole-dipole interactions or finite-size effects are required for long-range ferromagnetic order to occur. However, we have observed directly that magnetic anisotropy plays the crucial role in stabilizing long-range ferromagnetic order at room temperature since in the absence of the effective anisotropy we find that long-range ferromagnetic order collapses.

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