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Abstract. By using the magneto-optic Kerr effect (MOKE) we have investigated the coercivity of ultrathin epitaxial Co films grown by molecular beam epitaxy with the aid of Pb as the surfactant on Cu(111). We find two different regimes: below $\sim$6 ML (ML $\equiv$ monolayer), the coercive field H_c increases continuously, whereas for thicker layers it falls at a rate that is inversely proportional to the film thickness. While this latter behaviour is typical for bulk systems, we show that the initial one is an effect of reduced dimensionality, reflecting the variation of the Curie temperature of the magnetic films in the same thickness range. This phenomenon could have important implications for atomic-scale engineering of magnetic materials.

1. Introduction

Magnetic ultrathin films represent model systems for studying the influence of reduced dimensionality and symmetry on electronic properties. The coincidence of the nanometric scale of the films and particles with the characteristic magnetic lengths results in critical magnetic behaviour. The broken symmetry at surfaces and interfaces creates contributions to the magnetic anisotropy that can alter the easy axes of magnetization [1–3]; low dimensionality creates effects such as the thickness dependence of the Curie temperature [4–6].

Magnetization reversal and relaxation processes, in particular, are attracting a great deal of interest. Coercivity is one of the most critical parameters of any magnetic material; besides its fundamental importance to the understanding of relaxation processes at the atomic scale, its value determines the range of possible applications. High values of the coercive field H_c are required for magnetic storage media and for permanent magnets, while 'soft' materials are needed for sensors, reading heads, and transformer cores. In this paper we report on a study of the thickness dependence of the coercivity of thin Co films grown on Cu(111) with the help of a surfactant monolayer of Pb. Thanks to the latter, the structural quality is greatly enhanced, and the influence of increasing film roughness can be eliminated [3, 7]. We show that in the low-coverage range ($\theta_{Co} \leqslant 6$ ML), H_c gradually increases following the evolution of the film's Curie temperature with thickness, before a bulk-like behaviour sets in for thicker films.

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2. Experimental procedure

The experiments have been carried out in a UHV system with two sections: the first one is dedicated to the growth and structural characterization of the ultrathin Co films, and is equipped with MBE evaporators and facilities for LEED, AES spectroscopy, and neutral He-atom diffraction (TEAS). The second chamber contains a set-up for magneto-optic Kerr effect (MOKE) experiments: the measurements can be performed in both polar and longitudinal geometries, with a maximum applied field of 600 Oe. The substrate was a Cu(111) single crystal with a miscut angle of $\sim 1°$. It was mechanically and electrochemically polished prior to inserting into vacuum, and cleaned by *in situ* cycles of Ar$^+$ sputtering and annealing. Co was evaporated from a high-purity rod heated by electron bombardment, with the sample held at room temperature (RT). Pb was evaporated from a Knudsen cell; a full monolayer of surfactant was employed in all cases. Thicknesses were calibrated from TEAS intensity oscillations during surfactant-induced layer-by-layer growth. After preparation, the sample was transferred under vacuum to the MOKE chamber, where the hysteresis curves were measured at RT.

3. Results and discussion

The evolution of the coercive field H_c as a function of Co film thickness δ_{Co} is displayed with solid squares in figure 1(a). All the films were grown using 1 ML Pb as surfactant; they had in-plane magnetization, and the hysteresis curves were recorded in longitudinal geometry. Two very different regimes can be clearly distinguished: in the shaded region ($\delta_{Co} \leqslant 6$ ML), H_c rises steadily, while above this thickness it decreases at a much slower rate. In this paper we aim to demonstrate that the initial increase is a genuine effect of reduced dimensionality, associated with the changes in the film's Curie temperature. For larger
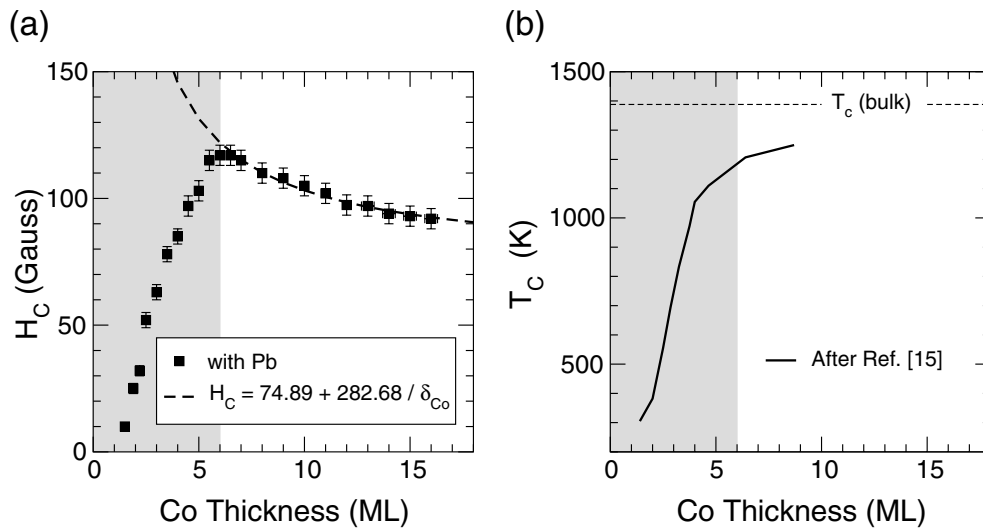
Figure 1. (a) Experimental values, obtained using the MOKE, of the coercive field H_c in Co films grown with Pb as surfactant (filled squares). The dashed line is a fit to the high-thickness data using equation (1) (see the text). (b) Predicted evolution of the Curie temperature of Co films grown on Cu(111) (after [15]). Most of the change in T_c occurs for the same range of Co coverages as the initial increase in H_c (shaded area).

thicknesses, three-dimensional behaviour sets in and the variation of H_c with Co coverage is provoked by the influence of this parameter on the structure and dynamics of magnetic domain walls.

Additional support for this interpretation is provided by the data depicted in figure 2. The filled circles show the values of the Kerr intensity measured at saturation (I_{sat}) for increasing Co film thickness. The solid straight line is a fit to the high-thickness data points. The linearity of I_{sat} with Co coverage indicates that the Kerr signal is proportional to the film's total magnetic moment. However, significant deviations are found for the lowest coverages; the inset in the figure shows a blow-up of this region. Below $\theta_{Co} \simeq 2.5$ ML, the Kerr intensity drops below the straight line, indicating that a fraction of the deposited Co is not contributing to the magnetic signal in our experiment.
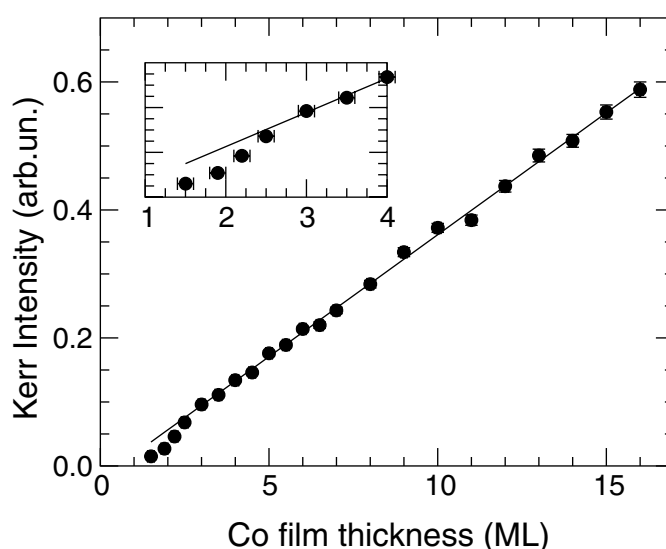
Figure 2. Evolution of the Kerr intensity measured at saturation, as a function of Co film thickness. The straight line is a linear fit to the high-coverage data. The inset shows a blow-up of the low-coverage region, where the data deviate from the linear behaviour.

The explanation for these features lies in the close relationship between the samples' morphology and dimensionality and their magnetic properties. It is therefore necessary to perform a careful characterization of the structure of the samples and its evolution during growth. Using He and x-ray diffraction, we have thoroughly studied the growth of Co on Cu(111), with special emphasis on the surfactant effect of a monolayer of Pb. These experiments will be presented in full detail elsewhere [8]; nevertheless, the most relevant results are summarized in figure 3. This graph shows the progressive filling of individual atomic layers during deposition. The Pb monolayer, predeposited on the clean Cu(111) substrate, is floating atop the growing Co film at all times. The high structural quality achieved in the epitaxial Co films thanks to the presence of the surfactant is evident from the figure; with Pb, Co grows in a nearly perfect layer-by-layer fashion. Above 2 ML Co, a steady state is reached such that the surface morphology continuously replicates itself without any apparent degradation as evidenced by the parallel lines indicating the evolution of individual layer occupations. In fact, during the growth of {Co/Cu} superlattices we have observed that the beneficial effect of the surfactant layer can be maintained at least up to 900 Å thickness [9], after which the well-ordered Pb layer can still be detected on the surface.
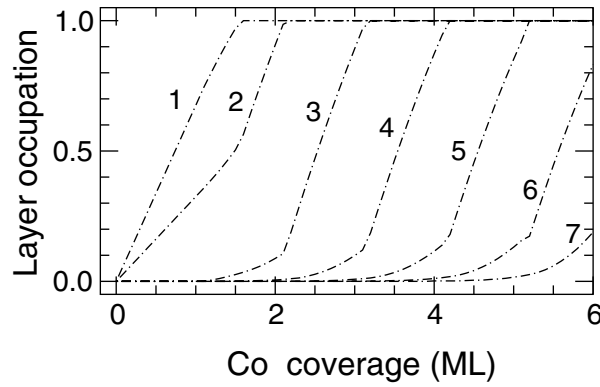
Figure 3. Progressive filling of atomic layers during Co deposition on Cu(111) at room temperature; the presence of 1 ML of Pb acting as surfactant induces layer-by-layer growth. These results have been obtained from the analysis of our TEAS and x-ray diffraction experiments [8].

Let us analyse in detail the information furnished by the data in figure 1. Coercivity is a property related to the rate at which magnetic relaxation between the remanent and demagnetized states takes place. At $T = 0$ the coercive force measures the height of the barriers that must be overcome by the magnetic moments to restore equilibrium, i.e., to reach the demagnetized state. This relaxation process involves displacements of magnetic domain walls and can therefore be strongly influenced by the film structure and thickness. In fact, the coercive fields typically measured in ultrathin magnetic films are orders of magnitude smaller than those of bulk samples of the same materials [10]. This discrepancy has been attributed to the effect of step edges, such as island borders [11]. Demagnetization effects can also influence the sample's coercivity via changes in the average local slope of the surface roughness [12]. For these reasons, it is extremely important to eliminate from the magnetic measurements any possible influence of topological defects. In our experiments, this goal is achieved thanks to the use of the surfactant. The data displayed in figure 3 demonstrate that the film's morphology does not change noticeably for Co thicknesses between 2 and 6 ML, since all atomic layers from the third onwards are filling at exactly the same rate. Furthermore, the same growth scheme, with equivalent layer fillings and constant diffracted intensity during deposition is maintained for film thicknesses higher than 6 ML, during the second regime of coercivity. The increase in H_c must therefore have a different origin.

In a simple model, we consider the energy γ per unit area stored in a magnetic domain wall; γ arises from exchange, anisotropy, magnetostriction, and magnetostatic contributions. The wall area is S. The total force acting on a wall under an external field H must be zero at its equilibrium position. Thus, the force exerted by the potential in which the wall moves, $d(\gamma S)/dx$, must exactly counterbalance the force applied on the wall by the field: $\mu_0 M_s H S(\cos\phi_1 - \cos\phi_2)$, ϕ_1 and ϕ_2 being the angles formed by the magnetization vector on both sides of the wall. H_c is the minimum field required to induce irreversible wall motion; therefore, using the approximation $\cos\phi_1 - \cos\phi_2 \simeq 1$, one gets

$$H_c = \left(\frac{1}{\mu_0 M_s}\right)\left(\frac{d\gamma}{dx}\right)_{max} + \left(\frac{\gamma}{\mu_0 M_s \delta}\right)\left(\frac{d\delta}{dx}\right)_{max}. \tag{1}$$

The second term in (1) varies as $1/\delta$. The derivative is practically constant, because it reflects changes in δ associated with roughness in the Co film, which does not undergo significant variations with increasing film thickness as we have seen from figure 3 and discussed in the previous paragraph. By using this expression we have been able to fit our experimental data

for Co films thicker than 6 ML; the theoretical curve is depicted in figure 1 with a dashed line. The resulting fit parameters are the following:

$$\left(\frac{1}{\mu_0 M_s}\right)\left(\frac{d\gamma}{dx}\right) = 74.89 \text{ G}$$

and

$$\left(\frac{1}{\mu_0 M_s a}\right)\gamma\left(\frac{d\delta}{dx}\right) = 282.68 \text{ G} \times \theta_{Co}$$

where we have substituted for the film thickness δ the product $\theta_{Co}a$, with a being the atomic interlayer spacing and θ_{Co} the Co film thickness expressed in monolayers. Typical values for these parameters are $\gamma \simeq 10^{-3}$ J m^{-2} and $1/\mu_0 M_s a \simeq 3 \times 10^9$ T^{-1} m^{-1}; hence, the fit indicates that $(d\delta/dx)$ is of the order of 10^{-2}, corresponding to thickness fluctuations of ~1 Å over typical distances on the surface, such as the mean terrace size (~120 Å). Such a value is quite reasonable, because the average roughness in these samples amounts to a single monolayer, as demonstrated by the data depicted in figure 3 and by additional STM experiments [13].

We will now focus on the low-coverage region. The initial rise of the coercivity with increasing film thickness appears quite unexpected in view of the above discussion. In order to understand this behaviour, it is worth noting that the coercive force is a decreasing function of temperature. Since all contributions to the magnetic anisotropy fall with increasing temperature faster than the spontaneous magnetization, the coercivity tends toward zero as the sample's temperature approaches T_c [14], vanishing at that point. This general rule applies also to thin films as demonstrated by the MOKE data presented in figure 4, which were obtained on a 3 ML Co film grown on Cu(100); the strong interdiffusion of Co on Cu(111) prevented us from performing equivalent measurements on this latter surface. Panel (a) shows several hysteresis curves obtained at different temperatures. On approaching T_c, both the coercivity H_c and the saturation magnetization M_s tend towards zero. These results are summarized in figure 4(b), where the temperature dependences of both magnitudes are displayed.
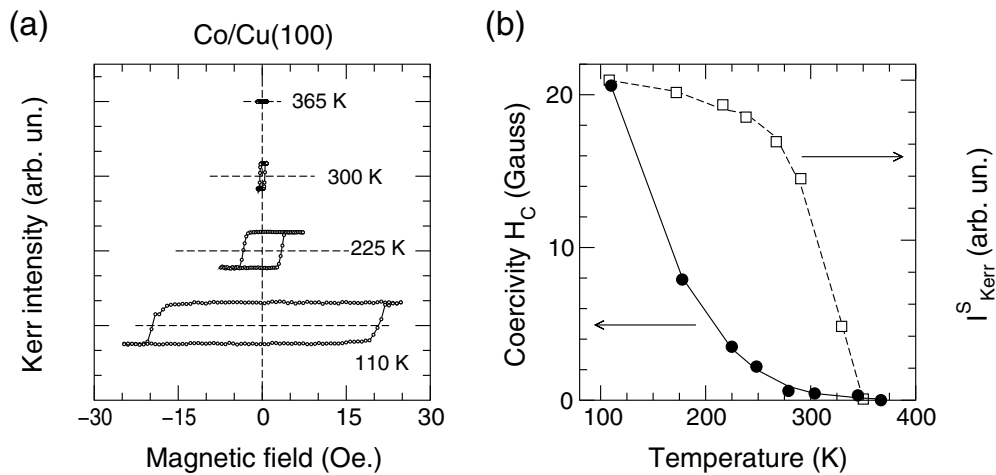
Figure 4. (a) Hysteresis curves measured using the MOKE for a 3 ML Co film grown on Cu(100), at different temperatures. (b) Temperature dependences of both the coercive field H_c and the saturation Kerr intensity (proportional to M_s). The two magnitudes vanish at the film's Curie temperature.

It is a well-established fact that the Curie temperature T_c of an ultrathin magnetic film depends on its thickness. This effect has been observed experimentally [4–6] and confirmed by theory [15] for Co on both Cu(100) and (111). The results of this latter calculation are depicted in figure 1(b). The scale of the ordinate axes has been chosen in such a way that the Curie temperature of the thinnest film for which we can detect remanence (1.5 ML) coincides with RT. From figure 3 one can see that it is at precisely this thickness that the first atomic layer fills up and the Co film becomes continuous. Below that point, it consists of small, isolated Co patches and islands; these particles must be superparamagnetic with a blocking temperature lower than RT. The transition region extends up to $\sim$2.5 ML, when the second atomic layer is completed and the whole sample first contributes to the magnetic measurement. Beyond this coverage every Co atom incorporated into the film adds the same amount to the sample magnetization, which explains the linearity of the data in figure 2. Nevertheless, the evolution of the film's T_c towards the bulk value takes longer to complete. This process cannot be observed experimentally by performing measurements at elevated temperatures due to the interdiffusion of the Co into the Cu substrate, and we can only rely on theoretical calculations.

Our interpretation of the $H_c(\delta_{Co})$ data relies precisely on this coverage dependence of T_c. Recalling that all of our measurements were taken at room temperature (T_R), we define a reduced temperature $\tau = T_R/T_c(\delta_{Co})$. Our experiments on films with increasing thickness probe the magnetic properties of these samples at different values of τ, down to 0.21 which corresponds to the Curie temperature of bulk Co. The dependence of H_c on the reduced temperature in ferromagnetic systems is well known [14, 16]. Thus, the increment of the coercive fields during the early stage of growth must be ascribed to the thickness dependence of T_c, induced by the reduced dimensionality of the Co films. A value of $T_c(\delta_{Co})$ close enough to T_c^∞ is reached at $\delta_{Co} \simeq 6$ ML [15]; from this point on, the Curie temperature of the Co films changes very slowly, and the $1/\delta_{Co}$ dependence of H_c described in the first part dominates. Finally, it should be mentioned that Co films grown without surfactant show also an increasing coercivity in the low-coverage region (2 to 4 ML), indicating that the presence of the Pb layer does not affect noticeably the Curie temperature of the magnetic films. Nevertheless, the actual values of H_c for those films are reduced with respect to those for the samples grown with surfactant, due to the large roughness and lack of continuity caused by the pyramidal growth of Co on clean Cu(111) [13], and their evolution with increasing thickness is irregular.

Results similar to ours have been reported for the coercivity of Co films grown on Cu(100). Weber *et al* [17] observe an initial increase of H_c up to $\approx$5–6 ML Co that closely follows the evolution of T_c for that system [5] and also our own results for Cu(111). For higher thicknesses these experiments differ from ours in that H_c continues to rise slowly, instead of decreasing as $1/\delta_{Co}$. The changes in the films' Curie temperature being negligible, this increase of the coercivity reveals a gradual build-up of surface roughness, since those Co films were prepared without surfactant. TEAS [18] and MEED [19] experiments on the epitaxial growth of Co on Cu(100) have shown layer-by-layer growth, with diffracted intensity oscillations superimposed on a slowly decreasing average level. This slow decay of the surface reflectivity points to the accumulation of some additional roughness during growth; the effect of this increment in the density of defects can easily outweigh the expected δ^{-1} dependence. In fact, Jiang *et al* [20] have measured simultaneously the evolution of both the sample coercivity and the surface roughness as functions of Co film thickness, growing also on a Cu(100) substrate. After an analogous initial rise of H_c below $\theta_{Co} < 7$ ML, they measure a slowly decreasing, nearly constant coercivity coincident with an increasing Co film roughness. Finally, similar results have been reported for Co films grown on a Cu-buffered Si(111) surface [21].

In summary, we have measured the evolution of the coercive field H_c with increasing thickness in epitaxial Co films grown on Cu(111). The remarkably high structural quality of

these films was achieved using a monolayer of Pb as surfactant. This allowed us to rule out any influence of structural or morphological features on the observed behaviour. We find that H_c first increases up to a Co coverage of $\sim$6 ML, and later falls as the inverse of the Co film thickness. While this latter dependence follows the expectations for thick, bulk-like layers, the initial increase is an effect of reduced dimensionality, and reflects the evolution of the Co films' Curie temperature in the same thickness range. Although similar results had been reported in other instances, this is the first time that the physical origin has been identified.

These observations have several important implications. The first one is that the measurement of H_c on ultrathin films of different thicknesses can serve as an indirect, and technically simple, method of monitoring, at least in a qualitative way, the evolution of their Curie temperatures. A direct measurement of T_c is frequently impossible, especially for magnetic superlattices, because they are destroyed by interdiffusion at elevated temperature. Besides, and perhaps more importantly, these findings open new possibilities for atomic-scale engineering of magnetic properties. By finely tuning the thickness of magnetic layers, it should be possible to control with great accuracy the magnetic hardness of any given material.

Acknowledgment

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