Effect of Substrate Symmetry on the Preferred Magnetization Orientation of Ni Films on Cu

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Effect of substrate symmetry on the preferred magnetization orientation of Ni films on Cu

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For ultrathin Ni films grown on Cu(100) and Cu(110), the easy axis of magnetization starts out in the plane of the films. Above a critical thickness, the preferred magnetization orientation changes to perpendicular to the films. This behavior is contrary to the spin reorientation transition reported for many thin film ferromagnetic systems where the preferred magnetization orientation starts out perpendicular to the film and then switches to in-plane above a critical thickness. We report in situ surface magneto-optic Kerr effect measurements of the magnetic anisotropy as a function of film thickness. The change in the predominant domain orientation distribution occurs at 8 monolayers (MLs) for Ni on Cu(100) and at 16 ML for Ni on Cu(110). For Ni on Cu(111) no such change is observed down to 3 ML thickness. These results are discussed in terms of competing surface anisotropy and dipole–dipole energy terms. © 1995 American Vacuum Society.

I. INTRODUCTION

For ultrathin magnetic films the competition between the spin–orbit surface anisotropy and the dipole–dipole interaction determines the direction of the easy axis of magnetization. The surface anisotropy depends on the symmetry properties of the film and the interfacial strain, while the dipole–dipole interaction increases with increasing volume. For films with an initial perpendicular easy axis, as the film thickness is increased the volume dependent dipole–dipole interaction increases with film thickness and overcoming the surface anisotropy. This forces the preferred direction of magnetization to in-plane, which is shown in Fig. 1(a). This transition has been observed for Fe/Ag(100),1,2 Fe/Cu(100),2 and Co/Au(111).3 The surface anisotropy is, however, sensitive to small changes in interatomic distances and the symmetry of the surface such that other possibilities exist [Figs. 1(b) and 1(c)]. For Ni films deposited on Cu(100) and Cu(110), the preferred direction of magnetization is in-plane, and it switches to perpendicular at switching thicknesses of 8 and 16 monolayers (MLs), respectively. For Ni films grown on Cu(111) the easy axis of magnetization lies in-plane up to at least 18 ML and down to 3 ML.4

Bulk Ni is the canonical Heisenberg ferromagnet with a spin moment of 0.6 bohr magneton,5 a Curie temperature $T_c$ of 627 K,6 and a weak magnetocrystalline anisotropy with the easy axis of magnetization in the [111] direction and the hard axis in the [100] direction.7 In the thin film limit we expect the surface anisotropy will be much greater than the bulk magnetocrystalline anisotropy. Theoretical studies of Ni monolayers predict that a (100) oriented Ni film has a preferred direction of magnetization in the plane of the film.8,9 To what extent these ground state calculations for monolayer thick films can be applied to thicker films is yet to be determined. Recent state-tracking, first-principles calculations demonstrate that the magnetocrystalline anisotropy energy for monolayers is dependent on band filling and strain.10 Any differences in film thicknesses for switching of the easy axis of magnetization could be due to a sudden change in the interfacial strain at the critical thickness for pseudomorphic growth.

In this article we show that the differences in the switching thickness for Ni films on Cu substrates are consistent with differences in the critical thickness for pseudomorphic growth. When Ni is grown on Cu(100), the film assumes the in-plane lattice constant of Cu up to a critical thickness of 10 ML beyond which the lattice constant relaxes to the value of bulk Ni (2.6% smaller than Cu). On Cu(110), we expect this critical thickness to increase since this surface has a higher corrugation than the (100) surface, which more effectively clamps the overlayer thin films. In contrast, a recent study by x-ray photoelectron diffraction shows that Ni on Cu(111) interfaces are not pseudomorphic lattice-matched interfaces, and Ni films thicker than 5 ML have a fully relaxed bulk Ni(111) in-plane lattice constant.11 Since films greater than 5 ML thick have an easy axis of magnetization in-plane, we conclude that this is the preferred direction for these strain-relaxed films.

II. EXPERIMENT

Ni thin films were grown epitaxially on single crystal Cu(100) and Cu(110) substrates at 400 and 300 K, respectively. The substrates were cleaned mechanically and electropolished, then after a few cycles of 500 eV Ar+ sputtering and subsequent annealing to 750 K, the substrates were free of contamination and display sharp $P(1 \times 1)$ low energy electron diffraction (LEED) patterns. The deposition rate of Ni was controlled to be approximately 1 ML/min for all the films. The composition of the films was determined by Auger electron spectroscopy (AES), with all of the films showing a small (<10%) but measurable amount of carbon and oxygen contamination. The surface structure of the films was monitored by low energy electron diffraction and reflection high energy electron diffraction (RHEED) patterns. The magnetic properties were studied in situ by the surface magneto-optic Kerr effect (SMOKE) with a HeNe laser $\lambda = 632$ nm incident at 70° from the surface normal. Our apparatus allows us to...
measure both the polar Kerr effect for perpendicular magnetization and the longitudinal Kerr effect for in-plane magnetization.

III. RESULTS

Figure 2 shows the hysteresis loops of both longitudinal and polar Kerr effects for Ni films grown on Cu(100) with film thicknesses of 6, 7, 8, and 10 ML. There is an abrupt increase in the polar Kerr signal at 8 ML while the coercivity for parallel direction increases beyond the maximum field of our apparatus. Meanwhile, the coercivity for perpendicular magnetization decreases to about half of the original magnitude, which indicates that the easy axis of spin now lies perpendicular to the thin film. There is similar behavior for Ni films grown
on Cu(110): the signal for polar Kerr effect has an abrupt increase at 16 ML, while the coercivity of in-plane magnetization also increases abruptly beyond the maximum field of our apparatus. These measurements show that the switching of easy axis of magnetization from parallel to perpendicular direction occurs at 16 ML in the Ni(110) films.

LEED measurements show that there is no gross structural change occurring at the switching thickness. However, a subtle structural change occurs at films thicker than 10 ML for Ni on Cu(100) where the film transforms from a strained face centered tetragonal structure at low thickness with an in-plane lattice constant identical to the Cu substrate to a relaxed face centered cubic structure with the in-plane lattice constant of bulk Ni. This structural relaxation change affects the anisotropy through the spin–orbit coupling, which is sensitive to small changes in distances of nearest neighbor atoms as well as the different symmetry properties of the overlapping wave functions for the two different structures. A recent calculation of this magnetocrystalline anisotropy energy shows that generally increasing the atomic spacing will make the crystal field weaker and the environment of atoms more isotropic, and this will result in a decrease of the energy which prefers a perpendicular magnetization.11

Some previous studies have shown that interfacial roughness has complicated effects on the strain and anisotropies of thin ferromagnetic films, so we have also studied the effect of interfacial strain on this particular spin reorientation transition by taking similar measurements on Ni(100) films grown on a Cu substrate with the surface roughened by Ar bombardment, which is expected to modify the interfacial anisotropy. We find that increasing interfacial roughness weakens the magnetization in both the perpendicular and parallel directions while the coercive field becomes larger than that of the smoother interface. However, the reorientation transition still occurs at the same film thickness for the rough as well as the smooth interfaces.

All of our measurements show that for Ni films grown on Cu(100) and Cu(110), there is a switching thickness below which the films have a dominant in-plane magnetization, while above it the easy axis of magnetization is out-of-plane of the films. This switching thickness is 8 ML for Ni on Cu(100) and 16 ML for Ni on Cu(110). Our measurements (Figs. 4 and 5) indicate that these films have a multidomain magnetic microstructure which is a combination of out-of-plane and in-plane domains, with in-plane domains dominating below the switching thickness, which is consistent with the theoretical predictions. This behavior can be interpreted as the dominance of surface anisotropy at the very low film thickness. As the film thickness increases, the magnetic anisotropy is affected by a small structural relaxation which changes both the atomic spacing and the symmetry properties of the overlapping wave functions. At much greater film

**FIG. 4.** Kerr intensity at remanence for Ni films grown on Cu(100) and Cu(110). There is an abrupt increase for both longitudinal and polar Kerr intensities at 8 ML for Ni/Cu(100) and at 16 ML for Ni/Cu(110).

**FIG. 5.** Coercive field for Ni films grown on Cu(100) and Cu(110). For Ni/Cu(100), there is an abrupt decrease in coercivity for perpendicular magnetization at 8 ML while coercivity for the parallel direction increases beyond the maximum field of our apparatus. For Ni/Cu(110), the coercivity is increasing monotonically for both parallel and perpendicular magnetizations.
thickness, Ni films on Cu(100) show a transition back to in-plane magnetization,\textsuperscript{17} which is due to the thickness dependent dipole–dipole interaction which favors an in-plane magnetization. In a previous publication,\textsuperscript{4} we have shown that Ni(111) films on Cu do not show this effect; the magnetization remains in-plane at these thicknesses.

These results show the qualitative behavior of the Ni films, since the magnitudes of the Kerr effects are not strictly proportional to the magnetizations. More detailed information can be obtained by performing wavelength-dependent SMOKE,\textsuperscript{18} such as how the spin–orbit interaction changes at the critical thickness.

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