Ferromagnetic order at surfaces of ultrathin Fe(100)/Ag(100) and Tb/Fe(100)/Ag(100) films determined by electron capture spectroscopy

C. Rau and G. Xing
Physics Department, Rice University, Houston, Texas 77251

(Received 7 October 1988; accepted 28 November 1988)

The temperature and magnetic field dependence of the electron-spin polarization (ESP) at atomically clean and flat surfaces of ultrathin (1–10 monolayers) bcc Fe(100)/Ag(100) and Tb/Fe(100)/Ag(100) films is studied as function of film thickness. For the investigation of the ESP, we use electron capture spectroscopy which enables us to study long-ranged ferromagnetic order at the topmost layer of a surface. The geometrical and chemical structure of the films is well characterized using Auger electron spectroscopy and low- and high-energy electron diffraction. Atomic flatness is studied using scanning tunneling microscopy. Nonzero ESP is found at the surface of all films. We find ambiguously that in-plane remanent ferromagnetic order exists at uncoated surfaces of 1 to 10 monolayer thick bcc Fe(100)/Ag(100) films for temperatures ranging between 173 and 303 K. It is further found that the Curie temperature of the Fe films depends on film thickness. Evidence is found for in-plane magnetic anisotropies which strongly depend on film thickness. In striking contrast to uncoated Fe films, large temperature-dependent perpendicular (out-of-plane) magnetic anisotropies exist at surfaces of Tb-coated Fe(100)/Ag(100) films.

I. INTRODUCTION

Recently, magnetic phenomena at surfaces, at interfaces, and in ultrathin films have received great attention. This stems from the fact that they are not only of pivotal importance in modern magnetic recording and memory devices, but also, if completely understood, provide a profound basis to our understanding of the effects of reduced symmetry on the properties of matter. At surfaces and interfaces, the crystalline symmetry is reduced and, consequently, intrinsic out-of-plane and in-plane magnetic surface anisotropies can be induced.

Leaving aside such intrinsic surface effects, elastic strain, existing at surfaces, at interfaces, and in films, such as artificial superlattices or sandwiched films, can cause additional strain-dependent, nonintrinsic magnetic surface anisotropies which can be profitably used to manufacture new magnetic materials where the easy axis of the surface magnetization points in a desired direction.

For the surface of a system of localized spins, first pioneering theoretical work was published by Néel.[1] For an itinerant-electron system first most promising work was done by Cooper and Bennett,[2] and by Takayama, Bohnen and Fulde.[3] Quite recently, Gay and Richter,[4] and Fritsche and Noffge[5] calculated the spin anisotropies for supported and unsupported ultrathin Fe films composed of only a few monolayers (ML).

Gradmann and co-workers[6] and Prinz, Rado, and Krebs[7] were among the first who successfully reported on the experimental determination of surface magnetic anisotropies. In the meantime, several further high-quality publications are available, some of them focussing on magnetic surface anisotropies existing in thin Fe films which either are uncoated,[8–10] coated,[11] sandwiched, or multilayered between nonmagnetic host layers.[12]

Comparing theoretical predictions[13,14] with experimental results and their, in a few cases, sophisticated interpretation, the situation at present can be characterized as being far from consensus.

Developing theoretical concepts for predicting spin anisotropies in specific systems, Gay and Richter[4] recently state that their preliminary calculations of the anisotropy of a monolayer of Fe on Ag indicate that the Ag substrate must be treated fully relativistically. Further, they report that the spin–orbit energy is slowly convergent in the number of **k** vectors used to approximate the Brillouin-zone integral and, therefore, depends strongly on the number of **k** vectors used.

As regards the published experimental work,[7–12] it has to be noted that the interpretation of these few experimental data is extremely complicated because of the present difficulty to separate a plurality of surface magnetic effects ranging from strain-induced effects to effects caused by coating a surface. Gradmann[6] recently pointed out that surface anisotropies of metal-coated surfaces seem to be lower than the anisotropies of free surfaces. Further, the various probing depths of the presently existing surface spectroscopies vary over a wide range, a fact which also has to be taken into account when "intrinsic" surface magnetic data are extracted from experimental data.

The major aim of the experiments, described in this paper, is to study the magnetic behavior of a truly two-dimensional magnetic system consisting of a few monolayers of bcc Fe(100) on Ag(100) substrates. Using electron capture spectroscopy (ECS), we investigate the long-ranged electron-spin polarization (ESP) at the topmpost surface layer of well-prepared and characterized, ultrathin (1–10 ML) Fe(100) films epitaxially deposited on Ag(100) substrate single crystals.

We report on the dependence of the ESP on film thickness,

 0734-2101/89/031889-04$01.00

substrate temperature, and applied magnetizing field. Special emphasis is given to the existence of magnetic surface anisotropies at uncoated and Tb-coated Fe surfaces. In all experiments, the magnetizing field is applied in the surface plane of the Fe(100) films along a [011] axis.

Using ECS,[15] we find unambiguously that in-plane remanent ferromagnetic order exists at uncoated surfaces of 1 to 10 ML thick bcc Fe(100)/Ag(100) films for temperatures ranging between 173 and 300 K. It is further found that the Curie temperature of the Fe films depends on film thickness. Evidence is found for in-plane magnetic anisotropies which strongly depend on film thickness. Next, and in striking contrast to uncoated Fe films, large temperature-dependent perpendicular (out-of-plane) magnetic anisotropies are found at surfaces of Tb-coated Fe(100)/Ag(100).

The preparation of atomically clean and flat substrates and the deposition of well-ordered and well-characterized films often represent major difficulties in experimental studies of ultrathin epitaxial films. These obstacles can now be overcome for an increasingly large number of interesting systems. The epitaxial growth of Fe(100) on Ag(100) is already well established.[8,11,16] Details will be described below.

II. EXPERIMENTAL

The present experimental results were obtained using electron capture spectroscopy. This method has been discussed in detail in Ref. 15, and we shall only briefly describe its essential features. ECS is based on the capture of one spin-polarized electron during grazing-angle surface reflection of fast deuterons D^+.[15] At specular reflection, the distance of closest approach of the deuterons is 1–2 Å, establishing that the ions probe only the topmost surface layer of the sample.

After reflection, the atomic part of the beam consists of D^0 atoms with polarized electron spin. The electrons then transfer their polarization to the D^0 nuclei by hyperfine interaction. The nuclear polarization thus measures the electron spin polarization, and is determined from the asymmetry in the angular distribution of the ^{4}He particles emitted in the reaction $T(d,n)^4$He.

Because the electrons captured by different deuterons originate at widely separated points on the target surface, observation of a nonzero spin polarization indicates the existence of long-ranged surface ferromagnetic order. Defining the electron spin polarization P along the direction of the magnetizing field yields

$$P = (n^{\uparrow} - n^{\downarrow})/(n^{\uparrow} + n^{\downarrow}),$$

with $n^{\uparrow}$ and $n^{\downarrow}$ the numbers of up(majority)- and down(minority)-spin electrons.

For the deposition of ultrathin Fe(100) films, atomically clean and flat Ag(100) substrate crystals are prepared under ultrahigh vacuum in a target preparation chamber.[15] The surface orientation of the Ag(100) crystals is better than 0.02°, and is monitored by use of a precision x-ray diffractometer.

Applying standard cleaning and annealing procedures developed in our earlier studies,[15,16] and using Auger electron spectroscopy (AES) with a cylindrical mirror analyzer, residual C and O contaminations are measured to be $< 1\%$ of a monolayer. The single-crystalline (1×1) state of the Ag(100) surfaces is detected by low-energy and high-energy electron diffraction (LEED, RHEED). The atomic flatness of the substrate surfaces is investigated by means of a scanning tunneling microscope (STM) and by means of grazing-angle ion reflection of deuterons at the Ag(100) surfaces. The ion reflectivity I (reflected beam intensity per solid angle/incoming beam intensity per solid angle) amounts to 95%, establishing the atomic flatness of the surfaces.[15] In previous work, it was already established that bcc Fe(100) on Ag(100) grows layer by layer for the first 3 ML.[8,11,17] We note that the lattice constants of bcc Fe and fcc Ag differ by a factor of $\sqrt{2}$, which results in a 45° rotation of the Fe surface net compared to the Ag surface net.

For the deposition of 1 to 10 ML Fe(100), we use electron beam evaporation at 5×10^{-10} mbar as described in Refs. 18–20. The atomic flatness of the films is revealed by ion reflectivity measurements, and the absence of surface contaminations is detected by AES.

Low-energy electron diffraction measurements show a $p(1\times1)$ structure for all films studied. No changes in symmetry of the electron-diffraction patterns are found, implying the epitaxial growth of bcc Fe(100) on fcc Ag(100). Above 4 ML, however, the LEED spots become very broad. The thickness of the films is determined with a calibrated quartz oscillator and with calibrated Auger electron signals.[19]

After preparation and characterization, uncoated and Tb-coated surfaces of Fe(100)/Ag(100) films are studied *in situ* at 6×10^{-11} mbar. Electron capture spectroscopy is used to measure the long-ranged surface ferromagnetic order, the films being magnetized along a [011] direction in magnetic fields up to 900 Oe, and the temperature of the samples being kept constant within 0.05 °C.

III. RESULTS AND DISCUSSION

Results of the ESP measurements for 1–10 ML thick bcc Fe(100) films epitaxially deposited on fcc Ag(100) substrates are shown in Table I. For temperatures ranging between 173 and 303 K, we find that nonzero in-plane remanent ferromagnetic order exists at the surface of all films. The applied mangetizing field was varied between 0 and 600 Oe, and a marked magnetic field dependence of the ESP was only found for Fe films consisting of 1 ML. Films with thickness D between 2 and 10 ML where already saturated in fields of a few Oe, whereas 1-ML thin films reached saturation only at ~300 Oe (see Table II).

It is interesting to note that all ESP data, obtained at film surfaces consisting of one single domain (magnetized to saturation) and measured at low reduced temperatures $t < 0.5$ $[t = T/T_c(D)]$, amount to about $(13 \pm 2)\%$. This directly indicates that for Fe(100)/Ag(100) films, the magnetic order at the topmost surface layer is hardly affected by a change in the Fe film thickness. This is in striking contrast to our ECS experiments with Ni(100) films on Cu(100) substrates,[18] where a strong dependence of the surface ESP on Ni film thickness, caused by the overlap of Ni and Cu d-

TABLE I. Electron-spin polarization $P(\%)$ at surfaces of ultrathin Fe(100)$p(1\times1)$/Ag(100); applied magnetic field $H = 0$–600 Oe.

Film thickness (ML)	Electron-spin polarization P (%)	Magnetic field dep.	Temperature (K)
1	+ (5–13) ± 1 (see Table II)	Yes	173–285
2	+ 14 ± 1	No	173
2	+ 13 ± 1	No	203
3	+ 15 ± 1	No	203
10	+ 12 ± 1	No	303

electron bands, is found. This directly implies the absence of band overlap for the Fe/Ag system and makes this system ideal to study intrinsic surface magnetism. Our present ESP data, obtained at surfaces of ultrathin Fe(100) films for $t < 0.5$, even equal the ESP of $(14 \pm 2)\%$ measured in plane at surfaces of bulk Fe(100).

Table II gives the magnetic field dependence of the ESP at surfaces of 1-ML thin Fe(100)/Ag(100) films. At $T = 173$ K, we observe a strong dependence of the ESP on the applied magnetic field H. At $H = 600$ Oe, the saturation ESP amounts to + 13%, remains constant until $H = 300$ Oe, and decreases down to a remanent ESP of 5% for $H = 0$ Oe. This behavior of the ESP, measured along the [011] direction in the (100) plane, can be explained by the existence of a magnetocrystalline anisotropy existing along [001] or [010] directions of easy surface magnetization being at 45° to the direction of the applied magnetic field. For $H = 0$, the remanent ESP along a [001] easy direction then amounts to $5\sqrt{2} = 7\%$. From an extrapolated plot of P vs H, we obtain a coersive field H_c of ~30 Oe which is much smaller than the field of 300 Oe needed to turn the ESP from the [001] directions in the [001] direction.

Increasing the temperature up to 285 K, we obtain a strong decrease of the ESP from 13% to 3% for $H = 600$ Oe. Reducing the field to zero, the ESP becomes zero (see Table II). For $T \geqslant 290$ K, the ESP remains zero at all applied fields which indicates that the Curie temperature $T_C(1)$ for a 1-ML thick Fe(100) film is located at ~290 K. This is in contrast to the ESP measured at 303 K at the surface of a 10-ML-thick Fe(100) film which still amounts to + 12% and reveals that $T_C(10)$ is far above room temperature.

TABLE II. Electron-spin polarization $P(\%)$ at surfaces of ultrathin Fe(100)$p(1\times1)$/Ag(100); applied magnetizing field $H = 0$–600 Oe.

Film thickness (ML)	Electron spin polarization P (%)	Magnetic field H (Oe)	Temperature (K)
1	+ 13 ± 1	600	173
1	+ 13 ± 1	300	173
1	+ 12 ± 1	150	173
1	+ 8 ± 1	30	173
1	+ 5 ± 1	0	173
1	+ 3 ± 1	600	285
1	+ 2.5 ± 1	450	285
1	+ 2 ± 1	300	285
1	+ 1.5 ± 1	150	285
1	0 ± 1	0	285

TABLE III. Electron-spin polarization $P(\%)$ at surfaces of ultrathin Tb films on 3-ML Fe(100) on Ag(100); applied magnetizing field $H = 0$–900 Oe.

Tb film thickness (ML)	Electron spin polarization P (%)	Magnetic field H (Oe)	Temperature (K)
2	0 ± 1	0	137
2	0 ± 1	150	137
2	0 ± 1	600	137
2	0 ± 1	900	137
2	0 ± 1	0	213
2	0 ± 1	300	213
2	+ 8.8 ± 1	600	213
2	+ 9.2 ± 1	750	213
2	+ 8.5 ± 1	300	303
2	+ 8 ± 1	300	323
2	+ 6.8 ± 1	300	333
2	+ 5 ± 1	300	348
2	0 ± 1	300	373

Increasing the film thickness from 1 to 2 ML (see Table I), we find that for $T = 173$ K, the strong magnetic field dependence of the ESP has disappeared and the surface is already saturated in fields of a few Oe. This directly implies the existence of a strong thickness dependence of the in-plane magnetic anisotropy. Increasing the thickness further from 2 to 3 and 10 ML causes no more change in the surface ESP of the Fe(100) films revealing that, for the investigated temperature range 173–303 K, the surface magnetization is always in plane independent of film thickness.

In a further ECS experiment, we have coated a 3-ML-thick Fe(100)/Ag(100) film with 2-ML Tb and find a behavior of the surface ESP, now measured at the surface of the Tb layers, that contrasts strikingly with that of the uncoated Fe surfaces. For $T = 137$ K, the ESP, measured in the surface plane, remains zero for all applied fields (up to 900 Oe) in accordance with the existence of a strong magnetic anisotropy perpendicular to the surface plane (see Table III). Increasing the temperature up to 213 K, we find, for applied magnetic fields larger than 600 Oe, an ESP of ~9%. This is in agreement with the existence of a strongly temperature-dependent out-of-plane magnetic anisotropy which, for instance, at 137 K, possibly cannot be overcome by magnetic fields of several kOe. Next, we keep the applied magnetic field at 300 Oe and increase T from 303 to 373 K. This causes the ESP to decrease from 8.5% at 303 K to 0% at 373 K indicating that the surface Curie temperature T_{Cs} of the Tb (2 ML) coated Fe/Ag films is at ~373 K.

Recently, we have determined T_{Cs} of a 50-Å-thick Tb(0001) film deposited on W(110).[21] We find $T_{Cs} = 250$ K which is far below 373 K. This directly reveals the influence of the underlying Fe layers on the Curie temperature of the topmost Tb layer.

We note that the Tb/Fe/Ag system provides a simple model system where large, strongly temperature-dependent perpendicular magnetic anisotropy can be easily produced. It is obvious that the surface Curie temperature can be shifted to higher or lower temperatures by varying the thickness of the Fe or Tb layers. We note that multilayered films composed of Fe films with periodically interlayered Tb monolayers might have interesting technological applications. From the results of our experiments, it is evident that the Tb/Fe/Ag system provides a simple most promising test case for future theoretical calculations where temperature-, layer-, and direction-dependent magnetic surface anisotropies are studied.

In conclusion, we remark that more refined theoretical studies and experimental work—using various spectroscopies possessing different probing depths—are needed to fully understand this most interesting area of surface and interface magnetism.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation, the Robert A. Welch Foundation, and by a Joseph H. DeFrees Grant of Research Corporation.

[1]L. Néel, C. R. Acad. Sci. **237**, 1468 (1953).
[2]B. R. Cooper and A. J. Bennett, Phys. Rev. B **12**, 4654 (1970).
[3]H. Takayama, K. P. Bohnen, and P. Fulde, Phys. Rev. B **14**, 2287 (1976).
[4]J. G. Gay and R. Richter, Phys. Rev. Lett. **56**, 2728 (1986); J. Appl. Phys. **61**, 3362 (1987).
[5]L. Fritsche and J. Noffke (to be published).
[6]U. Gradmann, J. Magn. Magn. Mater. **54/57**, 733 (1986); U. Gradmann, J. Korecki, and G. Waller, Appl. Phys. A **39**, 101 (1986).
[7]G. A. Prinz, G. T. Rado, and J. J. Krebs, J. Appl. Phys. **53**, 2087 (1982).
[8]B. T. Jonker, K. H. Walker, E. Kisker, G. A. Prinz, and C. Carbone, Phys. Rev. Lett. **57**, 142 (1986).
[9]M. Stampanoni, A. Vaterlaus, M. Aeschlimann, and F. Meier, Phys. Rev. Lett. **59**, 2483 (1987).
[10]J. Araya-Pochet, C. A. Ballentine, and J. L. Erskine (to be published).
[11]B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, Phys. Rev. Lett. **59**, 1756 (1987).
[12]N. C. Koon, B. T. Jonker, F. A. Volkening, J. J. Krebs, and G. A. Prinz, Phys. Rev. Lett. **59**, 2463 (1987).
[13]C. L. Fu, A. J. Freeman, and T. Oguchi, Phys. Rev. Lett. **54**, 2700 (1985); C. L. Fu and A. J. Freeman, J. Magn. Magn. Mater. **54/57**, 777 (1986).
[14]R. Richter, J. G. Gay, and J. R. Smith, Phys. Rev. Lett. **54**, 2704 (1985).
[15]C. Rau, J. Magn. Magn. Mater. **30**, 141 (1982).
[16]C. Rau and S. Eichner, Phys. Rev. Lett. **47**, 939 (1981).
[17]G. C. Smith, H. A. Padmore, and C. Norris, Surf. Sci. **229**, L278 (1987).
[18]C. Rau and G. Eckl, in *Nuclear Physics Methods in Materials Research*, edited by K. Bethge, H. Baumann, H. Jex, and F. Ranch (Vieweg, Braunschweig, 1980), p. 360.
[19]C. Rau, C. Schneider, G. Xing, and K. Jamison, Phys. Rev. Lett. **57**, 3221 (1986).
[20]C. Rau, G. Xing, and M. Robert, J. Vac. Sci. Technol. A **6**, 579 (1988).
[21]C. Rau, C. Jin, and M. Robert, Phys. Rev. Lett. (submitted).