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Direct observation of enhanced magnetic moments in Fe/Ag(100)

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The magnetic properties of ultrathin (1–5 monolayer) Fe films on Ag(100) substrates were investigated using SQUID magnetometry. Films were grown in pairs (one bulklike, the other thin) and characterized *in situ* by low-energy electron diffraction, Auger spectroscopy, and the surface magneto-optic Kerr effect. The films were then capped with Au and studied with a SQUID magnetometer over the temperature range 2–340 K. We report here a direct observation of enhanced magnetic moments for Fe on Ag(100), with interface moments enhanced as much as 29%.

Magnetic thin films have captured the interest of researchers due to the unusual behavior of such quasi-two-dimensional systems. The broken symmetry of the interfaces results in characteristic magnetic properties: predicted moment enhancements,[1,2] magnetic surface-anisotropy effects,[3,4] unusual spin-excitation spectra,[5] and reduced Curie temperatures.[4,6] Epitaxial Fe(100) on Ag(100) has been the subject of intense experimental study[7–9] due to the unstrained nature of the system and to extensive theoretical calculations[1,2] which predict an enhanced magnetic moment for one or two monolayers (ML) of Fe on Ag. Numerous experimental methods have been used to probe magnetic thin films, among them: spin- and angle-resolved photoemission,[3] ferromagnetic resonance,[10] and the surface magneto-optic Kerr effect (SMOKE);[4,11] these techniques only provide relative intensities, not absolute values of the magnetization. Recently, indirect evidence of ~14% moment enhancement for Fe(110) on W(110) was provided by extrapolating torsion magnetometer measurements[12] to low temperatures under the assumption of a common temperature dependence of the moment and the hyperfine field. Also, polarized neutron reflection[13] has provided evidence of moment enhancement, but such measurements require beam and background corrections, calculated asymmetries, and, as with all attempts,[14] very accurate film thickness determinations. We report here a *direct* observation of enhanced moments in ultrathin (1–5 ML) Fe on Ag(100) substrates through the use of SQUID magnetometry. We observe large moment enhancements (~30% above the bulk value) and provide layer-dependent evidence that the enhancement predominantly occurs in the interface layers.

The Fe/Ag(100) system presents a very attractive approximation of a two-dimensional system. The lattice constants of bcc Fe and fcc Ag differ by $\sim 2^{1/2}$; thus, a 45° relative orientation of the (100) surfaces results in almost perfect registry (0.8% mismatch). Since this is an unstrained system, theoretical calculations[1,2] are considered reliable. Furthermore, the overlayer and substrate bands have very little interaction; the only hybridization is through the Ag *sp* bands, and is nearly negligible. Finally, studies have shown that Fe grows in nearly layer-by-layer growth on Ag.[3,7]

Ag(100) substrates were cut from a single crystal (100) Ag boule and cleaned. Mechanical polishing was followed by electropolishing to remove surface impurities which may have been driven into the first few layers by the mechanical polish. Conventional Ne sputtering and annealing resulted in well-defined Ag(100) surfaces. Epitaxial layers of Fe were grown by electron beam evaporation from the tip of a 1.5-mm high purity Fe wire under UHV conditions (1×10^{-11} Torr), at a rate of 0.5 Å/min. Temperatures were held between 300 and 350 K to prevent diffusion of Fe into the substrate.

The samples were created in pairs, one bulklike (~ 25 ML), the other thin (1–5 ML), with a chosen ratio of growth time. Film growth was monitored using Auger spectroscopy and a calibrated quartz thickness monitor located at $\sim \frac{1}{3}$ the source-to-sample distance; however, the *absolute accuracy of the thickness monitor is unimportant.* Only the relative thicknesses of the two films are required for the moment calculations. The films were then capped with ~60 Å of Au to prevent atmospheric degradation upon transfer to the SQUID chamber. Auger analysis was used to monitor contamination (< 1%), and low-energy electron diffraction (LEED) data were used to study surface morphology before capping. On some films, SMOKE data were taken before and after capping with Au to verify that no measurable change in moment occurred due to capping. SMOKE data indicated that films thinner than about 2.5 ML exhibit perpendicular anisotropy; for our SQUID geometry, only in-plane moment measurements were practical, so no data for thicknesses $\leq$2.5 ML are reported here.

Hysteresis loops (magnetic moment versus field) were obtained using a Quantum Design MPMS SQUID magnetometer. The samples, cooled with He vapor, were mounted such that the applied field was parallel to the plane of the film and along the Ag(001) [Fe(011)] direction. Measurements were taken at high fields (±3500–5000 Oe) in order to determine the saturation moment of the films. The field was then stepped down from 2000 to 0 Oe to generate the first segment of a hysteresis loop and to measure the spontaneous moment. Complete loops were also taken to assess the overall shape of the hysteresis curve. Data were obtained at temperatures ranging from 2 to 340 K.

The Ag substrates provide a diamagnetic susceptibility which can be determined with a linear fit of the high

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field saturation region, as is evident in Fig. 1(a). Such a contribution was subtracted from the data to obtain the magnetic moment due to the Fe alone; examples are shown in Figs. 1(b) and 2. Point-by-point subtractions of data from bare substrates yielded the same results.

Figure 3 is a direct comparison between the saturation moments for a 4.4-ML film and its corresponding thick partner (23.5 ML). The data for the thick film have been scaled by the ratio of the number of atoms in the films (3:16); thus, the moment enhancement is readily evident. It is also obvious that, due to the nature of spin excitations in ultrathin films, low temperatures are *essential* for fully saturating the thin films in reasonable (~5000 Oe) magnetic fields.

The thin film moments are calculated using the saturation data and the relative thicknesses of the two films. Note that only the linearity of the quartz thickness monitor is important—the relative masking times determine the relative thicknesses. The number of atoms on the thick film may be well estimated from the thick film saturation moment and the bulk Fe moment, $\mu_{\text{bulk}} = 2.22\mu_B$ (μ_B is the Bohr magneton); this information and the area of the Ag substrate also provides an accurate determination of the number of layers. One may then calculate the number of atoms and number of layers present on the thin film using the ratio of growth times, or, equivalently, the thin film monitor ratios.

The moment per atom of the thin film is calculated by setting up a ratio of saturation moments, M_s:

$$M_{s,\text{thick}} = N_1 n_1 \mu_B, \quad M_{s,\text{thin}} = N_2 n_2 \mu_B \ , \qquad (1)$$

where n_1 and n_2 are the average number of Bohr magnetons per atom, and N_1 and N_2 are the number of atoms in the thick (1) and thin (2) films, respectively. The thin film moment per atom then depends only on the ratios of saturation moments and numbers of atoms. The average moment per atom deduced for a 2.9-ML film is $2.67\pm0.06\mu_B$, a 20% enhancement above the bulk value of $2.22\mu_B$. Figure 4 shows the average enhanced moment as a function of the number of monolayers, as obtained from three different sets of films. The slab calculations of Ohnishi, Freeman, and Weinert[15] have shown that the moment enhancement occurs predominantly in the interface layers of the films. Assuming that the moments in the interface layers of the *thick* films are also

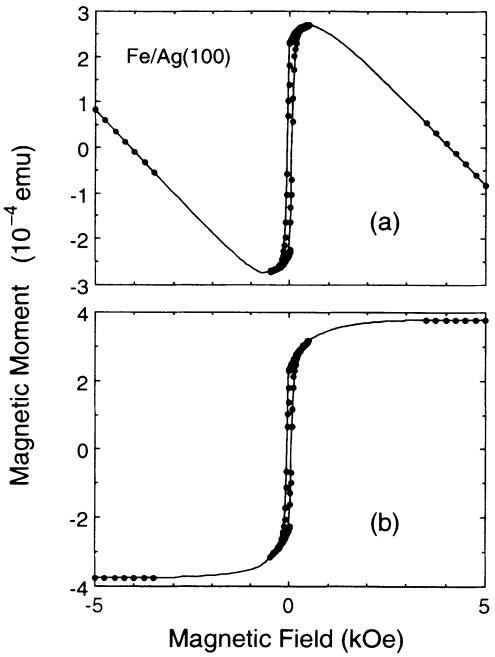

FIG. 1. Magnetic moment as a function of magnetic field for a 29-ML film of Fe(100) on Ag(100). (a) Raw moment data with contributions from both Fe film and Ag substrate. (b) Data for film alone after subtraction of the diamagnetic contribution from the substrate.

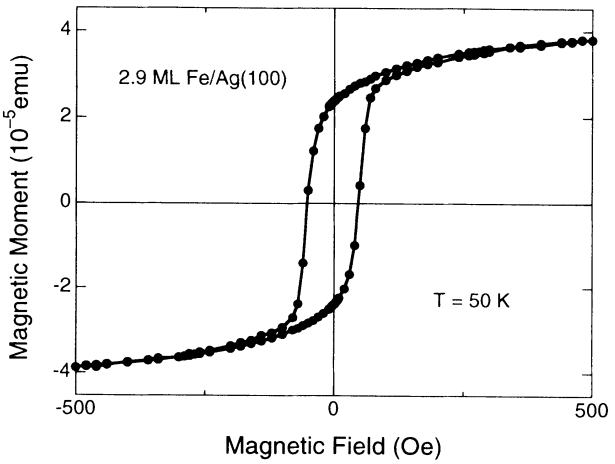

FIG. 2. Magnetic moment as a function of magnetic field for a 2.9-ML film of Fe(100) on Ag(100). Data have been corrected for the substrate contribution. Curvature is due both to in-plane anisotropy [rotation of the moment from the Fe(001) easy axis to the Fe(011) direction with increasing field] and to damping of spin excitations by the external field.

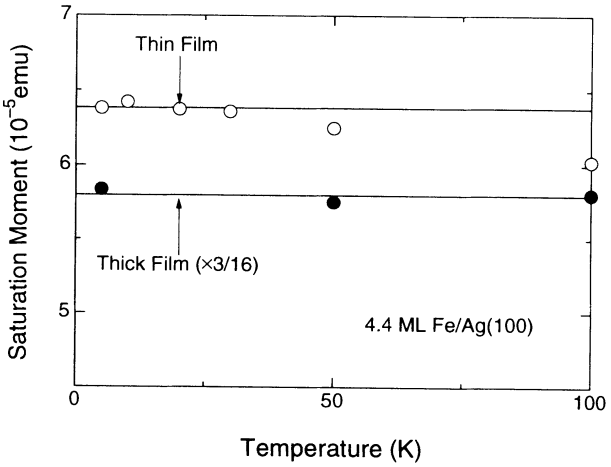

FIG. 3. Saturation moment as a function of temperature for a thin film (4.4 ML, open symbols) of Fe(100) on Ag(100) compared to that of a thick film (23.5 ML, solid symbols). The thick film data have been scaled by the ratio of the deposition times (number of atoms) in the films, so that the moment enhancement of the thin film is apparent. Note that the thin film moment is fully saturated only below 50 K for the fields used (3500–5000 Oe).

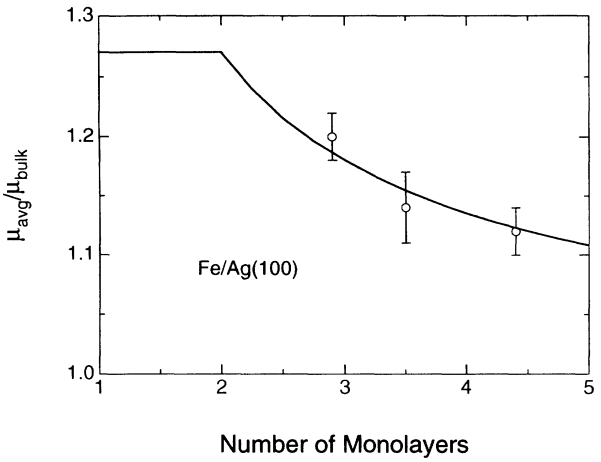

FIG. 4. Ratio of the average moment per atom to the bulk value ($2.22\mu_B$) as a function of film thickness (for 2.9, 3.5, and 4.4 ML films) as determined from three sets of Fe(100)/Ag(100) films. The solid line is a fit which assumes that all of the moment enhancement occurs in the interface layers.

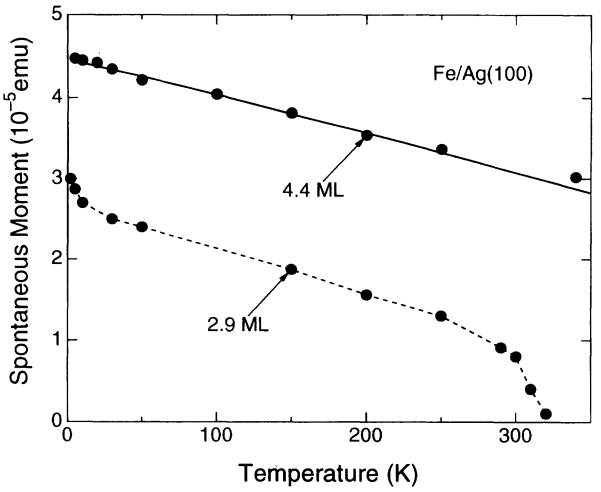

FIG. 5. Spontaneous moment as a function of temperature for two Fe(100)/Ag(100) films. The 4.4-ML film exhibits quasilinear behavior; the solid line is a fit to conventional thin-film spin-wave theory.

enhanced, a slight correction to the presumed bulk value for the thick film (e.g., an average of $2.25\mu_B$/atom instead of $2.22\mu_B$/atom) can be determined, resulting in an $\sim$ 2% correction to the thin film absolute moment per atom. For the 2.9-ML film, the approximation of interface-only enhancement implies that the $2.67\mu_B$ average moment corresponds to $2.87\mu_B$ interface moments, a 29% enhancement over the bulk value. The solid line in Fig. 4 merely assumes a constant interface moment enhancement, and that all of the enhancement occurs in the interface layers. From the fit we obtain an interface moment enhancement of 27%.

We have also measured the spontaneous moment $M(T)$ of the thin films to determine the behavior of the order parameter with temperature. Due to spin-wave excitations, bulk materials exhibit the well-known Bloch $T^{3/2}$ law at low temperatures, $M(T) = M(0)(1 - AT^{3/2})$. For thin films, normal modes perpendicular to the surface satisfy complicated boundary conditions. For a small number of layers p and $t \to 0$, one expects[16,17]

$$\frac{M(T)}{M(0)} = 1 - \frac{k_B T}{2\pi p D} \ln\left(\frac{k_B T}{\Delta}\right) , \qquad (2)$$

where D is the spin-wave stiffness and Δ is the energy gap induced by anisotropy. This is the origin of the oft-quoted linear dependence of $M(T)$ for thin films. Figure 5 compares the spontaneous moments of two typical films as a function of temperature. Linear behavior is especially evident in the 4.4-ML film throughout the temperature range; the solid curve for that film is a fit to Eq. (2) with $D = 30$ meV and $\Delta = 0.015$ K. This value of D is comparable to the bulk value, while the extracted Δ value may be compared with what one would expect if the spin-wave gap is attributed to the in-plane effective fourfold anisotropy field, $H_{\|}^{\text{eff}}$.[18] Extrapolating the data of Ref. 9, one has $H_{\|}^{\text{eff}} \approx 20$ Oe for 4.4 ML; here, one can estimate $H_{\|}^{\text{eff}} = \Delta/(da_0^2 M_s) = 19$ Oe, quite good agreement. The nonlinear behavior of the thinner film in Fig. 5 at very low temperatures is probably related to local excitations; the behavior correlates with surface morphology, since the terrace widths estimated from LEED are smaller for the 2.9-ML film ($\sim$60 Å) than for the 4.4-ML film ($\geq$ 100 Å, resolution limited). Perturbations to an ideal ferromagnetic spin-wave dispersion ($\hbar\omega = Da_0^2 k^2$) caused by surface topography with a typical length scale L would be expected to be most evident at low temperatures, i.e., for $k_B T \leq Da_0^2(\pi/L)^2$. Using the bulk value $D = 34.1$ meV and the LEED terrace width $L \approx 60$ Å, one obtains a crossover temperature $T \leq 9$ K, close to the temperature of the observed upturn in the spontaneous magnetization. The upturn and corresponding $M(T \to 0)$ value for the 2.9-ML film is in accord, however, with the saturation moment measurements. Such behavior highlights the need for further low-temperature measurements to understand the relation between surface topography and spin excitations in such nearly two-dimensional systems.

In summary, we have reported direct measurements of enhanced magnetic moments in ultrathin Fe films on Ag(100) substrates. The need for an accurate thickness measurement of a thin film was avoided by using a relative technique; the thin film moment calculation depends only on the ratio of saturation moments and the linearity (not accuracy) of the quartz oscillator microbalance. For the interface moments, a nearly 30% enhancement above the bulk value was determined. A correlation between surface structure and spin excitations was also noted.

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