Magnetic properties of ultrathin fcc Fe(111)/Ru(0001) films

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In situ polar and longitudinal Kerr-effect measurements have been used in conjunction with low-energy electron diffraction and Auger characterizations to study the magnetic properties of Fe(111)/Ru(0001). Ru(0001) provides the opportunity to grow fcc Fe with a dilated lattice spacing that is expected to be ferromagnetic. Ferromagnetism is detected, but only for films thicker than ~ 2 monolayers. The easy axis of magnetization is perpendicular for films <4.5 monolayers thick and becomes in plane for thicker films. The dead layers at the interface are attributed to *d*-band hybridization between the Fe and Ru.

The magnetic properties of ultrathin ferromagnetic films have attracted much attention recently because the remarkable predictions of perpendicular surface magnetic anisotropy[1] were closely followed by striking examples confirming its existence in a number of systems that consist of ferromagnetic 3*d* transition metals and Group Ib elements in epitaxial film, sandwich, and superlattice configurations.[2-11] These dramatic advances notwithstanding, numerous fundamental questions persist. The original predictions were directed to monolayer phenomena, while experimentally vertical easy axes have been found to persist well into the multilayer regime, especially[7-9] for fcc-Fe/Cu(100). It has been suggested that the vertical spin orientation is easy because of the unusual structural characteristics of the surface or interface.[8] For fcc-Fe/Cu(100) an extraordinary surface-layer expansion has been confirmed by three independent quantitative low-energy electron diffraction (LEED) investigations.[12] It has also been suggested that strain-layer epitaxy can induce a vertical easy axis. However, there is no difficulty in reconciling the persistance of vertical easy axes into the multilayer regime with the spin-orbit interaction at the surface.[13] In the light of these controversial issues it is of interest to further explore the origins of the vertical surface magnetic anisotropy.

In the present work Ru was chosen as a substrate because it has been suggested that transition-metal substrates with high surface free energy[14] σ might be superior candidates for achieving interfacial structures free of segregation[15] and intermixing,[16] and $\sigma_{Fe} < \sigma_{Ru}$, while $\sigma_{Fe} > \sigma_{Cu,Ag,Au}$. Basal plane Ru(0001) serves as a template for the growth of fcc-Fe(111) planes with the dilated interplanar lattice constant of 2.70 Å, compared to 2.55-Å epitaxy on Cu. The enlarged spacing should stabilize the ferromagnetic state of fcc Fe, which according to theoretical calculations can have either ferro- or antiferromagnetic ground states depending on volume.[17] Both states of fcc Fe have been grown by evaporation of Fe onto Cu(100) substrates. The ferromagnetic phase[7-9] is associated with the relatively discrete interface created by low-temperature growth conditions (≤ 300 K), while the antiferromagnetic phase[18] is created by high-temperature growth (~ 200° C) and consists of an intermixed Fe-Cu buffer layer separating the pure Fe and Cu regions. The preparation of antiferromagnetic fcc-Fe is also well documented by a different high-temperature method in the form of coherent precipitates in quench-condensed Cu-rich alloys.[19]

In the present study it is confirmed that Fe initially grows epitaxially on Ru(0001). The expected ferromagnetic state is obtained, but only for films thicker than ~ 2.5 and 3.0 monolayers (ML) for 100- and 300-K growth conditions, respectively. The magnetic dead layers at the interface are attributed to hybridization between Fe and Ru, as in dilute Fe alloys in a Ru host which do not possess a magnetic moment.[20] Vertical surface magnetic anisotropy is present and vertical easy axes are stabilized for Fe thicknesses ≤ 4.5 ML. However, the unusual surface-layer expansion[12] characteristic of fcc-Fe/Cu(100) is not expected to be present because the large in-plane expansion of fcc-Fe/Ru(0001) dictates a contracted (trigonally distorted) interlayer separation. This leaves the electronic origin of the anisotropy as a prime consideration for future theoretical evaluation.

The Fe films were grown by evaporation in 10^{-11}-Torr ultrahigh vacuum (UHV) and studied by *in situ* polar and longitudinal surface magneto-optic Kerr-effect (SMOKE) and LEED-Auger characterizations, as described previously.[9] The Ru(0001) substrate was mechanically polished to a 1-μ diamond-paste finish, followed by repeated Ar^+-ion sputtering and 700°C-annealing cycles until sharp LEED beams were observed. The only impurity found via Auger detection was $\leq 2\%$ oxygen. In the polar Kerr-effect configuration the applied magnetic field is perpendicular to the plane of the film, and in the longitudinal configuration the applied field is in plane. Thus, either the perpendicular or in-plane magnetization component is being probed in each respective case. A new near-infrared He-Ne laser (wavelength $\lambda = 1152$ nm) was added in the present study to check the Kerr-effect response at a different wavelength than that of the standard $\lambda = 632.8$-nm line. The signals from Fe/Ru(0001) are considerably weaker than expected compared to those measured for Fe/Cu(100), even after taking into account the different reflectivities of the two substrates.[21]

The film-growth mode was studied using Auger elec-

tron spectroscopy. Figure 1 shows Auger-intensity versus deposition-time curves for Fe grown on Ru(0001) at room temperature. The data could be satisfactorily fitted to exponential curves [Fig. 1(a)]; two breaks in both the Fe 645-eV and Ru 231-eV Auger signals can be readily identified [Fig. 1(b)]. The breaks indicate the completion of the first and second monolayers. Inelastic mean-free-path parameters deduced from the above fittings are 5.1 ± 0.5 Å for the Ru 231-eV and 11.8 ± 0.5 Å for the Fe 645-eV Auger electrons, which are close to the values obtained from the well-known universal curve[22] of 6.2 Å at 231 eV and 13.2 Å at 645 eV. These findings are indicative of a layer-by-layer growth mode.

LEED observations show clear but somewhat broader $p(1\times1)$ beams for the first two layers with some diffuse background compared to that of the substrate. These $p(1\times1)$ beams become weaker but are still visible in the background for film thicknesses of up to ~8 ML. This suggests that the first two layers of Fe are epitaxial and that disorder gradually develops. For the 100-K growth, the LEED beams were much broader, as expected. Our observations of the growth are similar to those reported for *Fe/Ru* $(10\bar{1}0)$ grown at room temperature.[23] Another report[24] of Fe/Ru(0001) grown at 520 K showed that the first layer followed the Ru(0001) structure but a new $(6\sqrt{3}\times6\sqrt{3})R30°$ superstructure developed for thicker films. No new diffraction beams were found at any coverage of Fe in the present study.

SMOKE measurements in both polar and longitudinal configurations were carried out immediately after Fe deposition. Results ($\lambda=632.8$ nm) for ultrathin Fe/Ru(0001) films grown and measured at room temperature are shown in Fig. 2 and compared with that for the Ru(0001) substrate. We did not detect any Kerr-effect hysteresis above 100 K for films less than 2-ML thick for both room-temperature and ~100-K growth under applied magnetic fields of ≤ 2 kOe. Measurements made at 1152 nm showed similar results.

The lack of a ferromagnetic signature for 1-ML and 2-ML Fe/Ru(0001) also appears in the thickness-dependent measurements. Figure 3 shows polar and longitudinal Kerr-intensity data plotted as the height of the hysteresis loop in the remanent state. This intensity is proportional to the Kerr rotation and to the sample magnetization. The films were grown at room temperature and measured there using a 632.8-nm He-Ne laser. The polar Kerr signal develops at a thickness of ~3.0 ML and increases linearly with thickness up to the critical thickness d_c where the easy axis reorients in plane. Straight-line interpolation of the polar signal to zero Kerr intensity shows the absence of remanent magnetization for films thinner than 2 ML. Data for films grown and measured at 100 K are consistent with this observation, as shown by the three solid-square symbols. Extrapolation of the longitudinal Kerr-effect data yields similar results. It should be emphasized that our SMOKE technique has the sensitivity to detect even submonolayer ferromagnetism.[9,25] This

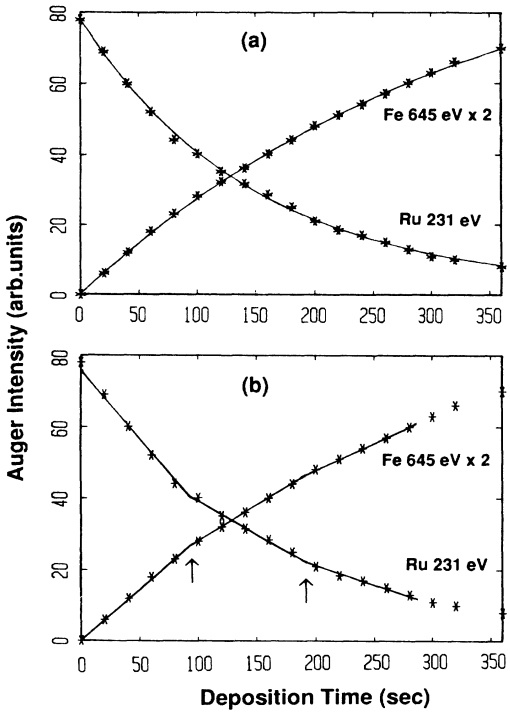

FIG. 1. Auger intensity vs deposition-time measurements for Fe grown on Ru(0001) at room temperature: (a) shows the exponential least-squares fit of the data, and (b) shows two breaks in each data set for the same data as in (a). The results are indicative of layer-by-layer growth.

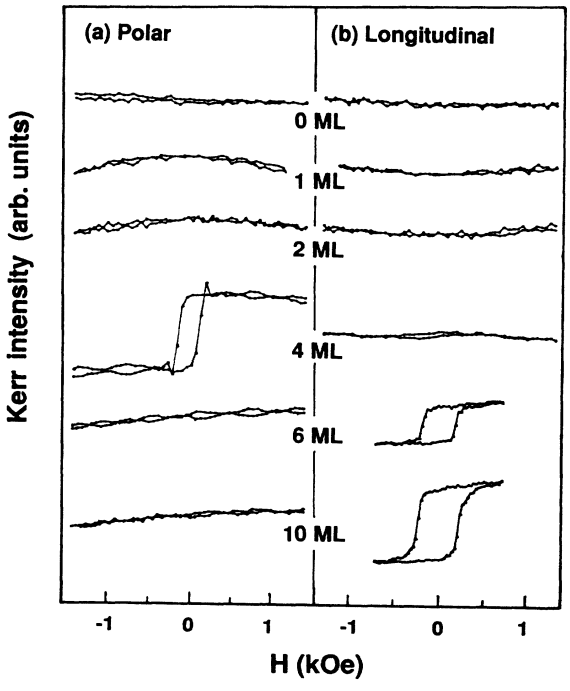

FIG. 2. Polar (a) and longitudinal (b) Kerr-effect signals for Ru(0001) substrate and ultrathin Fe/Ru(0001) films grown and measured at room temperature. The Fe thicknesses are indicated. No hysteresis is observed for films that are ≤ 2-ML thick.

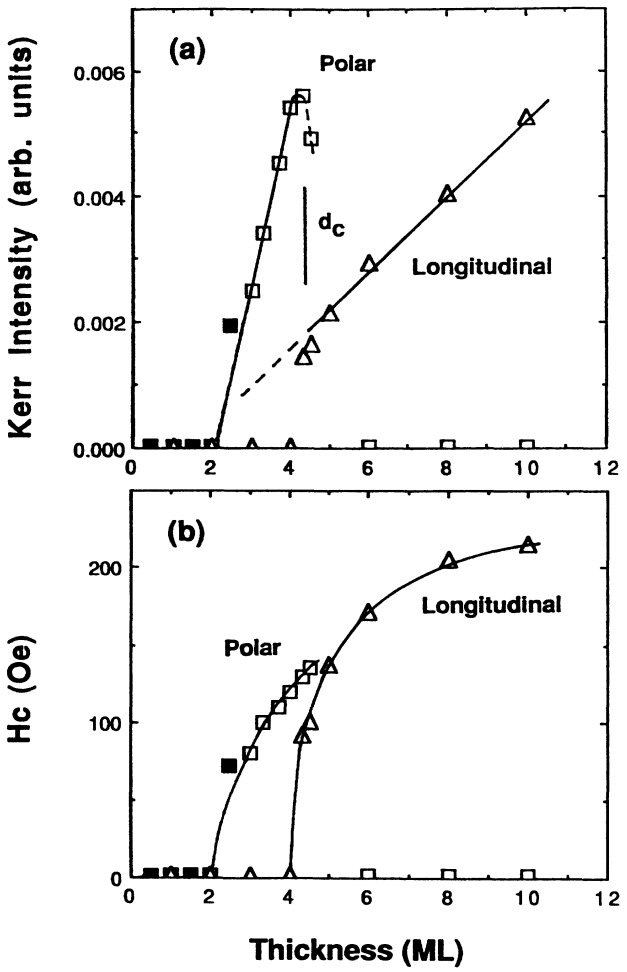

FIG. 3. Thickness dependence of the Kerr intensity (a) and coercivity (b) for Fe/Ru(0001) grown at room temperature (open symbols) and 100 K (solid symbols). Squares represent polar Kerr-effect signals and triangles represent longitudinal signals. The lines through the data are guides to the eye.

has been demonstrated again most recently in our study[26] of Fe/Pd(100), where Kerr-effect hysteresis loops have been obtained for ~0.4-ML Fe/Pd(100). It is not likely that the absence of hysteresis is attributable to high-coercivity values given the data trends shown in Fig. 3(b). Fe/Cu(100) (Ref. 9) and Fe/Pd(100) (Ref. 26) also exhibit modest coercivity values. It is also not likely that the lack of ferromagnetic signature is due to the formation of magnetic domains at the monolayer level, as has been recently investigated theoretically by Yafet and Gyorgy,[27] because a signal showing an unsaturated magnetization would still be expected to develop in a magnetic field. Thus, we conclude that there is no long-range ferromagnetic order for $\leq$2-ML Fe/Ru(0001) above 100 K.

Our LEED-Auger observations suggest that at least the first two layers of Fe/Ru(0001) grow epitaxially and follow the substrate lattice spacing. This implies that each of these layers corresponds to a (111) plane of dilated fcc iron with a lattice spacing of 2.70 Å. The enlarged lattice spacing should favor ferromagnetism according to the theoretical calculations.[17] However, both Fe and Ru have unfilled *d* bands; thus, the magnetic properties may be severely affected by *d*-band hybridization. It is well known that in dilute alloys Fe atoms lose their magnetic moment in a Ru host.[20] This fact and our SMOKE observations for Fe/Ru(0001) demonstrate that *d*-band hybridization can override the influence of the dilated lattice.

The Ru lattice spacing of 2.70 Å is considerably larger than that of fcc Fe (~2.54 Å). The large lattice mismatch (~6%) causes an elastic strain in Fe/Ru(0001). The elastic strain energy will build up with film thickness and gradually be relaxed. We estimate, using van der Merwe's model,[28] that for Fe/Ru(0001) an elastically stretched fcc-Fe film of ~2 ML can be obtained. This estimate is in good agreement with our LEED-Auger observations. Thicker films show increased LEED background and broadened $p(1\times1)$ beams, in contrast to the case of Fe/W(110), where the relaxation is reported as new superlattice structures.[29]

Since the *existence* of perpendicular surface anisotropy has been established in ultrathin films, the critical issue has become to identify the mechanism that causes it. Two have been proposed: one electronic and one geometric in origin. Gay and Richter[1] explain it as arising from the spin-orbit interaction at the surface. Although the numerical accuracy and the approximations invoked in Ref. 1 have been subjected to critical evaluation,[30] the electronic mechanism remains highly plausible, if not proven. Stampanoni *et al.*[8] suggest that for fcc-Fe/Cu(100) the persistance of the perpendicular anisotropy could be related to the anomalous lattice-constant variation along the film normal.[12] This distinguishes the vertical from the in-plane set of {100} directions. In the present case the vertical direction is already unique, and the [111] direction is a known easy axis for bulk fcc ferromagnets. However, a critical thickness for vertical easy axes still exists. Also, the large *intralayer expansion* associated with the Fe achieving epitaxy with Ru, should cause a substantial *interlayer contraction* rather than an expansion. Thus, the magnetic anisotropy behavior of Fe/Ru(0001) is not likely to be governed solely by its surface geometric properties.

In summary, fcc Fe films have been grown on Ru(0001) at room temperature and ~100 K. Direct, *in situ* SMOKE observations and thickness-dependent measurements indicate that there is no long-range ferromagnetic order for 1-ML and 2-ML Fe/Ru(0001) films above 100 K, and suggest the presence of magnetic dead layers. The observed polar Kerr-effect hysteresis and the development of longitudinal Kerr-effect signals for thicker films provide another documented case confirming the existence of perpendicular surface anisotropy in ultrathin films. The electronic and geometric structural origins of the anisotropy will undoubtedly be the topic of much work in the future.

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