Spin and angle-resolved photoemission studies of one to three monolayer films of $p(1 \times 1)$ Fe on W(100)

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The electronic and magnetic properties of thin (one to three monolayer) epitaxial Fe films grown on W(100) are studied using spin-polarized angle-resolved photoemission. Low-energy electron-diffraction studies of a number of samples confirm excellent epitaxy for monolayer films and suggest that two- and three-layer films begin to form strain-relieving dislocations. Submonolayer films manifest no in-plane remanent spin polarization at $T = 115$ K, but effectively quench W(100) surface states and surface resonances. Monolayer and thicker films yield spin-polarized photoelectrons under remanent magnetic conditions. Analysis of spin-polarized electron energy distribution curves (EDCs) corresponding to even-symmetry initial states along the $\bar{\Gamma}$-$\bar{M}$ direction of the two-dimensional Brillouin zone yields a flat majority-spin band at 0.95 eV and an exchange split pair of bands (ΔE_{ex} = 2.15 ± 0.2 eV at $\bar{\Gamma}$, 1.2±0.2 eV at $\bar{M}$) which exhibit significant dispersion. No significant changes in the binding energies of spin-up or -down features in EDCs obtained above the Curie temperature at $\bar{\Gamma}$ (normal emission geometry) were detected.

We report angle-resolved ultraviolet photoemission spectroscopy (ARUPS) studies, with and without spin detection, of one to three monolayer (ML) Fe films grown on W(100). This work was motivated by initial results of our magneto-optic Kerr effect studies[1] of the same system, which have revealed a number of novel magnetic properties, and by recent experimental[2] and theoretical[3] studies of a related system, Fe on W(110). The latter work has raised and addressed interesting issues pertaining to the role substrate electronic effects play in thin-film magnetism.

Recent spin-polarized inverse photoemission experiments[4] suggest that a 1-ML-thick Fe film on W(100) is magnetic, with zero-field remanent magnetization in the $\langle 11 \rangle$ plane. Film growth studies[5,6] have shown that despite the large (10%) lattice mismatch between bulk Fe and bulk W, thin Fe films grow pseudomorphically on W(100). Our low-energy electron-diffraction (LEED) studies suggest that monolayer films form a perfect two-dimensional net in registry with the W(100) surface. Thicker films show a decrease in the quality of LEED spots as evidence of dislocation formation to reduce strain. However, the epitaxy remains very good up to several ML; therefore, the Fe-on-W(100) system offers important opportunities for studying magnetic effects in a highly strained thin-film system.

The experimental[2] and theoretical[3] studies of Fe on W(110) referred to above have highlighted the effects of lattice strain and the effects of substrate-overlayer hybridization in affecting thin-film magnetic properties. One issue addressed by these studies is the opposing substrate-induced effects on the film magnetic moment resulting from (1) the increased lattice constant, and (2) hybridization between electronic states of the film and substrate. In the case of Fe on W(110), manifestations of these effects include a net increase in magnetic moment and a significant variation in substrate and thin-film electronic properties as a function of thickness in the 1–2-ML range. In the present case [Fe on W(100)], we have observed quenching of the W(100) surface states and surface resonances when as little as $\frac{1}{4}$ ML of Fe atoms is deposited. This effect represents a striking example of film-to-substrate electronic coupling.

Spin-resolved ARUPS experiments were carried out at the National Synchrotron Light Source (NSLS) U5U beamline.[7] The spin-resolved experiments were constrained to geometry which probed only even-symmetry initial states, at energy and angular resolution of 300 meV full width at half maximum (FWHM) and ±1.5°, respectively. Non-spin-polarized ARUPS experiments were conducted at U16A (also at NSLS) using both s-polarized (normal incidence) and p-polarized (beam incident angle of θ_{inc} = 50°) light. Photoelectron energy distribution curves (EDCs) were obtained using detection geometries which probed both odd- and even-symmetry initial states with energy and angle resolution of 140 meV FWHM and ±1.4° respectively.

The W(100) crystals were prepared using conventional techniques (x-ray Laue alignment, spark cutting, and repeated *in situ* annealing treatments in oxygen followed by flashing to 2500 K). Auger spectroscopy, surface-state photoemission signals[8] from W(100), and LEED observation of the reversible $c(2 \times 2)$ surface reconstruction at low temperatures were used as monitors of substrate cleanliness. Monolayer films for the spin-resolved experiments were grown at a substrate temperature of 1000 K; additional layers for thicker films were grown after the substrate was cooled to below 200 K. The thin-film sam-
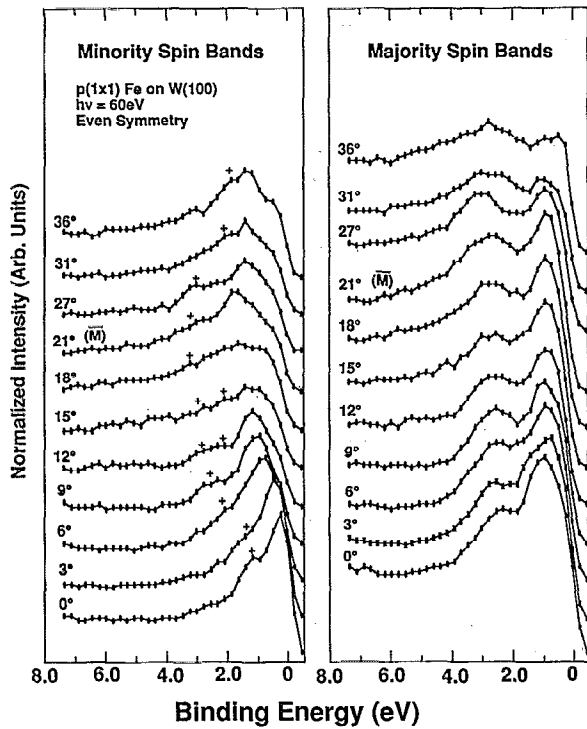

FIG. 1. Spin-resolved ARUPS spectra of 2 ML of Fe/W(100) with an angle of incidence of 35° and 60-eV photons. Spectra labeled by angle of emission from normal in even symmetry. Crosses mark contributions from the clean W(100) substrate.

FIG. 2. (a) Even-state bands: ▲ = majority, ▽ = minority, 2-ML Fe, $hv = 60$ eV, $\theta_{inc} = 35°$, data from spectra plotted in Fig. 1; ● = 1-ML Fe, $hv = 40$ eV, $\theta_{inc} = 50°$; ○ = 1.3-ML Fe, $hv = 40$ eV, $\theta_{inc} = 0°$; □ = 2.2-ML Fe, $hv = 60$ eV, $\theta_{inc} = 23°$; × = 1.0-ML Fe, $hv = 25$ eV, $\theta_{inc} = 0°$. (b) Odd-state bands ($\theta_{inc} = 0°$): ● = 1-ML Fe, $hv = 25$ eV; ■ = 1-ML Fe, $hv = 60$ eV; ◇ = 2.5-ML Fe, $hv = 21$ eV; ◆ = 1 ML-Fe, $hv = 25$ eV; □ = 2.0-ML Fe, $hv = 15$ eV. Shaded regions indicate the projection of W(100) bulk states (see Ref. 11).

ples were held at 115 K and were magnetized to a remanent state by application of a current pulse to a coil near the sample. Non-spin-polarized experiments were carried out at 300 K substrate temperature and with no efforts to create a single magnetic domain. Temperature-dependent studies of the spin-polarized bands at $\overline{\Gamma}$ of the two-dimensional Brillouin zone failed to detect any significant changes in initial-state binding energies.[9] As will be shown in following figures, excellent agreement was obtained from experiments conducted on the two beamlines where the parameters were equivalent.

Figure 1 displays spin-resolved EDCs corresponding to a 2-ML Fe film on W(100). The spectra cover a range of parallel momentum $k_{\parallel}$ extending from $\overline{\Gamma}$ to values significantly beyond the zone edge at $\overline{M}$. Systematic variation versus emission angle of several features can be recognized, which indicates dispersion of initial-state energies versus $k_{\parallel}$. Several of these features were also produced in EDCs of the clean W(100) surface and are denoted by crosses. Figure 2 displays peak binding energies versus parallel momentum for even and odd states. Spin-resolved data from Fig. 1 are plotted as open triangles (minority spin) and solid triangles (majority spin); non-spin-polarized data from dozens of EDCs obtained from films of different thickness in the 1–3-ML range are also plotted.

At normal emission, the majority-spin state at 0.85 eV exhibits p-polarization sensitivity, and a pair of exchange-split states at 0.30 eV (minority) and 2.45 eV (majority) exhibit s-polarization sensitivity. The exchange splitting of
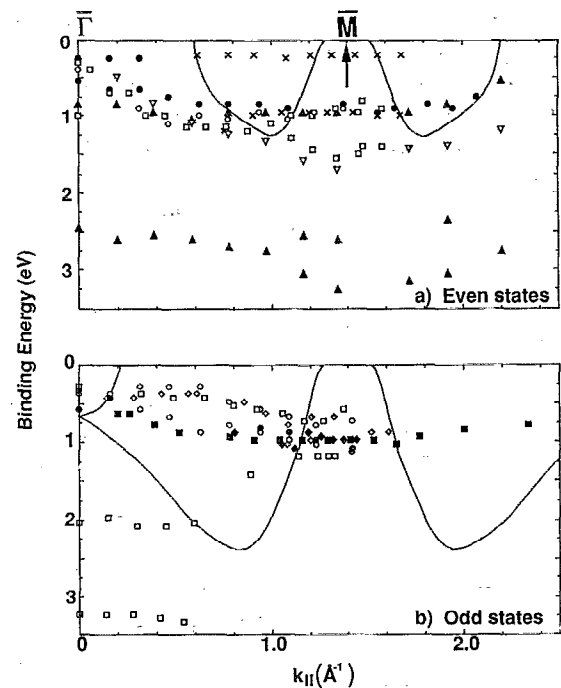
2.15 ± 0.2 eV compares very well with the measured splitting of 2.2 ± 0.3 eV of the Γ'_{25} states of bulk Fe(100).[10] Note that the minority-spin feature broadens considerably and shifts to higher binding energy as it disperses towards $\overline{M}$ (which occurs at 22° for $hv = 60$ eV). At the zone edge, the minority-spin state and higher-lying majority-spin-state peaks become more sensitive to p-polarized light and the lower-lying majority-spin state becomes more sensitive to s-polarized light, assignments opposite those made at the zone center. The slight dispersion of the broad majority-spin feature to 3.1 eV near $\overline{M}$ could result from contributions from the W substrate.

Contrary to the behavior[2] of Fe on W(110), the normal emission spectra show little change of character in either the majority- or minority-spin states for films between 1 and 3 ML thick. States at the $\overline{M}$ zone edge, however, do exhibit a strong thickness dependence. Smooth and relatively broad features that characterize the minority-spin spectra for 1-ML films sharpen and shift by ~0.50 eV to lower binding energy with increasing thickness. Majority-spin-state spectra, which are sharp and well developed even for the 1-ML film, are stable to within ~0.20 eV with increasing thickness.

Non-spin-polarized spectra of monolayer films show a prominent band at the zone edge at an initial state energy of 0.95 ± 0.10 eV, correlating well with the majority-spin band from the spin-resolved data. This band was not ob-

served to shift with changing photon energy. An additional feature having binding energy of 0.25 eV throughout the zone along $\bar{\Gamma}$-$\bar{M}$ was seen with $h\nu = 25$ eV for 1-ML films, but did not appear in EDCs for thicker films. Even-symmetry states mapped out using s-polarized light track the spin-resolved states of the same symmetry. The emergence of a second band at $\bar{M}$ in the spin-integrated EDCs for thicker films is consistent with the development of a sharp minority-spin feature that appears in the spin-polarized spectra. Excellent agreement is found between the data acquired under similar conditions on the two beamlines.

Odd-state bands for 1.0-ML (solid circles) and ~2.2-ML films (open circles) are mapped out in Fig. 2(b). Note that the binding energy (1.05 ± 0.10 eV, independent of photon energy) of the shallow state at $\bar{M}$ for 1-ML films and the emergence of a second band for thicker films is very similar to the behavior of the even-symmetry states at this high-symmetry point. Again, a broad band of states at ~2.6 eV stretches across the zone, although normal emission spectra taken with 15-eV photons partially resolve this feature into two bands at 2.1 ± 0.10 and 3.2 ± 0.10 eV binding energy.

The surface projection of the W(100) bulk states[11] (shaded region) for both even and odd states are also shown in Fig. 2. The issue concerning the effects of hybridization between substrate and overlayer becomes very interesting in this particular case since the even-symmetry Fe states lie mostly outside the gap of projected bulk W states, while the odd-symmetry Fe states lie mostly within the gap, except at the zone boundaries. Since the shallow majority-spin state at ~0.95 eV cuts through the even-symmetry gap without a significant change in character, it presumably is not hybridized strongly with the tungsten states outside the gap. On the other hand, the minority-

spin states appear to remain mostly outside the gap and exhibit the greatest dependence on film thickness.

In summary, we report a preliminary experimental study of the spin-resolved band structure of 1–3-ML Fe films on W(100), noting the strong thickness dependence of the minority-spin features at the $\bar{M}$ zone edge and possible effects of substrate hybridization.

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