

Surface-Step-Induced Magnetic Anisotropy in Thin Epitaxial Fe Films on W(001)

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Ultrathin epitaxial Fe films grown on W(001) surfaces prepared to yield uniform 25-Å-wide terraces are shown to exhibit layer-dependent in-plane uniaxial magnetic anisotropy having an easy axis perpendicular to the steps. Hysteresis loops measured by magneto-optical techniques when steps are aligned parallel to the applied field H manifest the flip of magnetization that occurs near $H=0$. The absence of magneto-optic effects for films less than or equal to one monolayer thick is consistent with recent *ab initio* calculations that predict significant moment reduction resulting from strong film-substrate hybridization.

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Epitaxial thin-film structures offer unique opportunities for exploring the relationship between structure and magnetism [1]. On an atomic scale, new phases of matter [2] (bcc Ni, fcc Fe, for example) can be stabilized as thin-film structures on suitable growth templates by molecular-beam epitaxy. *Ab initio* calculations [3–5] have predicted novel magnetic properties for these metastable phases and thin-film structures including dramatic variations in magnetic moments [3], ferromagnetic ordering in expanded lattice materials that are normally nonmagnetic [4], and unusual magnetic anisotropies [5] in ultrathin films. Thickness-dependent oscillatory magnetic coupling [6–8] has been observed through nonmagnetic Cr layers [6] and Cu layers [7,8] in thin-film sandwiches and superlattices. All of these results are manifestations of the intricate relationship between magnetic properties and various structural parameters.

A related topic that has received less attention from the viewpoint of thin-film structures pertains to the relationship between nanometer-scale lateral constraints and magnetic behavior. A well-developed subfield of micromagnetics exists [9], but few manifestations of the rich variety of effects that should exist in laterally constrained ultrathin-film systems have been observed or explored. Some of the difficulties in treating micromagnetics in ultrathin-film systems have been recently introduced [10] in relation to the role of pinning sites in reducing the coercivity in ultrathin films.

In this Letter, we report a new micromagnetic phenomenon associated with carefully prepared and characterized ultrathin epitaxial $p(1 \times 1)$ Fe films grown on a W(001) surface deliberately modified to yield uniform 25-Å-wide terraces along a (100) crystallographic direction. The nanometer-scale steps are shown to introduce an in-plane magnetic easy direction perpendicular to the steps. We also report experimental results that support the theoretically predicted [11] hybridization-induced quenching of ferromagnetism in one-monolayer (ML) Fe films on W(001).

Our experiments were carried out using 1-cm-diam \times 0.1-cm-thick W(001) substrates prepared as illustrated in Fig. 1. Half of the crystal was polished to yield

a surface normal within $\pm \frac{1}{2}^\circ$ of the (001) crystal direction; the other half of the crystal was polished to yield a 4° angle between the surface normal and the (001) crystal direction. This type of substrate permits comparison of thin-film magnetic properties on smooth and stepped surfaces at precisely the same temperature and film thickness. The step density on the two surfaces and the uniformity of the terraces was determined by analysis of the splitting and widths of low-energy electron diffraction (LEED) spots recorded both prior to and after growth of the epitaxial Fe films. The step density on the $\pm \frac{1}{2}^\circ$ (reference) half and 4° half of the crystal was $< (1 \text{ step})/(100 \text{ \AA})$ and $(1 \text{ step})/(25 \text{ \AA})$, respectively, with an rms variation less than $\pm 5 \text{ \AA}$.

The Fe films were grown and studied *in situ* at pressures below 1×10^{-10} torr. Standard techniques [12] were used to clean the W(001) crystal and to determine the contamination levels prior to and after film growth. Surfaces were judged clean when Auger analysis detected less than 1% total contamination (primarily C and O). Fe films were grown at a rate of approximately $0.5 \text{ \AA}/\text{min}$ by electron-beam evaporation from the tip of a high-purity 2-mm-diam Fe wire. The first Fe ML was grown with the W(001) crystal held at 900 K; subsequent layers

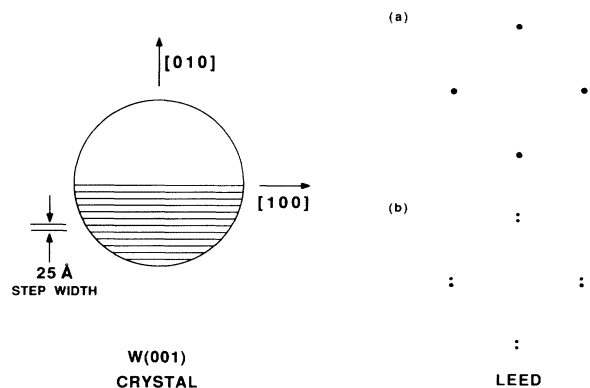


FIG. 1. Schematic representation of crystal showing smooth and stepped halves, and resulting LEED patterns for each half: (a) smooth half; (b) stepped half.

were grown after cooling the substrate to 250 K. Film thickness was monitored during growth by a quartz microbalance located at $\frac{1}{10}$ the source-to-sample distance and by measuring selected Fe and W Auger intensities. Thickness calibrations of the crystal monitor agreed with independent Auger electron spectroscopy calibrations obtained by conducting our own thermal desorption studies in which single atomic Fe layers on W(001) were prepared as described in previous work [13]. Thickness values quoted are believed accurate to $\pm 5\%$.

The W(001) surface offers several favorable properties for growing and studying epitaxial Fe thin films [12,14]. The lattice constants ($a_W=3.165 \text{ \AA}$, $a_{Fe}=2.866 \text{ \AA}$) result in a significant lattice expansion of a $p(1 \times 1)$ monolayer film ($\sim 9.4\%$), and the surface energies ($\gamma_W=2.9 \text{ J/m}^2$, $\gamma_{Fe}=2.0 \text{ J/m}^2$) strongly favor monolayer nucleation rather than other possible nucleation modes—three-dimensional island formation, for example. Extensive studies of the growth mode and monolayer stability of Fe on W(001) by our group [12,15], and of Fe on W(110) by others [13,14], have established that no interface mixing occurs at any temperature (Fe evaporates from the W surfaces before interdiffusion occurs) and that the first ML is stable and well ordered over a wide range of temperature. Our LEED studies [15] have established that the next few layers of Fe grown at 250 K are also in excellent registry with the W(001) growth template. However, changes in magnetic properties and LEED results for films greater than ~ 1 ML thick that accompany annealing to temperatures above 500 K suggest that these thicker films (grown at 250 K) are not thermodynamically stable.

No magneto-optic Kerr-effect signals were observed from Fe films on either the smooth or stepped W(001) surfaces for film thickness less than 1 ML. Consistent with this result are results of our previous spin-polarized angle-resolved photoemission studies [12] of $p(1 \times 1)$ Fe on W(001) which detected no remanent spin polarization at $T=115 \text{ K}$ for films less than 1 ML thick. Films having thickness greater than 1 ML were found to exhibit spin polarization under remanent conditions ($H=0$). This behavior implies that Fe/W(001) films just under 1 ML thickness are either nonmagnetic or antiferromagnetic at the temperatures ($T > 100 \text{ K}$) and applied fields ($H < 3000 \text{ Oe}$) accessible in the present experiments. This behavior is not a consequence of the well-established layer dependence of the Curie temperature (T_C) because 2-ML films have a relatively high T_C [$T_C(2) \sim 320 \text{ K}$]. Recent *ab initio* calculations [11] have explored the effects of both lattice strain and strong *d*-band hybridization on magnetism in the $p(1 \times 1)$ Fe on W(100) system. The giant ferromagnetic moment ($3.1\mu_B$) predicted for a free-standing Fe monolayer film is effectively quenched (to $< 0.1\mu_B$) by film-substrate hybridization effects. The energy separation between the ferromagnetic and antiferromagnetic ground state is found to be only 0.01

eV/atom. These results suggest that the magnetic ground state of 1-ML Fe on W(001) is unstable. The calculations also predict that strong ferromagnetism ($1.8\mu_B$ for interfacial Fe, $2.5\mu_B$ for surface Fe) is restored when a second Fe layer is added to the magnetically dead monolayer. These predictions are in good agreement with both our magneto-optic Kerr effect and spin-polarized photoemission experiments.

Figure 2 displays hysteresis loops, obtained using the magneto-optic Kerr effect, for $p(1 \times 1)$ Fe films of various thickness on W(001). The smooth half of the surface was found to always yield conventional polarization-independent hysteresis loops that exhibit very little variation with film thickness (for thickness > 1 ML) aside from an amplitude change which generally scales with film thickness. (Note that the hysteresis loops displayed in Figs. 1 and 2 are normalized.) The stepped half of the surface produced unusual hysteresis loop behavior. All hysteresis loops presented in this Letter were obtained using the longitudinal magneto-optic Kerr-effect configuration in which the applied field is in the plane of incidence. The hysteresis loops displayed in Fig. 2 were obtained with the sample step edges parallel to the applied

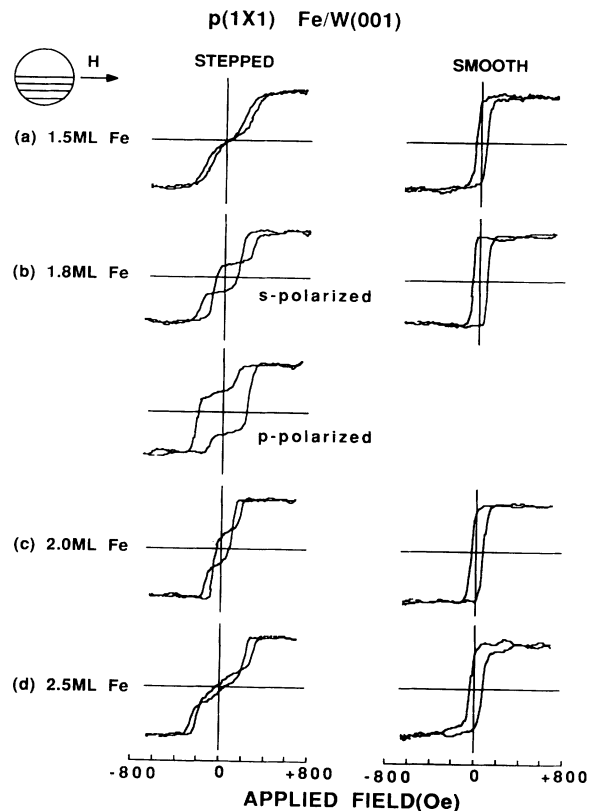


FIG. 2. Hysteresis loops obtained using the magneto-optic Kerr effect for epitaxial $p(1 \times 1)$ Fe films of various thickness on smooth (right panel) and stepped (left panel) W(001). Inset: Orientation of the crystal. All hysteresis loops are normalized to the same amplitude (vertical scale).

field, as shown by the inset. These films exhibit thickness-dependent anomalies in the hysteresis loops near $H=0$. Polarization dependences were observed in loops obtained from stepped surfaces. The polarization effect is illustrated in Fig. 2 for the 1.8-ML film. Corresponding results obtained when the crystal is rotated 45° and 90° are displayed in Fig. 3.

Several conclusions can immediately be drawn from these results. First, it is clear that the smooth half of the crystal behaves generally as an isotropic thin-film magnet. There is no significant variation of the coercive force (which is determined to be ~ 100 Oe) with either film thickness or crystal orientation. Polar magneto-optic Kerr-effect signals (H applied along the surface normal) were observed from some of the thin Fe samples (1.5 ML), but the saturation magnetization and coercive forces were large—typically over 1.0 kOe—an indication that there is no “perpendicular easy axis anisotropy” in the Fe on W(100) system over the temperature and thickness range of interest here. The film normal direction is a “hard” direction of magnetization, and therefore the novel magneto-optical effects observed for $H < 100$ Oe must be attributed to magnetization effects in the film plane. It is also apparent from Fig. 2 that higher fields are required to drive the films to saturation for H parallel

to the surface steps than for H perpendicular (or at 45° , Fig. 3) to the steps. This is a clear indication of an in-plane magnetic anisotropy.

Quantitative interpretation of magneto-optic effects in terms of local (Fresnel) optics [16–19] for magnetic films ~ 1 ML thick are subject to complications discussed in relation to “nonlocal” optical response [20]. However, explicit (Fresnel optics) formulas for magneto-optical intensities [18,19] that have been employed to interpret Kerr-effect microscope images and to study in-plane magnetic anisotropy of “thick” films remain approximately valid, and can be used to account qualitatively for the observed polarization-dependent behavior of hysteresis loops in terms of the macroscopic magnetization of the thin-film structure.

The conventional hysteresis behavior observed in all cases for the “smooth” surface and for configurations in which the applied field is primarily along the “easy” magnetization direction (*perpendicular* to the steps on the stepped surface) are expected. In these cases, the magnetization \mathbf{M} remains in the plane of incidence, and only the longitudinal magneto-optic Kerr effect contributes to the detected intensity. There are no anomalous “kinks” in the loop. When a sufficiently strong field is applied *parallel* to the steps, \mathbf{M} aligns in the plane of incidence, yielding the high-field saturation behavior observed for $H > H_{\text{crit}}$, where H_{crit} is the field at which rotation of \mathbf{M} begins to occur. For $H < H_{\text{crit}}$, the step-induced uniaxial anisotropy forces \mathbf{M} *perpendicular* to the plane of incidence (the transverse configuration [17]), and the longitudinal Kerr effect vanishes (or is diminished by the reduced projection of \mathbf{M} on the plane of incidence). The difference in hysteresis behavior determined using “s” or “p” polarized light for H parallel to steps occurs because when $H < H_{\text{crit}}$ both the transverse Kerr effect and longitudinal Kerr effect contribute to the measured intensity changes when *p* polarized light is used. Thus, all of the observed magneto-optical effects are accounted for by a surface-step-induced in-plane uniaxial anisotropy.

Annealing the films to 600 K or exposing the films to small amounts (0.80 L, where $1 \text{ L} = 1 \times 10^{-6}$ torrsec) of oxygen destroys the step-induced anisotropy. Changes in LEED patterns that occur after annealing *p*(1 \times 1) Fe films on W(001) suggest that three-dimensional island formation occurs at $T > 500$ K for thickness greater than 1 ML. Similar oxygen-induced behavior has been recently reported for *p*(1 \times 1) Fe on Ag(100) [21]. In this case small oxygen doses were found to change the magnetic anisotropy from perpendicular to the surface to an in-plane direction, and LEED spot-profile analysis, prior to and after the oxygen dose, suggested some degree of improvement in surface order resulting from the oxygen dose.

We have explored (to some extent) the step-induced magnetic behavior as a function of step density. There appears to be a range of step densities over which the

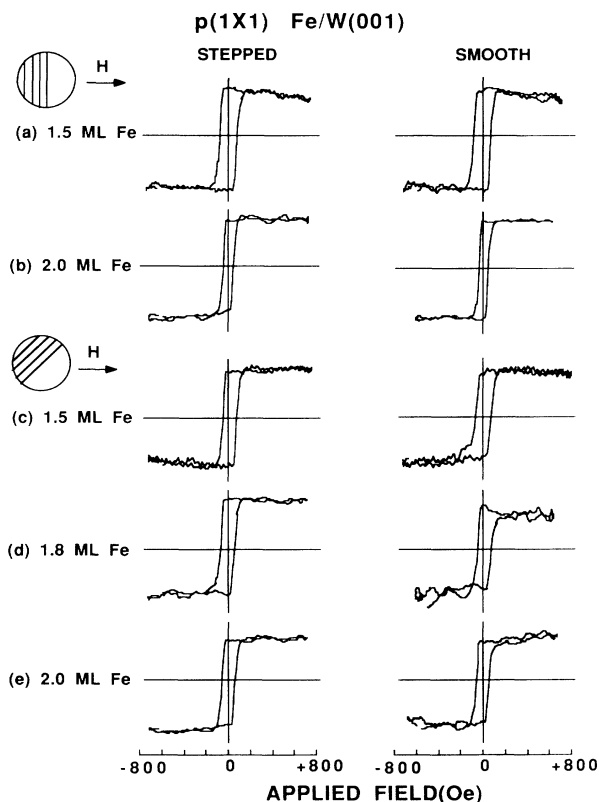


FIG. 3. Results corresponding to Fig. 1 with crystal oriented, as shown by the inset. All hysteresis loops are normalized to the same amplitude (vertical scale).

novel effects illustrated in Figs. 1 and 2 can be induced. We have not defined precise limits, but crystals polished 6° off of the (001) direction were not found to yield step-induced anisotropy, at least for the same growth parameters that yield the effect for 4° crystals. Smooth surfaces having surface normals within $\pm \frac{1}{2}^\circ$ of the (001) crystal axis also fail to yield the novel effects. These phenomena (step-density dependence, annealing dependence, and effects of oxygen adsorption on the stepped film structures) are being investigated using Kerr effects, polarized electron spectroscopy, and other techniques and will be reported later.

In summary, we have presented a new thin-film magnetic phenomenon: step-induced surface anisotropy. The established absence of interface interdiffusion in the W/Fe system rules out compositional effects as contributing factors. Our controlled experiments, in which the same film is studied on both flat and stepped surfaces, clearly establish the steps as the controlling factor. The novel magnetic effects are quite reproducible and exhibit systematic thickness variation, but the structural changes induced by annealing or chemisorption that extinguish the step-induced anisotropy are subtle. The easy magnetization direction on the stepped surface is perpendicular to the steps—*just opposite to what one would guess, based on shape anisotropy effects*. Our failed attempts to observe magneto-optic behavior in films just under 1 ML thick are consistent with recent *ab initio* calculations that predict magnetic quenching at 1 ML resulting from film-substrate hybridization effects. These results once again emphasize the importance of structure and interface coupling in controlling thin-film magnetic behavior.

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Note added.—It has been brought to the authors' attention that similar effects have been recently observed by U. Gradmann and co-workers in Clausthal. This work was reported at the ICMSF '91 (Glasgow) and at the ICM '91 (Edinburgh). The results will be published in the proceedings of these conferences (both in *J. Magn. Magn. Mater.*).

[1] L. M. Falicov, D. T. Pierce, S. D. Bader, R. Gronsky, K. B. Hathaway, H. J. Hopster, D. N. Lambeth, S. S. P. Parkin, G. Prinz, M. Salamon, I. K. Schuller, and R. H. Victora, *J. Mater. Res.* **5**, 1299 (1990).

- [2] *Novel Materials and Techniques in Condensed Matter*, edited by G. W. Crabtree and P. Vashishta (Elsevier, New York, 1982).
- [3] V. L. Moruzzi, P. M. Marcus, K. Schwarz, and P. Mohn, *Phys. Rev. B* **34**, 1784 (1986); V. L. Moruzzi, P. M. Marcus, and J. Kübler, *Phys. Rev. B* **39**, 6957 (1989); C. S. Wang, B. M. Klein, and H. Krakauer, *Phys. Rev. Lett.* **54**, 1852 (1985); S. Blügel, B. Drittler, R. Zeller, and P. H. Dederichs, *Appl. Phys. A* **49**, 547 (1989).
- [4] O. Eriksson, R. C. Albers, and A. M. Boring, *Phys. Rev. Lett.* **66**, 1350 (1991); M. J. Zhu, D. M. Bylander, and L. Kleinman, *Phys. Rev. B* **43**, 4007 (1991).
- [5] R. Richter, J. G. Gay, and J. R. Smith, *Phys. Rev. Lett.* **54**, 2704 (1985); J. G. Gay and R. Richter, *J. Appl. Phys.* **61**, 3362 (1987).
- [6] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, *Phys. Rev. Lett.* **57**, 2442 (1986); S. S. P. Parkin, N. More, and K. P. Roche, *Phys. Rev. Lett.* **64**, 2304 (1990).
- [7] B. Heinrich, Z. Celinski, J. F. Cochran, W. B. Muir, J. Rudd, Q. M. Zhong, A. S. Arrott, K. Myrtle, and J. Kirschner, *Phys. Rev. Lett.* **64**, 673 (1990).
- [8] W. R. Bennett, W. Schwarzacher, and W. F. Egelhoff, Jr., *Phys. Rev. Lett.* **65**, 3169 (1990).
- [9] W. F. Brown, Jr., *Micromagnetics* (Interscience, New York, 1963).
- [10] A. S. Arrott, *J. Appl. Phys.* **69**, 5212 (1991).
- [11] R. Wu and A. J. Freeman, *Bull. Am. Phys. Soc.* **36**, 702 (1991); (private communication).
- [12] R. L. Fink, G. A. Mulhollan, A. B. Andrews, J. L. Erskine, and G. K. Walters, *J. Appl. Phys.* **69**, 11 (1991).
- [13] X.-L. Zhou, C. Yoon, and J. M. White, *Surf. Sci.* **203**, 53 (1982).
- [14] U. Gradmann, M. Przybylski, H. J. Elmers, and G. Liu, *Appl. Phys. A* **49**, 563 (1989).
- [15] M. Drakaki, J. Chen, and J. L. Erskine (unpublished).
- [16] A. V. Sokolov, *Optical Properties of Metals* (American Elsevier, New York, 1967).
- [17] G. S. Krinchik and V. A. Artemjev, *J. Appl. Phys.* **39**, 1276 (1968); G. S. Krinchik and R. D. Nuralieva, *Zh. Eksp. Teor. Fiz.* **36**, 1022 (1959) [*Sov. Phys. JETP* **9**, 724 (1959)].
- [18] W. Rave, R. Schäfer, and A. Hubert, *J. Magn. Magn. Mater.* **65**, 7 (1987).
- [19] J. M. Florczak and E. D. Dahlberg, *J. Appl. Phys.* **67**, 7520 (1990).
- [20] P. J. Feibelman, *Prog. Surf. Sci.* **12**, 287 (1982); W. L. Schaich, *Phys. Rev. B* **37**, 6193 (1988); D. C. Langreth, *Phys. Rev. B* **39**, 10020 (1989).
- [21] Jian Chen, M. Drakaki, and J. L. Erskine, *Phys. Rev. B* **45**, 3636 (1992).