

## Curie Temperature Enhancement and Induced Pd Magnetic Moments for Ultrathin Fe Films Grown on Stepped Pd(001)

Hyuk J. Choi, R. K. Kawakami, Ernesto J. Escorcia-Aparicio, and Z. Q. Qiu  
*Department of Physics, University of California at Berkeley, Berkeley, California 94720*

J. Pearson, J. S. Jiang, Dongqi Li, and S. D. Bader  
*Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439*  
(Received 24 August 1998)

Fe films grown in ultrahigh vacuum onto a curved Pd(001) substrate with a continuous gradient in atomic step density were studied *in situ* via the surface magneto-optic Kerr effect (SMOKE). The steps induce an in-plane, uniaxial magnetic anisotropy with the easy axis perpendicular to the step edges, and whose strength scales linearly with step density. The Curie temperature of 1–2 monolayers of Fe is enhanced by the steps relative to that on a flat substrate. The enhancement is attributed to a step-induced Pd moment at step edges, as inferred from the enhancement of the SMOKE signal with step density. [S0031-9007(99)08552-X]

PACS numbers: 75.70.Ak, 75.30.Kz, 75.30.Pd, 75.50.Bb

An ordered array of atomic steps on a (001) substrate surface can induce an in-plane, uniaxial magnetic anisotropy in a ferromagnetic overlayer [1,2]. This anisotropy originates from the step edges due to the lattice symmetry breaking [3]. Studies involving step-induced anisotropy are greatly facilitated by the use of *curved* substrates to provide a continuous gradient in step density. In this manner step-induced anisotropies were found to depend *quadratically on step density* for the systems Fe films grown on stepped Ag(001) [Fe/stepped Ag(001)] [4] and Fe/stepped W(001) [5], but *linearly on the step density* for Co/stepped Cu(001) [6]. The quadratic and linear dependences are thought to arise from the symmetry properties of the bcc Fe and fcc Co lattices, respectively [4–6]. This dependence on lattice symmetry offers an opportunity to manipulate the magnetic anisotropy of a stepped thin film. For example, the quadratic and linear dependences should be interchangeable if the dominant contribution to the anisotropy can be altered between bcc and fcc structures. This fascinating possibility is addressed in this Letter. Another fundamental issue is that of magnetic long-range order (LRO) in a two dimensional (2D) Heisenberg system. It is well known theoretically that an isotropic 2D Heisenberg system does not exhibit LRO at finite temperature [7]; but a uniaxial anisotropy can stabilize the LRO [8]. Thus curved films can provide a good model system since the evolution can be followed from the weak bulk anisotropy of a flat surface to the strong uniaxial anisotropy of a stepped surface. Experimental results on the Fe/W(001) system, however, indicate no effect of the steps on the Curie temperature ( $T_C$ ) [5]. But the strength of the step-induced anisotropy in Fe/stepped W(001) is merely comparable to that of the bulk anisotropy of Fe, so it is unclear whether or not the step-induced anisotropy *should* have an appreciable effect on the 2D LRO. To test this idea, one needs a system

whose step-induced anisotropy is much stronger than the bulk anisotropy.

In this Letter we investigate the Fe/stepped Pd(001) system. Fe/Pd is chosen for two reasons. First, the strong spin-orbit interaction of Pd always produces a strong magnetic anisotropy in ferromagnet/Pd multilayers [9]. Thus, we expect a strong step-induced magnetic anisotropy in the Fe/Pd system. Second, ferromagnetic Fe polarizes the Pd at the interface of Fe/Pd(001) and causes it to become ferromagnetic [10]. This then provides a unique opportunity to explore the role of the lattice symmetry in the ferromagnetic bcc/fcc interface to the step-induced anisotropy. Such experiments may give information about the effects of the induced Pd moment to the step-induced anisotropy. We find that (1) the step-induced magnetic anisotropy in the Fe/Pd system is much stronger than that in both the Fe/Ag and Fe/W systems; (2) the  $T_C$  value of the Fe film on the stepped surface is higher than that on the flat surface for thickness  $< 2$  monolayers (ML); (3) there is an additional induced magnetic moment on Pd at the step edges; and (4) the step-induced magnetic anisotropy depends linearly on step density, as that was so for an fcc system [6].

The experiments were performed by first preparing the substrate, which was a 1-cm-diameter single crystal disk of Pd(001). It was mechanically polished into a curved shape with a 0.25- $\mu\text{m}$  diamond paste finish. The atomic steps are parallel to the [110] direction of the Pd, and the vicinal angle ( $\alpha$ ) ranges continuously from  $0^\circ$  to  $8^\circ$ . The substrate was cleaned, in an ultra high vacuum (UHV) chamber of a base pressure of  $9 \times 10^{-11}$  Torr, with cycles of 2–3 keV Ar ion sputtering and subsequent annealing at 700–800  $^\circ\text{C}$ . The cleaning process continued until no impurity was detected by Auger spectroscopy, and a sharp  $1 \times 1$  pattern was formed in low-energy electron diffraction (LEED). Fe wedges were grown on

the Pd substrate at room temperature typically using an evaporation rate of  $\sim 0.5 \text{ \AA}/\text{min}$ . Extensive studies on the pseudomorphic nature of the room-temperature growth of Fe films on Pd(001) can be found in the literature [11–13]. In particular, LEED I-V studies [13] show that the Fe films grow pseudomorphically with a body-centered tetragonal structure up to 65 ML. Magnetic measurements were carried out *in situ* by means of the surface magneto-optic Kerr effect (SMOKE) using a He-Ne laser. A slit was placed in the path of the reflected beam in order to improve the lateral resolution along the wedge, and hence the spread in the vicinal angle probed to  $\pm 0.2^\circ$ . The local value of the vicinal angle was determined by the reflection angle of the laser beam during the SMOKE measurement. No polar SMOKE signal was detected; the Fe magnetization is in the film plane, in agreement with a previous study [14]. All SMOKE measurements in this paper were taken in the longitudinal configuration.

Figure 1 displays hysteresis loops for a 2.9-ML Fe film grown on stepped Pd(001) with a vicinal angle of  $\alpha = 8^\circ$ , and with the magnetic field applied both perpendicular and parallel to the step edges. The full and zero remanences of the hysteresis loops along the two respective directions show that the atomic steps induce an in-plane, uniaxial magnetic anisotropy with the easy axis perpendicular to the step edges. The strength of the step-induced anisotropy can be estimated from the saturation field of the hard axis loop:  $K \sim MH_s = 1.3 \times 10^6 \text{ erg}/\text{cm}^3$ . This value is  $\sim 3$  times stronger than the volume anisotropy of Fe ( $\sim 4.5 \times 10^5 \text{ erg}/\text{cm}^3$ ), and also is much stronger than that found for both the Fe/stepped Ag(001) [4] and Fe/stepped W(001) [5] systems. We will come back to this point later.

The strong step-induced anisotropy of Fe/stepped Pd(001) makes it an ideal system to study the effect of

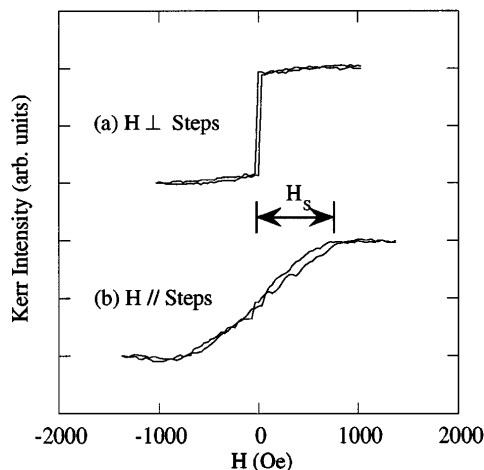


FIG. 1. Longitudinal hysteresis loops of a 2.9-ML Fe film grown on stepped Pd(001) (vicinal angle  $\alpha = 8^\circ$ ). The magnetic field is applied (a) perpendicular and (b) parallel to the step edges.

steps on the magnetic LRO in a 2D Heisenberg system. Figure 2 shows SMOKE loops at different temperatures for 1.6 ML of Fe grown on both flat and stepped Pd(001) ( $\alpha = 8^\circ$ ) with the magnetic field applied along the easy axis. The magnitude of the Kerr signals at 300 K were normalized to each other. The magnetic remanence of the 1.6-ML Fe film disappears between 394 and 401 K for the flat surface, and between 401 and 453 K for the stepped surface. Note that the Fe films were deposited on the two Pd(001) surfaces simultaneously, so the  $T_C$  difference in Fig. 2 cannot be attributed to thickness variations between the flat and stepped films. To understand the enhancement of  $T_C$  on the stepped surface, the thickness dependence of  $T_C$  of Fe films on both flat and stepped Pd was measured on wedged samples. At each temperature, hysteresis loops were measured along the Fe wedge to find the critical thickness at which the magnetic remanence vanishes. This critical thickness thus has its  $T_C$  equal to the given temperature. Figure 3 shows results for both flat and stepped ( $\alpha = 8^\circ$ ) surfaces. The  $T_C$  is higher on the stepped surface than on the flat surface for Fe film thickness less than  $\sim 2$  ML, but the  $T_C$  values approach each other above  $\sim 2$  ML. The equivalence of the  $T_C$  values for Fe thickness  $> 2$  ML indicates that the Fe-Fe interlayer interaction overwhelms the step-enhanced magnetic LRO. This is interesting because Fe/Pd(001) behaves 2D-like for thickness  $< 3$  ML Fe [14], and the step-induced anisotropy for 3 ML Fe is still much stronger than the bulk anisotropy. Thus, we would anticipate an enhancement of  $T_C$  up to at least 3 ML Fe if the cause were the step-induced uniaxial magnetic anisotropy. The absence of  $T_C$  enhancement in the 2–3 ML thickness range suggests that the observed enhancement of  $T_C$  is not caused by the step-induced magnetic anisotropy,

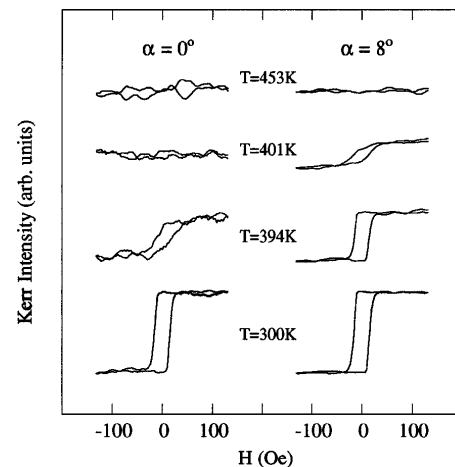


FIG. 2. Easy axis hysteresis loops of a 1.6-ML Fe film on flat ( $\alpha = 0^\circ$ ) and stepped ( $\alpha = 8^\circ$ ) Pd(001) at different temperatures. The magnetic remanence disappears on the flat surface at a lower temperature than on the stepped surface, indicating an enhancement of the Curie temperature on the stepped surface.

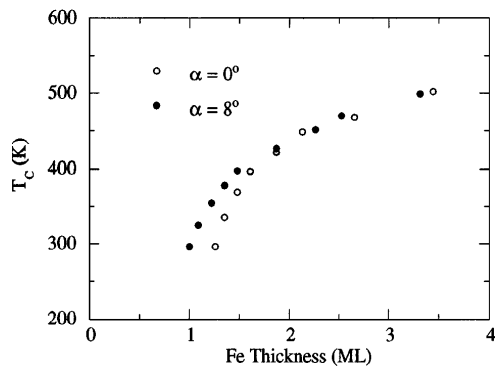


FIG. 3. Thickness dependence of  $T_C$  for flat ( $\alpha = 0^\circ$ ) and stepped ( $\alpha = 8^\circ$ ) Fe/Pd(001).

but is due to some other effect. One possibility is that annealing might increase interfacial alloying and diminish the influence of steps. However, SMOKE measurements after annealing demonstrate that the results are reversible up to  $T \sim 500$  K, so annealing can be ruled out as an explanation of our observations. Another possibility is step-induced roughness. Since the terrace length is much greater than the step height ( $L > 10a$ ), the majority of the stepped film is still Fe on flat Pd(001). Previous work on stepped Fe/Ag(001) [4], Fe/W(001) [2,5], and Co/Cu(001) [3,6] did not show evidence that the step-induced anisotropy arises from step-induced roughness. In particular, the Co/Cu system has been studied with scanning tunneling microscopy (STM) and step decoration to show that the step-induced anisotropy indeed comes from the symmetry breaking at the step edges. The fact that the easy magnetization axis could be either parallel (Fe/Ag and Co/Cu) or perpendicular (Fe/W and Fe/Pd) to the step edges is also hard to explain with step-induced roughness. Thus, we believe that the  $T_C$  difference and the step-induced anisotropy are unlikely to be caused by step-induced roughness.

It has been shown that Pd at the Fe/Pd(100) interface carries an induced moment due to the Fe polarization [10]. While this direct polarization occurs in the surface state of Pd in the (100) surface [10], it persists up to  $\sim 5$  ML of Pd on the (110) surface [15]. Hence, it is well recognized that the Pd near the Fe/Pd interface is ferromagnetic. The induced moment of Pd was also shown to enhance the  $T_C$  of the Fe film [14,16]. The Pd spin polarization comes from the electronic hybridization at the Fe/Pd interface, thus, it should depend sensitively on the local environment of the Pd. Therefore, we would expect a different induced moment of Pd at step edges relative to that at a flat surface because of the different Fe coordination at the two sites. To verify the existence of the additional Pd induced moment at step edges, we measured the magnitude of the SMOKE signal for a 1.8-ML Fe film grown on the curved Pd as a function of step density (Fig. 4). As shown in the figure, the SMOKE signal increases linearly with step density. The linear

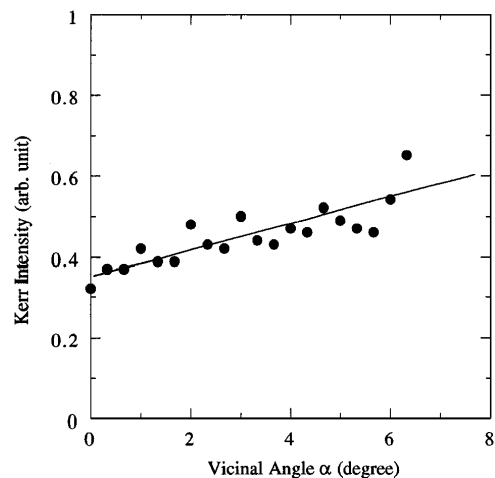


FIG. 4. SMOKE signal vs vicinal angle ( $\alpha$ ) for a 1.8-ML Fe film on Pd(001).

dependence indicates that the step-induced SMOKE signal is a local effect from the step edges. It is interesting to note that the SMOKE signal at  $\alpha \sim 6^\circ$  is  $\sim 1.6$  times the magnitude of that on the flat surface, i.e., the steps induce a SMOKE signal that is roughly equivalent to that from an extra monolayer of Fe. This significant enhancement of the SMOKE signal is not likely to be caused by an enhanced Fe moment, but more likely comes from the additional induced moment of Pd at the step edges. Pd provides a significant magneto-optic effect at around 1.9 eV due to its strong spin-orbit interaction [17]. We believe it is this additional induced moment of Pd at the step edges that further enhances the  $T_C$  of the Fe film on the stepped surface as compared to the flat surface. A firm verification of the additional induced moment of Pd at step edges will be undertaken in future work using element-specific magnetic circular dichroism.

The step-induced moment of Pd should also be reflected in the step-induced magnetic anisotropy. Since the magnetic anisotropy comes from the spin-orbit interaction, the much stronger spin-orbit interaction of Pd compared to Fe should generate an enhanced step-induced anisotropy. This would explain why the step-induced anisotropy in the Fe/Pd system is much greater than that in the Fe/Ag and Fe/W systems. Further evidence of the contribution of the Pd to the anisotropy comes from the dependence of the step-induced anisotropy on the step density. It was shown that the step-induced anisotropy depends quadratically on step density for bcc films [4,5], but linearly for fcc films [6]. Then, if the anisotropy in the Fe/Pd system is dominated by the ferromagnetic Pd near the interface, the fcc structure of Pd should produce a linear dependence of the step-induced anisotropy on step density rather than the quadratic dependence associated with bcc Fe film as observed in the Fe/Ag and Fe/W systems. Figure 5 shows the magnitude of the saturation field from hard-axis hysteresis loops, which measures the strength

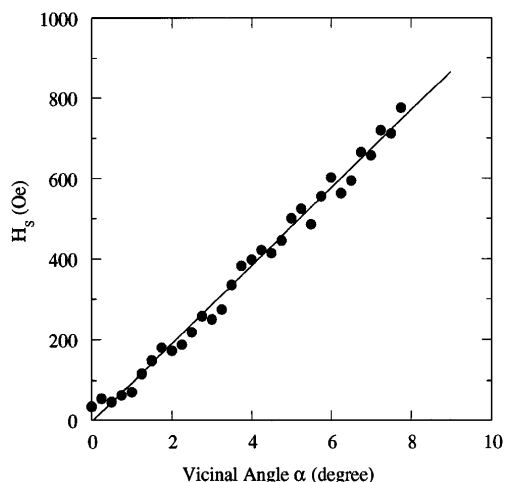


FIG. 5. The saturation field ( $H_s$ ), defined in Fig. 1, for 2.9 ML of Fe on stepped Pd(001).

of the step-induced magnetic anisotropy, as a function of step density for a 2.9-ML Fe film grown on the curved Pd. It is obvious that the step-induced magnetic anisotropy depends linearly on the step density. This linear dependence supports our hypothesis that the ferromagnetic fcc Pd near the interface dominates the step-induced magnetic anisotropy in this system. However, because of the Fe-Fe, Pd-Pd, and Fe-Pd interactions involved at the interface, one also needs to take into account the anisotropy energy introduced by the Fe(bcc)-Pd(fcc) pairs at the step edges. This remains as a challenge for future theoretical study.

In summary, we found that atomic steps along the Pd [110] axis in a Fe/Pd(001) system induce an in-plane, uniaxial magnetic anisotropy with easy axis perpendicular to the step edges. The Curie temperature of the Fe films is higher on the stepped surface than on the flat surface for thickness  $< 2$  ML of Fe. This enhancement of the  $T_C$  is attributed to the step-induced magnetic moment of Pd at the step edges. The existence of such a step-induced moment of Pd is verified by the linear increase of the SMOKE signal as a function of step density. The ferromagnetic nature of the fcc Pd at the interface is

further supported by the linear dependence of the step-induced anisotropy on the step density.

This work was supported in part by the DOE BES-MS under Contract No. W-31-109-ENG-38 (at Argonne), DE-AC03-76SF00098 (at LBNL), NSF DMR-9805222, and was also supported in part by the University of California for the conduct of discretionary research by Los Alamos National Laboratory.

- 
- [1] A. Berger, U. Linke, and H. P. Oepen, Phys. Rev. Lett. **68**, 839 (1992).
  - [2] J. Chen and J. Erskine, Phys. Rev. Lett. **68**, 1212 (1992).
  - [3] W. Weber, C.H. Back, A. Bischof, D. Pescia, and R. Allenspach, Nature (London) **374**, 788 (1995).
  - [4] R. K. Kawakami, Ernesto J. Escorcia-Aparicio, and Z. Q. Qiu, Phys. Rev. Lett. **77**, 2570 (1996).
  - [5] Hyuk J. Choi, Z. Q. Qiu, J. Pearson, S. J. Jiang, D. Li, and S. D. Bader, Phys. Rev. B **57**, R12713 (1998).
  - [6] R. K. Kawakami, M. O. Bowen, Hyuk J. Choi, Ernesto J. Escorcia-Aparicio, and Z. Q. Qiu, Phys. Rev. B **58**, R5924 (1998).
  - [7] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1996).
  - [8] M. Bander and D. L. Mills, Phys. Rev. B **38**, 12015 (1988).
  - [9] Brad N. Engel, Craig D. England, Robert A. Van Leeuwen, Michael H. Wiedmann, and Charles M. Falco, Phys. Rev. Lett. **67**, 1910 (1991).
  - [10] O. Rader, E. Vescovo, J. Redinger, S. Blügel, C. Carbone, W. Eberhardt, and W. Gudat, Phys. Rev. Lett. **72**, 2247 (1994).
  - [11] C. Liu and S. D. Bader, Phys. Rev. B **44**, 2205 (1991).
  - [12] E. Vescovo, O. Rader, and C. Carbone, Phys. Rev. B **47**, 13051 (1993).
  - [13] J. Quinn, Y. S. Li, H. Li, D. Tian, F. Jona, and P. M. Marcus, Phys. Rev. B **43**, 3959 (1991).
  - [14] C. Liu and S. D. Bader, J. Appl. Phys. **67**, 5758 (1990).
  - [15] W. Weber, D. A. Wesner, D. Hartmann, and G. Güntherodt, Phys. Rev. B **46**, 6199 (1992).
  - [16] K. Strandburg, D. W. Hall, C. Liu, and S. D. Bader, Phys. Rev. B **46**, 10818 (1992).
  - [17] W. Reim, H. Brändle, D. Weller, and J. Schoenes, J. Magn. Magn. Mater. **93**, 220 (1991).