Two-dimensional Ising transition of epitaxial Fe films grown on Ag(100)

Z. Q. Qiu,* J. Pearson, and S. D. Bader
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
(Received 12 August 1993)

Fe(100) films and wedges between 2 and 4 monolayers thick were grown onto a Ag(100) single-crystal substrate. High- and low-energy electron-diffraction observations confirm the epitaxy. The magnetic phase transition near the Curie temperature T_C was investigated *in situ* by means of the surface magneto-optic Kerr effect. The temperature dependence of the magnetization follows a universal power law of $M(T) \propto (1-T/T_C)^{\beta_c}$ with an effective value of $\beta_c \approx 1/8(0.124 \pm 0.002)$, which indicates that the phase transition is two-dimensional Ising class. This and the observed perpendicular easy axes are attributed to the surface magnetic anisotropy.

I. INTRODUCTION

The magnetic phase transition at the Curie temperature (T_C) in a two-dimensional (2D) system is a topic of recent experimental and theoretical interest. It is well known from the Mermin-Wagner theorem[1] that a 2D isotropic Heisenberg system does not have long-range magnetic order at finite temperature. However, the surface magnetic anisotropy can stabilize long-range order at a finite T_C value.[2] The uniaxial surface magnetic anisotropy originates from the broken translational symmetry along the normal direction of the film. The surface anisotropy usually consists of two terms: one is perpendicular and the other is parallel to the plane of the film. Based on a symmetry analysis, it is easy to show that the in-plane term vanishes if the surface has a more than a twofold rotational axis. The next higher-order term in the magnetic anisotropy is quadratic in the Cartesian components of the spin vector and is usually identified with the volume anisotropy. Therefore, magnetic thin films with cubic symmetry can be classified as follows:

Case I: There exists a uniaxial anisotropy, either perpendicular or parallel to the film plane;

Case II: There is no uniaxial surface anisotropy, so that the magnetization is in the film plane.

Obviously, case I can be described by a 2D Heisenberg model plus uniaxial anisotropy, while case II can be described by a 2D XY model plus a volume-anisotropy contribution. For case I, Bander and Mills showed[2] that the uniaxial anisotropy will make the magnetic phase transition Ising-like. For case II, however, José *et al.* showed[3] that the quadratic anisotropy will result in a nonuniversal character for the phase transition.

Experimental investigations of this subject usually involve temperature-dependent studies of the magnetization to determine the critical exponent β_c from the power law $M(T) \propto (1-T/T_C)^{\beta_c}$. Examples of systems studied include Fe/Au(100),[4] Fe/Pd(100),[5] Fe/Ag(111),[6] Co/Cu(111),[7] Ni/Cu(100),[8] Ni/Cu(111),[9] Ni/W(110),[10] Tb/W(110),[11] etc. The β values determined for various systems usually fall into one of two groupings: one has β close to the Ising value of $\frac{1}{8}$, and the other has a β value close to $\frac{1}{4}$. Almost all of the films with $\beta \approx \frac{1}{8}$ have either a perpendicular easy axis or an in-plane easy axis with in-plane uniaxial anisotropy. Therefore, they are representative of case I above. There are a few exceptions, of which Ni(111)/W(110) is an example. Ni(111)/W(110) has in-plane easy axes; but the Ni(111) surface has sixfold rotational symmetry, so the in-plane surface anisotropy should vanish. However, the W(110) substrate has only twofold rotational symmetry. We anticipate that the Ni(111) overlayer should be strained in one direction to destroy the sixfold rotational symmetry and result in in-plane uniaxial anisotropy. On the other hand, the films with $\beta \approx \frac{1}{4}$ belong to case II without exception, according to our knowledge. Although case-II films appear to exhibit universal behavior, the underlying character of their phase transition is still not well understood. Recent theoretical work on a finite-size XY model suggests the existence of a finite T_C value and a critical exponent $\beta = 3\pi^2/128 \approx 0.23$.[12] However, the role of the volume anisotropy in an XY system was not considered in Ref. 12. A fundamental issue that needs to be clarified is: what is the origin of the finite T_C value in this case? Does it come from the finite-size effect or from the volume anisotropy? Obviously, more experimental data relating to all cases are needed in order to understand more clearly the nature of 2D magnetic phase transitions.

In this paper, we study the magnetic phase transition in the Fe/Ag(100) system, which has perpendicular easy magnetization axes in the ultrathin regime.[13] This is a follow-up study to our recent investigation of the spin-reorientation transition for the Fe/Ag(100) system.[14] We chose this system also because the $3d$ electrons of Fe hybridize very little with the sp electrons of Ag, so the Fe film should form a nearly ideal 2D system.

II. EXPERIMENT

A. Growth and characterization

The Ag(100) substrate surface was prepared by vibratory mechanical polishing (Al_2O_3 powder) down to 0.05-μm finish, followed by chemical polishing with a solution of saturated chromic acid and ~1% hydrochlor-

0163-1829/94/49(13)/8797(5)/$06.00

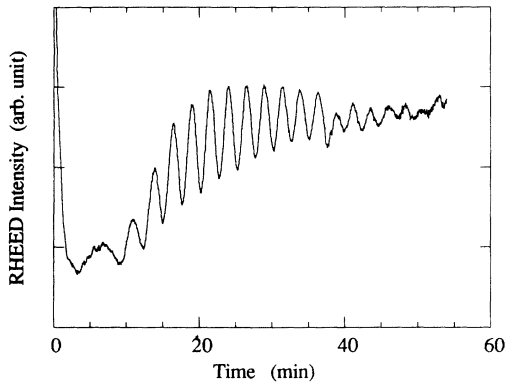

FIG. 1. RHEED intensity as a function of Fe deposition time. The strong oscillations imply a layer-by-layer-like growth of the Fe on Ag(100).

ic acid. The chemical polish was applied with a cotton swab (saturated with the solution) and immediately followed by a water rinse. In this way of the order of 100–200 Å of Ag was removed to eliminate the scratches produced by the mechanical polish. Then the substrate was ultrasonically cleaned in methanol before its introduction into the ultrahigh-vacuum chamber. Cycles of 2-keV Ar^+ sputtering followed by annealings at ~700 °C were undertaken then for the final cleaning of the surface. After these procedures a well-defined and clean Ag(100) surface was formed, as indicated by Auger-electron spectroscopy and reflection high- and low-energy electron diffraction (RHEED and LEED). The Fe film was grown onto the Ag(100) substrate at room temperature. The evaporation rate was monitored by a quartz thickness monitor and was typically ~0.6 Å/min. The RHEED and LEED patterns of Fe on Ag(100) have been reported previously.[14] The growth of Fe on Ag(100) also was studied by monitoring the RHEED intensity. Strong oscillations in the RHEED intensity were observed during the growth (Fig. 1), implying a "layer-by-layer-like" growth character. However, a recent STM study suggests that the existence of RHEED oscillations is not a sufficient condition for rigorous identification of *ideal* layer-by-layer growth.[15] After these growth-characterization studies, the film utilized for our magnetic studies was actually grown with a wedge shape to facilitate a continuous change of thickness. The slope of the wedge was ~0.2 ML/mm. The advantages of using wedge-shaped films have been demonstrated in a variety of magnetic thin-film studies recently.[16]

B. SMOKE measurements

The magnetic properties of the films were investigated *in situ* by means of surface magneto-optic Kerr-effect (SMOKE) measurements. To generate a SMOKE hysteresis loop, a beam of *p*-polarized He-Ne laser light ($\lambda = 6328$ Å) is reflected by the sample surface; the resultant beam intensity is recorded (after it is passed through a polarizer set ~1° from extinction) as a function of external magnetic field, which is applied either perpendicular (polar) or parallel (longitudinal) to the film plane.[17] The laser beam is focused to a spot size of ~0.2 mm diameter on the sample. Thus, the probed thickness variation due to the wedge shape is only ~0.04 ML within the laser spot; so the thickness probed can be virtually uniform. The temperature of the sample is measured by a thermocouple placed at the edge of the substrate a few mm from the laser spot. The temperature stability is controlled within ±0.2 K.

III. RESULTS AND ANALYSIS

We first confirmed that the easy axes of magnetization at room temperature for films less than ~7 ML of Fe are perpendicular to the film plane, as indicated by polar Kerr hysteresis loops with full remanence. The reorientation of the easy axis from perpendicular to in plane and the associated phase diagram for this system were investigated previously.[14] Here we will concentrate solely on the magnetic phase transition in the vicinity of T_C.

Polar hysteresis loops for a 2.5-ML film at different temperatures are shown in Fig. 2. Note that the polar signal results from the magnetization component perpendicular to the film plane. At low temperature, the loop is relatively square in shape and has almost full remanence, indicative of a well-established spontaneous magnetization perpendicular to the film plane. As the temperature increases, the remanence decreases and finally vanishes. We operationally define the temperature T_C^* at which the magnetic remanence vanishes as the Curie temperature. The relation between T_C and Fe film thickness then is determined by scanning the laser beam across the wedge at different temperatures to find out at what thickness the magnetic remanence vanishes. The result is shown in Fig. 3. As expected, T_C increases with increasing Fe thickness.

To study the magnetic phase transition in the vicinity of T_C we recorded the magnetic remanence for two Fe

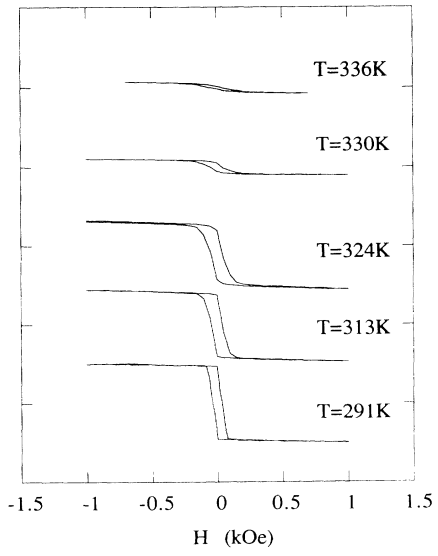

FIG. 2. Polar hysteresis loops for a 2.5-ML Fe film at different temperatures.

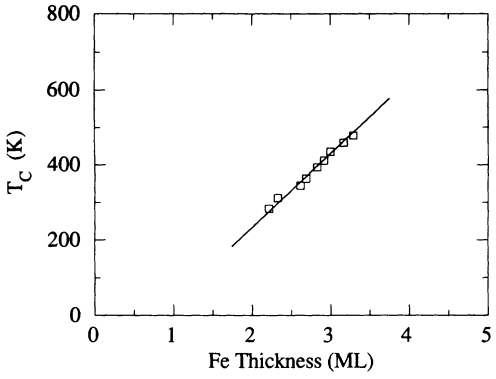

FIG. 3. Thickness dependence of T_C as a function of Fe film thickness.

films (2.5 and 2.7 ML) at different temperatures. The results are plotted in Fig. 4. It is obvious that the magnetization undergoes a second-order phase transition. Since the Kerr intensity is proportional to the magnetization M we can fit our data to the power law $M(T) \propto (1-T/T_C)^{\beta_c}$ in the vicinity of $T \leq T_C$, with T_C and β_c as fitting parameters. The results are shown as the solid lines in Fig. 4. As can be seen from Fig. 4, the power law fits the data except for the tail that is observed above T_C. The existence of a tail above T_C is found frequently in magnetic thin-film studies and is attributed to finite-size effects.[18] The finite size of the film limits the divergence of the correlation length and broadens the transition. The tail occupies a region $(T_C^*-T_C)/T_C \sim a/l$, where a is the lattice parameter and l is the length scale of the coherent regions of the film. The 3–5% tails above T_C imply a finite size of $l \sim 100$ Å. This value for l coincides with the typical terrace width of a metallic single-crystal surface. In addition, the fact that the value of l is much greater than the Fe film thickness ensures the two-dimensionality of the system.

To characterize the power law further, we constructed a log-log plot of the remanent magnetization vs reduced temperature (Fig. 5). The straight line in Fig. 5 depicts the power law and the slope yields an effective critical exponent $\beta=0.124\pm0.002$, in remarkable agreement with the Ising value of $\frac{1}{8}$. Regardless of the significant change in the T_C value between the 2.5- and 2.7-ML Fe films, $M(T)$ for the two films lies on the same straight line in the log-log plot. This is due to the universal character of the phase transition. It should be mentioned that the linear fitting in the log-log plot was performed by using data within 10% of T_C. This implicitly would suggest that critical fluctuations dominate $M(T)$ even 10% away from T_C. By using smaller temperature ranges for the fittings we obtain $\beta=0.123\pm0.003$ within 5% of T_C and $\beta=0.114\pm0.008$ within 2% of T_C. For intervals less than 2% of T_C, we do not have enough confidence to continue the procedure because there are so few data points and the systematic errors increase significantly.

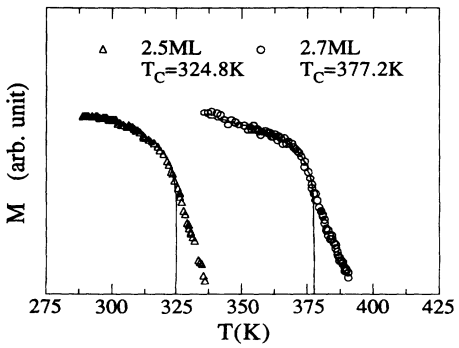

FIG. 4. Magnetic remanence of 2.5- and 2.7-ML Fe films as a function of temperature. The solid lines depict the theoretical power law $M(T) \propto (1-T/T_C)^{\beta_c}$, where T_C and β_c are fitting parameters.

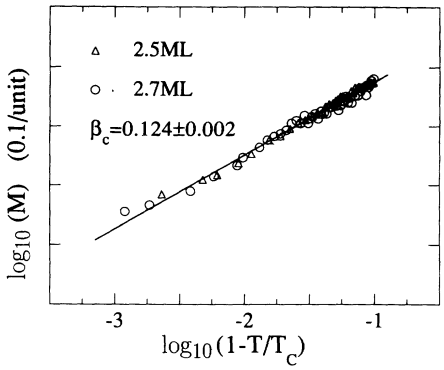

FIG. 5. Log-log plot of the magnetic remanence of the 2.5- and 2.7-ML Fe films from Fig. 4 as a function of reduced temperature. The straight solid line represents the power law. The slope of the line gives the value for the effective critical exponent of 0.124±0.002, which is remarkably close to the Ising value of 0.125.

IV. DISCUSSION

The methodology to determine β is worthy of discussion. There are two difficulties in the power-law fitting procedure. First, there is the problem that T_C itself is a fitting parameter, though a constrained one, due to the tail in $M(T)$. A small change in the assignment of the T_C value will, in general, affect the value of β. Recently, Farle, Lewis, and Baberschke determined the T_C value independently for a thick Gd film with an accuracy of ±0.1 K using the measured divergence in the magnetic susceptibility.[19] Then they fitted their Gd data to the power law with β as the only free parameter. Their result agrees very well with the 3D Heisenberg value for β, as anticipated. Unfortunately, their method is not very practical for ultrathin films because effects like the diamagnetism of the substrate tend to produce a background that limits the applicability of the method.

Another problem with the fitting methodology is that the power law should be valid only within a certain critical region ΔT below T_C. However, while the critical exponent β_c is a universal parameter, ΔT is not. Thus, the criterion adopted to fix a value for ΔT introduces uncertainties in the final analysis. In general, ΔT is expected

from theory to be larger for a system with a shorter correlation length.[20] For example, in a conventional superconductor, the large physical size of the Cooper pairs yields a ΔT(<1 mK) that is vanishingly small.[20] That is why superconducting critical phenomena have not been studied until the advent of the new short-correlation-length ceramic oxides. But it is not clear what the precise range of ΔT should be for a 2D magnetic thin film. Qualitatively, since the exchange interaction acts over (short) nearest-neighbor distances, ΔT should be "large." But, to attempt to quantify ΔT, either empirical guidelines are adopted, as in the analysis embodied in Fig. 5, or a realistic calculation needs to be performed to get at ΔT. Such a calculation would provide a welcome guide to experimentalists confronted with data-analysis tasks.

Kohlhepp *et al.*[7] recently used a clever approach to obtain insight into the problem. They used the full solution of the 2D Ising model that described $M(T)$ over its entire temperature range to determine the interval ΔT over which the power-law representation rigorously describes $M(T)$ near T_C. But the physical principle underlying this approach needs clarification. Recall that near T_C these epitaxial overlayer systems of interest (case I) have phase transitions that are of the 2D Ising universality class. But, the systems themselves are not 2D Ising systems; they are relatively ideal realizations of 2D Heisenberg systems with anisotropy. Thus, the literal use of a 2D Ising model to extract ΔT naturally has its limitations. For example, for $T \ll T_C$, while the 2D Ising solution gives an exponential form for $M(T)$, spin-wave excitations in 2D magnetic thin films usually give a quasi-linear $M(T)$. But, operationally, it is interesting to see where the approach of Kohlhepp *et al.*[7] leads. They find that, for example, over the intervals $\Delta T/T_C = 10\%$, 5%, and 2%, a force fit to the power-law expression can be used to extract an effective β value (β_{eff}) from the full Ising model $M(T)$ curve. The results are that $\beta_{\text{eff}} < \beta_c$, but $\beta_{\text{eff}} \rightarrow \beta_c$ as the fitting interval $\Delta T/T_C$ is reduced. $\beta_{\text{eff}} = 0.115$, 0.120, and 0.122 for 10%, 5%, and 2% intervals, respectively, while the ideal value of β_c, of course, is 0.125. This would suggest that the effective β value that we obtain in the present study of 0.124 ± 0.002 is closer to the Ising value of 0.125 than might otherwise be expected. It might be more proper for us to compare our determinations to the "effective" values quoted above, in which case the agreement remains quite good. But, more importantly, we can still conclude that our *Fe/Ag(100) phase transition belongs to the 2D Ising universality class.*

The exercise outlined by Kohlhepp *et al.*[7] illustrates that the value of $\Delta T/T_C \sim 10\%$ is beyond the rigorous confines of the critical region of the Ising model. But this is a formal exercise in the properties of mathematical functions. The physics that presents itself in our system includes a 3–5 % tail in the vicinity of T_C. This makes T_C itself somewhat indeterminate in that the critical fluctuations cannot diverge macroscopically; they are bounded by finite-size effects. Thus, rather than use the ideal Ising model, it might make more sense to follow the above methodology using a finite-size Ising model. For this purpose we can use Landau's simulations of finite-size 2D Ising models.[18] For the intervals within 2–10 % of T_C the effective "finite-size" β values (β_{FS}) deviate from the ideal Ising value such that $\beta_{\text{FS}} > \beta_c$. We believe that for the purposes of interpreting our data, the finite-size consideration that leads to $\beta_{\text{FS}} > \beta_c$ should outweigh the argument of Kohlhepp *et al.*[7] that led to the conclusion that $\beta_{\text{eff}} < \beta_c$. These opposing inequalities might help to explain the remarkable agreement of our experimental data with the ideal Ising value of β_c. It is obvious from this discussion that the subject of the width of the critical region ΔT would benefit from further investigations.

V. SUMMARY

Fe(100) films and wedges were grown onto a Ag(100) substrate at room temperature. RHEED and LEED patterns and RHEED intensity oscillations confirm the epitaxial nature of the growth. The SMOKE technique was applied to study the magnetic phase transition near the Curie temperature of the system. We found that T_C increases approximately linearly with Fe thickness in the limited region between ~ 2.2 and 3.4 ML Fe from a value near room temperature to just under 500 K. The temperature dependence of the magnetization near T_C follows a universal power law, $M(T) \propto (1 - T/T_C)^{\beta_c}$, with an effective exponent 0.124 ± 0.002 obtained within 10% of T_C, in agreement with the 2D Ising value of $\frac{1}{8}$.

ACKNOWLEDGMENTS

This work was supported by U.S. Department of Energy, Basic Energy Sciences–Materials Sciences, under Contract No. W-31-109-ENG-38. We benefited from correspondence with Professor U. Gradmann.

*Present address: Department of Physics, University of California, Berkeley, Berkeley, CA 94720.

[1] M. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[2] M. Bander and D. L. Mills, Phys. Rev. B **38**, 12 015 (1988).
[3] J. V. José, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, Phys. Rev. B **16**, 1217 (1977).
[4] W. Dürr, M. Taborelli, O. Paul, R. Germar, W. Gudat, D. Pescia, and M. Landolt, Phys. Rev. Lett. **62**, 206 (1989).
[5] C. Liu and S. D. Bader, J. Appl. Phys. **67**, 5758 (1990).
[6] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **67**, 1646 (1991).
[7] J. Kohlhepp, H. J. Elmers, S. Cordes, and U. Gradmann, Phys. Rev. B **45**, 12 287 (1992).
[8] F. Huang, G. J. Mankey, M. T. Kief, and R. F. Willis, J. Appl. Phys. **73**, 6760 (1993).
[9] R. L. Fink, C. A. Ballentine, J. L. Erskine, and J. A. Araya-Pochet, Phys. Rev. B **41**, 10 175 (1990).
[10] Y. Li and K. Baberschke, Phys. Rev. Lett. **68**, 1208 (1992).
[11] C. Rau, Appl. Phys. A **49**, 579 (1989).

[12]S. T. Bramwell and P. C. W. Holdsworth, J. Phys. Condens. Matter **5**, L53 (1993).

[13]B. T. Jonker, K.-H. Walker, E. Kisker, G. A. Prinz, and C. Carbone, Phys. Rev. Lett. **57**, 142 (1986); B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, *ibid.* **59**, 1756 (1987); M. Stampanoni, A. Vaterlaus, M. Aeschlimann, and F. Meier, *ibid.* **59**, 2483 (1987); J. Araya-Pochet, C. A. Ballentine, and J. L. Erskine, Phys. Rev. B **38**, 7846 (1988).

[14]Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **70**, 1006 (1993).

[15]J. A. Stroscio, D. T. Pierce, and R. A. Dragoset, Phys. Rev. Lett. **70**, 3615 (1993).

[16]J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **67**, 140 (1991); S. T. Purcell, W. Folkerts, M. T. Johnson, N. W. E. McGee, K. Jager, J. ann de Stegge, W. B. Zeper, W. Hoving, and P. Grünberg, *ibid.* **67**, 903 (1991); Z. Q. Qiu, J. Pearson, A. Berger, and S. D. Bader, *ibid.* **68**, 1398 (1992).

[17]J. Zak, E. R. Moog, C. Liu, and S. D. Bader, J. Magn. Magn. Mater. **89**, 107 (1990); S. D. Bader, *ibid.* **100**, 440 (1991).

[18]D. P. Landau, Phys. Rev. B **13**, 2997 (1976).

[19]M. Farle, W. A. Lewis, and K. Baberschke, Appl. Phys. Lett. **62**, 2728 (1993).

[20]V. L. Ginsburg, Fiz. Tverd. Tela (Leningrad) **2**, 2031 (1961) [Sov. Phys. Solid State **2**, 1824 (1961)].