Magnetometry of the Ferromagnetic Monolayer Fe(110) on W(110) Coated with Ag

H. J. Elmers, G. Liu, and U. Gradmann

Physikalisches Institut, Technische Universitaet Clausthal, D-3392 Clausthal-Zellerfeld, Federal Republic of Germany
and Sonderforschungsbereich SFB-126, D-5300 Bonn, Federal Republic of Germany

Magnetic moments per atom in the thermodynamically stable pseudomorphic monolayer Fe(110) on W(110), coated with Ag, were determined by magnetometry in UHV above 220 K. Extrapolation to $T=0$ using temperature-dependent magnetic hyperfine fields, measured by conversion-electron Mössbauer spectroscopy, results in ground-state moments enhanced by $(14 \pm 5)\%$ in comparison with bulk Fe.

The strong progress of self-consistent band theories in calculating the ground-state properties of ultrathin ferromagnetic films down to a monolayer[1] forms a challenge for experimental research on magnetic films in the monolayer regime, in order both to test these theoretical predictions and to study the basic phenomenon of two-dimensional magnetic order as a whole. Accordingly, ferromagnetism in ultrathin epitaxial Fe films on Cu(110),[2,3] Ag(100),[4-6] Au(100),[7] and W(110)[8-10] has been studied recently using advanced experimental techniques such as spin-polarized photoemission,[2,4] spin-polarized LEED,[7] conversion-electron Mössbauer spectroscopy (CEMS),[5,8-10] and Kerr magnetometry.[3] Reduced Curie temperatures and perpendicular anisotropies, which were detected previously in NiFe and Co films in a Cu matrix,[11,12] could be detected in these Fe films too.[3,4,6] Critical exponents were reported for Fe(100) films on Au(100),[7] and hysteresis for a monolayer of Co on Cu(100).[13] However, despite all of these sophisticated experiments, one most simple and basic piece of information is missing: The magnetic moment per atom in a Fe monolayer, which forms a central message of the extended theoretical work on monolayer magnetism,[1,14] has never been measured. The present paper reports a magnetometric study of the pseudomorphic monolayer Fe(110) on W(110), coated with Ag, providing just this magnetic moment per atom, for the first time. New information on unusual properties of the phase transition in this two-dimensional ferromagnet is included.

As for all other systems mentioned above, the monolayer Fe(110) is pseudomorphic to the W(110) substrate,[15] thus providing a common two-dimensional translational symmetry with the substrate. The homogeneous pseudomorphic strain caused by the negative misfit, $-f_{FeW}=9.4\%$ ($a_{Fe}=2.866$ Å; $a_W=3.165$ Å), imposes a strong but well defined deviation from bulk atomic distances. The unique advantage, however, of the Fe(110) monolayer on W(110) is its thermodynamic stability, which is caused by the high surface energy of the substrate ($\gamma_W=2.9$ Jm^{-2}) (Ref. 16) in comparison with that of the film ($\gamma_{Fe}=2.0$ Jm^{-2}) (a general discussion of monolayer stabilities is given in Ref. 17). The per-

sistence of this stability, despite the strong pseudomorphic strain energies, has been shown recently[8-10] by the fact that the Curie temperature of the Ag-coated monolayer, $T_{C,mono}=282$ K, is independent of annealing, up to 800 K, before coating. Even more, if Fe(110) monolayers are prepared on W(110) at 200 °C or above, T_C retains the monolayer value of 282 K in the submonolayer regime too, down to half a monolayer.[9] Obviously, attractive interactions between the Fe atoms on W(110) result in recrystallization into large-area monolayer patches, each of them representing the extended monolayer.[9] Accordingly, small deviations of the mean thickness from a monolayer do not disturb the magnetic monolayer properties. The stability of the present monolayer structure forms an appropriate precondition for straightforward magnetic analysis. Contrarily, disturbing tendencies for island formation or instabilities on annealing were observed in Fe films on Ag,[5] Cu,[18] and Au,[19] in the monolayer regime, as expected from the comparatively low surface energies of the substrates ($\gamma_{Ag}=1.1$ Jm^{-2}, $\gamma_{Cu}=1.6$ Jm^{-2}, $\gamma_{Au}=1.4$ Jm^{-2}). This imposes severe problems on straightforward magnetic monolayer analysis in these systems.

Preparation of Fe films on W(110) and magnetometry *in situ* were done in an UHV-compatible torsion oscillation magnetometer (TOM) described previously.[20] Fe films were prepared at pressures below 10^{-10} Torr and a preparation temperature $T_p=500$ K. The film thickness was measured using a quartz-oscillator monitor with a resolution better than 3% of a monolayer; it is given in the following in units of bulk monolayer equivalents, D (note that the pseudomorphic monolayer is given by $D=0.82$). Films were coated with Ag immediately after preparation, to prevent contamination.

For magnetometry, the sample is suspended on a torsion filament in a homogeneous magnetic field H; we measure the frequency of oscillation, ω, as a function of H and derive a (total) torque constant $R_{tot}=I\omega^2$ (moment of inertia I). The magnetic contribution R is determined by the magnetic moment m of the sample and by its anisotropy energy F, which for the present film geometry is given to a good approximation by $F=L_sA\cos^2\theta$, where θ is the angle between magnetiza-

tion J_s and film normal n, A the film area, and L_s an anisotropy constant (energy per area). R is connected with m, L_s, A, and H by

$$1/R = 1/mH + 1/2L_sA . \qquad (1)$$

Using an anisotropy field $H_L = 2L_sA/m$, Eq. (1) can be written alternatively as

$$m = (R/H)(1 + H/H_L) . \qquad (2)$$

In the low-field limit, $H \ll H_L$, J_s is locked to the sample, resulting in $R = mH$; in the high-field limit $H \gg H_L$, J_s is locked to the field and $R = 2L_sA$ is determined by the anisotropies only. A quantitative determination of both the magnetic moment m and the anisotropy field H_L results from a plot of $1/R$ vs $1/H$, which is shown in Fig. 1 for a sample with $D = 0.79$, just below the pseudomorphic monolayer value $D = 0.82$, measured at $T_m = 220$ K, giving $m = 7.0 \times 10^{-15}$ V s m from the slope and $\mu_0 H_L = 5.0 \pm 0.6$ T from the axial intersection. The excellent linearity proves excellent ferromagnetism of the sample at 220 K. The external field is directed along [1$\bar{1}$0], because the monolayer, like films below $D = 30$, is magnetized spontaneously along this direction as a result of strong in-plane surface anisotropies.[21] $\mu_0 H_L$ is composed of a shape anisotropy of roughly 2 T and a crystalline contribution of a further 3 T, which assists the shape anisotropy, contrary to the more spectacular case of perpendicular anisotropies,[3,4,6,11,12] which counteract the shape anisotropy. As a result of the high anisotropy field of 5 T, R/H equals m to a very good approximation;

compare Eq. (2).

Because the sample is suspended on the torsion filament during measurement, and thus thermally decoupled from the surroundings, measurements below room temperature had to be done at constant field H with slowly drifting temperatures ≥ 220 K (the measurement of T was done using eddy-current damping of the sample[20]). Measurements of this type are shown in Fig. 2 for the sample ($D = 0.79$) of Fig. 1. Note that R/H equals m to a good approximation; compare Eq. (2). The phase transition from the ferromagnetic to the paramagnetic state is indicated by the sharp drop of R/H with an inflection point at a temperature $T_{infl} = 291 \pm 1$ K which is independent of H. This is contrary to two-dimensional phase transitions in K_2CuF_4 layered compounds,[22] where T_{infl} increases monotonically with H, as in bulk materials. Further, T_{infl} is definitely higher than $T_{C,mono} = 282 \pm 3$ K as determined by Mössbauer spectroscopy.[9] Apparently, T_{infl} represents something like a critical temperature for saturation, different from $T_{C,mono}$, which in turn represents the onset of spontaneous magnetization. This unusual property of the phase transition
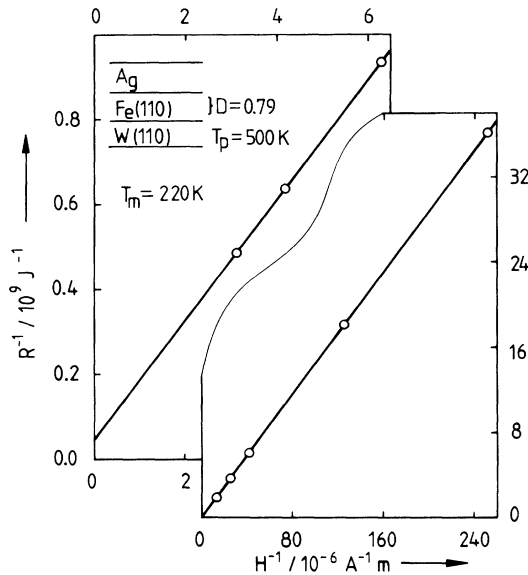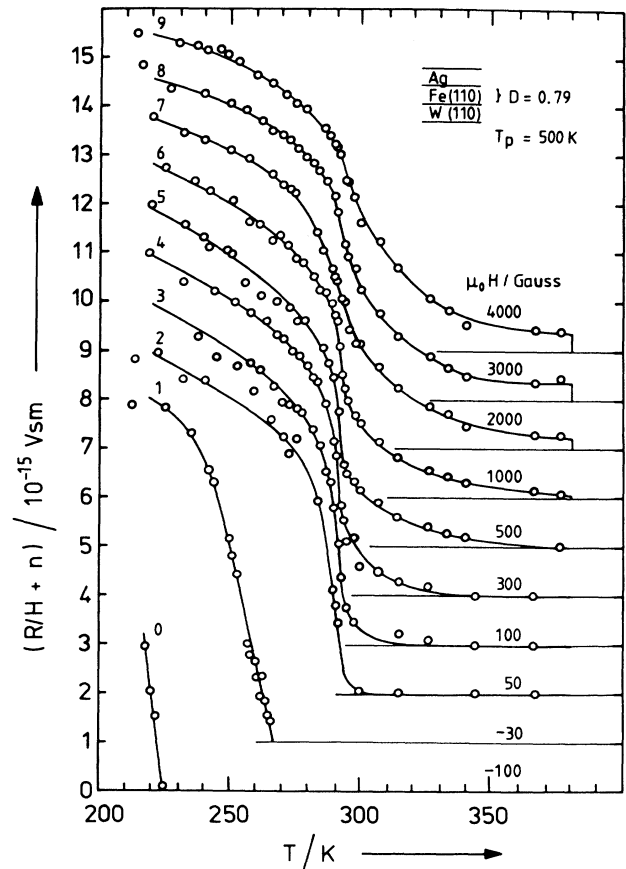

FIG. 1. $1/R$ vs $1/H$ for a pseudomorphic monolayer W(110)/0.79/Ag, consisting of 0.79 bulk monolayers of Fe(110) on Ag, prepared at $T_p = 500$ K, and measured at $T_m = 220$ K. Note the different scales for low fields (lower part) and high fields (upper part), respectively.

FIG. 2. R/H vs T, for constant magnetic fields as indicated, for the film of Fig. 1. Curves are shifted along the vertical axis for clarity.

remains to be explained. We suppose there is a connection with the strong surface-type anisotropies in the film;[21] the influence of the real structure of the substrate must be considered too. However, the transition shows the usual smearing by finite fields. For a further understanding of the transition, isothermal magnetization curves R/H vs H, obtained by replotting the data of Fig. 2, are shown in Fig. 3. A square magnetization curve is observed at $T = 220$ K $= 0.75 T_{C,\text{mono}}$ and remanent magnetization up to 270 K $= 0.96 T_{C,\text{mono}}$ [note that the decrease of R/H with increasing H, e.g., for $T = 220$ K, is simply a result of finite anisotropy; compare Eq. (2)]. Magnetic saturation at moderate fields is observed near $T_{C,\text{mono}}$ and indicated even up to the highest temperature, 380 K $= 1.35 T_{C,\text{mono}}$. For comparison, in bulk Fe, the J_s vs H curves become linear again at $T = 1.01 T_c$.[23] The extended width is a common feature of two-dimensional transitions; the indication of saturation far above $T_{C,\text{mono}}$ is an unusual feature of the present system. The correction of R/H for m, compare Eq. (2), does not change these qualitative findings; it was not performed because the temperature dependence of $\mu_0 H_L$ is not known.

Besides the critical properties near $T_{C,\text{mono}}$, we are interested mainly in the magnetic moment per atom, μ. It results from the magnetic moment of the sample, m, measured by TOM as shown in Figs. 2 and 3, divided by the number of atoms, determined from the quartz monitor and the film area. Separate calibrations of TOM (by determining the moment of inertia), of the quartz monitor, and of the film area resulted in an overall uncertainty of 10% for μ. In a second independent calibration, we used thick films, for which $\mu = \mu_{\text{bulk}}$ could be assumed, as calibration standards, resulting in an improved accuracy of 3% for μ. Both methods agreed within the limits of accuracy; the second one is used in the following.
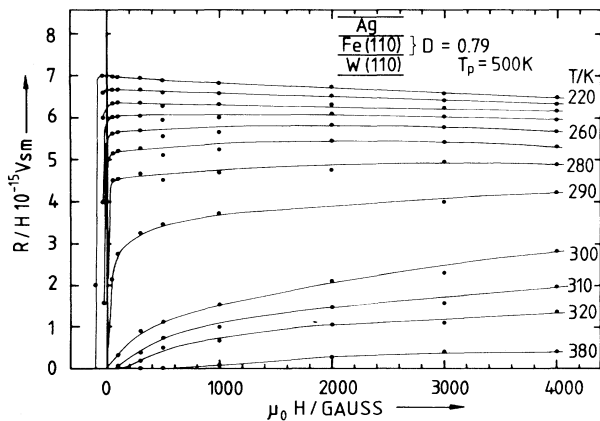
At the lowest temperature of measurement, $T = 220$

K, the magnetic moment per atom in sample $D = 0.79$ is found to be $\mu = (1.89 \pm 0.06)\mu_B$. Fortunately, μ can be extrapolated to $T = 0$ using Mössbauer data as shown in Fig. 4. The figure shows the remanent moment per atom for 220 K $\leq T \leq$ 250 K, temperatures where it could be determined reliably from Fig. 3, versus T, in comparison with the mean hyperfine field $B_{\text{hf}}(T)$ in a pseudomorphic monolayer $(D = 0.82)$ prepared at 475 K. Both quantities are normalized at $T = 220$ K; obviously, their temperature dependences agree in the limited temperature range available. This common temperature dependence of μ and B_{hf} is well established in bulk materials,[24] it has been confirmed recently[25] for ultrathin Fe(110) films on Ag(111), and it is reasonable because all contributions to B_{hf} are caused by μ^1 and therefore should scale with it. Assuming $\mu(T) \sim B_{\text{hf}}(T)$ for $T < 220$ K too, we could extrapolate μ to $T = 0$, as shown in Fig. 4. Taking into account some uncertainty in the extrapolation to $B_{\text{hf}}(0)$, we obtain $\mu(0) = (2.51 \pm 0.12)\mu_B$ for the film of Figs. 1–4. Taking the mean value from four similar films results finally in $\mu(0) = (2.53 \pm 0.12)\mu_B$ for the pseudomorphic monolayer of Fe(110) on W(110), coated with Ag. It is enhanced by 14% with respect to the bulk value $\mu_{\text{bulk}} = 2.22\mu_B$. In a similar way, we determined $\mu(0) = (2.46 \pm 0.15)\mu_B$ for the pseudomorphic double layer.

In sharp contrast to the enhancement of $\mu(0)$, the hyperfine field in this monolayer is reduced to $B_{\text{hf}}(0) = 11.9$ T $= 0.34 B_{\text{hf}}(\text{bulk})$.[8] Obviously, in different systems, e.g., in the monolayer versus bulk material, B_{hf} can be taken as a measure of μ only with respect to its temperature dependence, but definitely not with respect to its magnitude.

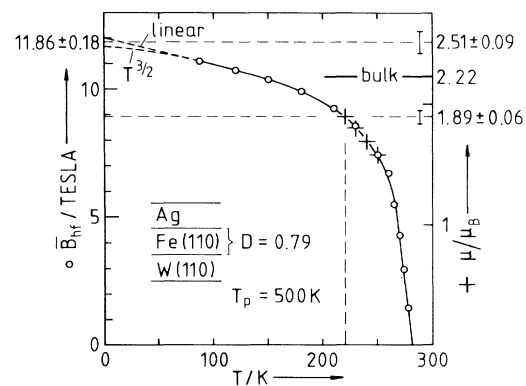Band-structure calculations just for our pseudomorph-

FIG. 3. Isothermal magnetization curves R/H vs H, obtained by replotting Fig. 2. Note that $m = (R/H)(1 + H/H_L)$ differs slightly from R/H [Eq. (2)].

FIG. 4. Magnetic moment per atom in the remanent state, in units of Bohr magnetons, μ/μ_B (+), vs T for the monolayer film of Figs. 1–3 in comparison with the mean hyperfine field B_{hf} (○) of a film W(110)/0.82/Ag, determined by CEMS. $B_{\text{hf}}(0) = 11.86 \pm 0.18$ T is taken as the mean value between a $T^{3/2}$ and a linear extrapolation. B_{hf} and μ/μ_B are normalized at $T = 220$ K.

ic monolayer Fe(110) on W(110), coated with Ag(111), even with consideration of relaxation of atomic layer distances to minimize the total energy, became available very recently,[26] after submission of this paper. In rough agreement with our experiments, theory yields $\mu(0) = 2.17\mu_B$ and $B_{hf}(0) = 9.3$ T. The slight enhancement of the experimental $\mu(0)$ is not reproduced by the theory. Similarly the theoretical $B_{hf}(0)$ is slightly smaller than the experimental one. The explanation of the small differences between theory and experiment remains as a task for future research.

In conclusion, we have shown that magnetic monolayers are accessible to straightforward magnetometric analysis. The ground-state magnetic moment per atom in the thermodynamically pseudomorphic monolayer of Fe(110) on W(110), coated with Ag, is enhanced by 14% in comparison with its value in bulk Fe. Interesting unusual features of the phase transition near $T_{C,mono} = 282$ K were observed.

[1]A. J. Freeman and C. L. Fu, J. Appl. Phys. **61**, 3356 (1987), and references therein.

[2]D. Pescia, M. Stampanoni, G. L. Bona, A. Vaterlaus, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 2126 (1987).

[3]C. Liu, E. R. Moog, and S. D. Bader, Phys. Rev. Lett. **60**, 2422 (1988).

[4]M. Stampanoni, A. Vaterlaus, M. Aeschlimann, and F. Meier, Phys. Rev. Lett. **59**, 2483 (1987).

[5]N. C. Koon, B. T. Jonker, F. A. Volkening, J. J. Krebs, and G. A. Prinz, Phys. Rev. Lett. **59**, 2463 (1987).

[6]B. Heinrich, K. B. Urquhart, A. S. Arrot, J. F. Cochran, K.

Myrtle, and S. T. Purcell, Phys. Rev. Lett. **59**, 1756 (1987).

[7]W. Duerr, M. Taborelli, O. Paul, R. Germar, W. Gudat, D. Pescia, and M. Landolt, Phys. Rev. Lett. **62**, 206 (1989).

[8]M. Przybylski and U. Gradmann, Phys. Rev. Lett. **59**, 1152 (1987).

[9]M. Prsybylski and U. Gradmann, J. Phys. C **8**, 1705 (1988).

[10]M. Przybylski, I. Kaufmann,. and U. Gradmann (to be published).

[11]U. Gradmann and J. Mueller, Phys. Status Solidi (b) **27**, 313 (1968).

[12]U. Gradmann, Appl. Phys. **3**, 161 (1974).

[13]T. Beier, H. Jahrreiss, D. Pescia, Th. Woike, and W. Gudat, Phys. Rev. Lett. **61**, 1875 (1988).

[14]J. Noffke and L. Fritsche, J. Phys. C **14**, 89 (1981).

[15]U. Gradmann and G. Waller, Surf. Sci. **116**, 539 (1982).

[16]L. Z. Mezey and J. Giber, Surf. Sci. **117**, 220 (1982).

[17]E. Bauer and J. H. van der Merwe, Phys. Rev. B **33**, 3657 (1986).

[18]D. A. Steigerwald and W. F. Egelhoff, Phys. Rev. Lett. **60**, 2558 (1988).

[19]R. Germar, W. Dürr, J. W. Krewer, D. Pescia, and W. Gudat, Appl. Phys. **A47**, 393 (1988).

[20]R. Bergholz and U. Gradmann, J. Magn. Magn. Mater **45**, 389 (1984).

[21]U. Gradmann, J. Korecki, and G. Waller, Appl. Phys. A **39**, 101 (1986).

[22]K. Hirakawa and K. Ubukoshi, J. Phys. Soc. Jpn. **50**, 1909 (1981).

[23]K. Heyner and R. Kohlhaas, Z. Agnew. Phys. **26**, 80 (1969).

[24]I. Vincze and J. Kollar, Phys. Rev. B **6**, 1066 (1972).

[25]G. Lugert and G. Bayreuther, Phys. Rev. B **38**, 11068 (1988).

[26]Soon C. Hong, A. J. Freeman, and C. L. Fu, Phys. Rev. B **38**, 12156 (1988).