Power laws of magnetization in ferromagnetic monolayers and the two-dimensional Ising model

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The temperature-dependent magnetization in anisotropic and in particular in uniaxial ferromagnetic monolayers should be compared with the exact solution of the two-dimensional Ising model rather than with the popular asymptotic power law, which equals the exact solution only in a very narrow critical range of temperatures. The saturation magnetization of a perpendicularly magnetized monolayer of Co on Cu(111) follows the Ising model in an extended range of temperatures.

I. INTRODUCTION

The two-dimensional Ising model (2DIM) and its exact solution given by Onsager[1] and Yang[2] is a cornerstone in the discussion of ultrathin ferromagnetic films. Its most popular result is that, near the Curie temperature T_C, the magnetization follows an asymptotic power law $M(T)\sim(T_C-T)^{\beta}$, with a critical exponent $\beta=\frac{1}{8}$. In recent experimental papers on ferromagnetic ultrathin films and monolayers,[3-6] their magnetization below the Curie temperature has been discussed in comparison with this critical power law. In the present paper, we will show that it makes more sense to compare with the exact solution of the 2DIM and that one should compare the saturation moment rather than the remanent moment with the model, as the two moments are not equal in real films. In addition, it will be shown that the saturation magnetization of a perpendicularly magnetized monolayer of Co(111) on Cu(111) fits the 2DIM to a remarkable approximation.

II. USE OF THE TWO-DIMENSIONAL ISING MODEL FOR ULTRATHIN MAGNETIC FILMS

The Ising model represents a ferromagnet with infinite uniaxial anisotropy. It is useful for the discussion of ultrathin films because strong magnetic anisotropies are common in ultrathin ferromagnetic films and monolayers (ML). Anisotropy energies of the order of a few 0.1 meV/atom, roughly 10^{-3} times the exchange constant J, corresponding to anisotropy fields of several T, are common for ferromagnetic surfaces and interfaces.[7] Anisotropies of the same order of magnitude have been measured in ferromagnetic monolayers of $Ni_{48}/Fe_{52}(111)$ on Cu (Ref. 8) and of Fe(110) on W(110).[9-11] In the case where the anisotropies favor out-of-plane magnetization, they may compensate shape anisotropy and result in spontaneous perpendicular magnetization, which has been observed in the monolayer regime for $Ni_{48}/Fe_{52}(111)$ in Cu(111),[8] Co(111) on Cu(111),[12] α-Fe(100) on Ag(100),[13] α-Fe(110) on Au(111),[14] Co on Au(111),[15] γ-Fe(111) on Cu(111) (Ref. 16) and Ag(111),[5] and γ-Fe on Cu(100).[17,5] The tight connections of these strongly uniaxial films with the 2DIM are obvious. In addition, it turns out that any two-dimensional anisotropic Heisenberg system becomes Ising-like near T_C. This was shown by Binder and Hohenberg[18] using Monte Carlo simulations and has been proven rigorously by Bander and Mills[19] using renormalization-group analysis. Long-range order in ideal ultrathin films can occur only if it is triggered by anisotropies or by extended fields, as shown rigorously by Mermin and Wagner[20] and discussed recently in detail by Erickson and Mills.[21] In this sense, any ferromagnetic monolayer which structurally represents the 2-d-translational symmetric models becomes Ising-like in some critical region. The question, of course, is the extension of this critical region and whether it can be observed near T_C of real monolayers.

The 2DIM provides interesting information on the extension of the critical regime in magnetic monolayers, as can be seen from Fig. 1, which shows a comparison of the exact solution for the spontaneous magnetization of the 2DIM,

$$M(T)=[1-\sinh^{-4}(2J/k_BT)]^{1/8}\ , \tag{1}$$

with its asymptotic power-law approximation,

$$\tilde{M}(T)=1.22(1-T/T_C)^{1/8}\ , \tag{2}$$

which equals $M(T)$ as $T\rightarrow T_C$. Visual inspection shows clear deviations outside a critical region of only a few percent of T_C. In order to characterize the meaning of

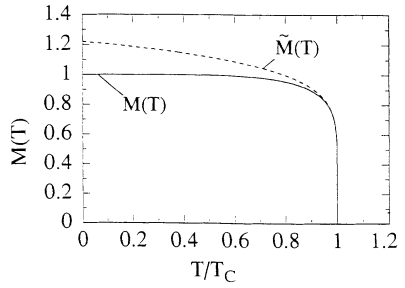

FIG. 1. Normalized magnetization $M(T)$ vs normalized T/T_C for the two-dimensional Ising model. Exact solution $M(T)=[1-\sinh^{-4}(2J/k_BT)]^{1/8}$ in comparison with the asymptotic power law $M(T)=1.22(1-T/T_C)^{1/8}$.

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any power-law approximation more quantitatively, and in the sense of a procedure used frequently in the evaluation of experiments, we fitted the exact solution in finite intervals $\tau_i < \tau < \tau_f$ of the reduced temperature $\tau = T/T_C$ by approximate power laws $M^* = A(1-T/T_C^*)^{\beta}$. Figure 2 shows the resulting parameter β^* for $\tau_f = 0.99$ and 0.999, respectively, as a function of τ_i. (T_C^* deviates from T_C by less than 1%, for the parameters of the figure; it is therefore not shown.) Obviously, in order to avoid deviations of β^* from $\beta = 0.125$ by more than say 2%, one should not exceed a critical region of 1% of T_C. If the temperature interval considered is extended below roughly $0.9T_C$, the notion of a critical exponent can no longer be used.

Apparently, the narrow extension of the critical regime of the 2DIM as visualized by Figs. 1 and 2 was not realized by the authors of several recent experimental papers[3-6] on the temperature dependence of magnetic order in ferromagnetic films in the monolayer regime. Dürr *et al.*[3] analyzed α-Fe(100) films on Au(100), magnetized spontaneously in the plane, with a mean thickness between 1 and 2.5 ML. They measured the *remanent* magnetization (in relative units) using spin-polarized electron techniques, fitted it by a power-law equation (2) and reported a "critical exponent" $\beta = 0.22(5)$, independent of the film thickness, for reduced temperatures τ between 0.25 and 0.999. Ballentine *et al.*[4] analyzed Ni(111) films on Cu(111), magnetized again in the plane, consisting of 2–3 ML. They fitted the *saturation* magnetization, measured by Kerr magnetometry, and reported $\beta = 0.24(7)$ for τ between 0.3 and 0.99. Liu, Moog, and Bader[6] analyzed perpendicularly magnetized base-centered tetragonal Fe(100) films on Pd(100) between 0.6 and 3 ML. They fitted the *remanent* magnetization, measured using Kerr magnetometry, and reported $\beta = 0.15(2)$ for τ between 0.4 and 0.998. Finally, Qiu, Pearson, and Bader[5] analyzed fcc Fe(111) films on Ag(111), between 1 and 3 ML, magnetized in the plane. They fitted again the *remanent* magnetization, measured using Kerr magnetometry, and reported $\beta = 0.137(8)$ for τ between 0.82 and 0.998. In all these papers, the power-law exponents were considered as critical exponents and compared with the prediction of the 2DIM, although the temperature intervals used were far outside the critical regime of the 2DIM. Further, the reported values of β were above the Ising value $\frac{1}{8}$, instead of being below as expected from the exact solution and its approximate power-law fits, as shown in Fig. 2. Therefore, it does not make sense to discuss the observed power laws in terms of the 2DIM. They present remarkable experimental properties of a group of ultrathin films which remain to be explained.

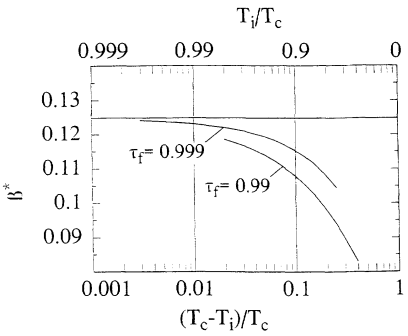

FIG. 2. Phenomenological exponents β^* of a power-law fit $M^* \sim (1-T/T_C^*)^{\beta^*}$ of the 2DIM, performed for $T_i < T < T_f$ vs $(T_C - T_i)/T_C$, with $\tau_f = T_f/T_C$ as the parameter.

The samples used in these experiments were by no means perfect monolayers. This showed up in two magnetic features which are not expected for ideal anisotropic monolayers. The first one is given by the appearance of magnetization tails above T_C, which were interpreted in terms of finite-size effects and necessitated the somewhat problematic determination of T_C (or strictly speaking T_C^* in our notation) as a parameter of the power law. The second feature is given by the deviation of the remanent magnetization M_r from the saturation magnetization M_s, which should equal in ideal films structures. This deviation, in turn, leaves it unclear whether to compare $M_r(T)$ or $M_s(T)$ with idealized models. The different choices of different authors are arbitrary. Obviously, both magnetic features reflect the unknown structural imperfections of the films in a complicated manner.

Apparently, nobody succeeded in finding ferromagnetic monolayers for which the temperature dependence of magnetization reproduces the exact solution of the 2DIM. To our knowledge the only film structure for which the properties of the 2DIM are indicated in some sense is contained in the work of Rau,[22] who found ferromagnetic order in a 5-ML-thick V(100) film on Ag(100). He reported a temperature dependence of magnetic order, measured using electron capture techniques in an external field of 100 Oe, which actually follows the exact solution of the 2DIM in the limits of accuracy for τ between 0.6 and 0.999, definitely better than the power law which Rau used for comparison.

III. MAGNETOMETRY OF A PERPENDICULARLY MAGNETIZED Co MONOLAYER ON Cu(111)

We performed some experiments with Co monolayers on Cu(111) which show remarkable accordance with the exact solution of the 2DIM. We choose this monolayer because its ferromagnetic behavior and perpendicular magnetization were known from early work[12] on ferromagnetic Co monolayers prepared on epitaxial Cu(111) films on mica. In order to avoid difficulties connected with the strong paramagnetization of the mica substrates, we switched to preparation on sapphire $(11\bar{2}0)$ surfaces. The sapphire substrates were cleaned by heating to 800 K in UHV (base pressure $< 10^{-10}$ Torr); they showed clear low-energy electron diffraction (LEED) patterns. Cu films roughly 250 nm thick were prepared on them at 800 K. The LEED pattern indicated a complete (111) texture with preference of spots in one special epitaxial orientation. Scanning electron microscopy (SEM) pictures showed Cu grains with lateral dimensions of some μm, separated by channels the area of which extended to some 10% of the film area. Accordingly, the films are far from

being perfect epitaxial structures. Nevertheless, a plot of the Auger amplitude of the growing Co film prepared on these Cu films versus film thickness showed the typical break which is usually taken as an indication of layer growth. Therefore, it can be assumed that rather good Co monolayer elements were formed on the Cu grains. They are then superimposed by other film components of ill-defined structure, situated in the channels between. The films were coated by Cu before magnetometry in order to avoid contamination from the atmosphere.

The magnetic moments and out-of-plane anisotropies of single monolayers were measured using a Torsion Oscillation Magnetometer[23] (TOM) working in air at temperatures between 10 and 700 K, capable of detecting less than 1% of a monolayer. As known from Co(100) films on Cu(100),[17,24,25] the magnetic properties of Co films in the monolayer regime are extremely sensitive to the preparation conditions. We observed this sensitivity, too. Films prepared at 450 K, in a similar mode as the previous films on mica,[12] showed roughly the same linear decrease of the magnetization with increasing temperature as observed previously, with Curie temperatures above 500 K which could not be reached without destroying the film structure by diffusion. Films prepared at 300 K showed a quite different magnetic pattern, which is documented, for the example of a film consisting of 0.97 pseudomorphic monolayers Co on Cu(111), in Fig. 3 by hysteresis loops and in Fig. 4 by the temperature dependence of the remanent moment m_r and the saturation moment m_s. Ferromagnetic hysteresis and perpendicular magnetization were observed for all temperatures up to a Curie temperature somewhere between 400 and 500 K. Three temperature intervals can be distinguished. Below 120 K, only minor loops could be observed, as shown in Fig. 3(a). The samples could not be saturated in the available fields of 0.6 T. Between 120 and 210 K, $m_r = m_s$ was observed as in Fig. 3(b). Above 210 K, m_r clearly differed from m_s, as shown in Figs. 3(c) and 4. The kinks in the temperature dependence of m_s in Fig. 4 indicate the presence of different structural elements in the films. We suppose that the smoothly varying m_s above 210 K represents the true monolayer component grown on top of the single-crystal Cu(111) grains. Apparently, additional components from the channels show up magnetically below 210 K only and disappear above 210 K by superparamagnetic or superferromagnetic instabilities. The saturation moment of the smooth monolayer component, 4×10^{-15} Vsm (1 Vsm$=7.96\times10^{8}$ G cm^3), amounts to only 55% of the moment of a complete monolayer with bulk magnetization, 7.3×10^{-15} Vsm. Band-structure calculations[26] indicate a slightly enhanced moment in the free Co(0001) surface. For our Co monolayer in Cu, a substantial reduction of the magnetic moment per atom is therefore improbable, and roughly bulklike atomic moments can be assumed. The reduced total moment of the true monolayer component then indicates that it contains only roughly 60% of the film volume, the residual 40% then being situated in the channels between the Cu(111) plateaus, in rough agreement with the estimate of their relative area from microscopical pictures.

Out-of-plane anisotropies were measured together with the magnetic moment. We use a quadratic approximation $f_L = L\cos^2\theta$ for the anisotropy energy density f_L, and replace the anisotropy constant L by the more visual anisotropy field $H_L = 2L/(\mu_0 M_s)$. (θ is the angle between M_s and the film normal.) For all temperatures, L and H_L were negative, indicating perpendicular magnetization in our notation. The magnitude of H_L vs temperature is shown in Fig. 5. Strong anisotropy fields above 0.5 T were measured up to the Curie temperature.

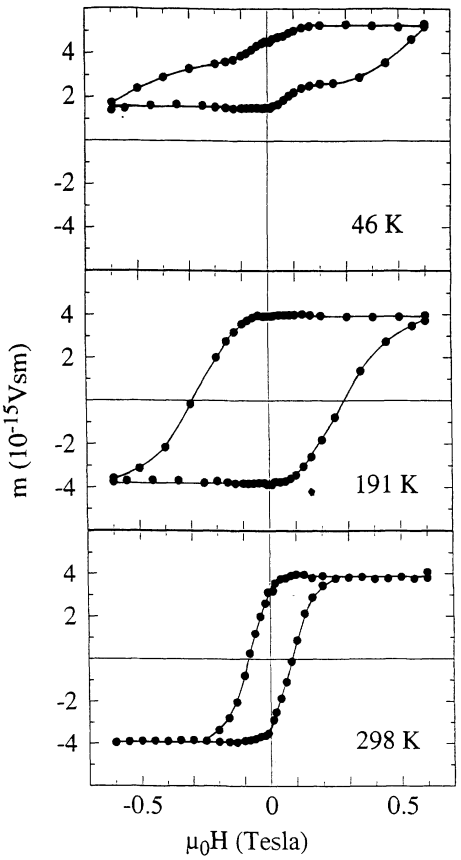

FIG. 3. Hysteresis loops for the perpendicular magnetic moment of a pseudomorphic monolayer of Co(111) on Cu(111), prepared at room temperature, coated by Cu, at temperatures of 46, 191, and 298 K, respectively. No in-plane moment could be observed at any temperature.

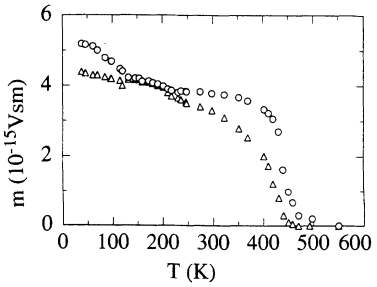

FIG. 4. Saturation moment m_s (○) and remanent moment m_r (△) vs temperature T, for the pseudomorphic Co monolayer of Fig. 3.

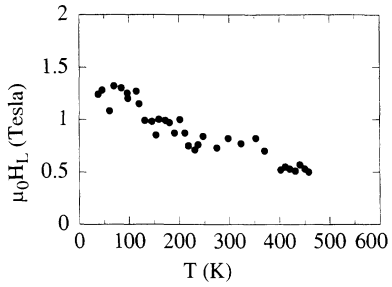

FIG. 5. Out-of-plane anisotropy field $\mu_0 H_L$ vs T for the pseudomorphic monolayer of Figs. 3 and 4.

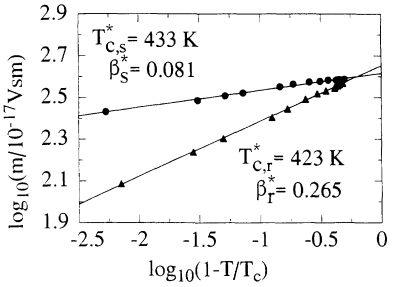

FIG. 7. Power-law fits for $m_s(T)$ (●) and $m_r(T)$ (▲) in a double-logarithmic representation.

IV. DISCUSSION

Following the interpretation that the part which remains magnetic above 210 K represents the true monolayer component, we fitted the saturation moment m_s, above 210 K, by the exact solution of the 2DIM. As shown in Fig. 6, we get an excellent fit if the open points in the tail are not used; the Curie temperature results as $T_C = 434$ K. An alternative fit by a phenomenological power law $m_s^* \sim (1 - T/T^*_{C,s})^{\beta^*_s}$ results in $T^*_{C,s} = 433$ K and $\beta^*_s = 0.081$ (Fig. 7), in excellent agreement with the expectation from Fig. 2; however, the exact solution fits the data better than the power law. The asymptotic power law $m_s \sim (1 - T/T_C)^{1/8}$, which is given for comparison, is inappropriate to explain the data. The remanent moment follows a phenomenological power law with $T^*_{C,r} = 423$ K, $\beta^*_r = 0.265$.

The remarkable result then is that the saturation magnetization of this properly prepared, yet imperfect, perpendicular magnetized monolayer follows the exact solution of the 2DIM in a wide range of temperatures, definitely far beyond the critical regime of the asymptotic power law with $\beta = \frac{1}{8}$, which obviously is inappropriate to describe the data. Accordingly, the phenomenological power-law approximation gives $\beta^*_s = 0.081 < \beta_{\text{Ising}} = 0.125$, in quantitative agreement with the data presented in Fig. 2. It might be surprising that it is m_s which follows the 2DIM instead of m_r, as assumed in several experimental papers. The preference of m_s must be seen in connection with the large out-of-plane anisotropy fields, in comparison with the external fields which are needed to saturate the samples, which were 1 order of magnitude lower, as shown in the hysteresis loop of Fig. 3(c). We suppose that the deviation of m_r from m_s, like the magnetization tails above T^*_C, result from the structural imperfections of the films. Obviously, the moderate external fields of about 0.05 T are sufficient to compensate for the small deviations of remanence from saturation. It is reasonable that these moderate fields have no essential influence on the magnetization, which is determined by the much larger anisotropy fields, in connection with the exchange interaction, and make the film Ising-like. We suppose that this forms a general rule: For the case that the remanent moment of an ultrathin film is lower than the saturation moment, but saturation is achieved in fields which are low in comparison with the anisotropy fields, one should interpret the saturation moment rather than the remanent moment in terms of spontaneous magnetization. It is a challenging question for forthcoming experiments whether in improved monolayer structures the magnetization tails and the deviations of m_r from m_s can be avoided.

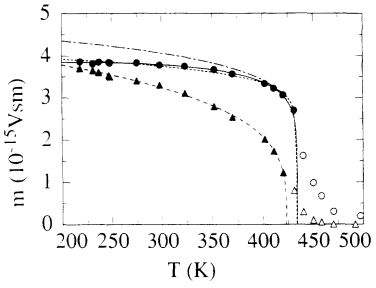

FIG. 6. Fitting of the magnetic moment of the pseudomorphic Co monolayer of Figs. 3 and 4 for temperatures above 210 K. The saturation moment m_s (●,○) is fitted by the exact solution of the 2DIM (——) with $T_C = 434$ K, and alternatively by a phenomenological power law (— — — —) with $\beta^*_s = 0.081$ and $T^*_{C,s} = 433$ K. The asymptotic power law (—·—·—·) is given for comparison. The remanent moment m_r (▲,△) is fitted by a phenomenological power law (— — —) with $\beta^*_r = 0.27$ and $T^*_{C,r} = 423$ K. Solid symbols only are used for the fits.

The measurement of the anisotropy offers the possibility to compare the measured Curie temperature with recent predictions of Erickson and Mills[21] (EM) who discussed the driving of long-range order in *sc* (100) monolayers by anisotropies. The anisotropy field of 0.5 T in our fcc(111) monolayer, just below T_C, corresponds to an anisotropy energy of 2.6×10^{-5} eV per atom. Neglecting the different symmetries, one may apply Eq. (5) of EM. Representing Co by $S = 0.85$, this results in an exchange constant of $J = 25.6$ meV/atom. Equation (4) of EM then results in $T_C^{(2)}/T_C^{(3)} = 0.22$. In view of the different symmetries, the agreement with the experimental value of $T^{(2)}_{C,s}/T^{(3)}_C = (435\text{ K})/(1388\text{ K}) = 0.31$ is surprisingly good. This, in turn, supports our interpretation of the magnetic component above 210 K.

The remanent moment follows a power law with $\beta^*_r = 0.27$ in a surprisingly wide range of temperatures, $0.5 < \tau_r < 0.99$. This is a remarkable phenomenological

result which, in our opinion, should not be discussed in terms of critical power laws. It might be seen, instead, in connection with the observation of phenomenological power laws of magnetization in other two-dimensional systems, like the monolayer examples discussed in Sec. II. Alternative examples are given by the $2d$ antiferromagnetic layered compound Rb_2MnF_4, where $\beta^*=0.18(1)$ was reported for τ between 0.1 and 0.999,[27] or by the thermodynamically stable ferromagnetic monolayer Fe(110) on W(110),[11] where power laws with β^* between 0.16 and 0.25 (depending on the covering metal) were observed for the magnetic hyperfine field B_{hf}, for τ between 0.3 and 0.99 (B_{hf} was measured by Mössbauer spectroscopy in a remanent state). Accordingly, the possibility to describe the remanent magnetization in a wide range of temperatures by a phenomenological power law with an exponent somewhere between 0.15 and 0.28 represents a frequently observed property of two-dimensional magnetic systems, which remains to be explained.

The temperature dependence of remanent and saturation magnetization should be distinguished carefully in two-dimensional ferromagnets. In particular, it turns out that in our example, the Curie temperature $T^*_{C,r}$, which can be derived in a phenomenological way from m_r, is by 10 K lower than the corresponding $T^*_{C,s}$ derived from saturation. A similar situation was observed for the monolayer Fe(110) on W(110), coated by Ag, where a Curie temperature of 291 K was derived from magnetometry in finite fields,[9] whereas Mössbauer spectroscopy in a remanent state resulted in a lower value of 282 K.[28,11] Apparently, a difference between phenomenological Curie temperatures, derived from saturation and remanence, respectively, must be considered generally in ultrathin films, the latter being virtually smaller than the first one.

In conclusion, we have shown that the saturation magnetization of a properly prepared, strongly anisotropic, perpendicularly magnetized ferromagnetic monolayer follows the exact solution of the 2DIM in a wide range of temperatures. A comparison with the asymptotic power law of the 2DIM is inappropriate. The Curie temperature shows good agreement with recent theories. Magnetization tails above T_C and deviations of M_r from M_s are supposed to result from considerable imperfections in the presently available films. Experiments with improved monolayers structures will be required for a detailed understanding of these systems.

ACKNOWLEDGMENT

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