Magnetic Phase Transition in Two-Dimensional Ultrathin Fe Films on Au(100)

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Thin Fe films on Au(100)—including one monolayer—exhibit long-range ferromagnetic order. The temperature dependence of the magnetization, as measured with spin-polarized low-energy electron diffraction and spin-polarized secondary electron emission, shows a second-order phase transition at a thickness-dependent Curie temperature. The critical exponent β for (1–3)-monolayer-thick films is 0.22 ± 0.05, independently of film thickness.

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It is theoretically well established[1] that second-order phase transitions can be classified into universality classes, depending on a small number of parameters, like the *dimensionality* of the system. A phenomenon typically covered by the universality hypothesis is the transition from a ferromagnetic to a paramagnetic state. Accordingly, the critical exponents should assume different values depending on whether the spins are arranged into a three-dimensional lattice or, for instance, are located at the very surface of a semi-infinite solid.

In a two-dimensional (2D) magnetic system, theoretical arguments suggest that either the long-range order parameter, i.e., the magnetization M, is always zero at any finite temperature—the Mermin-Wagner theorem for short-range isotropic magnetic interaction[2]—or M disappears at a finite temperature T_C according to $M = \text{const} \times (1 - T/T_C)^{\beta}$, where β is the corresponding critical exponent. Theory predicts β to be in the vicinity of $\frac{1}{8}$ for most 2D universality classes[3]; in particular $\beta = \frac{1}{8}$ for the 2D Ising model.[4]

Experimentally, a possible realization of a 2D system is a single, crystallographically well ordered monolayer (ML) of magnetic atoms atop a nonmagnetic substrate. The advent of modern vacuum science, particularly epitaxial-growth techniques, nowadays facilitates a reproducible preparation of well characterized monatomic overlayers.

While the occurrence of long-range order down to the monolayer range is indisputable,[5-11] the physical realization of a sharp phase transition for monatomic films is still far from ideal. The sharpness of the phase transition crucially depends on the perfection of the sample. Structural defects and impurities are hindrances to the long-range fluctuations that cause the order parameter to vanish as a power law at a well-defined temperature T_C.[12] As a consequence of these finite-size effects, a rounding of the transition occurs, with the order parameter showing a characteristic tail above T_C. For bulk samples, this tail only extends over a very limited range of the reduced temperature T/T_C, typically 10^{-3} to 10^{-4}, since they can be prepared with a contamination level as low as a few ppm and with perfect crystallinity extending over at least a few micrometers. Conditions are totally different at a surface: A 1% amount of contaminants is considered excellent by any modern standards. Moreover, the surfaces of metals in general and noble metals in particular have a high density of steps (mostly monatomic) separated by flat terraces extending over only a few 100 Å.[13] On these grounds, phase transitions in thin films are likely to be broader than bulk phase transitions, and considerable rounding has indeed been observed in some recent studies of truly two-dimensional phase transitions.[6,14]

In the present work we report on the temperature dependence of the long-range order parameter M in thin (1–3 ML) films of bcc Fe on Au(100), measured with two complementary techniques: spin-polarized low-energy electron diffraction (SPLEED) and spin-polarized secondary electron emission spectroscopy (SPSEE). We confirm the finding of Bader and Moog[5] that at sufficiently low temperatures all films thicker than 0.9 ML exhibit long-range ferromagnetic order with in-plane magnetization along [001]. Furthermore, we find (i) that all films undergo a second-order magnetic phase transition at a well-defined Curie temperature T_C, which depends on the film thickness, and (ii) that the critical exponent β is 0.22 ± 0.05, independently of the film thickness. This independence shows that the phase transition in this system is a truly two-dimensional one. The difference in the behaviors of T_C and β versus film thickness experimentally confirms the universality hypothesis.

We access the magnetization of the system by means of two complementary techniques involving spin-polarized electrons: SPLEED and SPSEE. Both techniques probe the first few surface layers and are therefore sensitive to magnetism in the monolayer range. In SPLEED,[15] electrons with well-defined spin state impinge on the surface. The intensity of the specularly reflected beam is measured for incident electrons with spin parallel ($R\uparrow$) and antiparallel ($R\downarrow$) to the in-plane magnetization. The exchange part of the Coulomb potential causes $R\uparrow$ to differ from $R\downarrow$ and the resulting

asymmetry $A_{ex}=(R\uparrow-R\downarrow)/(R\uparrow+R\downarrow)$ to be proportional to the long-range order parameter M in the surface sheet sampled by this technique. In SPSEE,[16] an unpolarized incident electron beam of typically 2.5 keV excites valence-band electrons in the sample to energies above the Fermi level through a cascade scattering process. A fraction of the excited electrons are emitted at low kinetic energies. The spin polarization P of these secondary electrons is defined as $P=(N\uparrow-N\downarrow)/(N\uparrow+N\downarrow)$, where $N\uparrow$ ($N\downarrow$) is the number of electrons with magnetic moment parallel (antiparallel) to the quantization direction given by the spin analyzer. P is proportional to the component of the sample magnetization M along the quantization axis. Although the proportionality of A_{ex} and P to M is well established,[17,18] multiple scattering and spin-flip scattering are often invoked to question it. The fact, however, that we find an identical temperature dependence of the two intrinsically different quantities A_{ex} and P to within experimental uncertainties is a compelling evidence that we are indeed measuring the long-range order parameter M.

We have conducted an extensive investigation of the growth mode of Fe films on Au(100),[19] using Auger-electron spectroscopy and low-energy electron diffraction (LEED). The Au substrate was prepared by the usual ion-etching and heating cycles and terminated by Au-onto-Au autoepitaxial evaporation, which yielded extremely sharp LEED patterns with the 5×20 reconstruction and no contaminants. The Fe films were then evaporated at 300 K at a rate of ~0.1 ML/min. The 5×20 reconstruction of the Au substrate disappears at ~0.2 ML of Fe, transforming into a sharp $p(1\times1)$ pattern indicative of crystallographic order, which persists up to at least ten layers of Fe. The Fe film growth was monitored with the Au $N_{67}O_{45}O_{45}$ Auger line at 69 eV. The Au signal decreases exponentially with evaporation time and, at closer inspection, exhibits a polygon shape with two kinks lying on an exponential, which unambiguously demonstrates layer-by-layer growth up to at least two to three layers. The polygon shape was determined by a computer fit procedure which minimized the deviations, using the kink positions as free parameters. The fit yielded exact periodicity of the polygon in evaporation time, which is a strong reliability test and provides an accurate thickness calibration. We note that the substrate temperature at which the films are grown is important: Deposition at 300 K is a necessary condition to obtain both layer-by-layer growth and single crystallinity and to avoid interdiffusion between substrate and adsorbate atoms.

For a magnetic characterization,[20] magnetic hysteresis loops were recorded with SPSEE at various temperatures. They exhibit sharp magnetization reversals with pronounced corners indicative of single-domain states at remanence with in-plane magnetization along the Fe(100) direction. Therefore, all the temperature dependences could be recorded at remanence, without any applied magnetic field. From spin-polarized Auger spectroscopy we find that no induced magnetic moment occurs in the Au subsurface, showing that the magnetism is strictly confined within the Fe films. Absolute values of P or A_{ex} are as large as 10% for 1 ML at low temperatures. Their interpretation in terms of, e.g., the value of the magnetic moments, although appealing, is complicated by the presence of a substrate contribution and is beyond the scope of this paper.

As a first result we now discuss the thickness dependence of the Curie temperature T_C, shown in Fig. 1. We wish to emphasize that from this magnetic behavior an independent, new, and strong evidence arises for layer-by-layer growth of Fe on Au(100). We find that the Curie temperature of the 1-ML film is strongly reduced with respect to the bulk value. This reduction, already observed in similar systems,[5,8,11] is consistent with the loss of nearest neighbors upon approaching the monolayer thickness. The key observation of Fig. 1, however, is the onset of magnetic long-range order at very low coverages, in the range between 0.6 and 0.9 ML. Evidently, almost complete wetting of the Au surface is necessary in order for a sizeable long-range magnetic order with remnant magnetization to occur at such low adatom concentrations. We suggest that the exact determination of the magnetic phase diagram in the submonolayer regime could provide insight into the physics of dilute magnetism and percolation phenomena. For thicknesses larger than 1 ML, the bulk value is rapidly approached. Data points about 2.5 ML could not be recorded because of interdiffusion and/or clustering at elevated temperatures.

Next, we report on the critical behavior of the order parameter. Figure 2 shows the temperature dependence of A_{ex} for 1 ± 0.1 ML Fe on Au(100) in the vicinity of T_C. Similar curves were recorded with SPSEE in the range $0.3\leq T/T_C\leq1.2$ for various thicknesses. For comparison, the spin polarization of the thickest film with an accessible Curie temperature, 2.5 ML, is included in Fig. 2. Beside the complete vanishing of A_{ex} or P

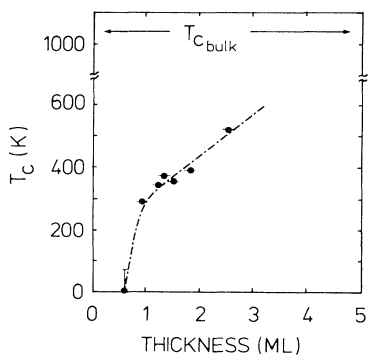

FIG. 1. Thickness dependence of the Curie temperature T_C as determined by spin-polarized secondary electron emission; the dashed curve is a guide to the eye.

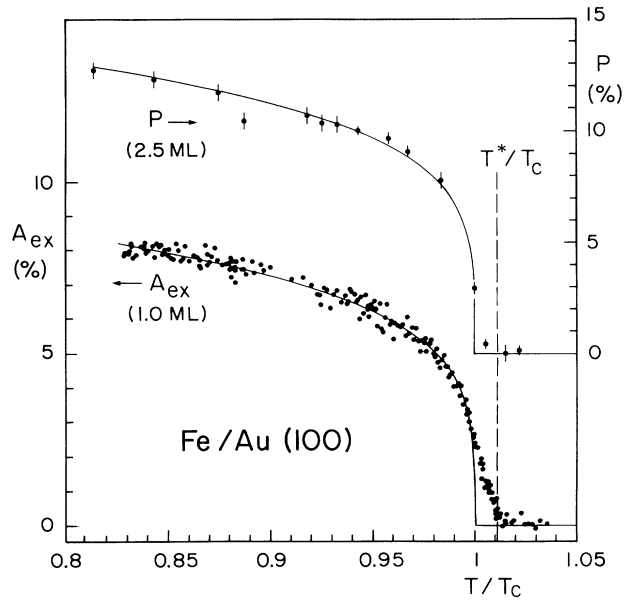

FIG. 2. Exchange asymmetry A_{ex} for 1.0 ± 0.1 ML and spin polarization P for 2.5 ± 0.2 ML of Fe on Au(100) vs reduced temperature T/T_C, with $T_C = 315$ and 521 K, respectively. SPLEED: electron energy 17.2 eV, angle of incidence 22°. SPSEE: primary energy 2500 eV, secondary energy 9 eV. T^* is the temperature at which A_{ex} and P vanish.

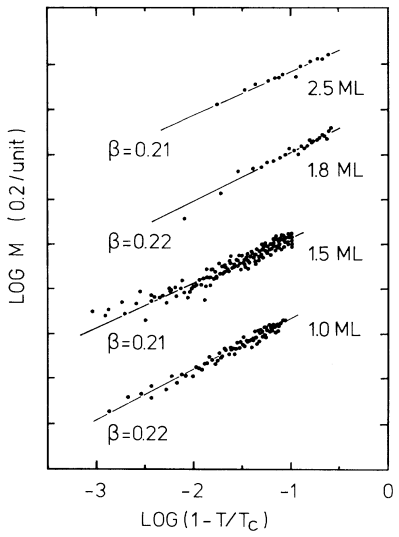

FIG. 3. Temperature dependence of P (2.5, 1.8, and 1.5 ML) and A_{ex} (1.0 ML) in a log$P$ or logA_{ex} vs log$(1 - T/T_C)$ representation.

at a certain temperature T^*, we observe the existence of an inflection point at some temperature below T^*—a clear indication that M does not exactly follow a power law. This leaves us with the problem of determining T_C, below T^*, in order to get a proper estimate of β. In the case of a rounded transition, the inflection point is often taken as the Curie temperature of the system. We prefer to adopt an alternative way of finding T_C which is in line with the stringent criteria for determining β given in Ref. 21. T_C is chosen by maximizing[22] the range of $\log(1 - T/T_C)$ over which the data points in a log$M$ vs $\log(1 - T/T_C)$ representation form a straight line. The slope of this straight line is the critical exponent β of the phase transition. This procedure implies the physically plausible assumption that our system follows a power law up to a certain temperature, above which a deviation occurs due to the finite-size effects. If the range of deviation, i.e., $(T^* - T_C)/T_C$, is a small number, this assumption is justified.[22] The solid line in Fig. 2 is a curve of the form $M = \text{const} \times (1 - T/T_C)^{\beta}$ with T_C and β determined as described.

Figure 3 shows log-log representations of the data points for four films of various thicknesses. From the slope of the straight lines we obtain $\beta = 0.22 \pm 0.05$, independently of film thickness. For all films T_C was determined according to the method described above. In all cases the tail of M was less than 3% in units of T/T_C. It was found to vary within a factor of 2 depending on the particular film quality, but no trend with film thickness was detected at the present state of film preparation. Notice that β turns out to be independent of the size of the tail as well, justifying our suspicion that, as long as the tail is small, power-law critical exponents can be derived reliably.

The key element in Fig. 3 is the thickness independence of β. Compared with the strong thickness dependence of T_C (see Fig. 1) it reveals an important aspect of the universality hypothesis, namely, that the critical behavior does not depend on the strength of the magnetic interaction. The average number of nearest neighbors and therefore the total exchange energy per spin increases with the film thickness. This causes T_C to increase with film thickness: T_C is evidently not a universal physical quantity. In contrast, the critical exponent β does not change with a variation of the total coupling strength per spin; i.e., it is a universal quantity, sensitive to the topological and spin dimensionality only.[1] Furthermore, the thickness independence of β can be regarded as proof of the two-dimensional nature of the phase transition. Two characteristic lengths can be defined: the thickness d, which limits the size of the fluctuations normal to the film, and the size Δ of the largest lateral fluctuations. Since $d \ll \Delta$, all films fall into one single two-dimensional universality class.

From the value of β and from the curves in Figs. 2 and 3 the following points might be addressed: (i) The critical exponent is slightly larger than typical values expected for two-dimensional universality classes (0.1–0.15); it is, however, out of the range of bulk (0.38) or surface (0.8) critical exponents. What is behind the larger value

of β is not clear at present. We note that the calculated value of $\frac{1}{8}$ is strictly valid for short-range interactions only. The experimental value of bulk Fe is also slightly larger than the best theoretical estimate for a Heisenberg ferromagnet[21] of 0.365. This leads us to suspect that the present finding is a further example of a systematic deviation, in band ferromagnets like Fe or Ni, from values calculated for short-range interactions. We bring to the reader's attention that β for an infinitely long-ranged interaction is just the mean-field value[4] of 0.5 and that long-ranged interactions do indeed exist: The Ruderman-Kittel-Kasuya-Yosida interaction is an example.[4] (ii) The order parameter does not vanish exactly at T_C but has a small tail of the order of 1% in units of T/T_C. Evidently, as pointed out before, the monatomic film is not perfectly flat over macroscopic distances.[23] The sharpness itself of the phase transition is, however, the most sensitive measure of the length scale over which the film approaches the perfectly flat layer. The size of the largest magnetic fluctuation which does not lose its coherency because of defects, i.e., the correlation length ξ, is given[12] by $\xi = a(1 - T^*/T_C)^{\nu}$, where a is the lattice constant. Using for the critical exponent ν the Ising value of 1, we obtain that the 1% tail corresponds to a 150-Å distance between coherence-breaking defects. This value is in good agreement with the distance between steps on the Au(100) surface observed with scanning tunneling microscopy.[13]

In conclusion, we have shown that Fe films on Au(100) represent a physical realization of a truly two-dimensional phase transition. The thickness independence of the critical behavior confirms the universality hypothesis. The critical exponent β is found to be 0.22 ± 0.05 This represents a relevant deviation from the expected value of $\frac{1}{8}$ for a two-dimensional Ising-type system. Further work on both itinerant- and localized-electron 2D ferromagnetic systems is needed in order to establish the true origin of this deviation.

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