Magnetic order and critical behavior at surfaces of ultrathin Fe(100)$p(1\times1)$ films on Pd(100) substrates

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The magnetic order and critical behavior at well-defined surfaces of ultrathin (1.9–2.1) monolayer Fe(100)$p(1\times1)$ films, deposited at room temperature on atomically flat Pd(100) substrate crystals, is investigated using electron-capture spectroscopy (ECS). Ferromagnetic order sets in at a surface Curie temperature $T_{C_s}=613.1$ K. At room temperature, the long-ranged electron-spin polarization (ESP) amounts to 34% and is oriented in-plane for well-annealed films. Near T_{C_s}, the ESP follows the power law $(T_{C_s}-T)^\beta$, with a critical exponent $\beta=0.125\pm0.01$ evaluated near T_{C_s}. This result is in good agreement with the exact value $\beta=\frac{1}{8}$ of the two-dimensional Ising model with short-ranged forces. For temperatures far from T_{C_s}, the ESP follows precisely the temperature dependence of the magnetization as predicted by Yang's exact solution of the two-dimensional Ising model including long-ranged forces.

INTRODUCTION

It is now well established, both from theory[1–3] and experiment,[4,5] that the magnetic properties of ultrathin films can differ strikingly from those of the corresponding bulk. Recent advances in experimental techniques not only enable us to prepare high-quality ultrathin magnetic films, but also allow us to study their magnetic properties with high-sensitivity spin-resolved spectroscopies.

These films might exhibit in-plane or out-of-plane magnetic anisotropies which makes them very attractive for fundmental studies on the influence of reduced symmetries on the properties of matter, which, if completely understood, can lead to the development of new magnetic systems possessing unusual magnetic properties. Moreover, magnetic materials such as ultrathin films allow us to study the universal properties of phase transitions in two dimensions.[6,7]

The purpose of this paper is to report on an experimental test of the predictions of the two-dimensional (2D) Ising model,[8] originally proposed by Lenz,[9] which describes the temperature dependence of the spontaneous magnetization of 2D ferromagnets with infinite uniaxial anisotropy. To this, we note that Mermin and Wagner[10] have shown that in isotropic 2D, continuous Heisenberg spin systems with short-ranged forces, no spontaneous magnetization can exist at nonzero temperatures.

Our experiments are not only performed in a temperature region very close to the critical point but also over a large range of temperatures which allows us to compare our experimental data with the predictions of the 2D Ising model far beyond the critical region.

It is interesting to note that the temperature dependence of the magnetization of the 2D Ising model with nearest-neighbor forces was already calculated exactly by Kaufman and Onsager[11] in 1949 and by Yang[12] in 1952 including long-ranged forces. It is well known that very close to the critical temperature T_c, the magnetization $M(T)$ can be described by the asymptotic power law, $M(T)\sim(T_c-T)^\beta$ with a critical exponent $\beta=\frac{1}{8}$. As an illustration, we give in Figs. 1 and 2 the exact solution for the temperature dependence of the spontaneous magnetization (solid lines in Figs. 1 and 2) and of the asymptotic power-law approximation (dashed lines in Figs. 1 and 2). From this it is obvious that measurable differences between the exact solution and the power-law approximation should occur already a few percent below the reduced temperature $t=T/T_{C_s}$ with $T_{C_s}(=T_c)$ being the surface Curie temperature. This clearly shows that the evaluation of β using the simple power law, shown above, is only useful for t being, within a few percent, close to 1.

To this, we note that in electron capture spectroscopy (ECS) experiments for ultrathin films of V(100) on Ag(100) (Refs. 13 and 14) and in surface magneto-optic Kerr effect (SMOKE) experiments for ultrathin films of Fe(100) on Pd(100) (Refs. 15 and 16) critical exponents of $\frac{1}{8}$ were already found.

Using ECS, we have studied the magnetic and critical behavior of truly 2D magnetic films consisting of ultrathin [1.9–2.1 monolayers (ML)] body-centered tetragonal

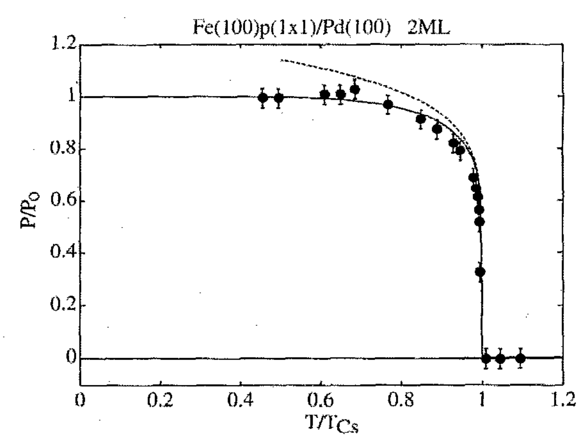

FIG. 1. Electron-spin polarization P/P_0 as a function of T/T_{C_s} for the surface of a 2 ML thin bct Fe(100)$p(1\times1)$/Pd(100) film. The solid and dashed lines represent, respectively, the exact solution of the 2D Ising model and the power-law approximation for $T\rightarrow T_{C_s}$.

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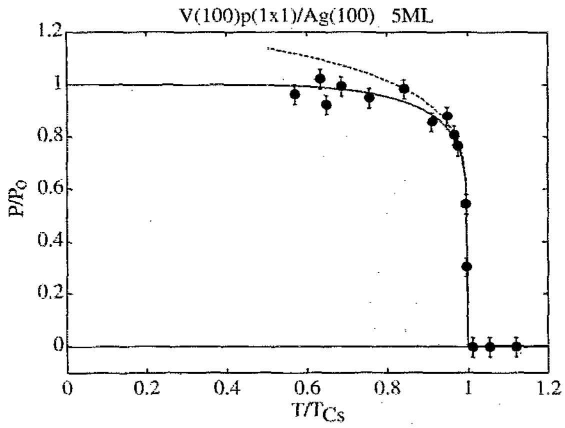

FIG. 2. Electron-spin polarization P/P_0 as function of T/T_{C_s} at the surface of a 5 ML thin $V(100)p(1\times1)/Ag(100)$ film. The solid and dashed lines represent, respectively, the exact solution of the 2D Ising model and the power-law approximation for $T\rightarrow T_{C_s}$.

(bct) $Fe(100)p(1\times1)$ films epitaxially grown on atomically flat Pd(100) surfaces.

It is found that the surface of these 2 ML thin films orders ferromagnetically at low temperatures. This already indicates the presence of magnetic anisotropies as discussed by Malyshev[17] and more recently by Bander and Mills[18] for the anisotropic 2D Heisenberg model which behaves like the 2D Ising model near T_{C_s}.[6]

Next, it is found that for t being within a few percent close to 1, the magnetization of the topmost surface layer behaves like $(T_{C_s}-T)^{\beta}$, with $\beta=0.125\pm0.01$ in excellent agreement with the exact result for the 2D Ising model for temperatures very close to T_{C_s}. Most important, we find that the temperature dependence of the surface ESP for t ranging from 0.5 to 1.0 follows precisely the exact solution of the 2D Ising model as given by Yang. Finally, we note that, above T_{C_s} there are no rounding-off effects of the surface ESP, a fact which points towards the high-quality of the deposited films.

EXPERIMENT

The present experimental results are obtained using electron-capture spectroscopy (ECS).[19] This method, which is discussed in detail elsewhere,[19] allows us to determine the existence of long-ranged ferromagnetic order which can be characterized by the spin polarization of electrons captured by fast ions at the topmost surface layer of a magnetic system.

Defining the electron-spin polarization (ESP), P along the direction of the magnetization yields $P=(n^+-n^-)/(n^++n^-)$, with n^+ and n^- the numbers of up(majority)- and down(minority)-spin electrons.

For the deposition of ultrathin iron films, atomically clean and flat Pd(100) substrate crystals are prepared in ultrahigh vacuum in a target preparation chamber.[19] The surface orientation of the Pd(100) crystals is better than 0.01° and is monitored by use of a precision x-ray diffractometer.

Applying standard cleaning and annealing procedures developed in our earlier studies, and using Auger electron spectroscopy with a cylindrical mirror analyzer, residual C and O contaminations are measured to be less than 1% of a monolayer (ML). The single-crystalline (1×1) state of the Pd(100) surface is detected by low-energy electron diffraction (LEED).

The iron films are deposited by electron-beam evaporation at 8×10^{-10} mbar. For a substrate temperature of 293 K and an evaporation rate of 0.02 Å/s, homogeneous and island-free growth of the Fe films is obtained. The island-free growth of the Fe films is checked by measuring Auger intensities as functions of deposition time and by monitoring the ion reflectivity of specularly reflected ions which is not reduced from its initial value measured at atomically flat Pd(100) surfaces. Further details of these procedures can be found in our earlier studies of epitaxial Fe films on Cu, Ag, and Au single-crystal substrates.[5]

LEED measurements show a $p(1\times1)$ structure for all films studied. No changes in symmetry and intensity of the electron-diffraction patterns are found, implying the epitaxial growth of bct Fe(100) on Pd(100) which was already extensively studied by Quinn *et al.*[20] The thickness of the films is determined with a calibrated quartz oscillator and with calibrated Auger signals.

After preparation and characterization, the Fe(100)/Pd(100) samples are studied *in situ* at 2×1^{-10} mbar. ECS is used to measure the long-ranged surface ferromagnetic order, the specimens being magnetized in magnetic fields ranging from 75 to 0 Oe, and the temperature of the samples being kept constant within 0.05°. Such applied fields have a negligible effect on the ESP in the investigated temperature range which shows that, within experimental errors, the remanent magnetization is equal to the saturation magnetization.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the normalized long-ranged electron-spin polarization P/P_0 at the surface of 2 ML thin Fe(100) films on Pd(100), as function of T/T_{C_s} with $T_{C_s}=613.1$ K being the surface Curie temperature of the films and $P_0=-33\%$ the calculated electron-spin polarization at $T=0$. The full line in Fig. 1 represents the temperature dependence of the magnetization as predicted by Yang for the 2D Ising model, and the dashed line in Fig. 1 gives the asymptotic power-law approximation as discussed before. It is obvious that the experimental data point can be precisely described with the exact solution of the 2D Ising model as given by Yang.

We note that from preliminary ECS experiments, there was evidence that all Fe/Pd films, deposited at room temperature, first showed a zero in-plane ESP, which, after annealing of the films to 350 °C ($\geqslant T_{C_s}$) increased up to 33%. This implies that metastable, 2 ML thin Fe(100)/Pd(100) films, grown at room temperature, possess an out-of-plane magnetization, and that only well-annealed Fe(100)/Pd(100) films are spontaneously magnetized in-plane.[15,16]

The surface Curie temperature T_{C_s} and the critical exponent β for well-annealed 2 ML thin films are determined by a linear least-squares fit of the polarization data under the assumption of a power law of the form $(T_{C_s}-T)^{\beta}$ for t between 0.97 and 1.0. The value of β is found to be $\beta=0.125\pm0.01$ giving the slope of a straight line in the log-log representation of the polarization data. The dashed curve in Fig. 1 corresponds to $\beta=0.125$ and fits the experimental data quite well for t between 0.97 and 1.0. The value we obtain for the critical exponent β agrees well with the exact value ($\frac{1}{8}$) of the 2D Ising model of a ferromagnet for $T\rightarrow T_{C_s}$. In addition, we have performed fits of the experimental data points for t values ranging far beyond ($0.75\leqslant t\leqslant 1$) the critical region. Within experimental errors, we again obtain $\beta=0.125$. We note that more experimental evidence is needed to establish if the critical region for 2D phase transitions extends over a larger range of temperatures compared to that for 3D phase transitions.

Comparing our experimental data points for $0.45\leqslant t\leqslant 1$, we find excellent agreement with the temperature dependence of the spontaneous magnetization as predicted by Yang for a 2D Ising magnet.

In order to obtain a deeper insight into these important findings, we have reevaluated earlier ECS data where we found an Ising-type phase transition ($\beta=0.128$) at the topmost surface layer of 5 ML, artificially structured, epitaxial $V(100)p(1\times1)/Ag(100)$ films.[13,14,21] Indeed, the data, given in Fig. 2 together with the exact solution of the 2D Ising model (solid line) and the power-law approximation (dashed line), show, over the measured temperature range, excellent agreement with Yang's exact solution of the 2D Ising model.[22] We note that also for this system, the remanent magnetization was equal to the saturation magnetization.

We note that our ECS results, which show the existence of ferromagnetic order at the topmost surface layer of these two systems are consistent with ground-state one-electron band-structure calculations of Li *et al.*,[23] Blügel, Wienert, and Dederichs,[24] Yokoyama *et al.*[25] and Fu, Freeman, and Oguchi.[1]

Work is in progress to study these two systems with spin-polarized electron emission spectroscopy (SPEES).[26] SPEES enables us to determine the layer dependence of magnetic properties and, therefore, allows a determination of the nature of the magnetic coupling between layers and also the detection of a possible nonzero induced spin polarization of the substrate electrons.

ACKNOWLEDGMENTS

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