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Paramagnetic-ferromagnetic phase transition during growth of ultrathin Co/Cu(001) films

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We have investigated the magnetic response of ultrathin paramagnetic Co/Cu(001) films in an applied field using the magneto-optic Kerr effect. We find that the paramagnetic susceptibility χ increases sharply upon Co deposition within a narrow thickness range close to the critical thickness at which the onset of ferromagnetism occurs. We attribute the observed behavior to a geometrical, or percolation phase transition, since the corresponding critical exponent $\gamma=\frac{43}{18}=2.39$ agrees well with the experimentally determined value of 2.40 ± 0.07.

I. INTRODUCTION

High-quality ultrathin films of Fe, Co, and Ni deposited on a nonmagnetic substrate provide an opportunity to study two-dimensional (2D) magnetic systems. Experimentally, effects such as the modification of the Curie temperature,[1,2] interface-induced magnetic anisotropies and enhancement of the magnetic moment[3,4] have been reported. The Curie temperature $T_C(d)$ is dependent on thickness in ultrathin films[1,2,5] and becomes zero below a finite thickness. In thermodynamic terms the thickness-dependent phase transition at fixed temperature is distinct from the temperature-dependent transition at fixed thickness. The temperature-dependent transition has now been extensively studied experimentally in ultrathin ferromagnetic films.[1,2,5] This is in contrast to the thickness-dependent transition where no critical exponents, which may indicate percolative behavior, have been published to our knowledge.

Since χ diverges at the phase transition, this is a natural quantity to study experimentally in investigations of the critical behavior. The strong magnetic response to an applied field of a 2D film in the paramagnetic phase has been reported by several authors.[6,7] In the present work we exploit this property in order to investigate the paramagnetic-ferromagnetic phase transition as a function of thickness d at fixed temperature T in the fcc Co/Cu(001) system. This extends an earlier study[8] of the thickness dependence of the coercive field H_c in ultrathin Co/Cu(001) films close to the phase transition, which revealed that H_c increased with thickness dramatically, following a power law. We choose this epitaxial system since good epitaxial growth is vital for a sharp phase transition; only recently has a deviation from perfect growth of Co on Cu(001) been detected in the early stages.[9-12] By comparing the measured and predicted critical exponents we are able to conclude that the onset of ferromagnetism with increasing thickness in the fcc Co/Cu(001) system occurs via percolation.

II. EXPERIMENT

The experiments were performed in a UHV chamber with a base pressure of less than 2×10^{-10} mbar, and a pressure of less than 5×10^{-10} mbar during Co deposition. The Cu(001) surface was cleaned by 1-kV Ar^+ sputtering and annealing to 700 K. This gave a sharp $p(1\times1)$ low-energy electron-diffraction (LEED) pattern and a contamination-free Auger scan. Samples were kept at 300 K throughout the experiment, in order to avoid Cu segregation that has been shown to be important at higher temperatures.[9] A Helmholtz coil was used to generate ±170 G along the easy [110] axis, and the magneto-optic Kerr effect (MOKE) in the transverse geometry[13] was used for magnetic measurements. M-H loops were recorded at 2 Hz. Only the shutter of the eva-

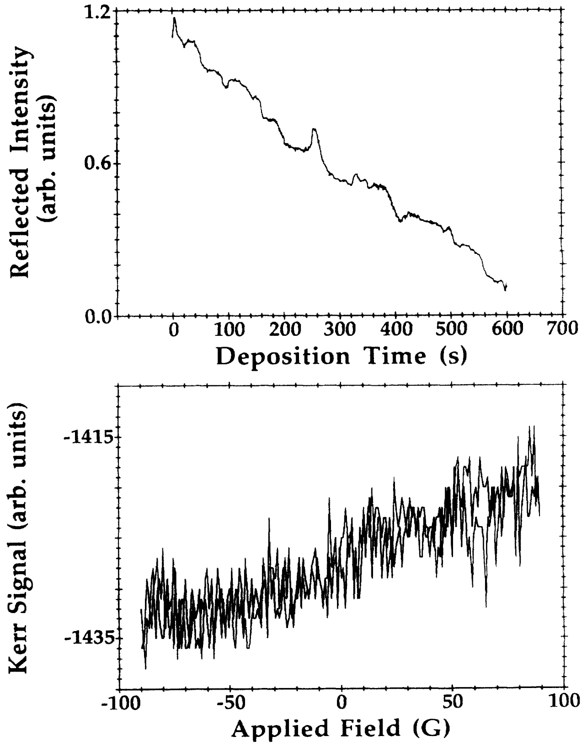

FIG. 1. The upper panel shows the reduction of the reflected intensity during Co growth; the associated paramagnetic M-H loop after 600-sec deposition is displayed in the lower panel.

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porator had to be moved during the experiment, thus eliminating experimental errors due to repositioning of the sample.

The deposition rate is controlled by the internal flux monitor of a commercial *E*-beam evaporator (Omicron EFM3), and by recording the decrease of the reflected laser intensity upon Co deposition. Figure 1(a) shows the drop in the reflected intensity during Co deposition, and the associated *M-H* loop can be seen in Fig. 1(b). It can be seen that a monotonic linear dependence is obtained over the thickness range shown, corresponding to approximately the critical thickness d_C at which long-range ferromagnetic order develops (at 300 K). There is still some disagreement over the absolute value of $d_C(T)$ but it is now generally accepted to lie between 1.0–1.7 ML (Refs. 13–17) at 300 K, which agrees with a value of 1.3 ± 0.3 ML from our own Auger study. The change in optical reflectivity results from the difference between the optical constants of Co and Cu. The noise observed corresponds to a thickness variation of approximately $0.1 d_C$ allowing the relative film thickness to be determined with this accuracy. We can therefore use the optical signal as an independent check on the deposition rate (typically of the order ~ 0.1 ML/min).

III. RESULTS

The initial *M-H* loops showed no detectable magnetic signal. Close to the critical film thickness (at 300 K) a signal is detectable which then rapidly increases in strength with further Co deposition. We have determined χ (defined as the gradient of the magnetization with respect to applied field evaluated at zero field) for such loops by fitting the gradient of the *M-H* loops in the linear region.[18] In principle, the gradient diverges at d_C and therefore we are able to determine the critical thickness with high accuracy by plotting χ as a function of thickness. In Fig. 2(a), the first detectable *M-H* loop is displayed, averaged over approximately 250 field sweeps. A straight line can be seen to provide a good fit to the data over the entire field range. With increasing thickness the signal-to-noise ratio is increased and the slope of the curve increases. In Fig. 2(b), the last paramagnetic loop is given and the region for which the linear fit is employed is reduced to a few Gauss. The distinction between paramagnetic and ferromagnetic loops close to the transition point can be made by plotting the thickness dependence of χ and comparing the observed behavior with a power law. Above a critical thickness we find that the data no longer conforms to a single power law and thus we can determine the boundary between the paramagnetic and ferromagnetic phases: we can therefore rule out the possibility that we are seeing a ferromagnetic response rather than the true paramagnetic signal beneath the critical thickness.

In contrast to Fig. 2(a) only part of the loop shown in Fig. 2(b) is linear: however, an interpretation of the nonlinear part of the *M-H* curve is not relevant as far as the determination of χ is concerned. From a theoretical viewpoint, although the exact field dependence of the magnetization is not known, it is clear that a "classical" Langevin or Brillouin function is not appropriate. Theoretical studies for an isotropic 2D Heisenberg model yield a linear $M(H)$ dependence and a $M \sim \ln(H)$ law as approximations for the low and medium field regimes, respectively.[19–21]

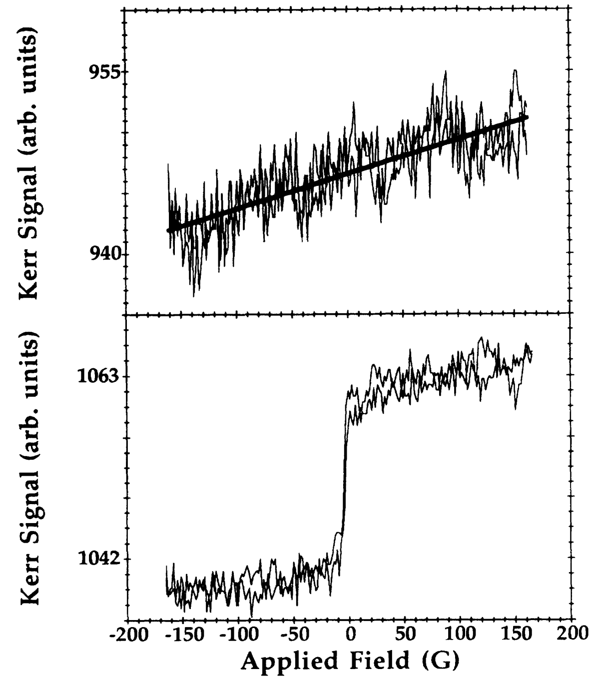

FIG. 2. The upper panel shows the first paramagnetic *M-H* loop with a linear fit, the slope of which is proportional to the susceptibility χ. The last loop of the growth sequence is displayed in the lower panel.

The results of the measured thickness dependence of χ are shown in Fig. 3 for two separate experiments (as indicated by circles and triangles). The main feature revealed by this plot is that within a narrow thickness range ($\sim 6\%$ of d_C), χ increases very sharply, indicating a well-defined magnetic transition. However, the determination of the critical exponent does not require

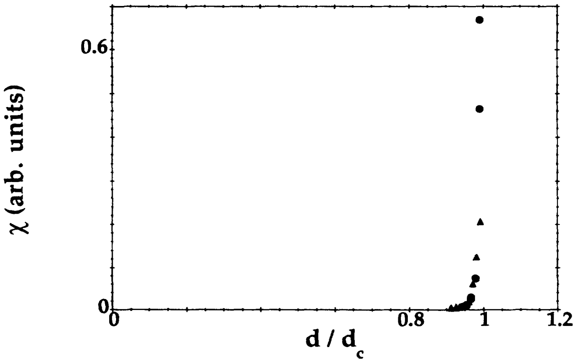

FIG. 3. The evolution of χ is shown as a function of the thickness in reduced units: for the experiment as in Fig. 1 (circles) and of a second experiment (triangles). The critical thickness was determined (in arb. units) as described in the text.

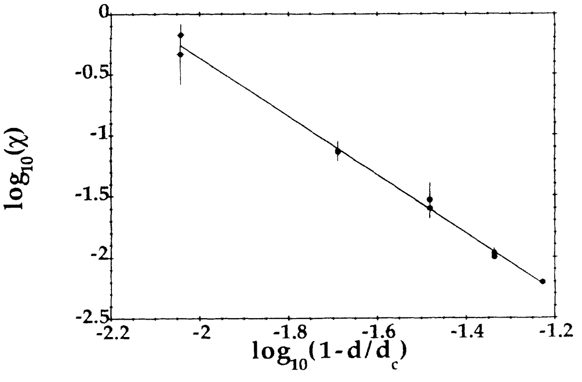

FIG. 4. The figure shows a log-log plot of the data shown as circles in Fig. 3. The slope of the linear fit is the critical exponent γ, which has been determined to be 2.41±0.07 in this case.

knowledge of the absolute value of the critical point. The measured thickness-dependent χ data are fitted to a power law of the form:

$$\chi \propto \left[1-\frac{d}{d_c}\right]^{-\gamma} = D^{-\gamma} .$$

We have determined the critical exponent γ for each of the data sets by plotting $\log(\chi)$ versus $\log(D)$ in the usual way[1] (Fig. 4). The value of d_C (in relative units) and the associated value for γ are determined by plotting the sum of the square of the errors versus d_C. The minimum of this curve refers to the best approximation for d_C and the experiments yielded $\gamma = 2.41 \pm 0.07$ (circles) and $\gamma = 2.38 \pm 0.07$ (triangles).

IV. DISCUSSION

Our results for γ are very close to the value of 2.389, calculated for the 2D percolation phase transition[22,23] and are not consistent with the value of 1.66 expected for the 3D case. Within the percolation model one assumes a statistical occupation of the lattice sites in 2D. In the percolation transition, at a critical 2D concentration (corresponding to a critical thickness in our experiment), exchange interactions between the atoms can extend across the whole sample. In other words the clusters coalesce and the size becomes infinite, resulting in one "giant" magnetic island. An experimental realization of this kind of transition is difficult, since good epitaxial growth requires finite temperatures (e.g., 300 K), but the "true" percolation transition occurs only at 0 K. However, the following considerations suggest that thermal effects are unimportant in influencing the value of γ:

(i) the thickness dependence of T_C of ultrathin magnetic films has been reported in several publications.[1,2,5,24] All of these results show that T_C initially rises sharply from zero to finite values with thickness, once a minimum thickness is exceeded.

(ii) Kerkmann, Pescia, and Allenspach[7] show that due to the strong response of 2D magnetic films in an applied field (100 G), it is possible to detect magnetic signals at temperatures 20% above the Curie temperature. Thus we can use this observation to estimate that T_C has increased by ~50 K between the first and last *M-H* loop shown in Fig. 2 despite not having measured T_C as a function of the thickness d directly. This estimate agrees with the finite-size scaling results of Huang *et al.*[5]

With these points in mind, it is reasonable to assume that our d_C is close to the percolation threshold, given the rapid variation of the transition temperature with d, which we have deduced from our measurements [point (ii) above]. Accordingly the behavior of this system at 300 K should approximate very well the true percolation at 0 K . Given the very good agreement between the measured and theoretically predicted critical exponents, we therefore believe that we have experimentally observed a percolation phase transition.

A possible objection to our explanation is that we observe a blocking of spins in superparamagnetic islands rather than a paramagnetic-ferromagnetic phase transition. The main obstacle for the observation of true 2D magnetism is the presence of superparamagnetic islands:[25] for example Mössbauer spectroscopy studies on Fe/Ag (111) superlattices revealed superparamagnetic features.[26] However, studies by Kerkmann, Pescia, and Allenspach[7] using MOKE and scanning electron microscopy with polarization analysis demonstrate that Co/Cu(001) films do not show any superparamagnetism, even in the vicinity of a temperature where the remanent magnetization vanishes. They could then identify this temperature with the Curie temperature T_C. Also Krams *et al.*[17] found that the anisotropy of Co/Cu(001) films (required for superparamagnetism) vanishes upon approaching d_C, as expected for a paramagnet.

Despite this, for completeness we will estimate the lateral size l of superparamagnetic islands[27] that would affect our measurements, assuming that the blocking temperature is 300 K and that the relaxation time is of the order of our data collection time (~ 1s). Assuming the relevant in-plane anisotropy for a Co thickness of 2.5 ML,[17] we estimate $l \sim 1000$ Å. Since our film is thinner than 2.5 ML this is a lower limit for the lateral island dimension.

Scanning-tunnel microscopy and spot profile analysis LEED studies of metal epitaxy of Fe and Co on Cu(001) give island sizes of the order of 10 Å (Refs. 28–30) before they coalesce, which is considerably smaller than the value of l we estimate. This would mean that the block spins of these superparamagnetic islands can fluctuate freely at room temperature and a magnetic measurement will yield a zero result, corresponding to our observations below $0.94d_C$. However with increasing coverage, the islands increase in size and begin to coalesce, and then island sizes of the order of 1000 Å can indeed occur; a magnetic measurement will then give a nonzero result.

A change of only 6% in the quantity of Co deposited rules out any dramatic change in the lateral dimension of islands, except at the point of coalescence itself. Nevertheless, the approach to the critical point follows the percolation model, and in the approach the response is entirely paramagnetic in its nature.

This sharp transition is strongly supported by our finding of a sharp onset of H_C during Co/Cu(001) growth[8] and the equally sharp onset of the remanance reported for the same system.[15] The growth of Co/Cu(001) films was regarded as a textbook example of layer-by-layer growth, but recent experimental studies, particularly the systematic investigation by Kief and Egelhoff, indicate a deviation from perfect growth in the early stages.[9,11,12] However, as long as these imperfections are statistically independent they should have no influence on the nature of the phase transition, since the only characteristic length is the correlation length, which diverges at the critical point. Therefore it is reasonable to assume, as suggested by our experiments, that the microscopic details (particularly deviations from perfect growth) can be neglected, as far as the critical behavior is concerned. In contrast, these microscopic details are known to affect such properties as magnetic anisotropies for example.

In summary, we have reproducibly determined the thickness dependence of the magnetic susceptibility χ during the growth of paramagnetic Co/Cu(001) films, and report the observation of a sharp increase of this quantity within a narrow thickness range (6% of the total thickness). By comparing the measured behavior with theoretical predictions we conclude that a 2D percolation phase transition occurs.

Note added in proof. Since this paper was written Elmers *et al.* have reported evidence of magnetic percolation in Fe(110) films [H. J. Elmers, J. Hauschild, H. Höche, U. Gradmann, H. Bethge, D. Heuer, and U. Köhler, Phys. Rev. Lett. **73**, 898 (1994)].

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