Magnetic Hysteresis Loop of One Monolayer of Co on Cu(100)

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We give experimental evidence that long-range ferromagnetic order exists at finite temperatures, over a macroscopic size, and in the absence of magnetic field for 1 monolayer of Co on Cu(100). The magnetization as a function of the applied magnetic field follows a hysteresis loop. At remanence (zero applied magnetic field) the film is in a single-domain state. The magnetization reversal occurs by the breaking down of the film into magnetic domains.

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In the history of solid-state physics, the magnetism of a monatomic thin film occupies an eminent position. A famous result by Mermin and Wagner[1] states that in a monatomic thin film no long-range order is allowed at any finite temperature for an isotropic short-ranged interaction. Despite this no long-range-order rule, Fu, Freeman, and Oguchi,[2] and simultaneously Richter, Gay, and Smith[3] succeeded in demonstrating that the ground state—the state at exactly 0 K—of a number of $3d$-transition-metal monolayers is ferromagnetic. Most intriguingly, when the thickness approaches 1 monolayer (ML), these authors found that the magnetic moment increases well above the bulk value, one of the most spectacular outcomes of computational physics. A remarkable exception is the case of 1 ML of Ni on Cu, for which some calculations predict the total loss of magnetism even at 0 K.[4] These theoretical results have a common denominator: They show that the magnetic properties are profoundly altered when the 1-ML thickness limit is approached. The peculiar position of the single monolayer is also evident at the level of magnetic anisotropy: The most advanced model calculations find a transition from perpendicular spin orientation to in-plane spin orientation when the film thickness increases from 1 to 2 ML (Ref. 5) and above.

Experimentally, a string of papers have appeared in recent times on the magnetism of thin films.[6-15] In particular, Mössbauer spectroscopy detected the nuclear hyperfine splitting for 1 ML of Fe on W(110),[6] a typical sign of ferromagnetism in this type of spectroscopy. Pescia *et al.*,[7] and successively Liu, Moog, and Bader[14] were able to detect *square* hysteresis loops in ultrathin Fe films on Cu(100). Curiously enough, however, the square hysteresis loop, observed by Liu, Moog, and Bader even for films as thin as 1.5 ML, could not be realized in the monolayer thickness limit—an experimental hint of the peculiarity of the 1-ML film, for which, in contrast to, say, the 1.5-ML film, *all* adsorbate atoms are in direct contact with the substrate. This outcome again left readers wondering whether some important mechanism—like, for instance, the reduced number of nearest neighbors or the hybridization with the substrate[2,4]—is not acting to prevent 1-ML films from having a square hysteresis loop, i.e., from showing long-range order over macroscopic distances.

The subject of the present paper is the existence of long-range ferromagnetic order in 1 ML of Co on Cu(100). Thin films of Co on Cu(100)—including the 1-ML film—were the subject of a recent study by Pescia *et al.*[8] The key element distinguishing these films from all other chemically similar systems is the perfection with which they can be manufactured. Sharp break points in the Auger signal versus deposition time, with straight line section in between, and a high-quality LEED pattern were observed by a number of authors.[8,16,17] The flatness of these films was the key element in the successful outcome of a very sensitive neutron-optics interference experiment recently performed.[18] There is therefore a wide consensus that this system fulfills the standard requirements for layer-by-layer growth and a sharp interface. In Ref. 8—the first reporting on the possibility of a finite magnetization in thin Co films on Cu(100)—the ferromagnetism of Co films on Cu was inferred from the low magnetic field necessary to bring the system into saturation. However, no remanence could be found, leaving the proof of long-range order over a macroscopic size without the decisive evidence of a *hysteresis loop*. The existence of a remanent magnetization (a finite magnetization in zero applied magnetic field) is particularly significant in truly two-dimensional systems. In two dimensions, in fact, a very small magnetic field is enough to bring about a long-range order which is strictly absent without magnetic field.[19] A rigorous test of the Mermin-Wagner theorem is therefore only possible in the absence of a magnetic field. In addition, the results in Ref. 8 left two possibilities open for the orientation of the spins in zero applied magnetic field: in-plane magnetization or per-

pendicular domains. In the present paper we measure the in-plane magnetization of 1 ML of Co on Cu(100) as a function of the applied magnetic field using the magneto-optical Kerr effect. We find the following: (a) 1 ML of Co on Cu has a square hysteresis loop; i.e., long-range ferromagnetic order exists at finite temperatures, over macroscopic size (precisely the size of the laser spot used for the measurements, ~ 3 mm^2) and in the absence of magnetic fields. (b) The spins lie in the film plane. This represents a clear and definite answer to the important question of perpendicular versus in-plane magnetization for a system whose perfection makes it a test case of the most advanced total-energy calculations on magnetic anisotropy (see, for instance, Ref. 5).

The experimental technique used to measure the magnetization is based on the magneto-optical Kerr effect. The possibility of using the Kerr effect in ultrahigh-vacuum experiments to measure the magnetization of very thin films was first demonstrated by Bader and Moog.[9] Here we use the *transversal* (or equatorial) Kerr effect: The plane including the surface normal and the incident and reflected light beams is perpendicular to the applied magnetic field. The magnetic field is *applied in the film plane.* The polarization of the light—a He-Ne laser—is parallel to this plane (p polarized light). In this geometry, the *reflected intensity* depends on the magnetic state of the sample[20]; i.e., by our sweeping the magnetic field the hysteresis loop can be recorded. Figure 1 shows the hysteresis loop recorded for the 1 ML of Co on Cu(100) at room temperature. The thickness was determined from Auger spectroscopy with use of the calibration curve published in Ref. 8. Similar hysteresis loops were measured up to 10 ML. No magnetic signal was observed for 0.6 ML. The main features of the hysteresis loop in Fig. 1 are the following:

(1) The magnetization M_R at remanence ($H=0$) is equal, to within experimental accuracy, to the saturation magnetization; i.e., the monolayer is in a single-domain state at $H=0$. This establishes unequivocally the existence of long-range order over macroscopic dimensions for a monatomic film. Moreover, apart from giving essential information on the magnetic state of the system, the observation of a square hysteresis loop adds to the evidence for epitaxial growth based on Auger and LEED spectroscopy,[8,16,17] the fascinating "magnetic" evidence of the 1 ML being so perfect that, after the field is removed, it survives in a single-domain state of macroscopic size!

(2) The magnetization reversal ($H<0$) does not occur abruptly at a certain field: The magnetization changes continuously between $+M_R$ and $-M_R$. Therefore coherent rotation or displacement of one single macroscopic domain wall can be excluded. Instead, the film splits up into domains carrying $+M_R$ or $-M_R$, the numbers of which are exactly equal at $H_C \cong -20$ Oe, where the resulting macroscopic magnetization disappears.

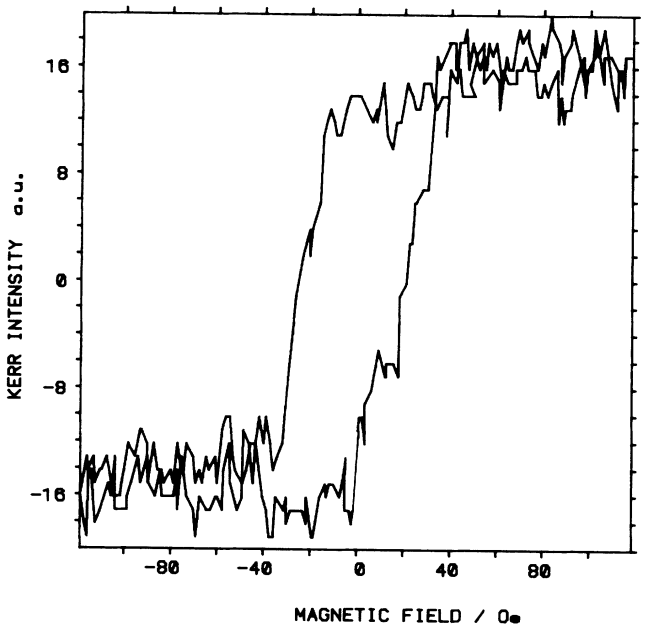

FIG. 1. Hysteresis loop for 1 ML of cobalt on Cu(100), averaged over twenty rapid field scans. The magnetic field, provided by two Helmholtz coils placed directly in the vacuum chamber, is applied parallel to the film surface. The coercive field is 20 Oe. During deposition of the Co film and the measurement of the hysteresis loop the substrate was held at room temperature. According to Refs. 8, 16, and 17 deposition at room temperature gives rise to layer-by-layer growth. In agreement with Ref. 8 we did not observe any change in the magnetic properties up to ~ 150 °C. The experiment was performed in a vacuum which never exceeded 5×10^{-10} mbar.

(3) The in-plane square hysteresis loop for films ranging from 1 to 10 ML and the absence of remanence perpendicular to the film plane observed in Ref. 8 demonstrate that the spins lie exactly in the film plane; i.e., no evidence of the transition[5] from perpendicular to in-plane magnetization upon increasing the thickness is observed.

Figure 1 shows that the conventional concepts of ferromagnetism, like hysteresis loop, coercive field, and domains can be straightforwardly extended to systems as thin as 1 ML. The size and shape of these domains remain a topic for future research.

The findings of this paper identify Co/Cu(100) as a truly epitaxial ferromagnetic monolayer. In the course of this work a similar system, fulfilling the criterion as well, has been discovered[21]: Fe/Au(100). Provided the substrate is held at room temperature during deposition, distinct breaks can be detected in the Auger signal of the substrate versus deposition time,[22] signaling the occurrence of layer-by-layer growth. In our laboratory the magnetization of the system was measured with the standard technique of spin-polarized LEED.[23] Because of

the exchange part of the Coulomb potential, low-energy (typically 10–50 eV) incident electrons with spin parallel to the sample magnetization and those with spin antiparallel are reflected with different intensities, $R_\uparrow$ and $R_\downarrow$, respectively. Therefore, *provided the film has a magnetization different from zero,* a spin asymmetry $A_{ex}=(R_\uparrow-R_\downarrow)/(R_\uparrow+R_\downarrow)$ results. Because of the use of low-energy electrons, measurements have to be performed in zero applied magnetic field; i.e., A_{ex} *measures the remanent magnetization.* As a consequence, a full hysteresis loop is not yet available. While the mechanism of magnetization reversal is not known, for 1 ML of Fe on Au(100) at 0 °C we measure (i) values of A_{ex} as large as $10\%\pm0.3\%$ (depending on the angle of incidence and the energy of the electrons)[24]; i.e., the film has a finite remanence M_R. Moreover, (ii) M_R could be switched to $-M_R$ by the application of a coercive field as low as 2 Oe. (i) and (ii) establish the ferromagnetism of a monolayer of Fe on Au(100) according to the stringent criteria used in this paper.

Evidently, provided the interface is sharp and the amount of defects in the film is low enough, the physical realization of ferromagnetic monolayers of 3*d* transition metals is within the reach of experimental physics.

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[22]As reported by Bader and Moog, Ref. 9, the Auger signal of the substrate—for the deposition conditions given in Ref. 9 (substrate temperature 230 °C)—does not decrease exponentially with deposition time, signaling the break down of layer-by-layer growth. We find that when the substrate is held at room temperature during deposition and not at 230 °C as in Ref. 9, layer-by-layer growth occurs; see R. Germar, W. Dürr, J. W. Krewer, D. Pescia, and W. Gudat, to be published.

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[24]Notice that, for the Kerr intensity I in Fig. 1, $[I(H=60\text{ Oe})-I(H=-60\text{ Oe})]/[I(H=60\text{ Oe})+I(H=-60\text{ Oe})]$ is about 2 orders of magnitude smaller than A_{ex}, because of the large probing depth of an optic experiment compared to the short probing depth (2–3 ML) of low-energy electron diffraction.