Two-Dimensional Magnet at Curie Temperature: Epitaxial Layers of Co on Cu(100)

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The response of magnetization to the application of a magnetic field has been measured above the Curie temperature T_C in epitaxial Co films on Cu(100). Magnetic domain images have been acquired up to T_C. The results suggest that for temperatures $T > T_C$ the system behaves like a two-dimensional Heisenberg magnet.

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When magnetic atoms are arranged layer by layer on top of a nonmagnetic substrate, they might become two-dimensional (2D) magnets. Indeed, numerous magnetic properties measured in such systems—like magnetic anisotropy [1], the reduced Curie temperature [2,3], or the enhanced magnetic moments [4]—confirm their 2D nature. Among the most extensively studied systems are Co films on Cu(100): The layer-by-layer growth of these films has been proven convincingly by various groups [3,5–7]. Our observation of the quantum interference from low-energy electron waves reflected by these thin films is further evidence that the films are spatially quantized [8].

Despite a wealth of results, one important observation is still missing: the physical realization of a 2D Heisenberg magnet. The reason for this is very simple: In such epitaxial structures, neither the magnetostatic dipole-dipole interaction nor "spurious" (albeit most important) magnetic anisotropies can be avoided, which break the continuous symmetry of the Heisenberg Hamiltonian. The direct result of this symmetry breaking is the occurrence of long-range order below a well-defined temperature T_C. Long-range order is forbidden in a Heisenberg magnet by the theorem of Mermin and Wagner [9].

In this paper we report on experiments to determine fundamental magnetic quantities in epitaxial Co/Cu(100) films, which in our opinion indicate that these films behave like Heisenberg magnets above T_C, the temperature at which long-range order vanishes. We have used two techniques, the magneto-optic Kerr effect [10] and spin-polarized scanning electron microscopy [11,12], to measure the temperature dependence of hysteresis loops and magnetic domain images. Thus we can establish the connection between macroscopic and microscopic quantities, in particular remanent magnetization and spontaneous magnetization, as well as the response of the magnetization to an applied field.

The steps leading to our experimental observations are as follows. Our experiment uses a Cu(100) single crystal surface prepared under UHV conditions (base pressure $< 2\times10^{-10}$ mbar) to give a very sharp low-energy electron diffraction (LEED) pattern. On top of this surface, Co films were grown at room temperature at a very slow evaporation rate [typically 0.1 to 0.2 ML/min (ML denotes monolayer)] and their growth was monitored *in situ* and in real time by the newly developed technique of low-energy electron oscillations [8]. All films were found to grow layer by layer as an almost perfect continuation of the underlying fcc (100) structure, in agreement with previous findings [3,5–7].

During growth of the film the presence of long-range order is detected by means of our transverse magneto-optic Kerr effect [13]. A magnetic field H is swept between $H=-8$ and $+8$ kA/m in the plane of the film along the [110] direction, and the intensity of the reflected light I is monitored. By recording the remanent magnetization $M_R \equiv I(H=0)$ versus deposition time we are able to stop deposition just as M_R becomes nonvanishing. The film thickness is then $\simeq 1$ ML as detected from the period of the LEED oscillations [14]. The sample is subsequently cooled. Upon warming from low temperatures, two quantities, remanent magnetization, $M_R \equiv I(H=0)$, and magnetization in an applied field of 8 kA/m, $M_H \equiv I(H=8$ kA/m), are measured in rapid succession at given temperatures. As a function of temperature we obtain the results shown in Fig. 1. We observe two remarkable features.

(i) The remanent magnetization M_R vanishes sharply at a well-defined temperature that we are tempted to identify with the Curie temperature T_C of the system. Slightly below T_C, M_R and M_H become identical within experimental accuracy.

(ii) Upon application of a "small" field, the magnetization attains sizable values even well above T_C. They are comparable to those assumed below T_C and extend to temperatures up to $T/T_C > 1.1$.

It is this second result, the observation of a large magnetic response above T_C, that led us to take magnetic domain images in the vicinity of T_C. Combining the results from these two experimental approaches, we were able to establish that remanent and spontaneous magnetization coincide. Therefore we suggest that the strong response to a small magnetic field is a consequence of the film becoming Heisenberg-like above T_C.

This conclusion does not follow trivially from Fig. 1, and some remarks might be appropriate at this point to

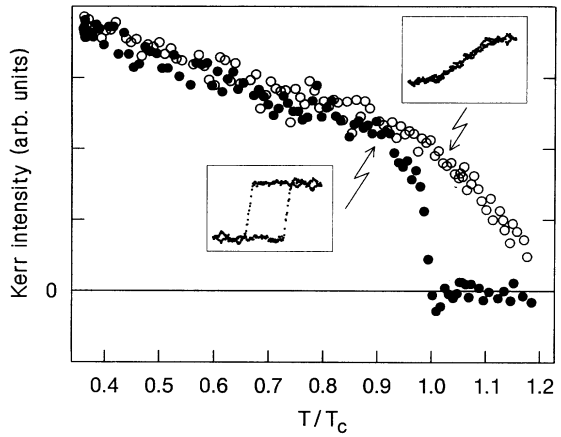

FIG. 1. Temperature dependence of the Kerr intensity I for a Co/Cu(100) film at an applied field of $H=0$ (M_R, solid circles) and in an applied field of $H=8$ kA/m (M_H, open circles); Curie temperature $T_C=289.5$ K. Insets: Two typical hysteresis loops at $T/T_C=0.90$ and 1.03, with field swept between ± 1.2 kA/m.

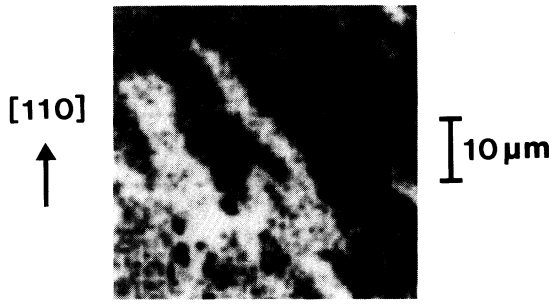

FIG. 2. Magnetic domain image for a Co/Cu(100) film at $T/T_C=0.7$ after demagnetizing in a magnetic ac field of decreasing strength. The in-plane magnetization component along the [110] direction is shown; the out-of-plane magnetization vanishes.

appreciate the subtlety of the argument. First, the Curie temperature is defined as the temperature at which the *spontaneous* magnetization rather than the *remanent* magnetization vanishes. Clearly the identification holds if square hysteresis loops are observed up to T_C. This is, however, not the case in our films; see Fig. 1. The hysteresis loops are square as long as M_R and M_H coincide, whereas near T_C their shape more closely resembles the ones observed in Fe/W(100) [15]. Second, a loss of the remanent magnetization concurrent with a sizable M_H could be caused by the occurrence of opposite static magnetic domains, which tend to reduce M_R. This then would imply that the Curie temperature in our films is much higher than our (in this case rashly defined) T_C. Indeed a theoretical argument exists suggesting that domain formation is a critical process [16]. There is, contrary to this explanation, an important observation by Gradmann *et al.* [17]. In Fe/W(110) films they detected magnetometrically a sizable magnetization in a small applied field at temperatures for which the hyperfine splitting in Mössbauer spectroscopy had already collapsed to zero. There is no way the presence of magnetic domains could make the hyperfine field vanish since Mössbauer spectroscopy is an ideal local probe. On the other hand, superparamagnetic response in epitaxial films has been demonstrated [18] to be important, and cannot be excluded based only on the Mössbauer spectra of Ref. [17]. If our Co/Cu(100) film behaves superparamagnetically then the temperature where M_R vanishes should be identified with a *blocking* temperature rather than with a *Curie* temperature.

We note that all three objections emerge from the same physical origin. We have to establish that our T_C defined in Fig. 1 is indeed the Curie temperature of the Co film, or—likewise—that the spontaneous magnetization M_S is identical to the remanent magnetization M_R in the entire temperature range, in particular near T_C. We used our spin-polarized scanning electron microscope [19] to extract precisely M_S. The spin polarization of the secondary electrons within a single domain locally measures a quantity proportional to the spontaneous magnetization for a given film with a lateral resolution of 20 nm.

The Co films as evaporated are in a single domain state over millimeter-sized areas, in perfect agreement with the earlier observations of the same system by Oepen *et al.* [20]. Following Ref. [20] we can generate domains of a few micrometers up to several 100 μm in size by demagnetizing the sample in a decreasing ac field. A typical domain pattern far below T_C ($T=0.7T_C$) is shown in Fig. 2, confirming the irregular domain boundaries and [110] as the easy magnetization direction.

The crucial experiment now consists of imaging the magnetic domains on the identical surface area versus temperature up to $T/T_C>1$. The difference in spin polarization between oppositely magnetized domains then yields a temperature dependence of polarization of the spontaneous magnetization $M_S(T)$. The result of this experiment is summarized in Fig. 3. For comparison the out-of-plane component of $M_S(T)$ is measured simultaneously to keep track of spurious instrumental asymmetries, and we deduce that they do not exist within experimental accuracy. Note that $M_S(T)$ of Fig. 3 and $M_R(T)$ of Fig. 1 coincide completely—despite the fact that two different techniques were used and two seemingly different quantities—M_R and M_S—probed. The domain images of the most interesting temperature range near T_C are given in Figs. 4(a)–4(d). For each image, temperature has been stabilized to ± 0.1 K, and the temperature increases from image to image by 1 K. We see that the contrast between oppositely magnetized regions becomes fainter and fainter, and no contrast is visible above T_C except statistical noise. The exact change in domain size versus temperature near T_C is a very interesting and complex issue in itself, but one that is clearly beyond the scope of this paper. The significance of

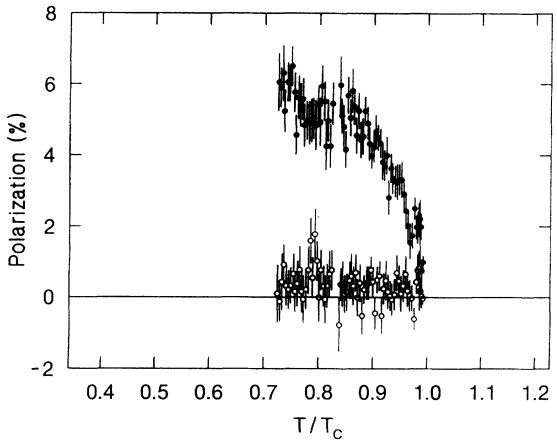

FIG. 3. Temperature dependence of the spin polarization (or spontaneous magnetization M_S) for a Co/Cu(100) film deduced from scans across oppositely magnetized domains. No field has been applied throughout the measurement. Solid circles: in-plane spontaneous magnetization; open circles: out-of-plane spontaneous magnetization (which vanishes completely). Note the perfect agreement between $M_S(T)$ determined by magnetic domain imaging and $M_R(T)$ in Fig. 1 from the Kerr effect.

studying magnetic domains in the entire temperature range from $0.7T_C$ up to $> T_C$ can be summarized as follows.

(i) To within 1 K, we have ruled out that the vanishing of the remanent magnetization at our T_C is due to the film breaking up into domains and hence masking the true Curie temperature.

(ii) We have established that it is correct to define a Curie temperature from the disappearance of the remanent magnetization measured by the Kerr effect by proving that the microscopic spontaneous magnetization is identical to the macroscopic remanent magnetization.

(iii) By observing the domains up to T_C we have disproved that superparamagnetism plays a role in our Co/Cu(100) films. The disappearance of M_R occurs in a temperature interval much sharper than what can be expected for a superparamagnet. Moreover, we see no relaxation of spontaneous magnetization or domains within the duration of our observations.

Therefore we conclude that the M_H curve in Fig. 1 is the strong response of the magnetization to an applied field above the Curie temperature of the system.

We suggest that the behavior above T_C is a consequence of the system becoming an almost ideal Heisenberg magnet, i.e., at T_C the symmetry-breaking interactions that are responsible for the existence of an ordered low-temperature phase are no longer important and cease to affect the behavior of the correlation functions, which are therefore able to display their pure Heisenberg character. The Heisenberg magnet in two dimensions was described by Pokrovskii [21] as consisting essentially of a set of large blocks with linear size R_c (in units of the lattice constant) and elementary spin about πR_c^2. R_c has the value (for a square lattice) of $R_c = \exp[2\pi\Gamma(T)/T]$, where $\Gamma = 2JS^2$, S is the spin per atom, and J is the exchange energy. The presence of these large spin blocks effectively enhances the "small" applied magnetic field by a factor of πR_c^2. At T_C, where $2\pi\Gamma(T_C)/T_C$ has the universal value of 4 (Ref. [21], p. 634), this enhancement factor attains a considerable value, $\pi R_c^2 \simeq 10^4$. As a consequence, the magnetization M at T_C calculated with the usual mean-field equation in this enhanced applied field yields $M(T_C)/M(T=0) \simeq 0.5$. This is roughly in agreement with the observation made in Fig. 1 and has to be compared with a value of $M(T_C)/M(T=0) \simeq 0.01$ obtained for a standard bulk ferromagnet, which does not have spin blocks. The anomalous response of the system to an applied magnetic field can therefore be ascribed to these 2D Heisenberg spin blocks.

Finally, we would like to comment on earlier investigations related to the present experiment and their interpretation. From the present study it becomes clear that owing to this giant magnetic response well above T_C, the results obtained from earlier attempts [7,22] to determine Curie temperatures with an applied field were incorrect. Indications of a 2D Heisenberg system are also found in Fe/Ag(100) multilayers [23]. Quite similar to the Fe/W(110) films, relaxation effects in Mössbauer spectra

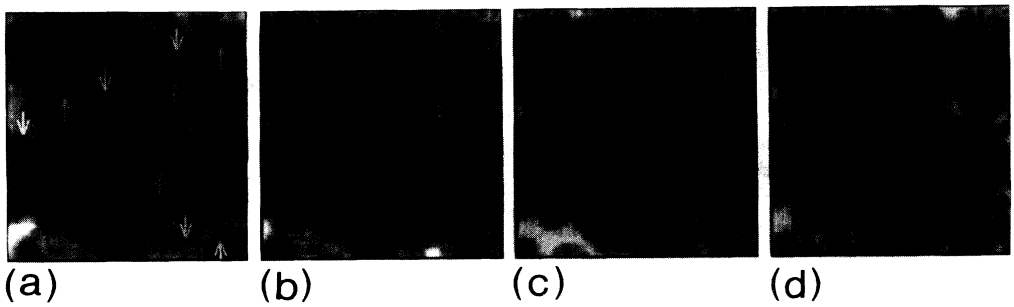

FIG. 4. Magnetic domain images near the Curie temperature on the identical 31-μm×33-μm surface area for temperatures of (a) T_C-2 K, (b) T_C-1 K, (c) T_C, and (d) T_C+1 K. The temperature has been stabilized for each image to within ± 0.1 K. The bright spot in the lower left corner is caused by a defect in the sample substrate. The magnetization direction within the oppositely magnetized domains is indicated in (a).

are observed, but in contrast to Ref. [17], superparamagnetic response is ruled out by magnetizing along easy as well as hard magnetization directions. Unfortunately the experiments on multilayers suffer from a gradual transition from 2D to 3D behavior with decreasing Ag thickness, which might mask the true 2D properties to be investigated. Nevertheless we argue that Fe/Ag(100), Fe/W(110), Fe/W(100), and Co/Cu(100) all behave essentially similar in terms of a proper Hamiltonian: Above T_C, a 2D Heisenberg model might be the most realistic one. A previous study succeeded in determining the critical exponents γ above and β below T_C in 1 ML Gd/W(110) by electron spin resonance [24]. This result seems to rule out an anomalous response of magnetization to applied field above T_C. Obviously Gd/W(110) behaves in a completely different way from Co/Cu(100). The former has a large out-of-plane uniaxial anisotropy and is therefore considered a 2D Ising system. Hence this deviating behavior above T_C is to be expected and corroborates our interpretation of Co/Cu(100) being a 2D Heisenberg system, in which no uniaxial anisotropy is present.

In conclusion, we have offered a consistent explanation for the anomalous response of magnetization to an applied field in a ML Co/Cu(100) film. It is based on joint experimental evidence from Kerr magnetometry and spin-polarized scanning electron microscopy, on the observation of the temperature dependence of remanent as well as spontaneous magnetization, and on imaging the static magnetic domains within 1 K around T_C. We point out that these epitaxial films appear to be the most perfect realization of a truly 2D ferromagnet. A straightforward extension of the present work will be to investigate the critical region above T_C, which has yet been undertaken experimentally only for one class of approximate 2D systems, the layered magnets developed in the 1970s [22,25].

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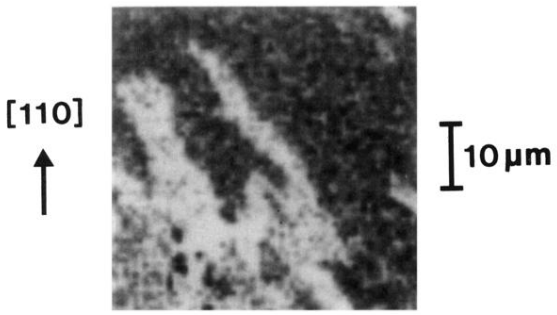

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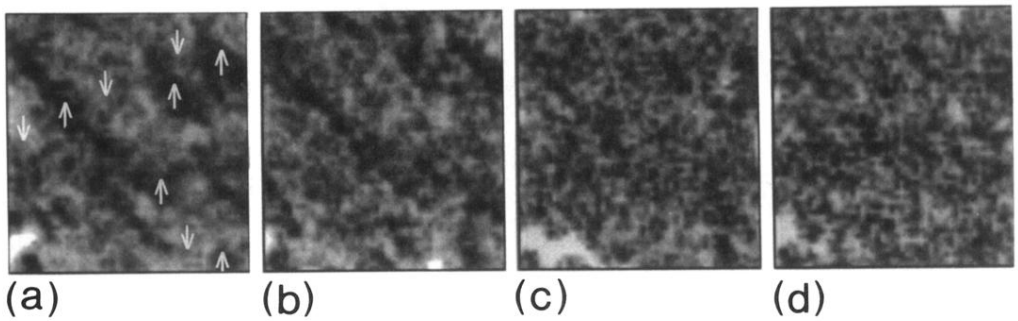

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