Evidence for domain formation near the Curie temperature in ultrathin Ni/Cu (001) films with perpendicular anisotropy

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Ni films, 8–10 monolayers thick on a Cu(001) substrate, are studied in UHV by the polar magneto-optic Kerr effect near the Curie temperature T_C. The films are magnetized normal to the surface. An unusual sharp drop of the remanent magnetization (M_r) and a large difference of about 10 K are measured between the temperature T_r at which M_r and the temperature T_H at which the magnetization $M(H)$ in a static field of 20 G vanishes. We show that at T_r the films break up into ferromagnetic domains with perpendicular magnetization components and that T_C is larger than T_r. The true T_C is deduced from field-dependent magnetization measurements. [S0163-1829(97)51718-8]

Tetragonally distorted fct Ni(001) ultrathin films on Cu(001) have been recently found to present an unusual spin-reorientation phase transition (SRT) from in plane to out of plane with increasing film thickness at about 7–8 monolayers (ML)[1–3] and increasing temperature.[4–7] Fourth-order anisotropy[4] terms make this unusual SRT a continuous (second- or higher-order) one, as it is proven by ferromagnetic resonance (FMR) measurements and magneto-optic Kerr effect (MOKE) experiments.[4–7] In both cases, thickness- and temperature-dependent SRT, no domain formation was evident near the borders of the SRT, in contrast to previous findings in ultrathin Fe/Cu(100)[8] and Fe/Ag(100)[9,10] films, where stripe-domain configurations were recorded at remanence.

For films with a thickness slightly larger than the critical one perpendicular anisotropy is small. As the temperature increases, anisotropy decreases with a high power of the spontaneous magnetization[11,12] and the question arises, what happens near T_C with the perpendicular magnetization. The competition between the perpendicular anisotropy and the demagnetization energy could result in a SRT from out of plane to in plane or, alternatively, in breaking the film into magnetic domains near T_C. Indeed, Yafet *et al.*[13] have proposed that a multidomain state with perpendicular magnetization could be stabilized in films with perpendicular anisotropy. Garel *et al.* have also shown that increasing spin fluctuations near T_C favor a multidomain state.[14] Upon describing a critical phenomenon one has to define an appropriate long-range order parameter. For the ferromagnetic to paramagnetic phase transition the spontaneous magnetization M_{sp} is the order parameter and the temperature, at which M_{sp} vanishes, defines the T_C of the magnetic system. However, in static magnetic and magneto-optic experiments, the observable that is measured is not M_{sp}, but the component of the remanent magnetization, M_r parallel to the direction of measurement. *M_r coincides with M_{sp} only if the system is in a single-domain state up to T_C and measurements are taken along an easy axis.* This is the case for ultrathin films with the easy axis in plane. Various temperatures may be distinguished in the experiment: T_r at vanishing remanence, T_H for $M(H)\rightarrow 0$ in small applied fields, and the proper T_C. For an in-plane magnetized 30 nm Gd(0001) film on W(110) a difference of about 2% (with $H=200$ G) between T_r and T_H has been shown to be consistent with a field induced magnetization.[15] The well-known Arrott-Kouvel plots[16] were used to determine the T_C of the sample. The T_C was found to be the temperature where M_r vanishes.[15] A difference of about 20% between T_r and $T_H$has been observed in an ultrathin 1 ML Co film on Cu(100)[17] in an applied field of 100 G. In that case no domain formation above T_r was observed within the spatial resolution of the experiment (20 nm), and T_C was determined to be identical with T_r. The large difference between T_C and T_H was attributed to purely two-dimensional behavior at temperatures higher than T_C.[18] Similar results were recorded later for 1–2 ML thick, in-plane magnetized Co/Cu(100) and Fe/W(110).[19,20] The question of domain formation in a perpendicular magnetized film near T_C stimulated our interest to investigate in detail the behavior of ultrathin Ni/Cu(001) films in the thickness range of 8–10 ML. The lower limit of 8 ML was determined by the onset of perpendicular anisotropy, while the upper limit of 10 ML was given by the T_C of the samples and the temperature up to which the samples could be heated without considerable interdiffusion.[21,22]

Ultrathin Ni(001) films were prepared in an ultrahigh vacuum on a Cu(001) substrate, as described earlier.[3–7] The thickness accuracy is better than 0.5 ML, and the temperature accuracy and stability is better than 0.1 K.

Magneto-optic hysteresis loops in polar geometry were recorded *in situ* by applying a magnetic field of ± 100 G. The Kerr signal (mrad) was calibrated to magnetization units (G) by comparison with the signal of a 20 ML Ni film considered to possess the magnetization of bulk Ni at room temperature and by correcting for the film thickness. ac-susceptibility measurements were performed via MOKE by applying a small oscillating magnetic field.[23] For all experiments the earth's magnetic field was compensated down to 10 mG.

In Fig. 1(a) the magnetization for an 8.4 ML film is shown measured in a static H of 20 and 0 G. The data were collected before (open) and after (closed symbols) 9 hours of continuous measuring at about 390 K. The effect of such a prolonged annealing on the magnetic properties of the film

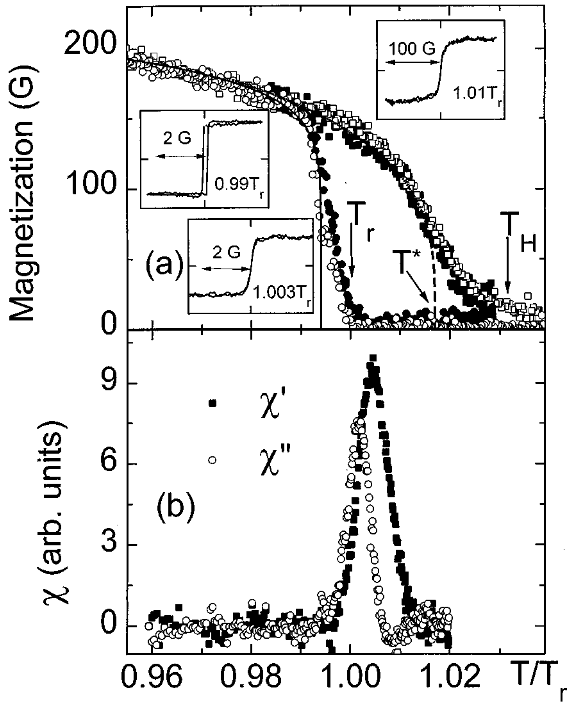

FIG. 1. (a) Magnetization at remanence (circles) and with an applied field of 20 G (squares) of 8.4 ML Ni/Cu(001). Solid and dashed lines are power-law fits according to $M_r \propto (1-1/T_r)^{0.12}$ and $M(H=20\ \mathrm{G}) \propto (1-T/T^*)^{0.28}$, respectively. Open (closed) symbols represent data taken before (after) 9 h of measuring at 390 K. The insets show typical hysteresis loops at the indicated temperatures; $T_r=381.5$ K, $T_H=391.2$ K. (b) The complex ac-susceptibility $\chi=\chi'+\chi''$ measured in polar MOKE geometry as a function of temperature for the same sample.

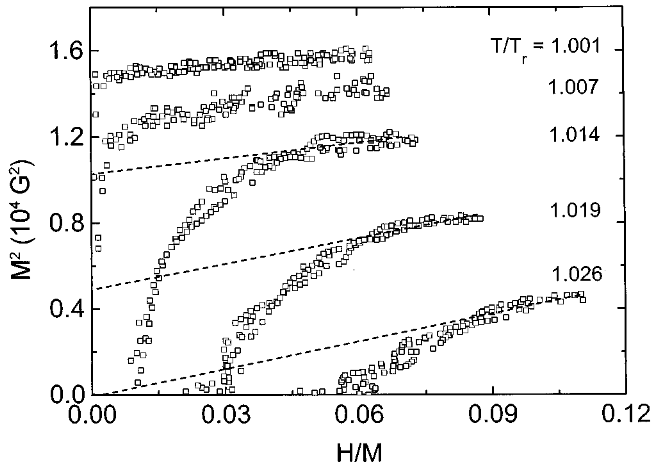

FIG. 2. Arrott-Kouvel plots for the 8.4 ML Ni/Cu(001) film at temperatures close to T_C with a maximum applied field of 100 G. The correct T_C is found by extrapolating the high-field data to zero (dashed lines).

was found to be negligible. The remanent signal decreases sharply near a temperature $T_r=381.5$ K (which we normalized to unity). There is quite a large difference of about 3% between T_r and the temperature T_H at which $M(H=20$ G) disappears. Even with a small applied field of 2 G this difference is larger than 1%, and it exceeds 5% for 100 G. Similar behavior has been recorded for all 8–10 ML thick films. Such a large temperature difference is not expected for homogeneous ferromagnets obeying the molecular field theory.[24] In the inset of Fig. 1(a) hysteresis loops are plotted at temperatures $0.99T_r$, $1.003T_r$, and $1.01T_r$. At temperatures just above T_r even a small field of 0.3 G is capable of driving the magnetization of the sample to saturation without any hysteresis effect. Such a sharp increase of $M(H)$ could be attributed to either 2D behavior or the change from a multidomain to a single-domain state. Superparamagnetism is totally inconsistent with the structure of our 8–10 ML thick Ni/Cu(001) samples.[21,22]

A power-law fit is depicted in Fig. 1(a) with M_r considered to be the order parameter (solid line). This fitting is carried out by choosing T_C to be the temperature that maximizes the range of a straight line in a $\log M$-versus-$\log(1-T/T_C)$ plot.[25] An *erroneous value of the critical exponent* $\beta=0.12$ is deduced this way, suggesting two-dimensional behavior for an 8.4 ML thick film. For 8–10 ML thick films it is rather unlikely to have two-dimensional behavior. A crossover from two- to three-dimensional behavior is usually found already at approximately 5 ML, for example by FMR measurements in Ni(111)/W(110),[26] or by MOKE in Ni/Cu(001) films.[1,27]

Even if one selects T_C to be equal to T_r, as in Ref. 15, the result of the fitting is improved, and the value of β would only slightly increase to 0.16. If we assume that domain formation occurs, then the remanence would not be the proper long-range order parameter. A better parameter would be, as a first approximation, the magnetization recorded slightly above the critical magnetic field, necessary for the formation of a single-domain state. Fitting $M(H=20$ G), a value of $\beta=0.28$ is obtained, while this power law vanishes at $T^*=388.0$ K, about 1.7% higher than T_r (dashed line). Also for perpendicular magnetized Co/Cu(111) films $M(H\neq 0)$ was found to be the better ''order parameter.''[28]

The real (χ') and imaginary (χ'') part of the susceptibility recorded with an ac field of amplitude 0.7 G is depicted in Fig. 1(b) for the same film. The two parts present a maximum at slightly different temperatures close to T_r.[23] A maximum of χ' is expected to occur at about T_C (Ref. 29) for $H\rightarrow 0$. It depends on the amplitude of H. As we will show below, the temperature at χ'_{max} does not reflect the true T_C in this case. χ'' peaks at a slightly lower temperature than χ' and is sensitive to the absorptive part of χ. An absorption signal is observed as long as the coercivity is smaller than the modulation amplitude.[23]

Further evidence for the existence of ferromagnetic domains above T_r is obtained by the Arrott-Kouvel analysis. In this analysis the $M^2(H/M)$ curve is known[16] to become a straight line passing through the origin exactly at T_C where the susceptibility $\chi= M/H$ diverges. Magnetization data up to $H=100$ G around $T^*$are shown in Fig. 2. The top data set corresponds to $T_r(T/T_r=1.001)$, while the bottom plot is recorded at $T_H(T_H/T_r=1.026)$. A downwards curvature is exhibited in all plots as H approaches zero. A similar curvature is expected to occur in the Arrott-Kouvel plots for temperatures just below T_C in the ferromagnetic region.[16] The intercept of such a plot with the Y axis has always to be positive. This condition is only fulfilled at $T/T_r=1.001$ and 1.007. For the rest of the plots neither the treatment of Arrott

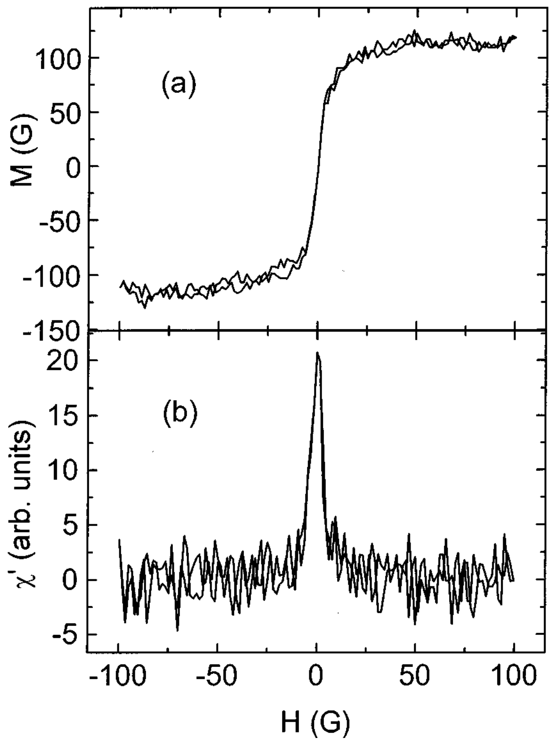

FIG. 3. Hysteresis loop (a) and the corresponding susceptibility $\chi(H)=\mathrm{d}M(H)/\mathrm{d}H$ (b) at 1.01 T_r for the 8.4 ML NI/Cu(001) sample. Note that χ' shows only one maximum value at zero magnetic field.

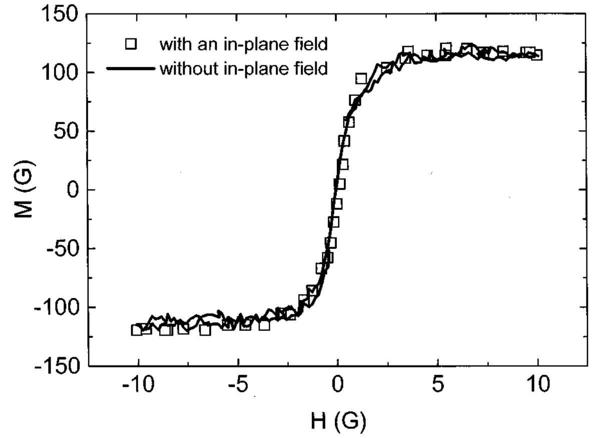

FIG. 4. Hysteresis loops at $1.005T_r$ with (open symbols) or without (solid line) an in-plane bias field of 16 G, for the 8.4 ML Ni/Cu(001) film.

and Kouvel[16] nor improved versions taking into account critical fluctuations[30] could give a satisfactory explanation. On the other hand, such an abrupt curvature could be explained if one considers domain formation at a critical field at temperatures below T_C. Then T_C would be correctly determined by extrapolating the high-field data to zero (dashed lines). The T_C evaluated this way is 2.6% higher than T_r (Fig. 2) and even 0.8% above the one evaluated by the power-law fitting. This suggests that the 20 G field is not capable of creating a single-domain state for all temperatures up to T^* [Fig. 1(a)]. The extrapolation of the $T/T_r=1.026$ data obviously has a large error bar. And it seems likely that the true T_C lies at a higher temperature. For a more accurate determination of T_C a larger H would have been necessary. However, the scope of the present work is to provide evidence that domain formation causes the abrupt disappearance of M_r at T_r and not the precise determination of T_C itself. It may be of some use for other discussions in the literature that $M^2=f(H/M)$curves, as in Fig. 2, for $T/T_r=1.001$ or 1.007 could also be observed for a two-dimensional ferromagnet exactly at T_C due to the high value of the critical exponent $\delta=15$ (Ref. 20) for $M(H)$ at T_C. In such a case T_C of the film would be equal to T_r. However, as discussed above, two-dimensional behavior for our 8.4 ML film is not likely.

Let us now come back to the ac-susceptibility results, shown in Fig. 1(b). At temperatures close to T_r the saturation field is very small, as is shown in the inset of Fig. 1(a) for $T=1.003T_r$. For such temperatures our ac-field amplitude of 0.7 G is capable of creating a single-domain state, and a maximum of the temperature-dependent susceptibility is recorded at about T_r. At higher temperatures the ac-field amplitude is not large enough to produce a single-domain state, and the susceptibility is almost equal to zero. This interpretation is supported by the extremely narrow ac-susceptibility χ' peak (less than 3 K) compared to the ones usually recorded at a real T_C.[29,31]

In Fig. 3 we show a typical hysteresis loop (a) and its derivative (b) recorded above T_r. It is interesting to note that there is a characteristic difference between the shape of the reversible hysteresis loop [Fig. 3(a)] recorded for our films and the ones recorded previously[10] for films exhibiting stripe-domain configurations. In the latter case the susceptibility $\chi(H)$ showed two sharp peaks at the positive/negative critical field for domain formation.[10] In our films $\chi(H)$ [Fig. 3(b)] has a single peak at zero field only. This behavior could indicate another type of domain configuration than stripe-domains with the spontaneous magnetization normal to the film plane. For example, a complicated canted domain configuration may exist.[3–5]

In order to find a further indication for the correct T_C of our film an additional experiment was performed. We recorded hysteresis loops at a temperature slightly higher than T_r in two ways. First, by varying the field normal to the film plane between ± 10 G. A small field of less than 2 G saturates the film magnetization while there is no remanence, as is shown in Fig. 4 (solid line). Secondly, with the same perpendicular field, but applying in addition a static magnetic field of 16 G parallel to the film plane (Fig. 4, open symbols) there is no difference between the two data sets. This suggests that the sample presents perpendicular anisotropy at temperatures higher than T_r, and the anisotropy field is much higher than 16 G. Since there is no isotropic (that is paramagnetic) behavior at temperatures higher than T_r, as one would expect if $T_r=T_C$, it seems more reasonable to consider the small saturation field of 2 G as a critical field for domain formation.

In the present work we show that domain formation at temperatures below T_C occurs in perpendicular magnetized Ni/Cu(001) ultrathin films and yields an abrupt decrease of the remanent signal far below T_C. In this case the remanent magnetization, which is usually taken as the order parameter, leads to *erroneous conclusions about T_C and eventually to an erroneous determination of a critical exponent β.* The magnetization recorded in a small field becomes a better choice for the order parameter.[28] Interestingly, the ac susceptibility recorded in polar geometry was found to present a maximum near T_r. However, the width of the peak is much smaller than the one usually found at T_C. This indicates that the maximum of the ac susceptibility is related to domain formation. Finally, we have shown that two-dimensional or multidomain behavior cannot be strictly distinguished in such cases. In our 3D case, where we find perpendicular magnetized domains just below T_C, the correct T_C needs to be determined by an extrapolation of the high-field (i.e., single-domain state) data to zero. In our particular film (Fig. 1) T_C seems close to $T_C \approx 398$ K, that is about 20 K above the apparent value at which M_r vanishes.

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