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Composition-driven change of the magnetic anisotropy of ultrathin Co/Au(111) films studied by means of magnetic-force microscopy in ultrahigh vacuum

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The magnetic anisotropy of ultrathin Co films on Au (111) changes with increasing film thickness in the regime of 3–5 monolayers (ML). This leads to a reorientation of the magnetization from the out-of-plane to the in-plane direction. We observed this transition by means of magnetic-force microscopy under UHV conditions. We found that by approaching a film thickness of 4 ML the average domain size shrinks from about 1 μm to 400 nm. For films thicker than 4 ML we have observed an in-plane domain structure. The magnetic structure of the ultrathin Co films was found to depend additionally on the chemical composition. For a Co film of a thickness just above the critical thickness for the reorientation transition the magnetic structure reverted from in-plane to out-of-plane due to carbon contamination of the thin films. We also found that the critical Co film thickness for the reorientation transition could be shifted even up to 18 ML by the presence of carbon. [S0163-1829(99)03106-9]

I. INTRODUCTION

In the past few years there has been growing interest in the magnetic anisotropy of ultrathin films. This attention originates both from fundamental physics and from applications in information storage technology. A strong effort has been made to discover and develop new materials which could be used in future recording media. Ultrathin Co films on Au (111) substrates reveal a magnetic reorientation transition from out-of-plane to in-plane anisotropy at a thickness between 3 and 5 monolayers (ML). This interesting property has been previously observed by scanning electron microscopy with polarization analysis (SEMPA).[1,2] Since the perpendicular anisotropy in magnetic thin films finds an application in perpendicular recording the observation is of great interest to explore in more detail.

In the present study we have applied magnetic-force microscopy (MFM) under ultrahigh vacuum (UHV) conditions in order to examine the magnetic domain structure of ultrathin Co/Au (111) at high spatial resolution as a function of film thickness and carbon content. While our MFM observations of the magnetic reorientation transition for clean Co films are in full agreement with earlier SEMPA studies we have found that the presence of carbon can significantly defer the reorientation transition by at least a factor of 4. This surprising result may have applications using ultrathin carbon-containing Co films for high-resolution perpendicular magnetic recording.

II. EXPERIMENTAL

We studied the magnetic structure of Co/Au (111) films by using UHV-MFM at a base pressure of 5×10^{-11} mbar. This technique allows for high lateral resolution, as well as for high sensitivity. The MFM was based on a commercial atomic-force microscopy/scanning tunneling microscopy (AFM/STM) system[3] with several operating modes available. We could perform standard STM using an etched tungsten wire or a sputtered silicon cantilever as the tunneling tip. The AFM operation mode allowed us to detect forces or force gradients. The latter is performed by vibrating the cantilever at its resonance frequency and detecting the frequency by FM detection.[4] We implemented a MFM scan mode allowing the measurement of force gradients at a constant tip-to-sample distance.[5]

The AFM/STM probe and the sample could be exchanged under UHV conditions. Thus, we were able to test the Fe-coated tips used as probes in this study on known samples before measuring Co/Au (111) thin films. Furthermore, we could measure the same Co film with several tips to confirm the results we have obtained. The maximum lateral scan range of the microscope is 6 μm.

The magnetic sensors for the MFM studies were prepared within the UHV system. Si cantilevers, which were specified to have spring constants of 1–3 N/m and resonance frequencys of 60–80 kHz, were cleaned by Ar^+-ion sputtering for 90 min, at a kinetic energy of 1.5 keV for the Ar^+ ions. Afterwards, the cantilevers were coated by a 5–10 nm thick film of Fe. Finally, the tips were checked by performing MFM on a Co/Pt-multilayer sample, which is known to have some hundreds nanometer wide domains, magnetized perpendicular to the surface.[6] The MFM images of this sample show a dark and bright contrast of the magnetic domains. This reveals the tip to be magnetized in the z direction. By this characterization, we could sort out our sensors, which gave a low signal or a bad resolution.

An Au (111) single crystal was used as the substrate. The miscut was specified to be better than 2°. Hence, the average size of the atomically flat terraces is at least 6 nm. The crystal was cleaned by cycles of Ar^+-ion etching at an ion energy of 600 eV and subsequent heating to 450 °C. The surface was checked by Auger, low-energy-electron diffraction (LEED) and STM. As soon as LEED indicated the $(23\times\sqrt{3})$ reconstruction of the Au (111) surface[7] and STM revealed a clean surface [Fig. 1(a)], the further preparation was stopped.

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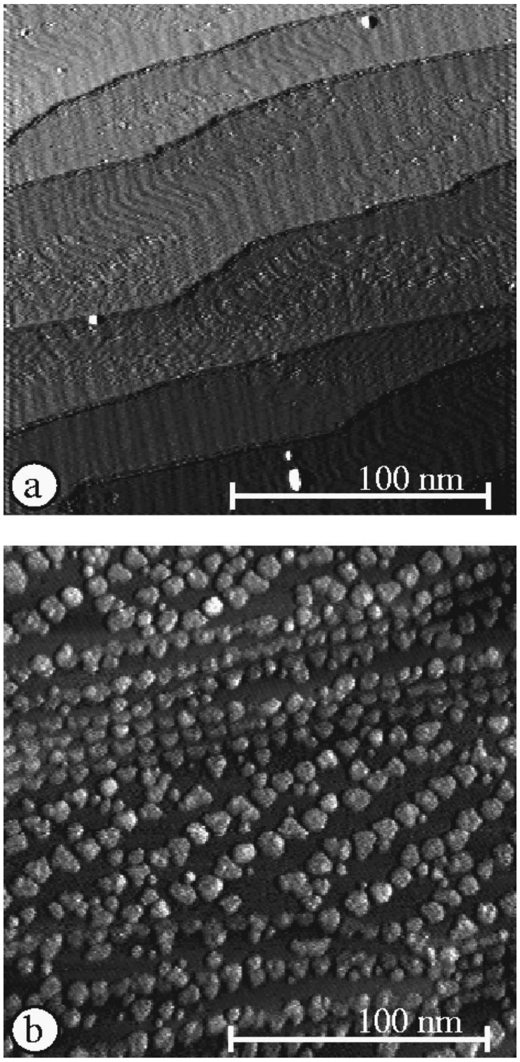

FIG. 1. (a) STM image of the clean Au (111) surface exhibiting the $(23\times\sqrt{3})$ *herringbone* reconstruction. (b) early stage of Co film growth, as used to estimate the evaporation rate of Co. The Co islands of a height of two monolayers decorate the elbow sites of the underlying Au(111) *herringbone* reconstruction. Scan size: 200 nm×200 nm.

We deposited the Co on Au (111), as well as the Fe on the Si cantilevers, by electron-beam evaporation. The flux rate of the Co or Fe, which is a measure for the evaporation rate, could be kept constant by automatically controlling the high voltage on the target. Hence, we determined the film thickness from the flux rate and duration of evaporation. For calibrating the Co evaporator, we prepared a submonolayer film on the Au (111) single crystal and measured the actual coverage by STM to get the evaporation rate [Fig. 1(b)]. Co starts growing in islands of 2 ML at the elbows of the $(23\times\sqrt{3})$ *herringbone* reconstruction.[8] The rate was found to be quite stable, if the filament current as well as the current between the filament and the evaporant was kept constant. The typical evaporation rate was about 0.6 ML/min.

III. RESULTS AND DISCUSSION

The magnetic anisotropy of ultrathin Co films on Au (111) changes with increasing film thickness in the regime of a few ML. We measured the thickness dependence of the magnetic reorientation using two ways of sample preparation. One way was to prepare the Co films starting from a clean Au substrate each time. The second method was to increase the film thickness in steps by evaporating Co on top of an existing film.

In the first case our results met the observation of the other authors, who found the transition to take place at about 4 ML of Co. Our results are displayed in Fig. 2. The images were obtained with several different cantilevers. The first five images show MFM data for a Co film thickness of 1.8, 2.0, 3.0, 3.2, and 3.8 ML. For 1.8 ML only a weak contrast is visible, which will be discussed later. The next four images show areas with dark and bright contrast. This feature can be interpreted as the magnetic domain structure of a film magnetized perpendicular to the surface. The size of the domains first grows (film thickness from 2 to 3 ML),[9] then shrinks as approaching the magnetic reorientation of the anisotropy, as shown for 3.2 and 3.8 ML.

For next three images, which represent a Co film thickness of 4.1, 4.3, and 4.5 ML, respectively, the appearance changes. Now, lines of dark and bright contrast can be seen. The contrast becomes more clear with increasing film thickness of 4.6, 4.8, 6.0, and 7.0 ML in Fig. 2. Since the tips were magnetized perpendicular to the sample surface, such a contrast indicates an in-plane magnetic domain structure. The image of the 6.0 ML film of cobalt shows an additional contrast within the magnetic domains which can be interpreted as a magnetic ripple structure.[10,11]

It is an interesting question for ultrathin Co films on Au (111) at which coverage the ferromagnetic order occurs and what type of domain structure could be observed. The first image in Fig. 2 shows the MFM data at the minimal thickness of the Co film, where we obtained a magnetic signal in addition to topographic crosstalk. We examined this sample in more detail. Figures 3(a) and 3(b) show STM data of a scan range of 400 nm×400 nm and 100 nm×100 nm. The Co film consists of islands of 2 ML height, where the third and fourth layer already started to grow. The islands began to coalesce, but there are still single islands as well as fractions of uncovered Au. Two MFM images of 3 μm×3 μm and 1.5 μm×1.5 μm scan area are displayed in Figs. 3(c) and 3(d). In the upper right corner of image 3(d), image (a) is displayed at the same scale as image (d) for comparing the sizes of the magnetic and topographic features. The MFM images reveal bright and dark stripes of about 50–100 nm width, running mainly from the bottom left to the upper right corner. The stripes follow basically the steps of the Au (111) crystal.

To confirm the magnetic origin of these stripes and exclude possible electrostatic effects, the same sample was measured using a nonmagnetic tip. We had to measure as close to the surface as possible to get any signal at all. The only structure found revealed features of significant smaller size than the magnetic structures and showed a stronger distance dependence. This structure is likely to originate from the *van der Waals* interaction between the tip and the sample surface. The difference in the structure as measured with the magnetic tip on the same sample affirm the magnetic origin of the contrast in Figs. 3(c) and 3(d).

The observation of the same Co film for a longer period

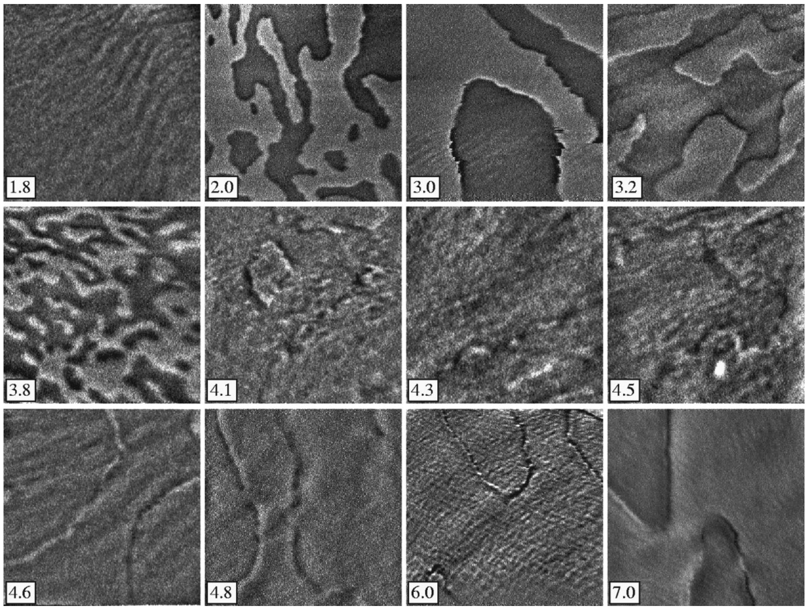

FIG. 2. MFM images obtained at increasing Co film thicknesses. The numbers denote the actual film thickness in monolayers. Scan size: 5 μm×5 μm.

of time revealed a change of its magnetic structure. For Co films at a thickness just above 4 ML we achieved a reversal of the magnetic anisotropy from the in-plane to the out-of-plane direction. Former work indicates two possible reasons for this change. Speckmann *et al.* showed for a Co wedge on Au (111) that heating the sample leads to a shift of the critical thickness for the reorientation transition. This was due to the migration of Au to the sample surface.[2] Hope *et al.* found for a Co film on Cu, that the easy axis of magnetization could be switched by 90° within the film plane by CO dosing.[12]

To find out the reason for the change of the magnetic structure in time in our case, we measured MFM and Auger spectroscopy on a 5 ML thick Co film for several days. From the Auger data the peak heights of two Auger lines, Au(165) (h_{Au}) and Co(656) (h_{Co}), were extracted. The quotient $h_{\mathrm{Au}}/h_{\mathrm{Co}}$ is plotted versus time in Fig. 4. Each mark represents the averaged data of five Auger spectra taken within 1 h of time at different spots on the sample. The error bars denote the statistical error. The straight line shows extrapolated data as a guide to the eye. For the migration of Au to the surface the $h_{\mathrm{Au}}/h_{\mathrm{Co}}$ signal should rise in time. This can clearly be excluded by the data. The second data set in Fig. 4 shows the relation of the C(272) Auger peak height to the Co(656) peak height ($h_{\mathrm{C}}/h_{\mathrm{Co}}$). Here we see that the Co surface becomes contaminated by C.

Along with the Auger spectroscopy MFM measurements were performed to observe the domain structure and thus to deduce the magnetic anisotropy. The letters in the lower part of Fig. 4 correspond to the MFM images in Fig. 5. The scan areas of the images are 5 μm×5 μm each. The images (a) and (b) show domain walls indicating that the film is magnetized in plane. For image (c) small perpendicular domains start to be seen, which became more pronounced in (d). The

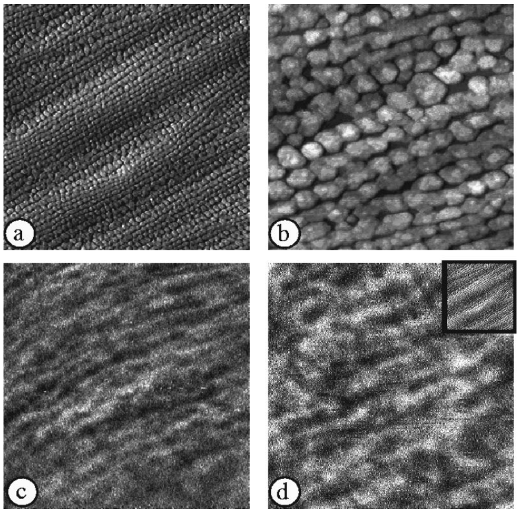

FIG. 3. STM and MFM images of a Co film of 1.8 ML thickness. The images (a) and (b) show the STM data of 400 nm ×400 nm and 100 nm×100 nm scan size. Images (c) and (d) display the MFM data of the same sample of 3 μm×3 μm and 1.5 μm×1.5 μm scan size. For comparison, image (a) is displayed as an inset in image (d) at the same scale.

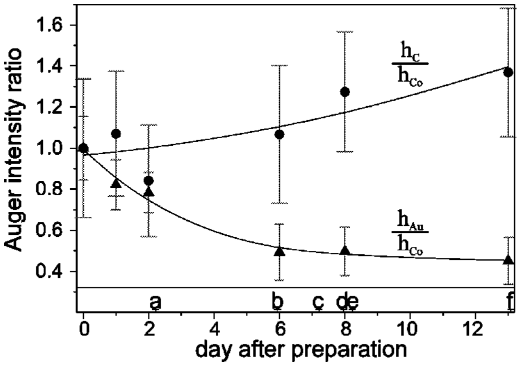

FIG. 4. Intensity of the C(272) and Au(165) Auger line with respect to the intensity of the Co(656) Auger line plotted against time for a 5 ML Co film.

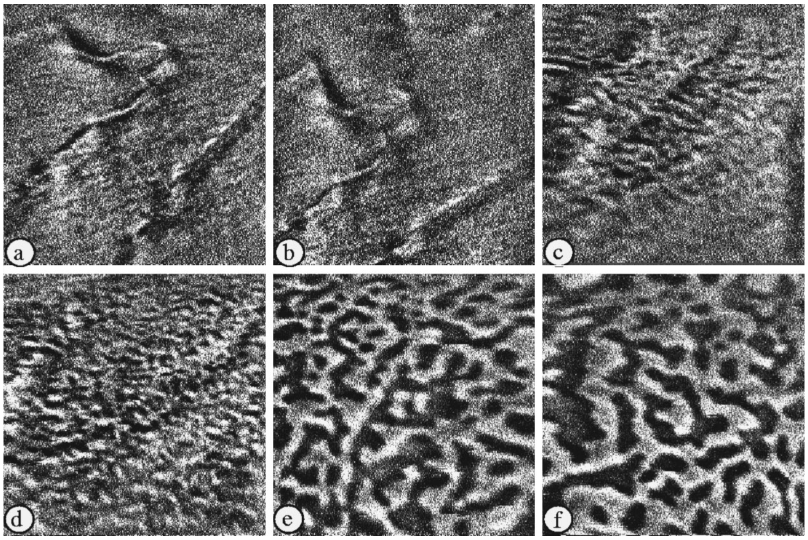

FIG. 5. Evolution of the magnetic structure of a 5 ML Co film in time due to C contamination. The images were taken about 2, 6, 7, 8, 8.2, and 13 days after the sample preparation. Scan size: 5 μm×5 μm.

sample ends up with a perpendicular domain structure as shown in (e) and (f). Comparing the MFM data to the plot in Fig. 4 one observes that the change in the MFM images corresponds to the rise of the C signal.

We studied the reversal effect in more detail by measuring the same area of 5 μm×5 μm on a 4.1 ML Co film sample several times. The CO concentration within the UHV chamber was increased during the experiment. The results of the MFM measurements are displayed in Fig. 6. The images (a)–(e) were taken 50, 90, 130, 170, and 210 min, respectively, after sample preparation. Image (f) was taken the next day, 630 min after preparation. The first image shows two domain walls running parallel from the bottom left to top right corner indicated by arrows. A third wall is visible in the lower right corner. The second image shows the same three domain walls. For the images from (c) to (e) bright and dark areas arise, indicating the nucleation and growth of domains magnetized perpendicular to the sample surface. Finally, a perpendicular domain structure is observed the next day at the same area, as shown in image (f). This time the magnetization reversal went much faster which is due to the increase of the CO concentration.

As mentioned before, we used a second method of sample preparation to study thickness-dependent effects. Starting from a 2 ML film we deposited Co on top, but without cleaning the Au (111) crystal between the evaporation steps. We found a change of the magnetic structure, but at a significantly higher thickness of the Co film. The series of MFM images (Fig. 7, 5 μm×5 μm each) starts at a Co film thickness of 13 ML. For each image an additional monolayer of Co was deposited. The first six images show clearly a perpendicular magnetization. The average width of the magnetic domains changes drastically from about 600 down to 100 nm. From 19 ML on, no clear perpendicular domain structure is visible. For thicker Co films we get some indication of a magnetic ripple structure (Fig. 7, 26–28 ML), but no clear indication of a domain wall is observed. The samples with a Co film thickness of 16–21 ML were also measured at a scan

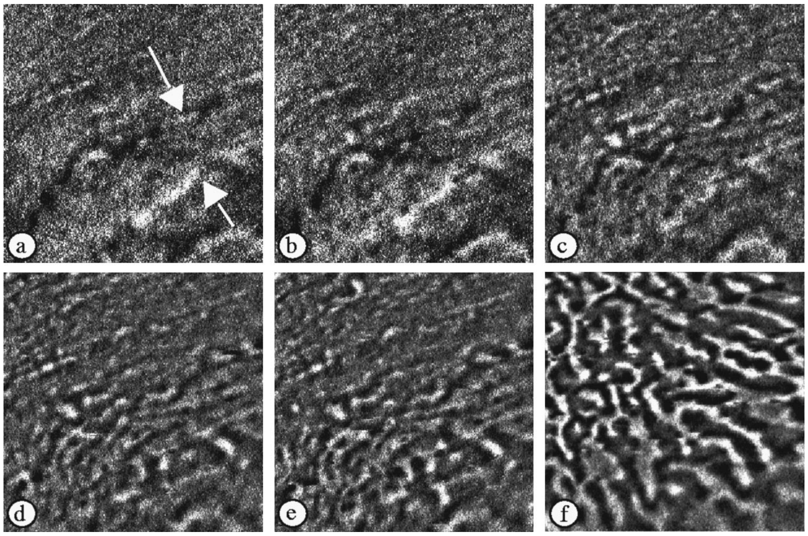

FIG. 6. Evolution of the magnetic structure of a 4.1 ML Co film in time due to C contamination observed at the same spot on the sample, 50', 90', 130', 170', 210', and 630' after sample preparation. Scan size: 5 μm×5 μm.

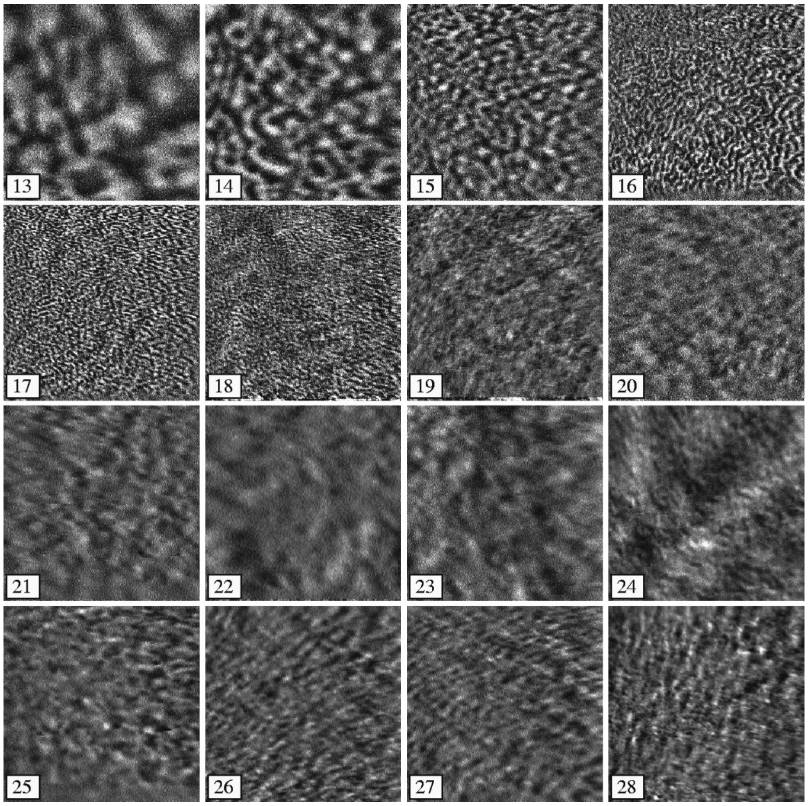

FIG. 7. MFM images of a sample for which the thickness of the Co film was increased in steps of 1 ML starting at 13 ML. Again, the numbers denote the Co film thickness in monolayers. The magnetization is perpendicular up to 18 ML due to C contamination. Scan size: 5 μm×5 μm.

size of 2.5 μm×2.5 μm. The result is displayed in Fig. 8. Up to 18 ML a domainlike pattern can be seen. The next image shows no structure from perpendicular domains, but some influence from the tip on the magnetic structure is present indicating that the Co film becomes magnetically soft at the critical thickness for the reorientation transition. After reaching a Co film thickness of 28 ML we performed Auger measurements. We see a huge C(272) peak, but still signatures of Co and Au. From the series of MFM data presented in Figs. 7 and 8 we conclude that the carbon contaminated Co film exhibits perpendicular anisotropy up to a thickness of at least 18 ML.

IV. CONCLUSION

We observed the reorientation transition of the magnetic structure of ultrathin Co films on Au (111) by means of MFM as a function of the Co film thickness. At a film thick-

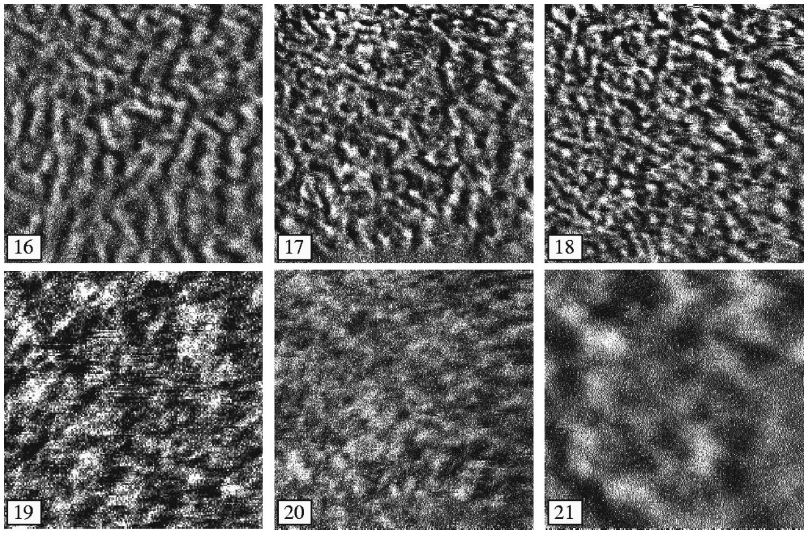

FIG. 8. Close up views of the deferred magnetic transition (cf. Fig. 7) for the 16 ML to 21 ML Co films contaminated by C. Scan size: 2.5 μm×2.5 μm.

ness of 1.8 ML we were able to detect a magnetic contrast of striped domains running parallel to steps on the Au substrate. The width of these stripes was about 50–100 nm. Increasing the film thickness leads to a growth of the average domain size up to several μm at 3 ML of Co. The domain size shrinks when approaching the critical thickness of 4 ML. For thicker films we observed a domain wall contrast indicating a magnetization in the film plane.

Carbon was found to have a huge influence on the magnetic domain structure of cobalt thin films. Due to the C contamination, the easy axis of the magnetization of in-plane magnetized Co/Au (111) films is switched back to the perpendicular direction. Furthermore, the presence of C stabilizes the perpendicular anisotropy up to a Co film thickness of at least 18 ML.

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