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Magnetic anisotropy of ultrathin cobalt films on Pt(111) investigated with x-ray diffraction: Effect of atomic mixing at the interface

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With standard and resonant x-ray magnetic diffraction we investigate the magnetic properties of Pt atoms in a Co/Pt(111) system. It is shown, that, depending on the thickness of the Co films, the magnetic anisotropy as monitored by the Pt magnetism changes from perpendicular to parallel, mimicking the behavior of the Co overlayer. Adsorption of carbon monoxide on very thin films has been found to change the easy magnetization axis. Moreover, the critical film thickness for the flipping of the easy direction of magnetization depends markedly on the growth temperature even in the vicinities of 300 K, where bulk alloying does not occur. A brief annealing to ~375 K of a parallel Co film suffices to reverse its easy direction to perpendicular. By analyzing the magnetic crystal truncation rods of the Pt surface, it is concluded that the change of anisotropy is due to site exchange between interfacial Co and Pt atoms. The site exchange which affects up to 4% of the interface atoms has been related to previous scanning tunneling microscopy findings on the same system. Comparison of magneto-optical and x-ray magnetic diffraction measurements on the same film show that the responses to the externally applied field are not identical. A possible explanation is suggested.

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I. INTRODUCTION

Multilayer and thin alloy films based on Co and Pt have attracted a large research effort in the last decade due to their technological importance in magneto-optic recording applications. Important aspects include the perpendicular anisotropy of very thin films and large Kerr rotations.[1–5] A large number of experimental and theoretical studies investigated the structural and electronic properties of Co/Pt interfaces in order to obtain insight into the origin of the magnetic properties. Well-established experimental facts include that the magnetic anisotropy in Co/Pt systems flips from perpendicular to parallel when the thickness of the Co films reaches ~1 nm; that interface Pt atoms, polarized by Co atoms at their vicinity, contribute to the perpendicular anisotropy; and that alloying also enhances the perpendicular anisotropy.[6,7]

However, in spite of the large research effort, much of the observed magnetic behavior remains unexplained, including a detailed understanding of the magnetic anisotropy and its change from perpendicular to in plane. The reason for this surely arises from the large sensitivity of the magnetic anisotropy to structural, morphological, and chemical details at the interfaces, which is at the origin of a variety of experimental results due to a lack of complete and accurate control of the fabrication parameters. A few examples include growth-induced perpendicular anisotropy in thin alloy films grown at 200−400 °C,[8] the existence of two distinct magnetic phases in multilayers prepared at 200 °C,[9] and the suppression of the perpendicular anisotropy by trace amounts of contaminants.[10]

In this paper we present results for ultrathin Co films grown on Pt(111), investigated with x-ray diffraction both in its standard form and also in resonant scattering, to sense the magnetism of the Pt atoms at the interface.[11] Our goal is to try to correlate structural properties at the interface with the magnetic anisotropy. The critical thickness for a flipping of the magnetization from perpendicular to parallel was found to depend markedly on the growth temperature. Also, adsorption of carbon monoxide on thin (~4 atomic layers) Co films changes their anisotropy. We also report the finding that a short annealing to about 100 °C changes the easy direction of magnetization of the film. A crystallographic analysis of the magnetic crystal truncation rods reveals that this is due to an atomic exchange process at the interface between Co and Pt atoms. Finally, we compare and discuss the magnetic response (magnetizations versus applied field intensity) of the same surface as measured by the Kerr effect and by magnetic diffraction.

In Sec. II the experimental details are given, including a short recapitulation of the basis of the resonant surface magnetic diffraction measurements. Section III deals with the results, which are split into four subsections, and Secs. IV and V contain a discussion and conclusions.

II. EXPERIMENT

The experiments were performed on the Surface Diffraction Beamline (ID3) at ESRF.[12] The Pt crystal was of (111) orientation, and miscut by less than 0.1°. It was prepared by standard methods resulting in surface mosaicities of 0.06°. The Auger spectra could be recorded to control the cleanliness of the films. The crystal was described with hexagonal bases $\mathbf{A}_1$, $\mathbf{A}_2$, and $\mathbf{A}_3$ $\{A_1$ and A_2 are equal to the nearest-neighbor surface distance $a_0/\sqrt{2}$, and $\mathbf{A}_3$ is perpendicular to the surface $(A_3=\sqrt{3}a_0)\}$. In this way the reciprocal space

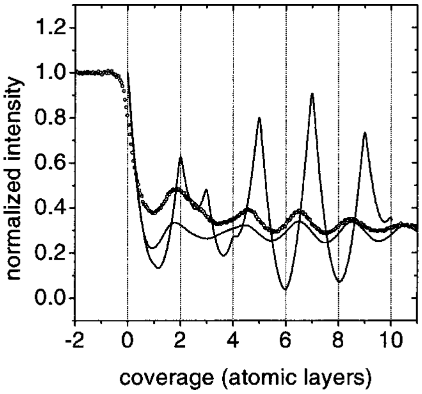

FIG. 1. Temporal evolution of the diffracted intensity at (0,0,1.5) during deposition of Co on Pt(111) at room temperature. The open circles are the experimental data points, and the continuous curves are calculated for a perfect layer by layer growth and for an imperfect one (see the text).

axes *H* and *K* are in the surface plane, and *L* is perpendicular to the surface plane. The photon energy was set to the Pt L_{III} atomic absorption edge (11.564 keV). Magnetic measurements were performed by applying a magnetic field from -1400 to 1400 G in the vertical direction parallel to the surface of the crystal. The field was created by an electromagnet located outside the vacuum chamber. The iron yokes of the electromagnet entered into the vacuum system by two flanges brazed to the iron yokes. Transverse magneto-optical measurements could be performed at the same time than the x-ray-diffraction measurements by using two view ports symmetrically located around the surface normal defining a horizontal reflection plane. Cobalt deposits were done with a water-cooled electron bombardment evaporator. During evaporation the pressure was in the low 10^{-10}-mbar range. The sample temperature was measured with a Pt resistor located in the vicinity of the Pt crystal.

We briefly recall the methodology of the measurements of the magnetic diffraction. Details may be found in Ref. 11.

The asymmetry ratio defined as $R(H,K,L)=(I\uparrow - I\downarrow)/(I\uparrow + I\downarrow)$, where $I\uparrow$ ($\downarrow$) stands for the diffracted intensity at (H,K,L) while applying a magnetic field to the sample pointing along the vertical direction upward (downward). In addition to *H*, *K*, and *L*, the atomic coordinates of the atoms in the crystal and, the magnitude of the applied field, *R* depends on a quantity n_m (negative with our sign convention) which is roughly proportional to the magnetic moment of the resonant Pt atoms. Moreover, *R* is nonzero only if the projection of the magnetization of the Pt atoms in the surface plane is nonzero, i.e., *R* ''senses'' the parallel magnetization of the Pt atoms.

III. RESULTS

A. Growth of Co on Pt(111)

The growth of Co on the Pt substrate was monitored *in situ* by measuring the temporal evolution of the diffracted intensity at (0, 0, 1.5), as depicted in Fig. 1. The oscillations of the intensity are characteristic of a layer-by layer growth. Their damping and small amplitude indicates that the growth is far from ideal. The dashed curve with large-amplitude oscillations is a calculation of a perfect layer-by-layer growth. Pronounced maxima occur at two, three, five, seven, and nine layers. The unequal amplitude of the oscillations arises from the different diffusion factors of Co and Pt. Roughly speaking, the minimum at a coverage close to 1 is due to the cancellation of the scattering amplitude of the substrate [proportional to $\frac{1}{2}\ Z(\mathrm{Pt})=39$] with that of one layer of Co [an amplitude proportional to $-Z(\mathrm{Co})=-27$]. This is in contrast to homoepitaxial systems, where the maxima occur at coverages of 1, 2, 3.... The solid curve with small oscillations is a calculation based on a theoretical model by Cohen *et al.*[13] where the accumulation of imperfections in the growth process is taken into account by a simple phenomenological model.

The measured amplitudes of the oscillations (data points in Fig. 1) depend on the growth temperature and also on the quality and preparation of the substrate. In different experiments with different crystals, we found intensities of the first maximum reaching 0.6. In the experiments described in the following, all the films were grown while monitoring the specular intensity as in Fig. 1. The thickness was deduced from the number of maxima and minima. As their precise positions depend on the quality of the growth which varies with the deposition temperature, an uncertainty of ±10% in the coverages has to be considered. The largely imperfect layer-by-layer growth has been previously reported in detailed scanning tunneling microscopy (STM) studies of the same system.[14,15]

B. Perpendicular and parallel magnetization of the films

The change in magnitude of the asymmetry ratio, when the intensity of the magnetic field varies, allows one to obtain information about the magnetic anisotropy of the surface. The asymmetry *R* gives a measure of the magnetization of the Pt atoms at the interface which is induced by the Co overlayer. It is natural to assume that changes in the magnetization of the Pt will be a consequence of changes in the magnetization of the overlayer. As the magnitude of the individual magnetic moments of the Pt atoms is not expected to change when the external applied field is varied, the changes in *R* will be due to changes in the magnitude of the projection of the magnetization of the Pt atoms in the direction of the applied field (which lies in the surface plane in our case). Co films with a hard axis in the perpendicular direction may result in low measured values of *R* if the external field is not intense enough to rotate the magnetization toward the surface plane, whereas films with an easy axis in the plane will result in larger values of *R*. Thus the dependence *R* vs field for parallel films is expected to consist of a rapidly raising curve, reaching a saturation value for values of the field that are not too high (a few hundred G). The data points for five and nine atomic layers in Fig. 2 have that expected behavior.

Conversely, for films with a perpendicular easy axis, *R* will increase slowly when the field increases. In Fig. 2, the data for the four layer film show this behavior: application of

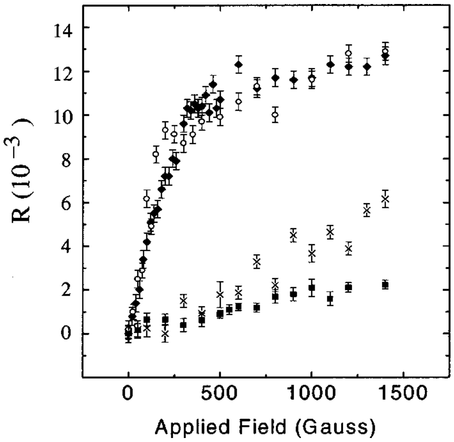

FIG. 2. Asymmetry ratio measured at $(H,K,L)=(1,0,2.85)$ vs the intensity of the applied magnetic field for Co films of different thicknesses: four atomic layers (filled squares), five layers (open circles), six layers (x), and nine layers (filled diamonds).

the maximum field intensity produces a value of R equal to only 0.17 times the value for a film of five or nine layers, which indicates that the magnetization rotates by $a\sin(0.17)\cong 10°$ from the surface normal towards the surface. Letting aside for a moment the data for the six-layer film, Figure 2 shows that the direction of the easy magnetization flips from perpendicular to parallel when the thickness changes from four to five atomic layers. However, in the course of many experiments, it was realized that the critical thickness was not always four layers but sometimes more or slightly less depending on the growth temperature. The data in Fig. 2 for four, five, and nine layers were taken from films grown at 300 K. The data for the six-layer film are from a film grown slightly above room temperature (~340 K). As shown, the anisotropy is not parallel but perpendicular, although less pronounced than that of the four-layer films grown at 300 K. This topic will be discussed in detail in Secs. III D and III E. Our results on the values of the critical thickness are in good agreement with previously published literature.[4,5,16]

C. Effect of the adsorption of CO on the anisotropy of the films

We found that the magnitude of R for very thin films of Co decreased with the time of exposure to the background gas in the vacuum system. The rate was $\sim 0{,}5\times 10^{-3}$ per hour in an ambient pressure of 5×10^{-10} mbar while keeping the sample at ~200 K. We investigated in detail the effect of carbon monoxide on the asymmetry ratio when applying our maximum field. The films were grown in all cases at temperatures in the range 200–230 K, and the exposures to CO were also done at these temperatures. The results are summarized in Fig. 3 which shows the variation of R caused by adsorption of CO on the cobalt surfaces. Before the exposure to the gas, the relatively high values of R of $(10-12)\times 10^{-3}$ indicate that the easy direction is parallel to the surface in all cases. As depicted in Fig. 3, only very thin Co films (3.2 and 4.0 layers) are affected by the CO exposure. The value of R drops almost to zero with 1 L of exposure to the gas, which according to Ref. 17, would correspond to a coverage of around 0.4 ML. An exponential fit of the decay results in $R\sim\exp(-L/0.23)$ (L is the gas exposure in L). Conversely, R does not change upon gas exposure on films of larger thickness (5.0, 6.0, and 8.0 layers) as indicated in the figure.

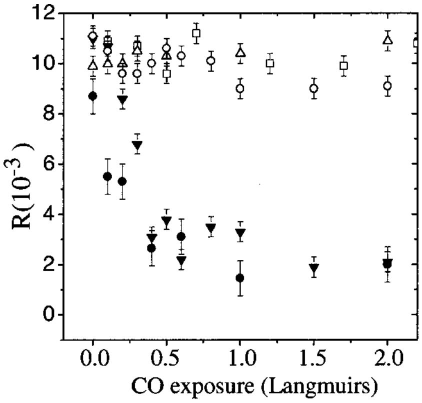

FIG. 3. Variation of R measured at $(H,K,L)=(1,0,2.85)$ when films of different thicknesses grown at low temperatures are exposed to CO. Filled symbols: 3.2 layers (downward triangles) and 4.0 layers (circles). Open symbols: 5.0 layers (circles), 6.0 layers (squares), and 8.0 layers (upward triangles).

The effect of CO on the surface magnetism of ferromagnetic metals has been known for several years. In the case on Ni, it is well established from experiments[18] and theory[19] that CO reduces the magnetic moment of the surface Ni atoms bound to the molecules. In addition, the hybridization of the $3d$ orbitals on Ni with antibonding orbitals of the CO is sensitive to the direction of magnetization of the substrate, as shown by circular magnetic dichroism experiments.[20] CO chemisorption has also been found to reverse the direction of easy direction of magnetization in Co/Cu(110).[21] In small Co particles, it was found that CO adsorption reduces the particles magnetization by quenching the magnetic moment of the outer atoms and leaving the inner atoms unaffected.[22] In line with previous literature, our results in Fig. 3 may easily be interpreted. For very thin films which have a thickness close to the critical thickness for magnetization flipping, the adsorption of CO reduces the magnetic moment of the Co atoms to which the gas molecules are bound. The magnetic part of the Co film is basically reduced in one atomic layer due to CO adsorption at saturation. The remaining magnetic Co film has a thickness smaller than the critical thickness, and therefore exhibits a perpendicular easy axis of magnetization and low values of R. For thicker Co films (open symbols in the figure) the quenching of the magnetism of the topmost Co layer does not induce a change in the easy direction of magnetization, since the films exceed the critical thickness by one layer or more.

An interesting result is that for thin films (~4 layers), a gentle annealing to ~290 K for a few seconds restores the values of R from the unexposed surface (probably due to CO desorption, as shown in Ref. 17), meaning that one can change the magnetization of the Co film from perpendicular to parallel in a controlled way.

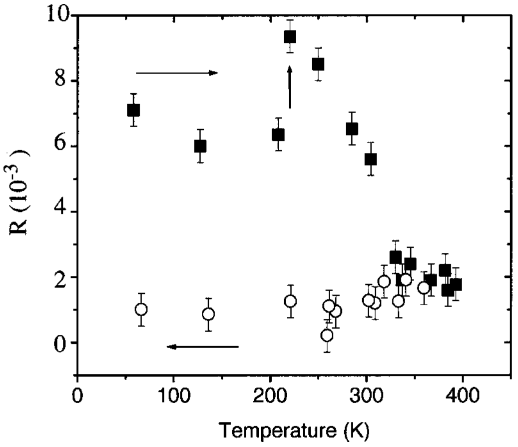

FIG. 4. Thermal evolution of the asymmetry ratio from a film of four layers of Co on Pt(111) deposited at low temperature, annealed (filled symbols), and subsequently cooled down (open symbols).

D. Effect of the temperature on the magnetization

1. Dependence of R on the surface temperature

In order to obtain further insight into the dependence of the critical thickness on the growth temperature, we investigated the effect of the temperature on the asymmetry ratio for a film of 4.0 layers of Co. Figure 4 depicts the results of the experiment. The film was grown at 230 K, and then the surface temperature was lowered to 55 K. The data point at 230 K (indicated with a vertical arrow) was taken immediately after growth. Upon cooling to 55 K, R decreased by about 20% due to some contamination effect associated to the relatively long time required to reach that temperature. From 55 K on, the temperature steadily increased while monitoring R. At 250 K an increase in R was observed, probably due to the desorption of some contaminant, although this point was not investigated further. The important result is that, at 300 K, R decreased noticeably and that at 320–330 K and above the values of R were close to zero. Cooling did not cause any recuperation of the value of R (white circles). The striking aspect of our result is the low-temperature value at which R vanishes. In other experiments where the heating was done more rapidly than in the experiment of Fig. 4, the temperature at which R dropped down almost to zero values could reach 400 K.

An explanation that one could propose for the observed drop in R is interface alloying. Co and Pt form several interface alloys, which display strong perpendicular anisotropy, upon heating Co/Pt heterostructures, as previously reported in the literature. However, the temperatures required to form the interfacial alloys are substantially higher than that at which the drop of R occurred. Some reported values are 773 K,[23] 600 K,[11] 750,[24] and 640 K.[25] Interface alloy formation may be monitored (among other ways) by a change in the Auger spectra or in the position of the diffraction peaks from Pt and Co upon annealing. Figures 5(a) and 5(c) in show the Pt and Co diffracted intensities at (H,0,0.5) (note the logarithmic scale) before and after the annealing. If significant alloying occurred, the Co reflection at $H=-1.09$ would have shifted towards the Pt reflection at -1.0.[11,25] No such shift can be detected on Fig. 5 within our sensitivity which is 3×10^{-3} r.l.u. The amplitudes of the Pt and Co Auger spectra depicted in Figs 5(b) and 5(d) are identical within experimental accuracy. Alloying would cause clear variations in the intensity of the low-energy Co peak, as previously reported.[24] The lack of observable changes indicates that, if alloying takes place upon a mild annealing of the film, its extent has to be very small.

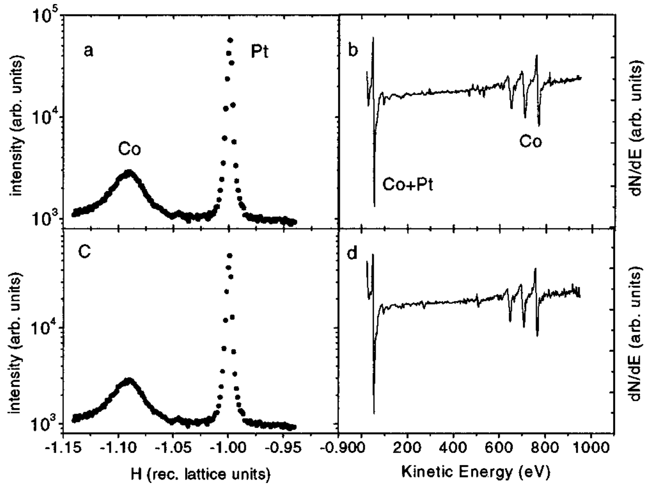

FIG. 5. (a) Diffracted intensities from Pt and Co for a film of six atomic layers of Co deposited at 230 K. (b) The corresponding Auger spectrum. (c) Diffracted intensities after annealing to 370 K for a few seconds. (d) Auger spectrum after annealing.

Figures 6(a) and 6(b) depicts the variation of the asymmetry ratio R [measured at (1,0,2.8)], vs the field intensity for a film of 6.0 layers deposited at 230 K and, after a brief annealing, at 370 K. The film at low temperature shows the characteristic R vs field behavior of a parallel magnetization, whereas the annealed film shows that of a perpendicular magnetization. Figures 6(c) and 6(d) show the transverse

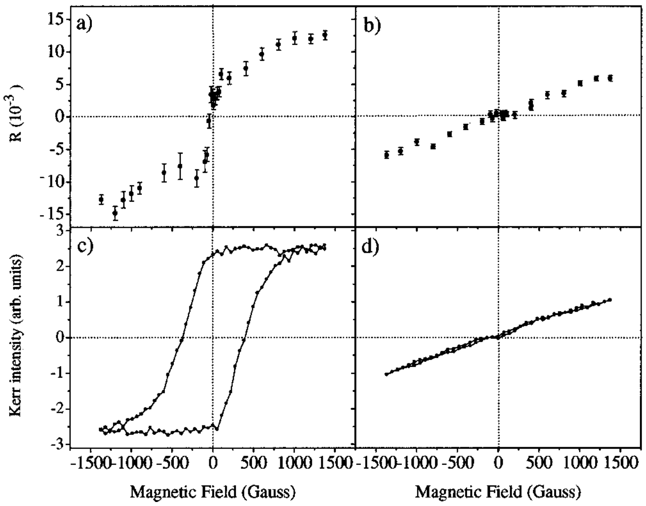

FIG. 6. (a) Dependence of R on the intensity of the applied magnetic field for a film of six atomic layers deposited at 230 K. (b) Same as (a) after a brief anneal of the film to 370 K. (c) Transverse Kerr signal of the same film. (d) Same as (c) after the anneal.

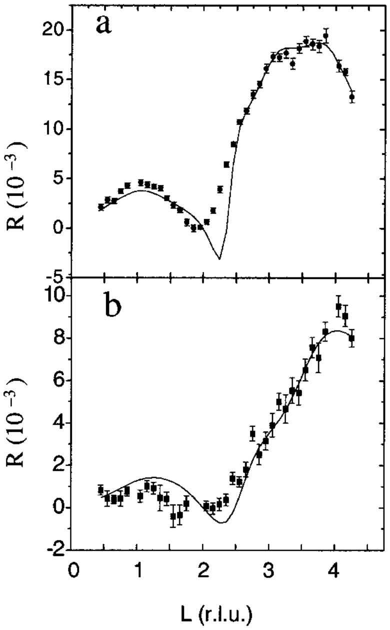

FIG. 7. Magnetic $(0,1,L)$ crystal truncation rod for a 6.0-layers Co film. (a) As deposited at 230 K. (b) After annealing to 375 K for few seconds. The continuous curves are the fits to the data (see the text). Note the change of scale of the ordinate axis between (a) and (b).

Kerr signals vs the field intensity in both cases. At the thickness we are considering, the Kerr signal is dominated by the magnetism of the Co atoms, since the contribution of the polarized Pt is only around 10%.[3] The as-grown film displays a hysteresis loop, evidencing an in-plane magnetization of the Co film; however, the absence of hysteresis in Fig. 6(d) is characteristic of perpendicular magnetization. From the above experiments it is clear that the decrease in R upon mild annealing is due to a flipping of the magnetization of the film from parallel to perpendicular.

2. Crystallographic studies

Several crystallographic data sets were collected to try to find some structural difference between the parallel and perpendicular films previously mentioned. The crystal truncation rods $(1,0,L)$ and $(0,1,L)$ from the Pt substrate were identical within experimental error. The specular rods were almost identical. The small differences among them did not allow one to obtain any conclusions about structural differences. (This point will be discussed in more detail below.) The Co rods at $(0,1.09,L)$ were slightly different before and after mild annealing. The annealing reduced the intensity by 10–20 % depending on the values of L. These changes are, however, too small to attempt a structural analysis. In contrast, the magnetic crystal truncation rods of the Pt were clearly different. Shown in Fig. 7 are the $(0,1,L)$ magnetic rods before (a) and after (b) the mild annealing (in both cases the applied field had the maximum intensity). As may be seen between $L=2.5$ and 4.5, the shapes of the $R(L)$ curves are clearly different. Let us first concentrate on the low-temperature measurements [Fig. 7(a)]. A simple structural model was assumed in order to fit the data based in the following: (1) As the Co grows with its own lattice spacing, which differs by ~10% from that of the Pt (see Figs. 5(a) and 5(c)), Co atoms do not contribute to the diffracted intensity along the Pt rods and may be ignored. (2) The three topmost atomic layers of the Pt crystal [layers 1, 2, and 3 in Fig. 8(a)] were allowed to relax in the surface normal direction. (3) the magnetizations of the Pt atoms near the Co film were allowed to vary. The continuous curve in Fig. 7(a) is the best fit obtained within the above hypotheses. The relaxations of layers 1, 2, and 3 were found to be 1.7%, 0.9%, and 0.5%, respectively (in units of the A_3 lattice parameter). The values of n_m were -0.65 and -0.10 for layers 1 and 2, respectively, as indicated in Fig. 8(a). The uncertainties in the above values of n_m were estimated to be 25% and 10% respectively. The results of the fit show that the relaxations of the Pt atoms are very small and that the magnetism of the Pt atoms is basically confined in the Pt layer in contact with the Co film, since the next Pt plane [plane 2 in Fig. 8(a)] has a magnetization reduced by a factor of 6.5. These results agree with our previously published work.[11]

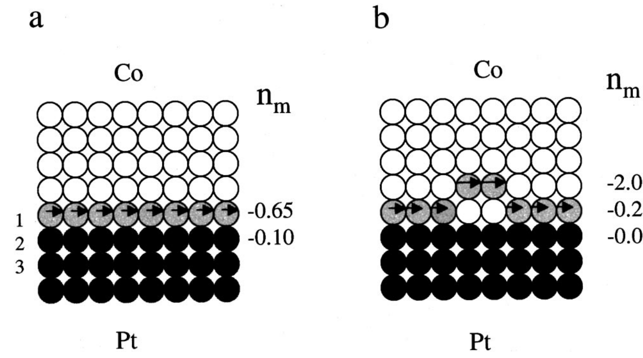

FIG. 8. Schematic representation of the Co/Pt interface. White circles represent Co atoms, black and gray circles are Pt atoms. (a) Abrupt interface corresponding to the film deposited at low temperature. The values of n_m (as obtained from the fit) of the Pt atoms in the two topmost Pt layers (layers 1 and 2) are indicated. (b) the interface after the annealing. The site-exchanged Pt atoms have an enhanced magnetic moment.

To fit the data in Fig. 7(b), we assumed that the atomic relaxations of the Pt surface layers were unaffected by the mild annealing, and we introduced in the model the possibility of atomic exchange between Pt and Co atoms. The continuous curve in Fig. 7(b) is a result of the fit. The atomic exchange between Pt atoms in layer 1 [see Fig. 8(b)] and the first Co layer was found to be 4 ± 1 % of an atomic layer. In our previous work on Co/Pt(110),[26] intermixing at the interface was also found for room-temperature growth.

The values of n_m were -2.0 ± 0.5, -0.25 ± 0.03, and -0.05 ± 0.01, as depicted in Fig. 8(b). The above values may be compared with -0.9 and -0.8 obtained in bulk CoPt alloys.[11] As a rule of thumb, $n_m=-2.4$ approximately corresponds to one Bohr magneton;[11] thus the exchanged Pt atoms on top of layer 1 in Fig. 8(b) have the relatively large polarization of 0.8 ± 0.2 μ_B. This value seems somewhat too high if compared with the usual magnitudes of the magnetic moment of Pt in ferromagnetic alloys. It is unclear to us at

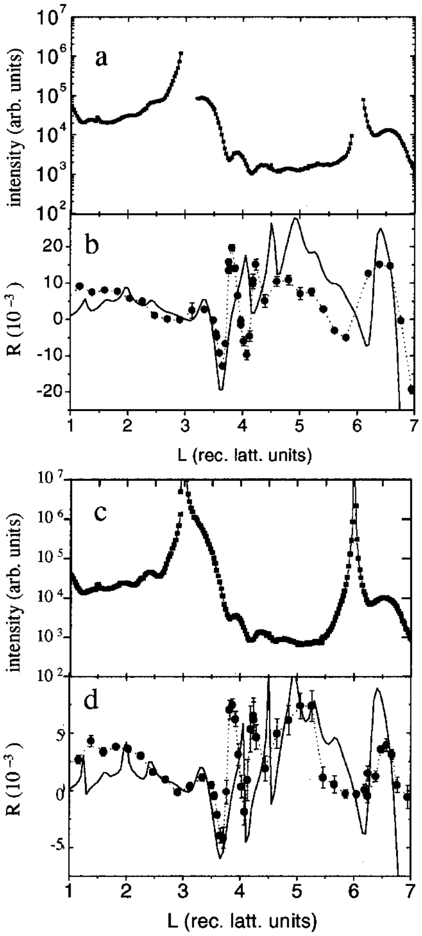

FIG. 9. Panels (a) and (c): $(0,0,L)$ scans (peak intensity) for a 6.0-layers Co film deposited at low temperature on the Pt substrate before and after a short annealing at 375 K. Panels (b) and (d): the corresponding specular magnetic rods $R(0,0,L)$. Note the change of scale of the ordinate axis between (b) and (d).

present if the above value corresponds to the actual magnetic moment or if the correct value is less due to the possible lack of proportionality between n_m and μ for ultradilute alloys.

The fact that the value of n_m of the Pt atoms in layer 1 in Fig. 8 is -0.65 before the annealing, and -0.25 after, does not mean that the magnitude of magnetic moment of the Pt atoms has changed but that the projection of the Pt magnetization in the surface plane has decreased. This is a consequence of the changing in the easy direction of magnetization from parallel to perpendicular: for the maximum applied field at which the magnetic rods are measured, the magnetization of the Pt is entirely in plane for the as-grown film, while it is at an angle to the surface plane for the annealed film.

3. Specular rods

In order to further illustrate the sensitivity of the magnetic rods to the crystal structure, we now present crystallographic and magnetic data measured on the specular rod $(0,0,L)$. Figures 9(a) and 9(c) show the diffracted peak intensities for a 6.0-layer Co film deposited at 230 K before and after short annealing, respectively. Both scans look very similar. The modulations at the high-L side of the Bragg peak at $L=3$ are slightly more pronounced in the film before the annealing (a). On the low-L side, the modulations are slightly enhanced in the film after the anneal (c). Figures 9(b) and 9(d) display the magnetic specular rods, i.e., $R(0,0,L)$. As may be seen, at the position of the ‘‘wiggles’’ of the standard specular scans, the magnetic rods show pronounced oscillations. This is clearly noticeable at $L\sim3.5$, 4, 4.3, and 6.5. These oscillations which often involve a change of sign of R, are sensitive to structural details.

Since in the $(0,0,L)$ rods, all the atoms of the surface region contribute to the diffracted intensity, even those with disorder on the surface plane, a crystallographic fit has to contain a large number of parameters to model the interface Pt and Co atoms which are distributed in about ten or 12 levels, as mentioned in Sec. III A and nicely illustrated in Ref. 15. As a large number of parameters for a limited data set results in very inaccurate results, no serious attempt to fit $R(0,0,L)$ was undertaken. In spite of these limitations, the simple model discussed in Sec. III D, that adjusts the magnetic data of the Pt rods, with the addition of a rough Co layer and an overall small relaxation of the whole Co film, produces the solid curves in Figs. 9(b) and 9(d), which have a remarkable qualitative similarity to the measurements. (As a matter of fact, the calculated solid curves in Fig. 9 are both scaled by a factor of $\frac{1}{2}$ to obtain values similar to the measurements.) As may be seen from the figure, the oscillation at $L=3.5$ is well reproduced in both cases; those at 4 and 4.3 in the data are reproduced in the calculations, but appear to be shifted. The maximum at $L\sim6.5$ is also reproduced by the simulation.

The magnitudes of R are also fairly well reproduced by the simulation. The calculated curve before the annealing is about four times that after the annealing [note the different scales in the ordinate axes on Figs. 9(c) and 9(d)], similar to the experimental results.

Although in the present case the crystallographic structure of the interface is too complicated to accurately fit the data in Figs. 9(b) and 9(d), most likely for less complex interfaces, or for much thinner films, measurements of $R(0,0,L)$ may be useful to obtain a precise description of the structure and of the Pt magnetism.

IV. DISCUSSION

The interatomic exchange between Co and Pt mentioned above was already suggested in the literature. Previous STM measurements[27,28] on Co/Pt(111) showed that submonolayer deposition of Co atoms, at substrate temperatures around 400 K, resulted in a very characteristic dendritic growth morphology associated with dislocation lines in the substrate. Detailed atomic scale imaging allowed to propose that the dendrites consist mainly of Pt atoms that have exchanged sites with Co atoms located below the Pt atoms. This atomic site exchange process between Co and Pt agrees with our results in Sec. III D 2, schematized in Fig. 8. Therefore, it seems clear that either growth or thermal treatments of Co/Pt interfaces at temperatures around 400 K or even less (as shown in the results of Fig. 4) result in interfaces where an atomic exchange between Co and Pt atoms has taken place. An important point here is that the above temperatures are well below the accepted temperatures of formation of Co-Pt alloys. The consequence of this atomic exchange on the magnetic properties of the interface is the flipping of the easy direction of magnetization from parallel to perpendicular (at least for very thin films), as shown in Fig. 6.

It is well established that the origin of the perpendicular magnetization on ultrathin Co films on Pt is due to the relatively large magnitude of the spin-orbit coupling in the Pt atoms which forces, via hybridization of the $5d$ Pt orbitals with $3d$ states in the Co film, the Co magnetic moment to point toward the film normal. The atomic exchange in Fig. 8(b), causes an enhancement of the number of Pt-Co bonds along the surface normal, and thus enhances the perpendicular anisotropy.

An interesting result from the fit discussed in Sec. III D 2 is that the induced magnetic moment ($\sim n_m$) of the Pt atoms embedded in the Co matrix is more than twice than that of the Pt atoms at the interface. Most likely this is due to their very different atomic coordinations. A Pt atom inside the Co films has 12 Co atoms as nearest neighbors, whereas if it is located at the interface as in layer 1 of Fig. 8(a), it has only three Co atoms as nearest neighbors. The induced magnetism of the Pt atoms due to Co atoms in its neighborhood will be more important in the former case.

As the magnetism of the Pt atoms is induced by the Co, to a first approximation the magnetic response of a film (i.e., magnetization vs applied field) should be the same whether viewed by Pt atoms (through the values of R) or Co atoms (via a magneto-optical signal). This is, at first sight, the case in Fig. 6. The parallel easy direction of the film is shown either by R [Fig. 6(a)] or the Kerr signal [Fig. 6(c)], and the perpendicular direction either by R [Fig. 6(b)] on by the Kerr signal [Fig. 6(d)]. An experimental detail to mention here is that R vs H can give only half a branch of the hysteresis curve due to the way that R is measured. The data in Fig. 6(a) are to be compared with the left branch of the hysteresis cycle in Fig. 6(c). Closer inspection reveals that the remanences and coercitive fields deduced from R and from the Kerr intensities are different. For example for a field of 0 G, the Kerr measurement gives a magnetization of the Co film very close to the saturation value, while R is close to zero, much less than its saturation value. At a field of -250 G, the Kerr signal is positive whereas R is negative.

The thickness of our Co films (1 nm or less on average) is certainly smaller than the exchange length $L=\sqrt{A/k}$ (A is the exchange stiffness, and k is the anisotropy constant) which for bulk Co is about 5 nm.[29] This excludes, in principle, the existence of more than one magnetic domain in the direction of the film normal. Recent studies on Fe deposited on vicinal W surfaces[30] show that, in fact, the spin rotation on the surface occurs over a length of 6 nm, which is sensibly smaller than that derived from the bulk properties of Fe. In spite of this, it seems unlikely that a domain wall plus two magnetic domains might exist in our ultrathin films in the normal direction.

Several recent results were published which somewhat resemble our findings.[9,31] In Co/Pt multilayers the authors found a bimodal hysteresis cycle which was due (as evidenced by Kerr microscopy) to the existence of two distinct magnetic phases on the surface. Both phases had different magnetic responses to applied fields. As previously mentioned, in our Co/Pt measurements the growth results in a multileveled surface. Thus, although the average thickness of the Co films in the experiments displayed in Fig. 6 is six atomic layers, the surface morphology consists of regions of ten or even 12 atomic layers, forming pyramidal types of mounds surrounded by areas of much less thickness.[15] Therefore, the surface consists of areas with parallel anisotropy (the mounds) coexisting with areas with subcritical thicknesses (i.e., with perpendicular anisotropy). The distances between mounds, in a film grown at room temperature, are in the range 30–40 nm.[15] In our films, which were usually grown at lower temperatures, that distance will be smaller but probably still larger than the exchange length. Therefore, it may well be that the surface has to be visualized at remanence as consisting of magnetic domains with parallel magnetizations surrounded by areas with perpendicular magnetizations. In this situation the intensity of the Kerr signal (which arises essentially from the Co atoms, as mentioned in Sec. III D 2) will be close to its maximum value, since the volume of the mounds will largely dominate over that of that of the Co atoms in the perpendicular areas. The measured asymmetry ratio R (relative to its saturation value for a fully magnetized parallel film) will be the result of a weighted average of Pt atoms with parallel magnetizations (those below the mounds) and Pt atoms with perpendicular magnetizations (those in the subcritical areas). The data in Fig. 6(a) indicate that R, at zero field, has about 27% of its maximum value, which should be related to the fraction of surface with parallel magnetization.

At a field of -250 G, Pt shows a negative magnetization of $\sim$58%, and Co a positive one of 40%. This indicates that the average coercitivity of the Pt atoms at the interface is smaller than the average one of the cobalt atoms. As most of the Co atoms are in mounds, it appears that the coercitivity in the mounds is larger than that in the subcritical areas.

The above discussion is somewhat speculative. Future work will be done to establish the validity of the above ideas.

V. CONCLUSIONS

The main conclusions of this paper are the following

(1) The critical thickness for flipping the easy direction of magnetization from parallel to perpendicular depends on the growth temperature or thermal treatments of the Co films.

(2) Adsorption of CO gas on the Co film causes a flipping of the easy direction of magnetization from parallel to perpendicular for films of about four atomic layers in thickness.

(3) Annealing of a parallel Co film to $\sim$375 K or even less causes the flipping of the magnetization to perpendicular.

(4) The change in the easy direction of magnetization is due to an atomic exchange process between Co and Pt at the interface. Its extent depends on the details of the thermal treatment. It has been found that 4% of site exchange suffices to reverse the easy direction.

(5) The polarization of Pt atoms in the Co matrix is about two times larger than that at the interface.

(6) The magnetic responses of the film as viewed from the Pt polarization (via R) of from the Co magnetization (via the Kerr signal) are not identical. A possible explanation based on the morphology of the film is proposed.

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