Topology-dependent interface contribution to magneto-optical response from ultrathin Co films grown on the (001), (110), and (111) surfaces of Pd

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We have grown ultrathin epitaxial Co films on three low-index surfaces of Pd. Their magnetic properties studied by the magneto-optical Kerr effect are correlated with the surface morphology analyzed by scanning tunneling microscopy. A perpendicular magnetic anisotropy in the films appeared after exposure to residual gas atmosphere at low temperature, after coverage with an Au overlayer or after annealing at 370 K and above. For one- and two-monolayer-thick films the hysteresis loops are reversed with respect to those of thicker Co films. This is due the negative polar Kerr rotation contribution from the Co/Pd interface dominating over the positive polar Kerr rotation contribution from the noninterface part of the Co film. Reversed loops are not seen when the Co films grow in a three-dimensional mode. This indicates a clear correspondence between the mode of growth and the magneto-optical response. Near the film thickness where the Kerr rotation changes sign a remarkable temperature behavior is observed which is discussed as a superposition of two different magneto-optical contributions. The results qualitatively agree with the available *ab initio* band-structure calculations for the Co/Pd multilayer structures with variable thickness of the Co layers.

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I. INTRODUCTION

Since the observation of perpendicular magnetic anisotropy in Co/Pd multilayers, numerous studies have been devoted to this system,[1–3] because it exhibits perpendicular magnetic anisotropy together with large magneto-optical Kerr rotation in the ultraviolet spectral range which is favorable for high-density magneto-optical recording. It is well known that the physical properties of layered structures are strongly influenced by the details of the interface structure which determine the overlap of the Co and Pd electronic orbitals. Thus, a number of investigations have been dedicated to the influence of the interface structure on perpendicular anisotropy (e.g., Ref. 4). It has been found that some theoretical predictions are not entirely consistent with experimental results. For example, layer-resolved self-consistent electronic calculations of magnetic anisotropy energy showed that for Co films on Pd(111) the easy magnetization direction lies in the film plane and turns out of plane only after Pd capping.[3] However, experimental observations for uncovered Co films grown on Pd(111) show perpendicular anisotropy up to a thickness of 5.5 monolayers (ML) (Ref. 5) or 4 ML (Ref. 6) or 2 ML (Ref. 7). *Ab initio* tight-binding linear muffin-tin orbital calculations showed that nonmagnetic Pd tends to become ferromagnetic at the Co/Pd interface and the magnetic moments of Pd atoms can reach up to $0.5\mu_B$ (Ref. 8). Obviously, the local spin polarization of the Pd interface atoms should also influence the magneto-optical response of Co/Pd structures.[9,10]

The correlations between film morphology, structure, surface alloying, magnetic anisotropy, and local spin polarization of Pd are complex and not fully understood. Moreover, nominally identical experiments do not necessarily have to yield the same results. The magneto-optical Kerr effect (MOKE) is a well-established technique to study the magnetism of ultrathin ferromagnetic films.[11] It is widely accepted that the Kerr signal in magnetic saturation depends linearly on the film thickness in the so-called ultrathin-film limit.[12] For a given magneto-optical geometry (i.e., angle of incidence and incident light polarization), the slopes of the Kerr observables, rotation and ellipticity, versus thickness of the ferromagnetic film grown on a nonmagnetic substrate originate first in the magnetism and optics of the film itself and, second, in the optical permittivity of the substrate. Extrapolation of the linear variation of the Kerr observables to zero thickness sometimes results in an offset signal which can be positive (i.e., of the same sign as the signal from thicker films) or negative (i.e., of opposite sign with respect to the signal from thicker films).[2,13–16] Positive offset means an additional contribution to the total Kerr signal from the film/substrate interface. Negative offset signal means either an absence of magneto-optical signal from magnetically dead interface layers or a negative magneto-optical contribution from the film/substrate interface. Any change of the magneto-optical response of the interface region is caused by a modified local atomic arrangement resulting in a modification of the local electronic structure (e.g., Ref. 17). Such interface effects were clearly demonstrated in multilayers with a considerable number of interfaces.[18] The MOKE is sensitive to the modified ground and excited states, and thus it is not easy to separate magnetism from optical effects. In particular, the Kerr signal from the film/substrate interface does not necessarily scale with magnetic moment in the same way as for a several-ML-thick film. This is because the op-

tical permittivity tensors of the film and of the film/substrate interface region are in general different. A negative interface contribution to Kerr rotation has been observed for several systems like Pd/Co/Pd(111) (Ref. 2) or Au/Co/Au sandwiches.[14] Later studies for ultrathin Co films grown on Pd(111) confirmed the negative interface contribution.[5,7] These observations were interpreted as possibly being due to opposite directions of the polarized Pd moments and the Co moments.[5]

In our previous work[13] we studied the longitudinal Kerr rotation of *p*-polarized light incident onto a Co film of variable thickness, grown on a Pd(001) substrate under identical geometry as was used in the longitudinal experiments of the present work. The offset Kerr rotation obtained by extrapolation to zero Co thickness exhibited the same sign as the contribution from the thick film and was equivalent to the Kerr rotation from 2–3 ML of Co.[13] It is commonly accepted that this offset is due to the Co/Pd interface which is formed during growth of the first atomic layers.[2,14–16] The fact that the contribution to the Kerr rotation from the Co/Pd interface exceeds the value expected for the same thickness of pure Co film should be related to the interface local neighborhood (chemical composition and order) which differs from the neighborhood in a pure Co film. Besides a possible Pd polarization and its contribution to the Kerr rotation from the Co/Pd system,[9] a changed value of the magneto-optical Voigt parameter for the Co atoms within a Pd surrounding may also be expected.[13]

By certain sample treatments we obtained a change in the direction of easy-axis magnetization from in plane to out of plane.[19] To analyze the interface effect it is preferable to compare results where the easy axis does not change and thus the problem of properly relating longitudinal and polar Kerr signals to each other does not arise. In order to test how the interface component is sensitive to the interface structure, one needs to change the local atomic arrangement at the interface and to correlate it with the Co/Pd interface contribution to the overall Kerr signal. This could be done by annealing which usually changes the film topology and enhances interface alloying. Unfortunately, in the Co/Pd system, annealing immediately switches the easy axis from in to out of the plane and a saturated longitudinal Kerr signal cannot be reached anymore with the magnetic field available in our setup.[19] Thus, the only way for us to determine a relation between the interface formation, alloying, and corresponding magneto-optical response was to analyze systems which show perpendicular magnetic anisotropy already before annealing by the polar (instead of longitudinal) Kerr effect. That is why we concentrate on the effects observed with samples with out-of-plane magnetization.

Finally, it had to be decided which magneto-optical observable, rotation or ellipticity, can provide more information about the interface contribution. The main argument supporting our choice of polar Kerr rotation was based on available theoretical calculations performed for Co/Pd multilayers.[20] For the experimental photon energy of 1.84 eV, the polar Kerr rotation depends strongly on the multilayer composition, whereas polar Kerr ellipticity remains almost independent of the composition. Exactly the same conclusion applies for Co/Pd alloys for which the ellipticity is almost constant irrespective of the alloy composition.[21]

In this article we report on the correlation between magneto-optical properties and details of the growth of Co films on clean and atomically flat Pd(001), Pd(110), and Pd(111) surfaces. Our aim was to perform MOKE experiments sensitive to the specific interface contribution and to correlate this contribution to the growth mode in order to detect tiny differences caused by changes of the growth conditions. We show that the negative contribution to polar Kerr rotation is a property of the Co/Pd interfaces. It is demonstrated to exist for all three low-index Pd surfaces investigated and can be related to the contribution of Pd. The interface Pd layer is spin polarized by the neighboring Co film and thus contributes to the magneto-optical response from the Co/Pd system. In particular, we show that negative polar Kerr rotation can be measured in the low-coverage limit depending on the mode of growth and resulting film topology. This demonstrates a clear relation between polar Kerr rotation and the local atomic arrangement at the interface. We support our experimental analysis by the available *ab initio* band-structure calculations performed for the Co-Pd alloys and Co layers on the Pd substrates.

II. EXPERIMENTAL DETAILS

The experiments were carried out in a multichamber ultrahigh-vacuum (UHV) system with a base pressure $<5\times10^{-11}$ mbar and less than 2×10^{-10} mbar during deposition. Prior to deposition the Pd substrates were cleaned by cycles of Ar^+ sputtering followed by annealing at 950 K until only Pd peaks were detected in the Auger electron spectra, sharp low-energy electron diffraction (LEED) patterns, and atomically flat terraces under the scanning tunneling microscope (STM) were observed. All STM measurements were performed in the constant current mode at 0.2–0.5 V positive tip bias voltage and 0.1–0.5 nA tunneling current. The magnetic properties were studied by longitudinal and polar magneto-optical Kerr effects where a *p*-polarized laser beam with a wavelength of 675 nm was used. For the longitudinal MOKE, the incidence angle (with respect to the surface normal) of the probing laser beam was 67°, whereas for the polar MOKE the incidence angle was 5°. The physical quantity we measured using the modulation technique (according to Ref. 22) was the Kerr rotation which was measured at the second harmonic of the modulation frequency.

III. RESULTS

A. Magnetic anisotropy

The growth of Co on Pd(001) and Pd(110) at room temperature starts with the formation of single-layer islands. Figure 1(a) shows a STM image taken from a 1.1-ML-thick Co film grown on Pd(001). The first layer is not fully formed while already second-layer nucleation starts. The typical island size ranges from 3 to 5 nm. Upon annealing, all species of the second layer move towards the step edges which become frizzy. Moreover, there is no more evidence of the former positions of the originally straight substrate step edges. This can be interpreted as a hint towards an alloy formation, because a growing stripe along a previous step

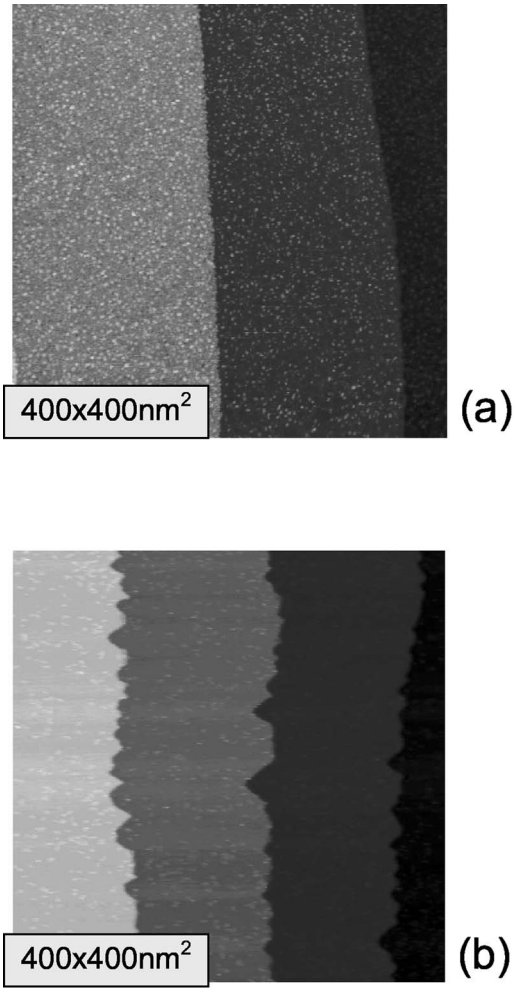

FIG. 1. STM images of 1.1-ML-thick Co film as grown on Pd(001) at room temperature (a) and after a final 20-min annealing at 600 K (b) following two preceding annealing steps at 400 K and 500 K.

edge composed of pure Co would show up in a clear topographic step [Fig. 1(b)].

Co films grown on Pd(001) and Pd(110) do not show perpendicular magnetic anisotropy, and no polar Kerr signal is detected immediately after deposition. Nevertheless, we found that the films deposited at 300 K exhibit a perpendicular magnetic anisotropy after exposure to residual atmosphere at low temperature (for a sufficiently long time) due to residual gas adsorption (for more details see, e.g., Refs. 23 and 24). For Co on Pd(111) in previous reports perpendicular magnetic anisotropy was observed up to at least 2 ML.[5–7] Our as-grown Co films on Pd(111) thicker than 1 ML are magnetized in plane, and the adsorption of residual gas is necessary to induce perpendicular magnetic anisotropy.

In order to be independent of time (i.e., of residual gas adsorption) during our MOKE experiments, we induced perpendicular magnetic anisotropy in a better-defined way: namely, by covering the surface with 20 ML of Au. Independently of the crystallographic orientation of the Pd substrate, covering of the Co films with Au results in perpendicular magnetic anisotropy, however up to a thickness which varies

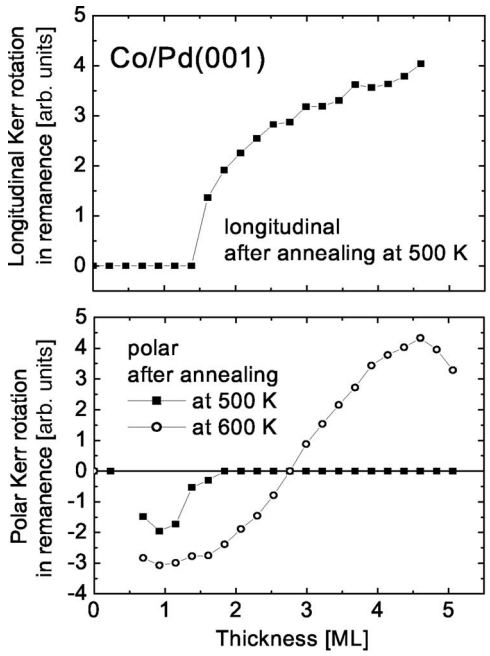

FIG. 2. Thickness dependence of longitudinal and polar Kerr rotation in remanence measured at 70 K for Co films grown on Pd(001) after annealing at 500 K and 600 K for 20 min. Perpendicular magnetization is achieved after annealing at 500 K, however in a relatively narrow thickness range (up to 1.5 ML). Only after another annealing at 600 K does perpendicular anisotropy extend over a Co thickness range of up to 4.5 ML. In this case, the Kerr rotation changes sign at the Co film thickness $t_c \approx 2.7$. Below about 0.7 ML the loops could not be measured due to the large coercivity exceeding the maximum field of 0.8 T which can be applied in our experimental setup.

for different Pd surfaces.[24] We believe that covering with Au is not a destructive method for the local atomic arrangement in the Co/Pd interface in the sense of the Co film topology. Actually, the maximum Co film thickness for which perpendicular magnetic anisotropy persists due to covering with Au is near the maximum thickness for which perpendicular anisotropy is induced by residual gas adsorption. For example, in the case of Co films grown on Pd(001) and covered with Au, the maximum Co thickness before the easy axis goes in plane does not exceed 1.1 ML (and 1.1 ML by adsorption observed after a long exposure to the residual atmosphere).

Annealing above 370 K induces perpendicular anisotropy in the Co films as well. The thickness range in which the out-of-plane magnetization can be kept extends to larger values in comparison to the maximum thickness at which perpendicular magnetic anisotropy is induced by residual gas adsorption.[24] However, upon annealing, the magnetic anisotropy of the system increases irreversibly due to structural changes experienced by the system (i.e., due to interface alloying). In the case of Co on Pd(001), 20 min annealing at 500 K results in perpendicular anisotropy persisting up to a thickness of 1.5 ML, which is increased up to 5 ML after additional annealing at 600 K (Fig. 2). The annealing also results in an increased coercivity which is remarkable at low Co coverage. As a consequence, below a thickness of about 0.7 ML, the polar loops could not be measured even when a maximum magnetic field of 0.8 T is applied.

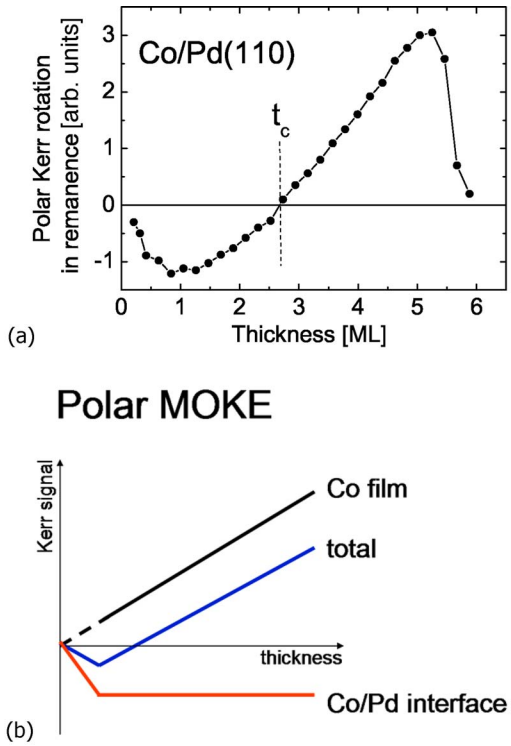

FIG. 3. (Color online) (a) Thickness dependence of polar Kerr rotation in remanence measured at 120 K for Co films grown on Pd(110) after an exposure to residual atmosphere at 120 K for 20 h (saturation state). The Kerr rotation changes sign at the Co film thickness $t_c \approx 2.7$ ML. The easy axis turns into the plane above a Co thickness of 5 ML. (b) Schematic representation of the superposition of the negative Co/Pd interface and the positive Co film contributions to the total polar Kerr rotation signal.

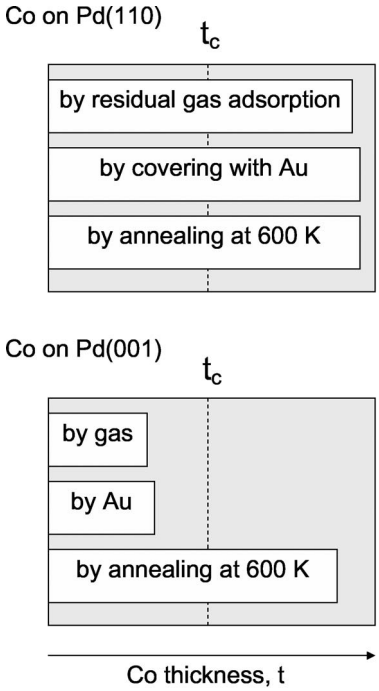

FIG. 4. The relation between the thickness range in which perpendicular magnetic anisotropy is induced (by different recipes) and t_c schematically displayed for the Co films grown on Pd(110) and Pd(001). Without annealing the transition from reversed to normal loops can be detected only for Co films grown on Pd(110). In the case of the Pd(001) substrate, without annealing the maximum thickness to which perpendicular magnetic anisotropy persists is smaller than t_c.

For the purpose of the present work it was important to be able to reproducibly induce perpendicular magnetic anisotropy and to obtain a polar MOKE signal from Co films grown on Pd both before and after annealing. The details concerning perpendicular magnetic anisotropy appearing due to residual gas adsorption, capping with Au, or annealing will be discussed elsewhere.[24]

B. Magneto-optical properties

For simplicity, in the following text we will refer to thin Co films on Pd in the thickness range of 5–10 ML as the "reference sample" for which the measured polar Kerr rotation loops we call "normal" by definition. These loops are of the same sign for (001), (110), and (111) surfaces of Pd. This means that for normal loops the value corresponding to saturation Kerr rotation at positive field is set to be positive. The loops with negative saturation Kerr rotation at positive field are then called "reversed."

Polar Kerr rotation loops measured at the lowest Co coverage on Pd(001) and Pd(110) after the appearance of perpendicular anisotropy are reversed with respect to the reference sample. The thickness range for which negative Kerr rotation occurs for the Co films grown on Pd(001) can be seen in Fig. 2. Independently of the mechanism responsible for the induced perpendicular magnetic anisotropy (residual gas adsorption, annealing, or coverage by Au), the polar MOKE loops then change sign at a certain thickness, denoted as t_c in the following, and become normal above t_c.

Figure 3 shows the variation of the polar Kerr rotation signal in remanence as a function of thickness for the Co films grown on the "open" Pd(110) surface. It is seen that reversed polar MOKE loops are detected for very low Co coverage whenever the polar signal appears. Normal polar loops are detected above $t_c = 2.7$ ML. The thickness t_c does not depend on the thickness range in which perpendicular anisotropy persists. Also, in the case of Co on Pd(001), when the films are magnetized out of plane due to residual gas adsorption or Au coverage, the polar loops are reversed for low Co thickness. The switching to normal loops is not observed due to the narrow thickness range in which perpendicular magnetic anisotropy is detected (up to 1.1 ML). In the case of Co on Pd(001) the loops switch to normal ones above $t_c = 2.7$ ML. However, polar magnetization can be achieved in this case only by annealing. The relation between the thickness range in which perpendicular anisotropy is induced (by different recipes) and t_c is schematically displayed for the Co films grown on Pd(110) and Pd(001) in Fig. 4.

In order to elucidate the origin of the disappearance of the polar Kerr signal at t_c, we prepared a uniform Co epitaxial film with 2.5 ML thickness—i.e., a little bit below t_c—on the Pd(110) surface. The film was covered by Au to induce perpendicular magnetic anisotropy. The polar MOKE loops

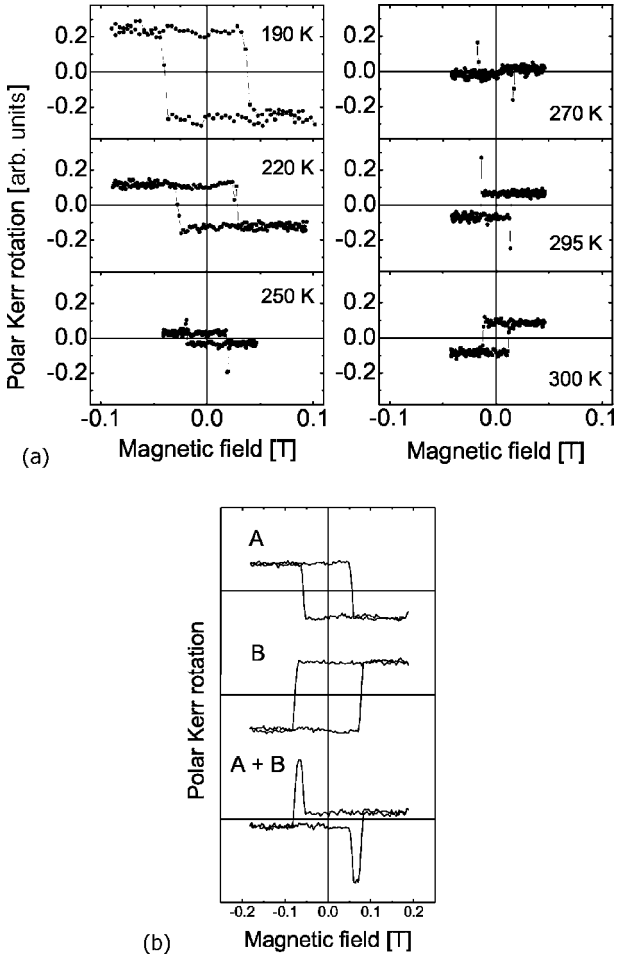

FIG. 5. Polar MOKE loops measured for a 2.5-ML-thick Co film on Pd(110) at different temperatures (a). The loops evolve from negative below to positive above 270 K. Loops measured around 270 K can be reproduced as a sum of two loops, negative (A) and positive (B), of a little different coercivity (b).

measured at different temperatures are shown in Fig. 5(a). It is clearly seen that at low temperatures reversed loops are measured. However, the Kerr rotation in remanence and saturation is very small. From the thickness dependence of the polar Kerr rotation measured for Co films on Pd(110) at 120 K, shown in Fig. 3(a), it can be proved that this film is a little bit thinner than t_c=2.7 ML. With increasing temperature the Kerr rotation in remanence decreases and approaches zero at a temperature of about 270 K. Above this temperature normal loops appear. However, their shape is a little bit strange similar to the shape obtained for the loops measured just below 270 K. Upon careful inspection of these loops, one finds that their shape can be reproduced by superimposing two square loops, one reversed and one normal, of slightly different coercivity [Fig. 5(b)].

Residual gas adsorption results in perpendicular magnetic anisotropy for Co films grown on the most densely packed Pd(111) surface as well.[24] However, there is no change of the hysteresis loop sign in this case and normal loops are measured independently of the Co film thickness. Contrary to previous observations of other groups,[5,25] our STM measurements performed for Co on Pd(111) showed that at 300 K the

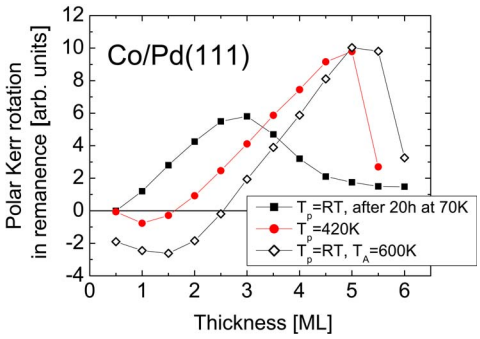

FIG. 6. (Color online) Thickness dependence of polar Kerr rotation in remanence measured at 80 K for Co films grown on Pd(111) at 300 K or at 420 K as well as for the Co films grown at 300 K and annealed at 600 K. At low Co coverage, the initially positive Kerr rotation signal becomes negative after annealing. At a thickness of about 5 ML the Kerr signal is the same independently of the initial mode of growth.

film growth starts by a formation of double-layer islands.[26] The fourth layer starts to grow already at a Co coverage of about 1.6 ML, clearly confirming three-dimensional growth. In agreement with our model, even for the lowest Co coverage, a positive Kerr rotation is measured for Co films on the Pd(111) substrate. Note that the thickness dependence of the polar Kerr rotation can be measured in remanence for the as-grown sample only after covering with Au or long exposure to residual atmosphere (necessary in order to induce sufficiently large perpendicular magnetic anisotropy). For the annealed sample this treatment is not necessary. Obviously, a quasilinear increase of the polar Kerr rotation signal (Fig. 6) starting at the lowest Co coverage is due to a nonideal double-layer growth. In the case of an ideal double-layer growth—i.e., completion of the first two atomic layers before the third one starts to appear—the positive signal should be measured only after the third layer is completed enough to overcome the negative signal of remaining double-layer patches. After a gentle annealing (up to 400 K), the out-of-plane hysteresis loops appear. Reversed loops are detected in this case at low Co coverage. With increasing deposition or annealing temperature, t_c shifts to higher values. Finally, after annealing at 600 K, reversed Kerr rotation loops clearly appear up to the thickness $t_c \approx 2.5$ ML as it is shown in Fig. 7 (the corresponding Kerr rotation is plotted in Fig. 6). Note that at the Co thickness of 5 ML, the Kerr signal is almost independent of the details of the sample thermal treatment (annealing temperature); i.e., it is independent of the initial mode of growth. This magnetic behavior is associated with a morphological reconstruction of the films during annealing. The STM images show drastic changes of the surface morphology. A monolayer of Co appears which is continuous and covers most of the sample area. In Fig. 8 we show the STM images corresponding to the Co film 1.5 ML thick, before [Fig. 8(a)] and after [Fig. 8(b)] annealing at 450 K for 20 min.[26]

IV. DISCUSSION

A. Co films on Pd(001) and on Pd(110)

A simple rule was derived for the polar and longitudinal MOKE geometries starting from the electromagnetic theory

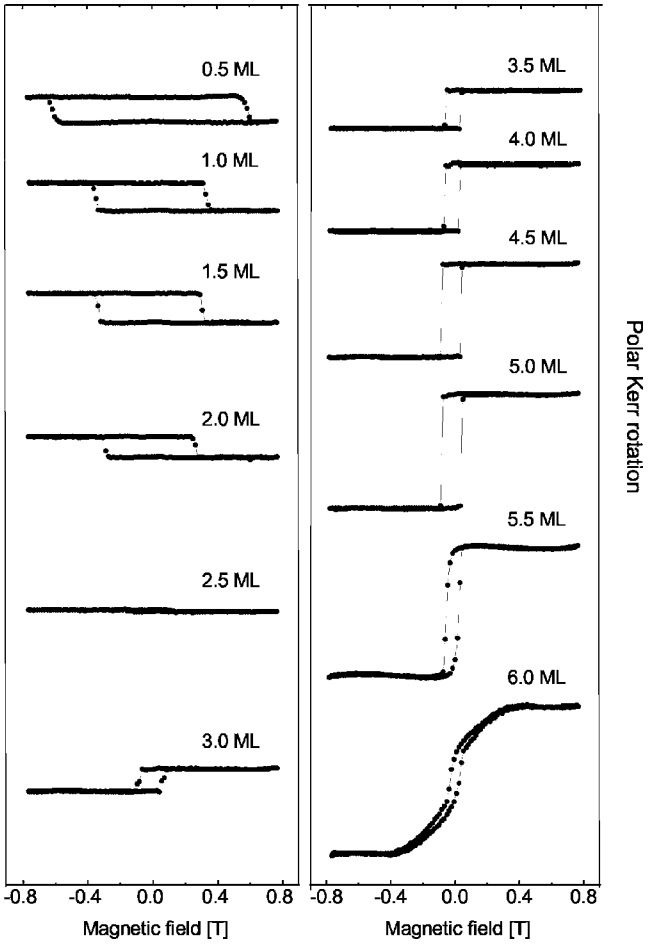

FIG. 7. Polar MOKE loops measured at 80 K for Co films of varying thickness grown on Pd(111) at 300 K and then annealed at 600 K for 20 min. The loops reverse (Kerr rotation changes sign) at a Co thickness of 2.5 ML.

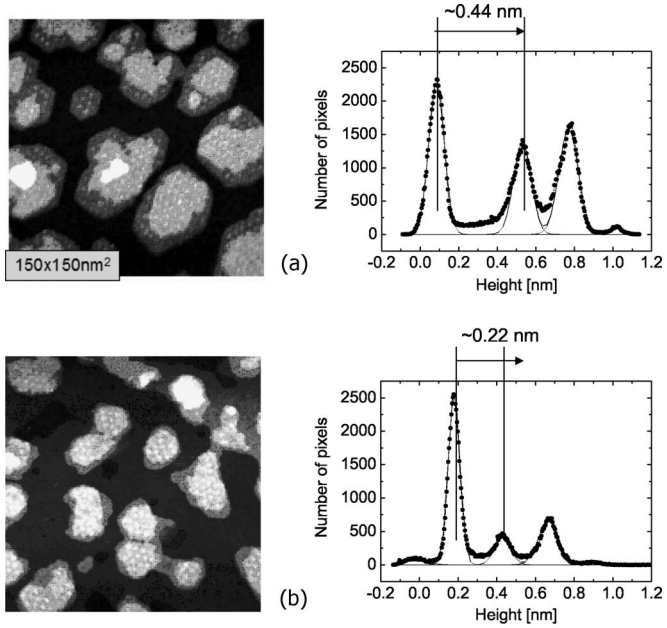

FIG. 8. Film topographies (and related thickness histograms) obtained by STM for 1.5 ML of Co on Pd(111), as grown at 300 K (a) and after annealing at 450 K (b). The distance of 0.22 nm corresponds to the height of one Co atomic layer on Pd(111).

for those magnetic films (or multilayer structures) which can be considered ultrathin.[12] In this case, the amplitude and phase of the optical wave during its propagation through the multilayer stack does not change considerably. Then the rule states that the magneto-optical observables, like the magneto-optical Kerr rotation, can be considered as sums of contributions originating in all magnetic layers involved. For Co films on Pd(110) and for annealed Co films on Pd(001), the polar MOKE loops change sign at a certain thickness t_c and become normal only above t_c. This effect is attributed to the competition between two magneto-optical components of the Kerr rotation signal having opposite signs [Fig. 3(b)]. One takes origin at the Co/Pd interface and the other one in the remaining part of the Co film (i.e., without the interface region). The Co/Pd interface is considered as consisting of the first 1–2 ML of Co on Pd and 1–2 topmost atomic layers of the Pd substrate—i.e., those Co and Pd atoms which are in a very close contact at the interface. At t_c these two contributions cancel each other [Fig. 3(b)]. Following the notation for the reference sample, the polar Kerr rotation (for *p*-polarized light, wavelength of 675 nm and incidence angle of 5°) is negative below t_c and positive above t_c. For as-grown Co films, the thickness t_c can be determined only for the Pd(110) substrate. Only in this case does the thickness range in which the perpendicular magnetic anisotropy is observed (by adsorption or covering) exceed t_c. For both (110) and (001) crystallographic orientations of the Pd surface, the thickness t_c is independent of the thickness range over which the out-of-plane magnetization persists (Fig. 4). This range can be eventually manipulated by exposure to the residual gas. We therefore conclude that appearance of the reversed loops results from the contribution of the Co/Pd interface.

For the Au-coated 2.5-ML-thick Co films on Pd(110), as shown in Fig. 5(a), with increasing temperature the negative Kerr rotation in remanence becomes less negative and approaches zero at a temperature of about 270 K. Above this temperature normal loops appear. This means that the Kerr rotation becomes positive. Such kind of temperature dependence, with changed sign of the loops at a given temperature, could arise only from different temperature dependences of the reversed and normal components to the total MOKE signal. Below the temperature of 270 K the negative signal (reversed loop) dominates over the positive signal (normal loop), whereas above this temperature the positive signal dominates over the negative one. This also means that at 270 K both components of the total Kerr signal balance to zero. In Fig. 9 we show how both components could depend on temperature. This dependence would lead to the above described behavior of the overall signal. Now one has to keep in mind that our film is not of uniform thickness over the whole sample area. This means that the vertical axis of Fig. 9 shows polar Kerr rotation which is averaged over the sample area from which the total Kerr signal is detected. Such an area has a certain topology where the Co thickness is locally changing. This could mean that the temperature dependences for the two components shown in Fig. 9 (i.e.,

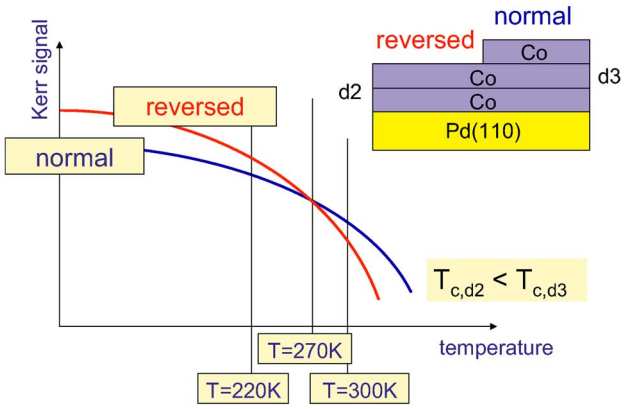

FIG. 9. (Color online) Expected temperature dependence of the positive and negative components contributing to the Kerr rotation signal from the 2.5 ML thick Co film on Pd(110). The schematic structure diagram in the inset shows how these components are related to the local Co film thickness (2 and 3 ML).

for two different values of the Kerr rotation related to two different thicknesses of the Co film) are very similar. However, one component contributes more to the overall Kerr signal due to the larger population of the corresponding film thickness sites. In particular, we expect that the negative saturation signal (i.e., reversed loop) corresponds to a thickness d_2 of 2 ML or less, whereas the positive signal (normal loop) corresponds to the thickness d_3 of 3 ML or more (as it is schematically shown in Fig. 9). This is in agreement with our interpretation that for 2 ML the total polar Kerr rotation is still negative, however very close to zero. For 3 ML it is still quite small, however already positive. In a very simplified picture, at a thickness of about 2.5 ML the population of 2-ML-thick areas and 3-ML-thick areas of the sample should be almost the same. Thus both components of the total Kerr signal should balance to the value which depends mainly on how negative is one component and how positive is the second one. The fact that coercivity of the normal loop (positive signal attributed to 3-ML-thick film) is larger than the coercivity of the reversed loop (negative signal attributed to 2-ML-thick film) agrees with what is usually expected for different thicknesses and thus supports our interpretation. This is due to the temperature dependence of coercivity which usually increases when temperature decreases with respect to the Curie temperature T_c. The temperature of 270 K is well below the T_c for 3 ML of Co and quite close to the T_c of 2 ML of Co. However, the character of both dependences could be slightly different. This is well known that the magnetization depends on temperature more strongly when the film thickness is reduced. More linearlike dependence in the case of 2-ML-thick film in comparison to more $T^{3/2}$ dependence for 3 ML and more, multiplied by their contribution to the total Kerr signal, can result in a crossing of both dependences at 270 K. It is also suggested from Fig. 9 that both signals, negative and positive, can disappear at different temperatures. Again, according to the arguments of reduced thickness and magnetic size effects, the Curie temperature of 2 ML should be smaller than that of 3 ML. This is why only the positive component (corresponding to 3-ML-thick Co film) is still visible at 300 K whereas the negative component (corresponding to 2-ML-thick film) already disappeared at lower temperature. Finally, the transition from negative to positive Kerr signal with increasing temperature can be interpreted as a result of a noninteger number of monolayers building up the film. The total Kerr signal comprises two components, negative and positive, corresponding to thicknesses of 2 and 3 ML, respectively [i.e., to the thickness distribution expected even for the layer-by-layer growing film which is 2.5 ML thick (Fig. 9)].

To summarize this part of the discussion, for Co films grown on Pd(110) substrates, the negative Kerr rotation (reversed loops) is detected for 1-ML-thick (large negative) and 2-ML-thick (still negative but smaller) film sites. For layer-by-layer growth this means that the negative rotation approaches a maximum when the first Co monolayer is completed. With further completion of the second atomic layer the negative signal decreases, approaching a small, however still negative, value at 2 ML. The positive components to the Kerr rotation signal averaged over different areas of the surface are introduced only after the third layer starts to grow. This is why the critical thickness at which the Kerr rotation changes sign exceeds 2 ML. This also means that the critical thickness corresponds to the thickness at which the small negative contribution from 2-ML-thick parts of the film is exactly compensated by the small positive signal from the parts of the Co film with 3 ML thickness. In reality, the growth never proceeds in an ideal layer-by-layer mode. For example, the film thickness could be distributed by a log-normal law. The exact Co thickness distribution would influence the value of t_c as well. Complementary superconducting quantum interference device (SQUID) experiments on Au-coated 2.5-ML-thick Co film on Pd(110) confirmed the pure magneto-optical origin of the effect of reversed Kerr loops as well as of the disappearance of polar Kerr rotation at t_c. The SQUID loop shows the magnetization proportional to the thickness of magnetically alive 2.5 ML of Co and a value of magnetic moment per atom exceeding the bulk value of $1.7\mu_B$.

B. Co films on Pd(111)

The most straightforward verification of the model can be performed for the system which does not grow in the layer-by-layer mode. When multiple-layer-high islands form in the initial stage of growth, the negative Kerr component should not appear. This is due to the fact that the Co film thickness is very inhomogeneous in this case: a film 1 ML thick on average over the surface is composed of islands which may be even a few monolayers high. The negative Co/Pd interface contribution for such high islands is dominated by the positive Co film contribution, resulting in a total positive polar Kerr rotation from each island. Surface flattening due to annealing should enhance the negative component. This is the case for Co films grown on Pd(111). The quasilinear increase of the positive polar Kerr rotation signal starting at the lowest Co coverage is measured for the films as grown at room temperature. This corresponds to $t_c=0$ as is shown in Fig. 10. After annealing at temperatures above 400 K, reversed Kerr rotation loops clearly appear up to a finite thick-

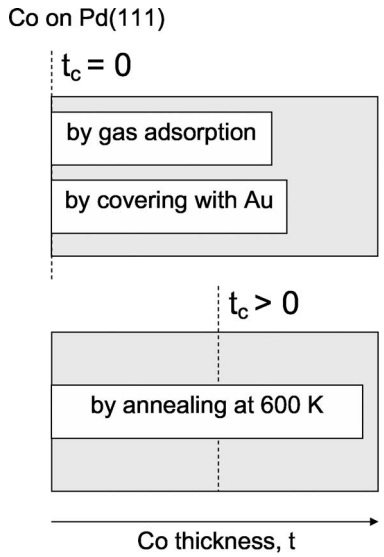

FIG. 10. The relation between the thickness range in which perpendicular anisotropy is induced (by different recipes) and t_c schematically displayed for the Co films grown on Pd(111). Without annealing, only normal loops can be detected. With increasing annealing temperature t_c increases and the transition from reversed to normal becomes detectable.

ness $t_c > 0$ which depends on the annealing temperature (Fig. 10). The t_c value approaches ≈ 2.5 ML after annealing at 600 K [Figs. 6 and 7]. At low coverage the islands spread over the whole surface and form 1-ML-thick areas [Fig. 8(b)].[26] This Co surface arrangement results in much smaller polar Kerr rotation with respect to the same coverage when the Co film is grown at 300 K [Fig. 8(b)], due to the larger negative interface component contributing to the total Kerr signal. In other words, with increasing deposition or annealing temperature, a thicker Co film has to be deposited to result in the same polar Kerr rotation as for the film grown at 300 K.

This confirms that in the low coverage limit the film topology mostly determines its magneto-optical response. The film topology at initial stages of growth very strongly depends on the growth conditions influenced partly by a local pollution of the Pd crystal surface on the atomic scale. Thus, tiny differences in the mode of growth can result in nonreproducible properties of the Co/Pd(111) systems reported in the literature, as the thickness range in which the negative Kerr rotation is observed.[5,7] However, a negative component observed after the treatment at elevated temperatures may appear not only due to the more two-dimensional-like morphology in contrast to the double-layer morphology of the as grown films at 300 K. It could be also caused by an intermixing and a deeper penetration of Co atoms inside the Pd substrate which would result in an increasing number of Co atoms in a Pd surrounding enhancing the negative Kerr rotation. Thus, the critical thickness of about 2.5 ML in the case of annealed Co films on Pd(111) cannot be quantitatively compared to t_c of the Co films grown at 300 K on the Pd(110) substrates. For thicker Co films, the increasing negative Co/Pd interface contribution results in lower positive total Kerr signals. This means a shift of the linear thickness dependence of the polar Kerr rotation on the Co thickness to lower signals which is equivalent to the increased value of t_c.

C. Origin of the negative Kerr rotation from the Co/Pd interface

The results discussed show the complicated relation between the interface and film magnetism on the one hand, and the polar Kerr rotation signal measured for the Co films grown on Pd single crystal substrates on the other. The negative polar Kerr rotation contribution from the Co/Pd interface (i.e., for ultrathin Co films on Pd substrates) is not surprising. Calculations using the spin-polarized relativistic linear muffin-tin orbital method performed for Co-Pd alloys[21] clearly show that at a photon energy of 1.84 eV (which is equivalent to a wavelength of 675 nm) the polar Kerr rotation is of different sign depending on the alloy composition. In the sign convention chosen by the authors of Ref. 21, the Kerr rotation is negative for low Pd concentration and increases towards positive values when the Pd concentration is increasing. At the $CoPd_5$ composition the polar Kerr rotation becomes positive and saturates at small positive values. Qualitatively one can say that the increasing concentration of Pd is equivalent to the reduced thickness of Co film grown on the Pd substrate. Moreover, previously a similar result has been obtained for a multilayer structure. While keeping the thickness of the Pd layer constant (8 ML) and reducing the thickness of the Co layer, originally the negative polar Kerr rotation increased and became positive at a Co thickness of 1 ML.[20] This is in agreement with earlier results[21] showing that the interband magneto-optical transitions in spin-polarized Pd give a positive Kerr rotation in the energy range below 2 eV, whereas the transitions between the electronic states with a mixture of Co 3*d* states result in a negative contribution to the Kerr rotation in this range, even when the Co states are considerably modified by the hybridization with Pd states.[20,21] The tendency was increased—i.e., the polar Kerr rotation was even more positive—when the Co monolayer was replaced by a $CoPd_3$ alloy layer. This scenario is equivalent to a nonperfect interface for which a kind of "alloying" can be expected.[20] Note, however, that we measured a negative Co/Pd interface component for the ultrathin Co films grown at room temperature on the Pd(001) substrate. It is reported recently that no intermixing occurs for Co films grown by standard thermal deposition technique.[27] The calculations of the magneto-optical properties are verified by calculations of the spin magnetic moments in the Co/Pd multilayer structure[20] resulting in almost perfect agreement with calculations of other groups.[8] Taking into account that the sign convention chosen in Refs. 20 and 21 is opposite to our coordinates, the above result means qualitative agreement between their calculations and our experimental data.

V. CONCLUSIONS

In conclusion, we stress the fact that the Co films show very similar magneto-optical behavior independently of the crystallographic orientation of the Pd surface. The negative

polar Kerr rotation was observed at low Co coverage for less than 2-ML-thick Co film sites where it is not overridden by the positive Kerr rotation from the thicker areas of the specimen. A relation to the real sample topology was also addressed through the temperature variation of the Kerr signal which comprises the components related to local variations of the film thickness. The experimental results were verified by predictions of the phenomenological structural model as well as by comparison with published electronic structure calculations. In particular, the sign reversal of the polar Kerr rotation predicted theoretically with decreasing thickness of Co film is confirmed and shown as limited to a maximum of the first two Co atomic layers on the Pd substrate. More general, it is claimed that the magneto-optical response from the sample should be always considered as a sum of contributions arising from different parts of the film structure that in general can be of different sign.

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