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A mode-of-growth-dependent magneto-optical response from ultrathin Co films on Pd surfaces

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Abstract

We have grown ultra-thin epitaxial Co films on (0 0 1) and (1 1 1) surfaces of Pd. Their magnetic properties are studied by magneto-optical Kerr effect (MOKE). They are correlated with surface morphology analyzed by scanning tunneling microscopy (STM). In agreement with theoretical predictions, reversed polar Kerr rotation loops with respect to those of thicker Co films were observed below the thickness of 2.5 monolayers. This is interpreted as being due to a dominating negative contribution of Co/Pd interface with respect to the positive contribution from the non-interface part of the Co film. There is no change of sign of the polar Kerr rotation at low Co coverage on Pd(1 1 1) due to the growth which starts by formation of double-layer patches in this case.

Keywords: Magnetic properties of thin films; Co on Pd; Magneto-optical effects; Surface magnetism; PMA

Since the discovery of a perpendicular magnetic anisotropy (PMA) in Co/Pd multilayers, a lot of effort has been devoted to this system [1,2]. Nevertheless, details of the correlation between the film morphology, structure, surface alloying and magnetic anisotropy still remain not fully understood and unequivocally evidenced by experiments. In particular, it has been shown that in such systems, originally nonmagnetic Pd metal tends to ferromagnetism and magnetic moments of the Pd atoms at the Co/Pd interface can reach up to 0.5 μ_B [3]. Obviously, the local spin polarization of the Pd interface atoms influences magnetic and magneto-optical properties of the Co/Pd structures [4,5].

The magneto-optical Kerr effect (MOKE) is a well established technique to study magnetism of ultra-thin ferromagnetic films and nanostructures [6]. For a given magneto-optical geometry (i.e., the angle of incidence and the incident polarization of light), the slopes of the Kerr observables, rotation and ellipticity, versus thickness of the ferromagnetic film grown on a nonmagnetic substrate originate first in magnetism and optics of the films itself, and second in the optical permittivity of the substrate. Extrapolation of the linear variation of the Kerr observables to zero thickness results in an offset signal which can be either positive or negative, which means, respectively, of the same or opposite sign with respect to the signal from the thicker films [7–9]. The offset magneto-optical signal is attributed to a contribution from the interface region between the film and the substrate. It is related to a change of local atomic arrangement, related hybridization effects and resulting modified electronic structure. For the Co/Pd system, which is the subject of our interest, the relation between the interface component and the interface structure can be understood by correlating the Co/Pd inter-

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face contribution to the overall Kerr signal with the changes of local atomic arrangement at the interface either due to different mode of growth or due to annealing.

In this contribution we report on the relation of magnetic and magneto-optical properties to growth details of ultrathin Co films prepared on clean and atomically flat Pd(0 0 1) and Pd(1 1 1) surfaces. We show that the negative Co/Pd interface contribution to both longitudinal and polar Kerr rotation occurs for both Pd surfaces and can be related to the contribution of the Pd interface layer which is spin-polarized by the neighboring Co film. In particular, we show that the sign of polar Kerr rotation in the low coverage limit depends on the growth mode and resulting film topology.

The experiments were carried out in a multi-chamber ultrahigh vacuum (UHV) system with a base pressure $<5\times10^{-11}$ mbar. All scanning tunneling microscopy (STM) measurements of the film topology were performed in a constant current mode at 0.2–0.5 V positive tip bias and 0.1–0.5 nA tunneling current. The magnetic properties were studied by longitudinal and polar MOKE at low temperature (70 K), by using a p-polarized laser beam with 1.84 eV photon energy. The incidence angle was set to 67° for the longitudinal MOKE and to 5° the polar MOKE. To avoid problems with sign convention, in this contribution we will always refer to the Co films with thickness of 5–10 ML on Pd substrate as to a 'reference sample'. For this sample, the value corresponding to saturation Kerr rotation for positive magnetic field is set to be positive by definition ('normal' hysteresis loop is observed). For some other values of Co film thickness this saturation Kerr rotation can turn negative (observed 'reversed' loop).

The growth of Co on Pd(0 0 1) is found to be pseudomorphic and close to the layer-by-layer mode up to three monolayers (ML) [9,10]. At the initial stage of growth, ML-thick Co patches are formed, as it is confirmed by Fig. 1 showing the surface topography of the 1.1 ML of Co on Pd(0 0 1). The Co films on Pd(0 0 1) do not show PMA and the polar Kerr signal is not detected for uncovered Co films. In our previous work we studied longitudinal Kerr rotation with p-polarized incident light for Co films grown on the Pd(0 0 1) substrate [9]. The present longitudinal experiments were performed for the same photon energy of 1.84 eV and for the identical geometry (i.e., for the p-polarized laser beam incident at 67°). For both substrates, Pd(0 0 1) and Pd(1 1 1), the signal increase with Co thickness is rather rapid below about 2 ML. Above 2 ML of Co, the slope of this dependence decreases and the Kerr rotation signal varies linearly with the Co thickness, as demonstrated in Fig. 2a for Co on Pd(0 0 1). The offset signal obtained by extrapolation of the Kerr rotation measured above the thickness of 2–3 ML to zero Co thickness is of the same sign as the rotation measured on the 'reference sample' (i.e., of the 5–10 ML of Co on Pd). This offset is equivalent to the Kerr rotation from 2–3 ML of Co [9]. The larger offset signal (equivalent to the Kerr rotation from 3.4 ML of Co) was detected for the Pd(1 1 1) substrate. The smaller one (equivalent to 2.5 ML of Co) was

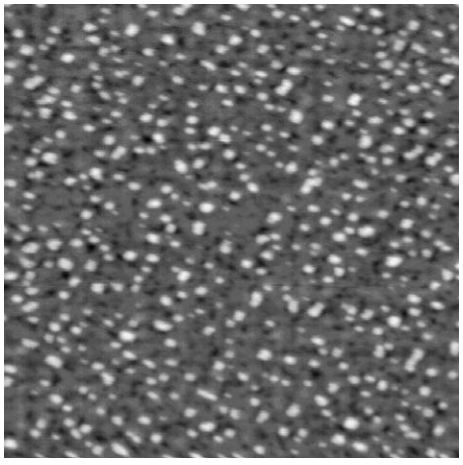

Fig. 1. STM image (100×100 nm^2) of the 1.1 ML-thick Co films grown on Pd(0 0 1) at 300 K. A layer-by-layer growth within the low coverage limit is confirmed.

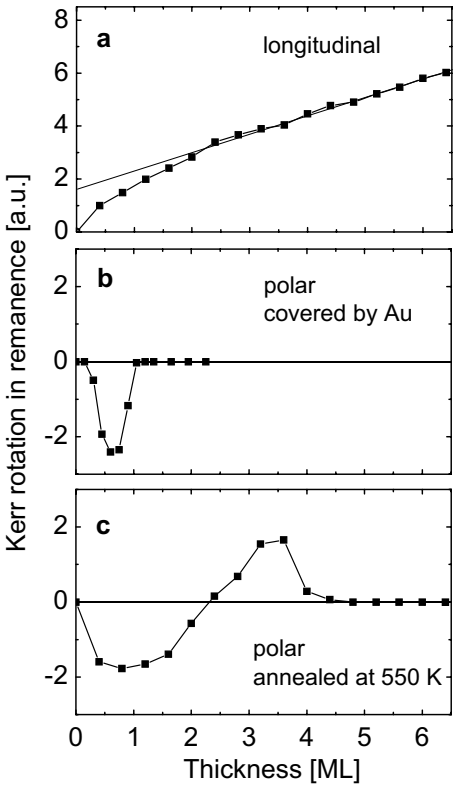

Fig. 2. Thickness dependence of Kerr rotation in remanence at 70 K for Co films grown on Pd(0 0 1) at 300 K: (a) longitudinal Kerr rotation for as grown films immediately after deposition, (b) polar Kerr rotation for the films covered with Au immediately after deposition, and (c) polar Kerr rotation for the non-covered films annealed for 20 min at 600 K. The longitudinal offset Kerr rotation (after extrapolation of the linear variation of the signal above 3 ML of Co to zero Co thickness) corresponds to the Kerr rotation from 2.5 ML. The initially negative polar Kerr rotation signal becomes positive after the PMA is forced (by annealing at 600 K) to persist for the thicker Co films.

found for Pd(0 0 1) surface (Fig. 2(a)). This offset originates in the Co/Pd interface formed during the growth of the first MLs of Co [8,9]. Besides a possible Pd polarization and its contribution to the Kerr rotation from the Co/Pd system [4], the changed value of the magneto-optical Voigt parameter for the Co atoms adjacent to Pd can be also expected [9].

We found that the Co films deposited at 300 K, which are magnetized in plane immediately after deposition, exhibit a PMA after an exposure to the residual atmosphere at low temperatures (70 K) for sufficiently long time. Similar effect can be observed when covering the Co films with 20 ML of Au, independently of the crystallographic orientation of the Pd substrate (Fig. 2(b))[10]. The maximum thickness up to which the PMA persists due to covering with Au is exactly the same as when it is induced by exposure of Co/Pd to the residual atmosphere. Thus we believe that covering with Au is not a destructive method for the local atomic arrangement in the Co/Pd interface. Consequently, we claim that just the same Co/Pd interface contributes to the polar Kerr rotation as the interface which was contributing to the longitudinal Kerr rotation before covering the film with Au. Also annealing above 300 K supports the PMA of the Co films grown on Pd substrates (Fig. 2(c)). However, upon annealing the PMA of the system increases due to structural changes and/or interface alloying. The Co/Pd interface does not remain the same in this case.

The polar Kerr rotation loops measured at the lowest Co coverage on Pd(0 0 1) after appearance of PMA are 'reversed'. The same effect was previously reported for Co on Pd(1 1 0) [10]. In already mentioned Fig. 2(b), the PMA of the Co films on Pd(0 0 1) was induced by covering with Au. It is seen that the 'reversed' polar Kerr rotation loops are detected for the Co coverage up to about 1.1 ML. Above 1.2 ML of Co, the PMA vanishes and it is impossible to reach the critical thickness t_c at which the polar Kerr rotation loops become 'normal'. The polar magnetization above 1.2 ML can be achieved only by annealing. Then the 'normal' polar loops are detected above the Co thickness, indicated in the following as critical thickness t_c, which equals ~2.5 ML (as shown in Fig. 2(c)). We note that for the Co films on Pd(1 1 0) surface, the PMA could be achieved by covering with Au up to the thickness of about 5 ML [10]. The 'normal' polar loops were detected above the critical thickness $t_c = 2.7$ ML in that case [10].

Independently of the mechanism responsible for the induced PMA (residual gas adsorption, annealing or coverage by Au), the polar MOKE loops measured in our experiments are 'reversed' below t_c and become 'normal' above t_c. This is attributed to the competition between two magneto-optical components of the Kerr rotation signal having opposite signs. One takes origin at the Co/Pd interface and the other one in the remaining part of the Co film (i.e., in that without the interface region). At t_c these two contributions cancel each other. Obviously, the Co/Pd interface contribution to the magneto-optical response which was seen as an offset in the longitudinal geometry (Fig. 2(a)) should be seen in the polar MOKE as well. The negative polar Kerr rotation measured for the Co/Pd interface agrees with the available theoretical calculations performed for Co/Pd multilayers [11]. The calculations show that for the experimental photon energy of 1.84 eV the polar Kerr rotation depends strongly on the multilayer composition, changing sign for the thinnest Co layers.

Note, that our polar Kerr experiments on the Co/Pd systems were performed within the same experimental setup as the longitudinal ones. The only difference concerned the incidence angle of the laser beam (because the longitudinal effect cancels at normal incidence). As it is plotted in Fig. 3, electromagnetic theory calculations described in Ref. [12] show that for Co films on Pd, for present experimental conditions, the polar Kerr rotation of the 'reference sample' should be of the same sign as that for bulk Co (Fig. 3(a)), whereas for the longitudinal geometry the signs should be opposite (Fig. 3(b)). This is caused by different development of the Kerr phase (given by arc tan of the ellipticity/rotation ratio) with the Co thickness in both cases. In experiments we indeed observed a sign change of the longitudinal Kerr rotation between the values mea-

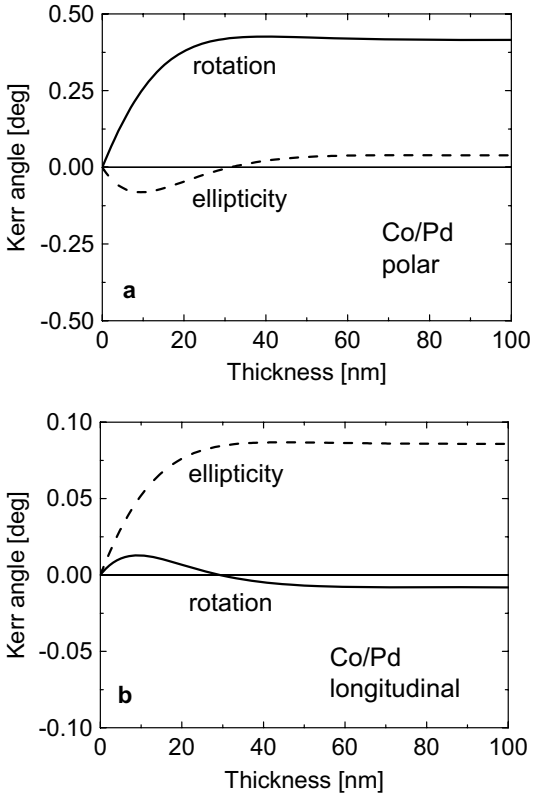

Fig. 3. Theoretical thickness dependence of Kerr observables (rotation and ellipticity) for incident p-photons with an energy of 1.84 eV for Co films on Pd substrate: (a) for out-of-plane magnetization–polar effect, 5° incidence angle and (b) for in-plane magnetization–longitudinal effect, 67° incidence angle.

sured for our reference sample (5–10 ML thick Co film on the Pd substrate) and for bulk Co (measured in our case for a 30 μm thick Co foil). This agrees with Fig. 3(b), solid line. However, note that this sign change has nothing to do with the Co/Pd interface effect discussed in this article.

Interestingly, the Co/Pd interface contribution to the polar Kerr rotation in our case (i. e., for photon energy of 1.84 eV and incidence angle of 5°) is of a different sign with respect to the contribution from the remaining part of the Co film. On the other hand, the same sign is observed when the longitudinal Kerr rotation is probed (incidence angle of 67°). Actually speaking, in both cases, i.e., for longitudinal and for polar Kerr rotation, the Co/Pd interface contribution is opposite with respect to the rotation signal from bulk Co. In the case of the longitudinal Kerr effect the rotation signal changes sign at the Co film thickness of 29.3 nm, i.e., about 140 ML (Fig. 3(b)). At this thickness the longitudinal Kerr phase reaches 0.5π (dominating ellipticity signal–Fig. 3(b)), while the polar phase is 0.85π (dominating rotation signal–Fig. 3(a)). On the other hand, for the 'reference sample' both phases are close to 0.65π and both MOKE angles are detectable.

When more-ML islands would be formed in the initial stage of growth, the negative component to the polar Kerr rotation should not appear. This is due to the fact that 1 ML of deposited Co in average over the specimen surface is composed of islands which may be even a few monolayers high. The negative Co/Pd interface contribution for such high islands would be overwhelmed by the positive non-interface Co contribution, resulting in the positive polar Kerr rotation from each such an island. For Co films on Pd(1 1 1), in contrast to Pd(0 0 1), at 300 K the growth starts by a formation of double-layer patches (Fig. 4(a)). The PMA for the Co films grown on the Pd(1 1 1) surface can be achieved by gas adsorption or by Au-covering as well. However, as it is shown in Fig. 4(b), there is no change of the polar Kerr rotation hysteresis loop sign in this case. Even for the lowest Co coverage, the positive rotation is measured for the Co films on Pd(1 1 1) substrate. This means that the negative Co/Pd interface contribution to the total polar Kerr rotation in the Co/Pd(1 1 1) system is overwhelmed by the positive contribution from the non-interface Co already at its thickness of 2 ML. However, this could also happen in the case of a non-ideal double-layer growth, i.e. when the third layer starts to appear before completion of the first two atomic layers. Then a positive signal would be measured due to the positive contribution from the 3 ML thick film areas. Note that exposure

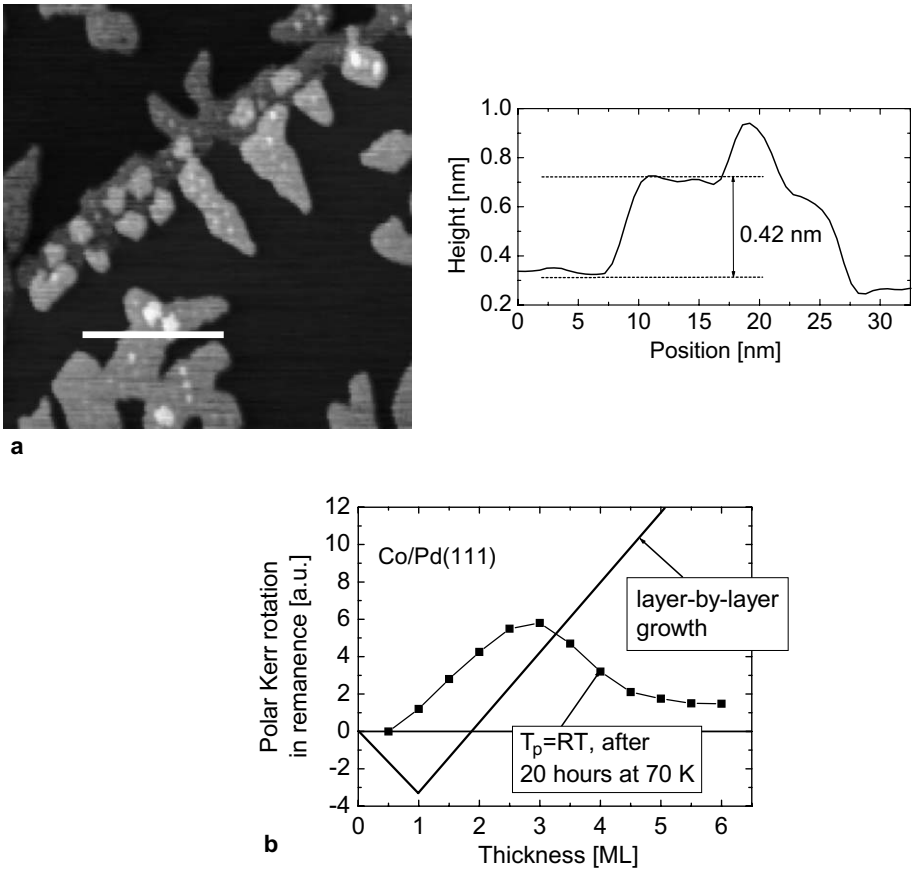

Fig. 4. (a) STM image of the 0.4 ML thick Co films grown on Pd(1 1 1) at 300 K, showing a clear double-layer growth mode – see a line profile along the white line, where the distance of 0.42 nm corresponds to 2 ML of Co. (b) Corresponding experimental thickness variation of polar Kerr rotation in remanence at 70 K for the Co films on Pd(1 1 1) exposed to a residual atmosphere for 20 h at 70 K (points) and expected variation of polar Kerr rotation for an ideal layer-by-layer growth mode (thick solid line).

to the residual atmosphere (or covering with Au) was necessary to induce the PMA to be able to measure the polar Kerr rotation.

'Reversed' out-of-plane hysteresis loops should appear in polar Kerr rotation at low Co coverage in the case of layer-by-layer growth, as it is schematically shown in Fig. 4(b) by the solid line. The gradual appearance of the Co monolayer would result in increasing negative polar Kerr rotation which would be balanced to about zero when the second atomic layer would start to complete as it happens for Co on Pd(0 0 1). This is due to the large negative Kerr rotation measured only for the 1 ML-thick Co layer on the Pd surface.

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 non-reproducible results for Co/Pd(1 1 1) systems reported in literature concerning, for example, the thickness range in which the negative Kerr rotation is observed [13,14].

In conclusion, we stress the fact that on the Pd(0 0 1) surfaces the negative polar Kerr rotation was observed at low Co coverage, i.e., for less than 2.5 ML-thick Co films. Above this thickness the interface contribution is overridden by the positive Kerr rotation from the non-interface Co atoms. For Co on Pd(1 1 1), due to the three-dimensional growth mode which starts from the formation of 2 ML-thick patches, the negative Kerr rotation is not observed even for very low coverage. The experimental results of the negative Kerr rotation contribution from Co/Pd interface with respect to that of Co agree qualitatively with published electronic structure calculations.

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