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The influence of interface structure on the magneto-optical properties of Co/Pd multilayers

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Abstract. Magneto-optical Kerr effect (MOKE) spectra of Co/Pd multilayered structures (MLS), measured in the polar geometry over the spectral range 0.8–5.5 eV, are presented and compared to spectra obtained from *ab initio* band-structure calculations. It is shown that the MOKE spectra of the Co/Pd MLS calculated using the spin-polarized relativistic linear-muffin-tin-orbital method reproduce the experimental spectra only moderately well if ideal multilayer structures with sharp interfaces are assumed. Calculations of the magneto-optical (MO) response of MLS with imperfect interfaces modelled by introducing ordered compounds at the interface have also been performed. The results obtained reveal that the MOKE spectra of the Co/Pd MLS can be adequately reproduced using a model of limited interdiffusion within one or two atomic planes at the interface which results in the formation of a Pd-rich alloy. The role of the Pd spin polarization in the formation of the MO spectra as inferred from first-principles calculations is examined and discussed. Finally, the influence of thermal annealing on the MO properties of Co/Pd MLS is studied both experimentally and theoretically. The good agreement between the calculated spectra of the MLS and experimentally observed ones demonstrates the validity of the proposed models. The results obtained demonstrate the crucial role played by the interface microstructure in the formation of the MOKE spectra of the multilayered systems studied.

1. Introduction

In recent years, artificial magnetic layered structures have been investigated extensively due to their unique physical properties. In particular, Co/Pd and Co/Pt MLS attract a lot of interest because they exhibit simultaneously large magneto-optical Kerr rotation in the uv spectral range [1] and perpendicular magnetic anisotropy [2]—a combination of properties that allows the use of these materials as a recording medium in a new generation of storage devices. It is well known that the physical properties of wide class of layered structures depend in a critical way on the quality of the interface microstructure. The influence of the interface structure, its thickness and its composition on the perpendicular magnetic anisotropy, giant magnetoresistance [3], or oscillatory interlayer exchange coupling [4] is of key importance and is the subject of a large number of studies. Recently, some investigations on this subject have been performed using *ab initio* calculations [5–7]. However, as far as we are aware, the influence of the interface structure on the magneto-optical properties of multilayered systems has not yet been studied from first principles.

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In spite of the large number of experimental and theoretical investigations [1, 8, 9, 14, 16], the mechanism responsible for the MO properties of Co/Pd MLS is far from being completely understood. The magnetic and MO properties of Co/Pd MLS depend on the individual Co and Pd layer thicknesses and are strongly affected by the interface microstructure. Experimental investigations of the MOKE spectra of Co/Pd MLS revealed that the spectra cannot be explained by scaling the fcc Co spectrum according to the amount of cobalt in the MLS. The additional contribution is commonly interpreted as arising from the exchange polarization of Pd 4d states [1] induced by adjacent Co layers. On the other hand, formation of an alloy layer at the MLS interface has also been suggested as the origin of this contribution. The extent of the so-called 'magnetic Pd' estimated via a widely used phenomenological model based on electromagnetic wave theory [1, 9] is of the order of several Pd atomic planes. However, it is impossible to derive from this model information concerning the microscopic origins of the interface magnetism and MO activity. The phenomenological approach does not account for subtle electronic effects at the interface such as 3d–4d hybridization, and the magnitude and range of the exchange polarization. This information can now be obtained due to the development of *ab initio* methods based on the band theory of the MOKE which have allowed the investigation of the microscopic origin of the MO effects for a wide class of metallic systems [10–18]. Until recently, the MO effects in complex structures could not be treated via *ab initio* calculations, due to computational limitations. Recent achievements for Co/Cu MLS [19] and Co–Pd alloys [20] have demonstrated, however, that the systems can be successfully modelled by means of first-principles band-structure calculations.

In the present work, Co/Pd structures are chosen as the model system for demonstrating and discussing the possible mechanisms of MOKE spectra formation. The aim of this paper is a detailed experimental and theoretical study of the electronic structure and MO properties of Co/Pd MLS using MOKE spectroscopy and first-principles band-structure calculations. The magneto-optical polar Kerr rotation and ellipticity spectra of a set of Co/Pd multilayers were measured over the spectral range 0.8–5.5 eV and compared to the MOKE spectra obtained from the *ab initio* calculations. The calculations were performed for ideal Co/Pd MLS with sharp interfaces as well as for MLS with imperfect interfaces of various thicknesses and compositions, with Co and Pd layer thicknesses close to the experimental ones. Special attention was given to the investigation of the different mechanisms responsible for the observed MOKE spectra, especially Pd spin polarization and alloying at the Co–Pd interface.

The paper is organized as follows. Section 2 describes experimental and computational details. The measured MOKE spectra of the Co/Pd MLS are described in section 3. In section 4.1 the MOKE spectra of the ideal model Co/Pd MLS calculated for different sublayer thicknesses are presented and discussed. In section 4.2 the results of the MOKE calculation for the model MLS structures with imperfect interfaces are compared to the experimental spectra. Concluding remarks are made in section 5.

2. Experimental and computational details

2.1. Experimental procedure

The Co/Pd multilayers were prepared on water-cooled glass substrates using the dc magnetically enhanced face-to-face sputtering system described in reference [21]. The chemical compositions of the films were determined by x-ray fluorescence analysis from the peak

intensities of the characteristic fluorescence radiation. Structural studies were performed by conventional x-ray diffractometry. Formation of multilayer structures was confirmed by low-angle x-ray diffraction and the modulation period was deduced from the position of the Bragg peaks. The periods determined from the x-ray measurements agree with those evaluated from the fluorescence analysis. The films were found to be of fcc (111)-textured structure. The parameters of the samples studied are given in table 1.

Table 1. The parameters of the Co/Pd MLS studied. The sample numbers are given, together with N_{layers} , the number of bilayers, and t_{Co} and t_{Pd} , the measured thicknesses of the Co and Pd sublayers.

| No | N_{layers} | t_{Co} (Å) | t_{Pd} (Å) |
|----|---------------------|---------------------|---------------------|
| 1 | 52 | 3.5 | 18.3 |
| 2 | 47 | 5.2 | 17.6 |
| 3 | 43 | 6.4 | 18.2 |
| 4 | 40 | 8.4 | 18.5 |
| 5 | 33 | 13.8 | 19.8 |
| 6 | 28 | 17.4 | 20.6 |

The polar Kerr rotation (θ_K) and ellipticity (η_K) spectra were measured by means of the polarization modulation method using a piezobirefringent modulator. Besides high sensitivity approaching 10^{-4} degrees, this method has the advantage that the Kerr rotation and Kerr ellipticity can be determined simultaneously. A detailed description of the MOKE spectrometer system used is given in reference [18]. All of the spectra were measured at room temperature over the photon energy range $0.8 < h\nu < 5.5$ eV. The optical properties were measured directly by spectroscopic ellipsometry using the rotating-analyser method over the same spectral range as for the MO spectra [18].

It is well known that the MOKE properties are determined by both the diagonal and the off-diagonal components of the optical conductivity tensor $\sigma_{\alpha\beta}$ of a material. For a solid with at least threefold rotational symmetry and in the polar geometry, the complex magneto-optical Kerr angle $\phi_K = \theta_K + i\eta_K$ is given by [22, 23]

$$\phi_K = \frac{-\sigma_{xy}}{\sigma_{xx}\sqrt{1 + i(4\pi/\omega)\sigma_{xx}}}. \quad (1)$$

The conductivity tensor $\sigma_{\alpha\beta}$ is related to the dielectric tensor

$$\varepsilon_{\alpha\beta} = \delta_{\alpha\beta} + i(4\pi/\omega)\sigma_{\alpha\beta}.$$

The complex quantities are defined as $\sigma_{\alpha\beta} = \sigma_{\alpha\beta}^{(1)} + i\sigma_{\alpha\beta}^{(2)}$, $\varepsilon_{\alpha\beta} = \varepsilon_{\alpha\beta}^{(1)} + i\varepsilon_{\alpha\beta}^{(2)}$, and the diagonal element $\varepsilon_{\alpha\alpha} = (n + ik)^2$, where n and k are refraction and extinction coefficients, respectively.

2.2. Computational details

Self-consistent energy band-structure calculations for Co–Pd compounds were performed by means of the spin-polarized fully relativistic (SPR) LMTO method [15, 24–26] with the combined correction terms to the LMTO Hamiltonian and overlap matrices taken into account. The calculations were based on the spin-density-functional theory with von Barth–Hedin parametrization [27] of the exchange–correlation potential. Core charge densities were recalculated at every iteration of the self-consistency loop. The Brillouin zone (BZ) integrations were performed using the improved tetrahedron method [28].

The absorptive part of the optical conductivity tensor components was computed on the basis of the linear-response expressions [29]. The combined correction terms were included in relativistic expressions for evaluation of the optical transition matrix elements starting from the SPR LMTO eigenfunctions [13, 16, 18, 30]. The dispersive part of the conductivity tensor was obtained via the Kramers–Kronig transformation. Finite-lifetime effects were simulated by broadening the calculated spectra with a Lorentzian of width 0.7 eV. An intraband contribution to the diagonal part of the optical conductivity was taken into account via a phenomenological Drude term, with the plasma frequency computed through Fermi surface integration [30, 31].

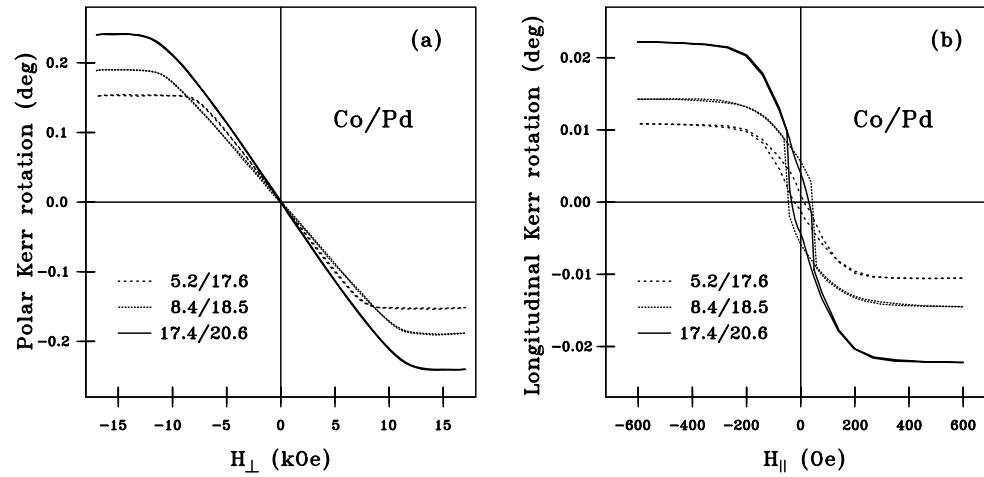


Figure 1. Polar (a) and longitudinal (b) Kerr rotation hysteresis loops for the selected Co/Pd MLS films measured at a photon energy of 2.27 eV. The Co and Pd sublayer thicknesses are given in the keys in units of Å.

3. Experimental results

In Co/Pd multilayered structures a large proximity magnetization of the Pd spacer, which is in contact with the magnetic Co, via a hybridization mechanism is expected. To study the origin of the magneto-optical spectra in the Co/Pd MLS and as a basis for discussion of the underlying mechanism, we measured the MOKE spectra for a set of multilayer films with different Co sublayer thicknesses. The total thicknesses of the samples studied were kept over 1000 Å, to avoid optical interference with the substrate. Representative results of the hysteresis investigations for selected MLS films are presented in figure 1. The hysteresis loops, measured in the longitudinal Kerr geometry with in-plane orientation of the applied magnetic field and in the polar Kerr geometry with out-of-plane orientation of the applied magnetic field, H_{\parallel} and H_{\perp} , respectively, show that the MLS studied were obtained with their easy-magnetization axes in the film plane.

The polar Kerr rotation (θ_K) and ellipticity (η_K) spectra of the Co/Pd MLS measured under saturation conditions are shown in figures 2(a) and 2(b), respectively. For comparison, spectra of 2000 Å thick fcc Co film, multiplied by a factor of 0.7, are also plotted in figure 2. It can be seen that the MLS Kerr spectra never exceed in magnitude those of the fcc Co over the whole spectral range. The prominent features of the θ_K -spectra are a broad negative peak at ~ 3 eV and a shoulder at ~ 4.3 eV followed by a θ_K -slope varying as a function of

the Co layer thickness. The θ_K -peak, centred at 1.5 eV, characteristic of fcc Co, is strongly reduced with decreasing MLS Co sublayer thickness, and the prominent minima at ~ 3 eV in θ_K and at ~ 5 eV in η_K diminish in amplitude. The shift of the zero-crossing of η_K up to ~ 3 eV is accompanied by the formation of a positive two-peak structure with decrease of the amount of Co present. Similar spectral features of the Co/Pd MLS, although within smaller spectral ranges and for other sublayer thicknesses, were observed in references [1, 8] and [9]. Inspection of the θ_K - and η_K -spectra of the MLS leads one to the conclusion that there is a MO contribution independent of, or weakly dependent on, the cobalt sublayer thickness. These effects that are independent of the Co content may arise from Pd spin polarization induced by the proximity of Co layers and/or from the polarization caused by roughness and limited alloying at the Co–Pd interface.

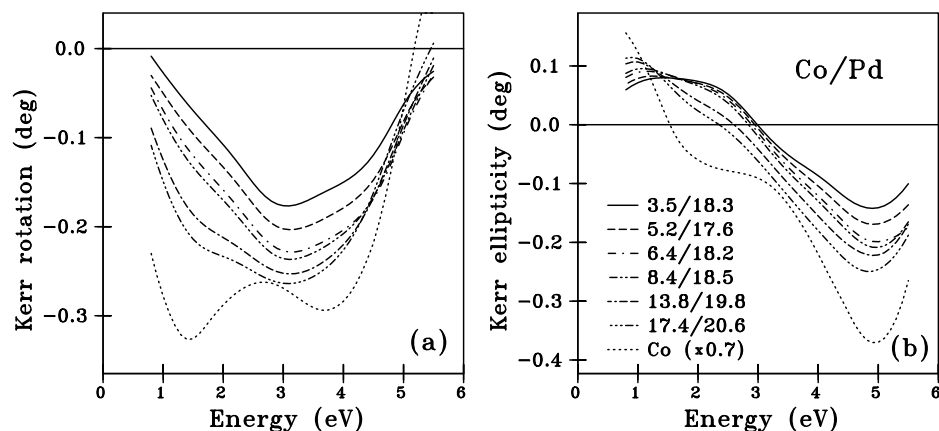


Figure 2. Experimental polar Kerr rotation (a) and ellipticity (b) spectra of Co/Pd MLS and fcc Co film. The Co and Pd sublayer thicknesses are given in the keys in units of Å.

To study the mechanism responsible for the MOKE in the Co/Pd MLS and to enable us to discuss the magneto-optical response of the films in terms of separate contributions of the Co and Pd sublattices, the data should be, in principle, analysed using the off-diagonal optical conductivity tensor components σ_{xy} , which are directly related to the magneto-optical transitions between the spin-polarized electronic states. From the measured θ_K - and η_K -data, and the diagonal σ_{xx} -tensor component, the off-diagonal σ_{xy} -component can be evaluated according to equation (1). The values of σ_{xx} for the fcc Co and Pd films and the Co/Pd MLS under consideration were obtained by an ellipsometric method for the range 0.8–5.5 eV. It was found that the σ_{xx} -spectra of the multilayers are close to each other and do not exhibit fine structure, except a small broad peak at about 4–5 eV.

For all of the Co/Pd MLS studied, it was found that the denominator in equation (1) evaluated from the optical data is a monotonic and structureless function over the whole energy range and depends weakly on the film composition. Consequently, all of the peaks in the polar Kerr rotation and ellipticity spectra originate from the corresponding features of the absorptive and dispersive parts of $\sigma_{xy}(\omega)$, respectively. The energy dependence of the denominator in equation (1) only changes the relative amplitudes of the uv and ir peaks of the spectra, without producing any additional spectral features. It can be concluded that in the Co/Pd MLS the MOKE is governed by the off-diagonal part of the optical conductivity tensor.

4. Theoretical results and discussion

4.1. Multilayers with perfect interfaces

In an attempt to reproduce and to explain the experimental MOKE spectra, band-structure calculations of the MO properties of some model MLS were performed. As the influence of the structure of the Co–Pd interface on the MOKE spectra is the subject of this study, in the first step an idealized model with a sharp interface was adopted in the calculations. The numbers n of Co and m of Pd atomic planes in the model $n\text{Co}/m\text{Pd}$ MLS were chosen as close as possible to the experimentally measured ones. For all of the MLS, an abc stacking sequence of closely packed Co and Pd planes was assumed. In reference [34] it was shown that for Co sublayers that are not very thick (less than 30 Å, with the Pd sublayer thickness being 12 Å), the in-plane lattice spacing in Co/Pd MLS is almost independent of the Co sublayer thickness and only $\sim 2\%$ less than the lattice spacing in fcc Pd. Therefore the lattice constant $a = 2.694$ Å of the hexagonal lattice was chosen. The interplane spacings in the Co and Pd sublayers and at the interface were chosen to be equal, and to correspond to the ideal c/a ratio ($c/a = q\sqrt{2/3}$, where $q = n + m$ is the number of close-packed atomic planes). The sphere radii of Co and Pd were both taken as equal to the average S_{WS} -value of 1.49 Å.

Table 2. Calculated Co and Pd spin magnetic moments (μ_B/atom) in the Co/Pd MLS. The number after a chemical symbol denotes the number of corresponding atomic planes starting from the Co–Pd interface. For fcc Co with S_{WS} set at the same value as for the MLS, the value of $1.72\mu_B/\text{atom}$ was obtained.

| MLS | Co2 | Co1 | Pd1 | Pd2 | Pd3 |
|---------|------|------|------|------|------|
| 1Co/8Pd | | 1.94 | 0.26 | 0.12 | 0.04 |
| 2Co/7Pd | | 1.84 | 0.23 | 0.10 | 0.03 |
| 4Co/8Pd | 1.72 | 1.83 | 0.23 | 0.11 | 0.04 |
| 6Co/9Pd | 1.72 | 1.83 | 0.23 | 0.12 | 0.04 |
| 9Co/9Pd | 1.72 | 1.83 | 0.23 | 0.12 | 0.04 |

The distributions of the spin magnetic moments in the Co and Pd atomic spheres derived from the band-structure calculations are given in table 2. For all of the MLS studied, the dependence of the magnetic moments on the distance from the interface is the same. The Co magnetic moment is enhanced at the interface and is already approaching the bulk value of $1.72\mu_B/\text{atom}$ in the second plane. The same value of the Co moment ($1.72\mu_B/\text{atom}$) was obtained from the calculations for fcc Co with the lattice constant expanded so that the value of S_{WS} was equal to that for the MLS. The value of the magnetic moment induced in Pd rapidly decreases with the distance from the interface. In the MLS with a Pd sublayer consisting of more than six Pd atomic planes, the magnetic moments induced in the middle of the Pd sublayer are smaller than $0.015\mu_B/\text{atom}$ and are not shown in table 2. The rapid decrease of the Pd magnetic moment with the increase of the distance from the interface can be easily understood if one recalls that the moment is mainly due to Co 3d–Pd 4d hybridization. A Pd atom, even in only the second atomic plane from the interface, has no Co atoms even amongst its second-nearest neighbours, and interacts with the exchange-split Co 3d states only indirectly.

The calculated MOKE spectra of the Co/Pd MLS are shown in figure 3. For the sake of comparison, spectra of fcc Co (calculated with the same value of S_{WS} as the MLS), multiplied by a factor of 0.7, are also shown. From the comparison with the experimental

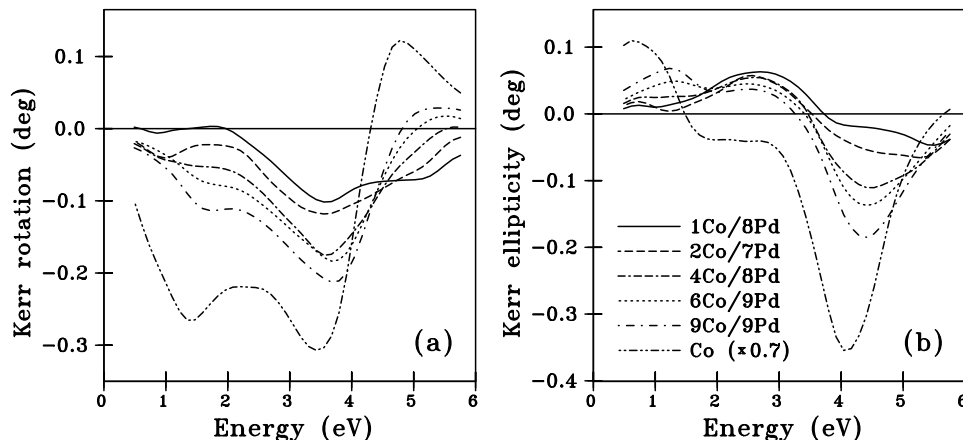


Figure 3. Calculated polar Kerr rotation (a) and ellipticity (b) spectra for the model $n\text{Co}/m\text{Pd}$ MLS with sharp interfaces (n and m are the numbers of Co and Pd atomic planes, respectively). For comparison, spectra of fcc Co (calculated with the same value of S_{WS} as the MLS), multiplied by a factor of 0.7, are also shown.

spectra, it is seen that in the uv range the shoulder at ~ 5 eV, which is a characteristic feature of the experimentally measured polar Kerr rotation spectra of the MLS, manifests itself only in the spectra of the 1Co/8Pd MLS and, to a lesser extent, those of the 2Co/7Pd MLS. For the MLS with larger numbers of Co atomic planes, the magnitude of the Kerr rotation spectra in the uv range increases with increase of the Co sublayer thickness, whereas the shape of the spectra changes only slightly. As can be seen from figure 3(b), the features of the Kerr ellipticity spectra, corresponding to the peaks at 1.0 and 4.3 eV in the fcc Co spectrum, become more pronounced as the number of atomic planes of Co increases.

Such a dependence of the calculated MOKE spectra of the MLS on the Co sublayer thickness is explicable. As the magnetic moment induced in the Pd atoms located far from the interface is very small, one can expect these atoms to give only a small contribution to the off-diagonal optical conductivity. Consequently, if the model with a sharp interface is adopted, the MO properties of the MLS are determined by the Co sublayer and one, or perhaps two, adjacent Pd atomic planes. Also, from the calculated DOS curves and Co magnetic moments it follows that only the electronic states in the two Co atomic planes nearest to the interface are modified strongly by the hybridization with the Pd states. Hence, as the Co sublayer thickness increases, the MOKE spectra of the MLS should become closer to that of pure Co. This is exactly what can be seen in figure 3. As the number of Co atomic planes increases, the relative contribution of the interfacial layer to the spectra becomes smaller and a shoulder appears at 1.5 eV in the spectra which is evidently related to the corresponding peak in the fcc Co spectrum.

A comparison with the experimental spectra of the MLS (figure 2) shows that the calculations reproduce only moderately well the main peculiarities of the MOKE spectra of the MLS and tendencies in the modification of the MLS spectra with the variation of the Co sublayer thickness, and the quantitative agreement is not satisfactory.

The calculated spectra are of smaller magnitude; the decrease of the magnitude for the MLS with a small number of Co atomic planes is larger than in the experiment. The feature at 1.5 eV in the calculated spectra of the MLS with a thin Co sublayer disappears totally and the magnitude of the spectra goes almost to zero in this energy range. The loss of

the amplitude at this energy, although observed experimentally, is not so drastic. Also, the calculations predict a rapid decrease of the Kerr rotation amplitude of the shoulder at 5 eV with respect to that of the main minimum as the number of Co atomic planes increases, while the shoulder is observable in the experimental spectra.

It is worth recalling that the spectra of fcc Co shown in figure 3 were calculated with the enlarged lattice constant used for the model MLS and, consequently, they should not be compared directly to the experimental Co spectra (see figure 2). The position of the uv minimum in the polar Kerr rotation spectrum of Co, calculated using the experimental fcc Co lattice constant (i.e., for $S_{\text{WS}} = 1.395 \text{ \AA}$), is shifted to higher energies by $\sim 1 \text{ eV}$ (see reference [18]). Thus, the shift of $\sim 0.7 \text{ eV}$ in the energy position of the uv minimum observed in the experimental MLS spectra with respect to that for the fcc Co film is reproduced—although it is overestimated—by the calculations.

There are several possible sources of these discrepancies. First of all, the peaks in the polar Kerr rotation spectra calculated for either fcc or hcp Co are shifted to higher energies with respect to the measured ones and the magnitude of the minimum at 1.5 eV is too small. This discrepancy is apparently due to a failure of the LDA-based calculations to predict correctly the MOKE spectra of the ferromagnetic 3d metals that has recently been extensively discussed in the literature [13, 18, 32]. A plausible cause for this lies in the approximate description of the exchange and correlation of rather localized 3d electrons. If the calculated magnitude of the ir peak of fcc Co were larger, it would bring the theoretical MOKE spectra of the MLS into closer agreement with the experimental ones for the energy range below 2 eV. In the uv range, however, a shift of the Co Kerr rotation peak to lower energies would hardly improve the agreement between the theory and experiment at all.

The shift of the calculated peak positions may also be caused by the difference between the experimental interatomic distances and those used in the calculations. Determination of the in-plane and interplane distances in an individual sublayer of the MLS or overlayers structures is an onerous task and the experimental data available for Co/Pd MLS differ significantly [34–36]. To estimate the sizes of the possible effects of the interplane relaxation, test calculations using the unmodified in-plane interatomic distance but a smaller c/a ratio were performed for Co (with a hexagonal supercell) and 4Co/5Pd MLS. It was found that the theoretical MOKE spectra are rather insensitive to the interplane distance. A decrease of the c/a ratio by 5% results in a small increase in amplitude of the polar Kerr rotation spectra, but the peak positions remain unchanged. Thus, though a contraction of interplane distances in the Co sublayer was derived from structural investigations [35, 36], it is unlikely to be responsible for the discrepancies between the theoretical and experimental MOKE spectra of the Co/Pd MLS. An in-plane relaxation in a thick Co sublayer might affect the calculated MOKE spectra strongly, but performing such a calculation is beyond the capability of our facilities. Moreover, there is no experimental evidence that relaxation really occurs in the MLS studied.

The idea of enhanced spin polarization in the Pd planes adjacent to the Co was also considered. For some of the MLS studied, we performed test calculations in which an additional exchange splitting was introduced for one or two of the Pd atomic layers nearest to the interface. As a result of the additional spin polarization of the Pd atoms, the intensity of the calculated polar Kerr rotation spectra in the uv range increases. However, the strong positive rotation at low photon energies worsens the agreement with the experiment in this energy range.

It can be concluded that the main cause of the differences between the theory and experiment lies in the choice of a model with a sharp interface as a basis for the calculations. An intermixing at the interface, even within just one or two atomic layers, can appreciably

change the shape and magnitude of the MOKE spectra of the MLS, especially those with small Co sublayer thicknesses. The next part of this paper is devoted to a quantitative study of this problem.

4.2. Multilayers with imperfect interfaces

From the discussion above, it follows that interfacial intermixing should be taken into account if one is to describe the MO properties of real Co/Pd MLS systems. A first improvement that takes us closer to describing 'real systems' is that of considering an ordered compound at the interface. To examine the consequences of interface intermixing for Co/Pd MLS MOKE spectra and their relation to the chemical nature of the interface, a model of multilayer supercells with alloyed interfaces was considered. The study was performed with the use of the above-described formalism of *ab initio* band-structure calculations. As the LMTO method requires a crystal lattice with specified translational and point symmetry, special model supercells of the multilayered structures with alloyed interfaces and fcc (111) texture were constructed. In the [111] direction, for all model MLS an abc stacking sequence of closely packed atomic Co and Pd planes was assumed. As an interface between the neighbouring Co and Pd layers, one or two Co–Pd mixed atomic planes were introduced. To satisfy the requirement of overall D_{3d}^3 lattice symmetry of the supercells and to minimize their volume, the unit cell was doubled in the basal plane. As a consequence, interfacial planes in these supercells can consist of mixed atomic planes composed of an ordered Co–Pd compound with 1:3 and 3:1 stoichiometry and threefold rotational symmetry. Three types of multilayered structure with different interfacial atomic planes were considered. The $n\text{Co}/\text{A}/m\text{Pd}$ - and $n\text{Co}/\text{B}/m\text{Pd}$ -type structures contain Pd-rich Co_1Pd_3 (A) and Co-rich Co_3Pd_1 (B) ordered interfacial planar alloys, respectively. The structure $n\text{Co}/\text{C}/m\text{Pd}$ with $\text{C} = \text{Co}_3\text{Pd}_1/\text{Co}_1\text{Pd}_3$ has a two-layer interfacial ordered alloy composed of consecutive B- and A-type planes.

To understand the relationship between the interfacial structures and the MOKE spectra, a systematic study was performed for Co/Pd MLS with a period of six atomic layers. Calculations for larger supercells are extremely time consuming and were performed only for some selected cases. It was found that the calculations performed are sufficiently representative and illustrate the main trends in the modification of the MO spectra with interface imperfection. In all of the calculations, the same lattice constant as for the ideal model MLS was assumed.

The calculated polar Kerr rotation spectra for the Co/Pd MLS, where the Co and Pd layers are separated by three types of interface alloy layer (A, B and C), are shown in figures 4(a)–4(c). For comparison, the spectrum of fcc Co calculated with the same value of S_{WS} and the spectra of ideal 1Co/5Pd, 2Co/4Pd and 3Co/3Pd MLS are also included. The spectra calculated for the supercell structures in which the Pd layers are separated by alloy layers only are presented in figure 4(a). The supercells 0Co/A/4Pd and 0Co/B/4Pd consist of a two-atomic-layer thickness of alloy, of either Co_1Pd_3 or Co_3Pd_1 composition, separated by a four-atomic-layer thickness of Pd spacer. The supercell 0Co/C/2Pd consists of repeated sequences of four alloy atomic planes, $\text{Co}_1\text{Pd}_3/\text{Co}_3\text{Pd}_1/\text{Co}_3\text{Pd}_1/\text{Co}_1\text{Pd}_3$, separated by two Pd atomic planes. These structures can be regarded as limiting cases, where Co atoms are present in alloy planes only.

As is seen from figure 4(a), in the uv range the main features of the 0Co/A/4Pd spectrum are very similar in shape to those of the 1Co/5Pd MLS with the sharp interface. It is interesting that the amplitude of the 0Co/A/4Pd spectrum is about 70% higher than the amplitude of the 1Co/5Pd spectrum, despite the higher Co content in the latter. However,

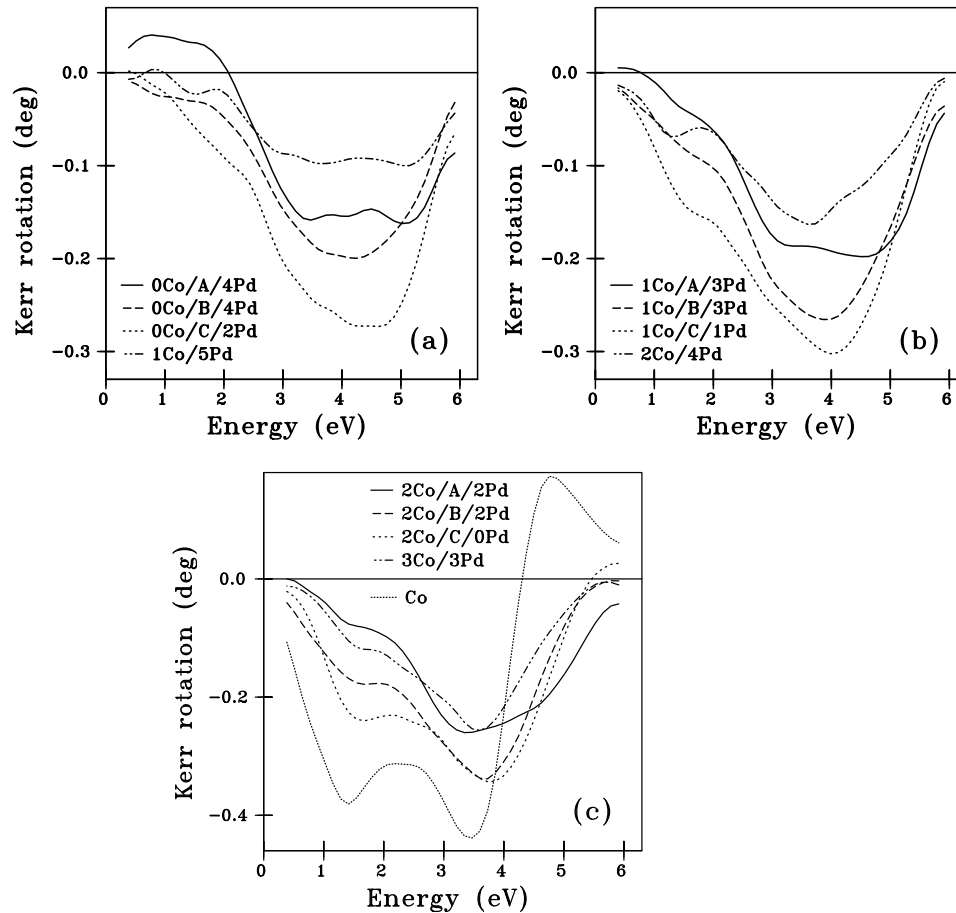


Figure 4. Calculated polar Kerr rotation spectra for the model $n\text{Co}/X/m\text{Pd}$ multilayered structures with imperfect interfaces. n and m are the numbers of Co and Pd atomic planes, respectively. $X = A, B, C$ denotes the interfacial planar ordered alloys Co_1Pd_3 , Co_3Pd_1 and $\text{Co}_3\text{Pd}_1/\text{Co}_1\text{Pd}_3$, respectively. For comparison, the corresponding MLS spectra for the ideal model $n\text{Co}/m\text{Pd}$ and the spectrum of fcc Co with the same value of S_{WS} as for the MLS are also shown.

in the energy range below ~ 2 eV positive rotation is observed for the $0\text{Co}/\text{A}/4\text{Pd}$ MLS. It should be noted that the overall shape of the $0\text{Co}/\text{A}/4\text{Pd}$ Kerr rotation spectrum is very close to that calculated for homogeneous diluted Co–Pd alloys [20]. Thus, the conclusions are that the Kerr rotation spectrum of the $0\text{Co}/\text{A}/4\text{Pd}$ structure is determined to a great extent by the MO properties of spin-polarized Pd. The spectrum of $0\text{Co}/\text{B}/4\text{Pd}$, in which the interface region is formed by the Co-rich alloy, differs significantly from both the $0\text{Co}/\text{A}/4\text{Pd}$ and the $1\text{Co}/5\text{Pd}$ spectra. The two-peak uv structure transforms into a broad minimum centred at 4.2 eV and in the ir region the rotation becomes negative. The structure with a C-type interfacial layer is particularly interesting, as it can be obtained from the ideal $2\text{Co}/4\text{Pd}$ multilayer structure simply by interchanging the positions of every fourth Co and Pd atom in the neighbouring interfacial Co and Pd layers. As a result, Co-rich Co_3Pd_1 and Pd-rich Co_1Pd_3 planar alloy layers appear around the border. The composition of the $0\text{Co}/\text{C}/2\text{Pd}$ MLS corresponds to that of $2\text{Co}/4\text{Pd}$; however, it should be pointed out that the change

of the interface microstructure leads to very large changes in the shape and magnitude of the MO spectra, as can be seen from the comparison of the 0Co/C/2Pd and 2Co/4Pd Kerr rotation spectra in figures 4(a) and 4(b), respectively.

The Kerr rotation spectra are modified appreciably upon addition of one (see figure 4(b)) or two (see figure 4(c)) Co atomic planes to an MLS with an imperfect interface. In the supercells, the Co and Pd planes are separated by A-, B- and C-type interfacial layers. The essential points as regards the modification of the spectra can be summarized as follows.

(i) The amplitude of the spectra increases as the amount of Co increases. In the ir spectral range, the characteristic features of the Co spectrum become more pronounced.

(ii) For the supercells containing one planar alloy of Co_1Pd_3 composition treated as an interface region, the change of the relative amplitudes of the characteristic peaks in the uv region upon addition of two Co atomic planes is clearly visible.

(iii) For the supercells containing as an interface region a single Co_3Pd_1 atomic plane or two $\text{Co}_3\text{Pd}_1/\text{Co}_1\text{Pd}_3$ atomic planes, the main effect of increasing the number of Co atomic planes is a shift of the uv peak position to lower energy. This effect is accompanied by a narrowing of the band width.

(iv) The amplitudes of the spectra in the energy range above 4 eV are up to 100% higher than those of the ideal 2Co/4Pd and 3Co/3Pd MLS.

From the band-structure calculation results it follows that the Co and Pd electronic states in the alloy interface layers are strongly modified by the hybridization. As the volume in which the hybridization occurs is significantly larger in a MLS with an imperfect interface, the contribution of these hybridized states to the MO spectra increases compared to that for a MLS with an ideal interface. Hence, the similarity between the $n\text{Co}/A/m\text{Pd}$ MLS spectra and the spectra of Pd-rich CoPd alloys [20] becomes closer, especially for the MLS with a small Co sublayer thickness. Another effect of interfacial alloy formation is stronger—compared to that for the ideal MLS—spin polarization of the whole Pd spacer. The averaged magnetic moment per Pd atom calculated for the different structures approaches values up to 80% larger than those for the ideal MLS models. One can expect that as a result of the increased magnetic moment induced on the Pd atoms, they will make larger contributions to the MO spectra. This can be clearly seen in the energy range below 2 eV, where the Kerr rotation spectra are very sensitive to the Pd contribution. As was established earlier [20, 33], the interband magneto-optical transitions in spin-polarized Pd give a positive Kerr rotation in the ir energy range, whereas the transitions between the electronic states with a large degree of admixture of Co 3d 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. As a result of the two compensating contributions, the amplitude of the Kerr rotation spectra of the MLS is suppressed in the ir energy range and the rotation becomes positive in the case of 0Co/A/4Pd MLS (see figure 4(a)). As the number of Co atomic planes increases, their relative contribution to the MOKE spectra becomes higher and the Kerr rotation in the ir range becomes negative, which brings the spectra closer to the fcc Co spectrum, as can be seen in figure 4. A peak appearing at 1.5 eV in the MLS spectra is evidently related to the corresponding peak in the fcc Co spectrum.

As is seen in figure 4(a), in the uv spectral range the contribution to the MLS spectra coming from the interface alloy region and strongly polarized Pd atomic planes is also large enough, contrary to the case for the ideal model of the MLS, and controls the spectral shape in that energy range. The observed overall increase of the rotation amplitude along the sequence 0Co/A/4Pd, 0Co/B/4Pd, 0Co/C/2Pd is related to an increase of the relative Co content in the structures, which changes from 8% in 0Co/A/4Pd up to 33% in 0Co/C/2Pd.

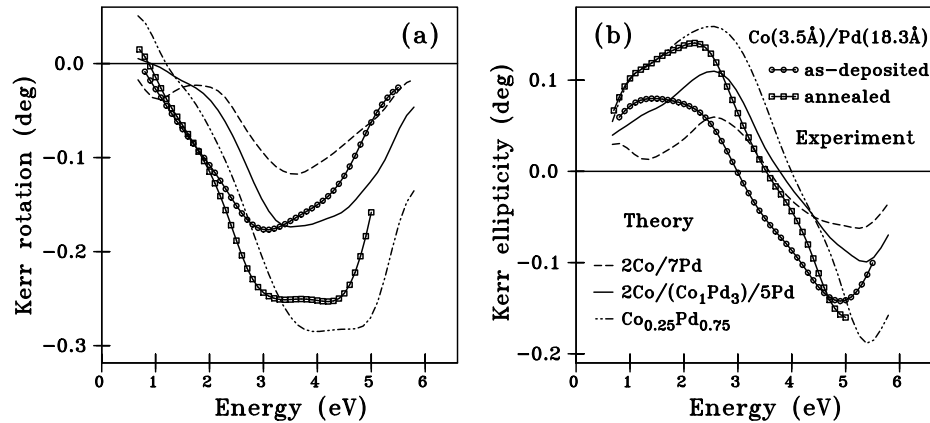


Figure 5. Comparison of the measured and calculated polar Kerr rotation (a) and ellipticity (b) spectra of the as-deposited 3.5 Å Co/18.3 Å Pd MLS film and the model 2Co/(Co₁Pd₃)/5Pd as well as the ideal 2Co/7Pd MLS model. The spectra of the film annealed at 800 K and calculated for the model of fcc disordered Co_{0.25}Pd_{0.75} alloy are included.

If one or two Co atomic planes are included in the structures, the relative contribution of Co to the total Kerr spectra increases. Hence, the shape of the spectra becomes closer to that of the fcc Co spectra and the enhancement of the Kerr rotation peak amplitude in the uv range can be seen in figures 4(b) and 4(c). It is worth pointing out, however, that the MOKE spectra are not simple superpositions of the contributions arising from the constituent sublayers.

We have found that of all the model MLS considered, the models in which the interface alloy layer is a Pd-rich alloy reproduce better the main spectral features of the experimentally studied Co/Pd MLS and the tendencies in the modification of the spectra as the thickness of the Co sublayer increases. In figure 5, the MOKE spectra calculated for the ideal 2Co/7Pd MLS and the 2Co/(Co₁Pd₃)/5Pd model structure are directly compared to the experimental spectra of the 3.5 Å Co/18.3 Å Pd MLS with the sublayer thicknesses closest to the model structure. It is clearly seen that, if the interfacial alloying is taken into account, the characteristic shape of the measured spectrum with the shoulder in the uv range is reproduced well by the calculations. Also, the amplitude of the calculated Kerr rotation is close to the experimentally studied one, as opposed to the ideal model 2Co/7Pd MLS with the same period for which the amplitude is 50% smaller. The enhancement of the calculated amplitude and better agreement with the experiment can be observed also for the Kerr ellipticity spectra.

To study the effect of total interlayer intermixing on the MOKE spectra and as a final test of the assumed interface model, we annealed the 3.5 Å Co/18.3 Å Pd MLS film under high vacuum at 800 K for 5 h. At such high temperatures the diffusion processes are very intensive and the whole layered structure is uniformly homogenized, becoming a disordered fcc alloy film of effective composition Co_{0.2}Pd_{0.8}, as was controlled by our structural study. The huge changes of the MOKE spectra after annealing can be seen from figure 5. The Kerr rotation and ellipticity spectra amplitudes increase significantly and the spectral shapes become close to those observed for Co–Pd alloys of comparable composition [20]. The calculated MOKE spectra of disordered Co_{0.25}Pd_{0.75} alloy taken from reference [20] are also included in figure 5 for comparison. As can be seen, the agreement of the calculated and measured Kerr rotation and ellipticity spectra of the alloyed structure is excellent (the

systematic shifts of the spectra on the energy scale are commonly seen for LDA-based calculations, as was discussed in section 4.1) Thus, on the basis of the results obtained for the Co/Pd MLS, the following conclusions have been reached:

- (i) the MLS studied have relatively sharp interfaces of the 'chemical thickness' of the order of one atomic layer; and
- (ii) they can be well described by a model MLS structure with an ordered Pd-rich interfacial alloy.

In the real Co/Pd MLS, the interface microstructure may be more complicated and probably only partial chemical ordering occurs. As was inferred from the x-ray diffraction data, the Co/Pd MLS studied have well-defined layered structure. Nevertheless, some intermixing at the interface during the samples' deposition process is very probable. This effect leads, in turn, to higher induced spin polarization in a larger volume of Pd and makes the whole volume magneto-optically active. The results obtained allow us to explain the origin of the observed MOKE spectra of the MLS and to understand why the thickness of the so-called 'magnetic interface' can be larger than that of the 'crystallographic' one, as was recently reported for other Pd-based multilayered systems [37]. The conclusions are also consistent with the results derived from the phenomenological multireflection approach, not presented here, which indicate the Pd spin polarization to extend through a depth of the order of four Pd atomic layers.

5. Summary

A detailed comparative experimental and theoretical study of the electronic structure and the magneto-optical properties of Co/Pd multilayers has been performed. The polar Kerr rotation, ellipticity and optical spectra for a set of Co-Pd MLS were measured over the spectral range 0.8–5.5 eV. The MOKE spectra were compared to the spectra of model MLS structures calculated by the SPR LMTO method. It was found that in the Co-Pd systems the MOKE is governed by the off-diagonal part of the optical conductivity tensor.

First, *ab initio* calculations were performed for model Co/Pd MLS with sharp interfaces and sublayer thicknesses close to those in the experimentally studied MLS. It was found that these calculations reproduced the main peculiarities of the experimental Co/Pd MOKE spectra only moderately well.

Then, Co/Pd MLS with the interface microstructure modelled by ordered planar alloys of different compositions were examined. The MOKE spectra, calculated from first principles for these MLS, differ considerably from those obtained for the models with sharp interfaces. It is shown that the main peculiarities and the tendencies in the modification of the MLS spectra with the variation of the Co sublayer thickness are adequately reproduced when alloying, even limited to one atomic plane, is taken into account. The MOKE spectra calculated for the model structures with ordered Co₁Pd₃ interfacial planes reproduce the measured spectra best. Very good agreement between the calculated and measured MOKE spectra demonstrates the validity of the adopted model and approach. The results obtained demonstrate that interface microstructure plays a crucial role in the formation of the MOKE spectra of the Co/Pd layered structures.

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