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Influence of alloy composition and local environment on the magneto-optical properties of Co_xPd_{1-x} alloys

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Results of *ab initio* calculations performed to investigate the dependence of magneto-optical properties of Co_xPd_{1-x} alloys on chemical ordering and alloy composition are presented. A procedure of averaging the calculated spectra for compounds with respect to the orientation dependence of the gyration vector is proposed that makes it possible to account for the effects of random orientation of microcrystals in polycrystalline samples. Good agreement between theoretical and experimental spectra for varying alloy composition is found demonstrating the applicability of the theoretical approach. [S0163-1829(98)09503-4]

I. INTRODUCTION

Transition-metal systems consisting of a ferromagnetic $3d$ element and nonmagnetic $4d$ or $5d$ elements have drawn a lot of attention over the past years because of their favorable magnetic and magneto-optical (MO) properties (see, e.g., Refs. 1 and 2). In particular, Co-Pd and Co-Pt alloys as well as related multilayered structures received special interest because they exhibit simultaneously large magneto-optical Kerr rotation in the uv spectral range[1] and perpendicular magnetic anisotropy[2]—the combination of properties that allows us to use these materials as a recording medium in a new generation of storage devices.

A large number of studies have been performed for these compounds using various experimental methods.[1–4] In particular, it has been shown that in such systems normally nonmagnetic Pd metal orders ferromagnetically and its magnetic moment can reach up to $0.5\mu_B$.[5] The magneto-optical Kerr effect (MOKE) spectroscopy is a particularly valuable tool to study the spin-polarized electronic structure of magnetic materials, and a number of magneto-optical discoveries including the quantum confinement effects[6] and orientation dependence of the Kerr effect in epitaxial Co films[7] have been established. These results have stimulated theoretical investigations of the magnetic and electronic structure of these systems.

Progress in the *ab initio* band-structure calculations, especially in the past decade, allows to obtain a reliable microscopical description of the experimentally investigated MO effects for various transition metal compounds (see, e.g., the review in Ref. 8 and references therein). However, all these calculations were carried out so far for ordered crystalline solids. On the other hand, the wide and important class of disordered alloys could not be treated until now because of technical difficulties.

The description of the magneto-optical properties of a disordered system on a microscopic level and understanding the effect of chemical order and crystal symmetry on these properties are of great importance. The most powerful approach to calculate the electronic structure of randomly disordered solids is without doubt the coherent potential approximation (CPA) in connection with the Korringa-Korr-Rostoker method of band-structure calculation (KKR-CPA).[9] Within this formalism a quantitative description of such phenomena as magnetoresistance[10] and x-ray dichroism[11] was obtained. In both cases the central quantity to be calculated is the optical conductivity tensor.[12] Unfortunately, the techniques to deal with that quantity for finite frequencies in the optical regime of light have not yet been developed. For this reason calculations of optical and magneto-optical spectra of disordered alloys could not be done so far.

To make, nevertheless, some progress in the description of the MO properties of disordered alloys, we performed investigations of the influence of composition and local environment on the MOKE spectra of Co-Pd alloys using supercell calculations. To test this theoretical approach, we have also measured the MOKE spectra for a set of Co_xPd_{1-x} polycrystalline alloy films.

The paper is organized as follows. In Sec. II A the theoretical approach as well as structure models are described. A short description of experimental techniques applied is given in Sec. II B. The results of our theoretical treatment are shown and discussed in Sec. III. Conclusions are presented in Sec. IV.

II. THEORETICAL AND EXPERIMENTAL APPROACH

A. Theory

To investigate the influence of composition and local environment on the MO properties of disordered alloys, *ab initio* band-structure calculations for *ordered* Co–Pd alloys (Co_1Pd_{15}, Co_1Pd_{11}, Co_1Pd_7, Co_2Pd_{10}, Co_4Pd_{12}, Co_3Pd_9,

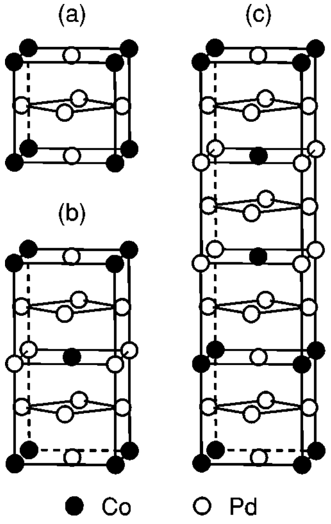

FIG. 1. Unit cells used for (a) Co_1Pd_3 ($L1_2$), (b) Co_2Pd_6 ($D0_{22}$), and (c) Co_4Pd_{12} ($D0_{23}$) alloys.

Co_2Pd_6, Co_1Pd_3, and Co_1Pd_1) have been performed. This sequence corresponds to a variation of the Co content in the range of 6–50 %. For Co_1Pd_1 and Co_1Pd_3 alloys we assume AuCu ($L1_0$) and $AuCu_3$ ($L1_2$) structure, respectively. Supercells for all other alloys, except for Co_1Pd_{11}, consist of two or more adjacent fcc unit cells with Co and Pd atoms distributed over nonequivalent atomic positions according to the specific alloy composition. As an example, the unit cells used for Co_2Pd_6 and Co_4Pd_{12} tetragonal crystals of $D0_{22}$ and $D0_{23}$ structures, respectively, are shown in Fig. 1. For the Co_1Pd_7 and Co_1Pd_{15} alloys both cubic and tetragonal supercells were constructed, whereas for the Co_1Pd_{11} alloy a hexagonal unit cell was used. The lattice constant for all the alloys has been chosen to correspond to the experimental data.[13]

All calculations were carried out using the spin-polarized relativistic linear muffin-tin orbital method[14] within the framework of spin-density-functional theory. For the exchange and correlation potential the parametrization of von Barth and Hedin[15] was used. On this basis the dispersion of the dielectric tensor $\varepsilon_{\alpha\beta}(\omega)=\delta_{\alpha\beta}+i(4\pi/\omega)\sigma_{\alpha\beta}$, where $\sigma_{\alpha\beta}$ is the optical conductivity tensor, has been calculated with both intraband and interband contributions taken into account in the way described in Ref. 16.

Magneto-optical effects are determined by the dielectric tensor components $\varepsilon_{\alpha\beta}$. This tensor can be represented as a sum $\varepsilon_{\alpha\beta}=\varepsilon^s_{\alpha\beta}+\varepsilon^a_{\alpha\beta}$ of two parts that are symmetric (optical) and antisymmetric (magneto-optical) with respect to the magnetization direction. That is, the relations $\varepsilon^s_{\alpha\beta}(\mathbf{M})=\varepsilon^s_{\alpha\beta}(-\mathbf{M})$ and $\varepsilon^a_{\alpha\beta}(\mathbf{M})=-\varepsilon^a_{\alpha\beta}(-\mathbf{M})$ hold, where $\mathbf{M}$ denotes the magnetization vector. The antisymmetric part $\varepsilon^a_{\alpha\beta}$ can be expressed as $\varepsilon^a_{\alpha\beta}=ie_{\alpha\beta\gamma}g_\gamma$, where $e_{\alpha\beta\gamma}$ is the antisymmetric pseudotensor and g_γ is a component of the gyration vector $\mathbf{g}$.[17]

For most materials the relation $|\mathbf{g}|\ll\det||\varepsilon^s_{\alpha\beta}||$ holds and expressions for all nonreciprocal magneto-optical effects, measured usually as a difference quantity with respect to a reversal of the magnetization, can be decomposed into the product of the complex number $\mathbf{g}\hat{\mathbf{g}}$ and some function depending on the optical part of the dielectric tensor $\varepsilon^s_{\alpha\beta}$ as well as the light propagation and magnetization directions (see, e.g., Ref. 18).

It should be noted that, although the optical part of the dielectric tensor may play a crucial role in the MO spectra formation as, for example, in CeSb,[19] or may exhibit a noticeable anisotropy,[20] this is not the case for the Co-Pd compounds investigated here. We found that in the energy range $0.5 < h\nu < 6$ eV the calculated components $\varepsilon^s_{\alpha\beta}$ are smooth and exhibit only a very weak dependence on both the chemical ordering and the alloy composition. Moreover, for the uniaxial model structures the optical anisotropy defined as $\delta_A=(\varepsilon_\perp-\varepsilon_\parallel)/\epsilon$ does not exceed 5%. Here we denoted $\epsilon=(2\varepsilon^s_\perp+\varepsilon^s_\parallel)/3$, where $\varepsilon^s_\perp$ and $\varepsilon^s_\parallel$ stand for the main optical dielectric tensor components corresponding to the light polarization perpendicular and parallel to the n-fold symmetry axis (c) of the crystal. Thus, all features of the MO spectra of the Co-Pd compounds are determined by the $g(\omega)$ function.

As mentioned above, our calculations involved both cubic and uniaxial models. For the crystal with cubic symmetry the dependence of g on the magnetization direction $\hat{\mathbf{M}}$ is negligible,[21] whereas for uniaxial crystals it can be noticeable. Uspenskii, Kulatov, and Halilov[22] have shown that, up to the second order in spin-orbit coupling strength, this dependence can be expressed in terms of the gyration vectors $g_\parallel$ and $g_\perp$ calculated for the cases of $\hat{\mathbf{M}}\parallel c$ and $\hat{\mathbf{M}}\perp c$, respectively:

$$\mathbf{g}=\hat{\mathbf{M}}(g_\parallel\cos^2\theta+g_\perp\sin^2\theta)+\hat{\mathbf{n}}(g_\parallel-g_\perp)\sin\theta\cos\theta, \qquad (1)$$

where $\hat{\mathbf{n}}$ stands for the unit vector, orthogonal to $\hat{\mathbf{M}}$ and lying in the same plane as $\hat{\mathbf{M}}$ and $\mathbf{c}$, and θ is the angle between the magnetization vector $\mathbf{M}$ and n-fold symmetry axis c.

B. Experiment

To allow for a direct comparison of our theoretical results with corresponding experimental data, Co_xPd_{1-x} alloy films were prepared for $x=0.13$, 0.22, 0.31, and 0.44 by the electrodeposition method.[23] This preparation technique resulted in polycrystalline samples with random orientation of crystallites.

The polar Kerr rotation θ_K and ellipticity η_K spectra were measured using a MOKE spectrometer based on the polarization modulation technique with a piezobirefringent modulator. Besides high sensitivity approaching 10^{-4} deg, 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 Ref. 16. All the spectra were measured at room temperature in a saturating external field up to 1.8 T within the photon energy range $0.8 < h\nu < 5.5$ eV.

III. RESULTS AND DISCUSSION

In Fig. 2 we present the calculated $\mathrm{Im}\,\omega^2 g_\parallel(\omega)$ and $\mathrm{Im}\,\omega^2 g_\perp(\omega)$ spectra for a selected set of uniaxial crystals. One can see that for the Co_1Pd_1 and Co_2Pd_6 the orientation dependence of the gyration vector is rather strong and exhibits itself in changes of the relative magnitude and energy

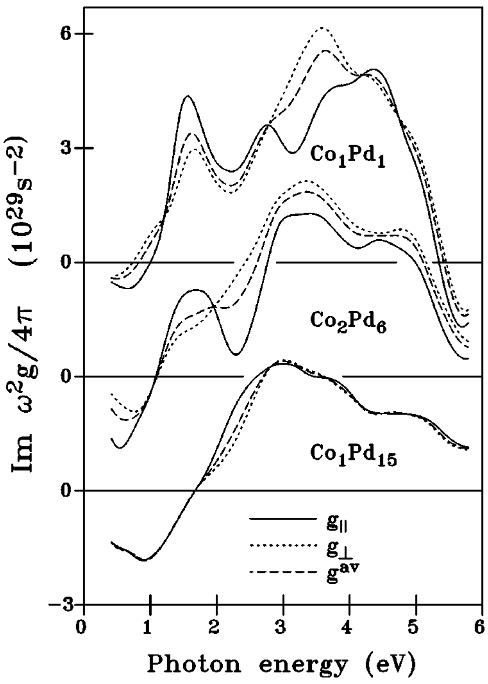

FIG. 2. Orientation dependence of the $\mathrm{Im}\omega^2 g(\omega)$ spectra for the Co_1Pd_1, Co_2Pd_6, and Co_1Pd_{15}. All spectra were convoluted with a Lorentzian of width 0.5 eV.

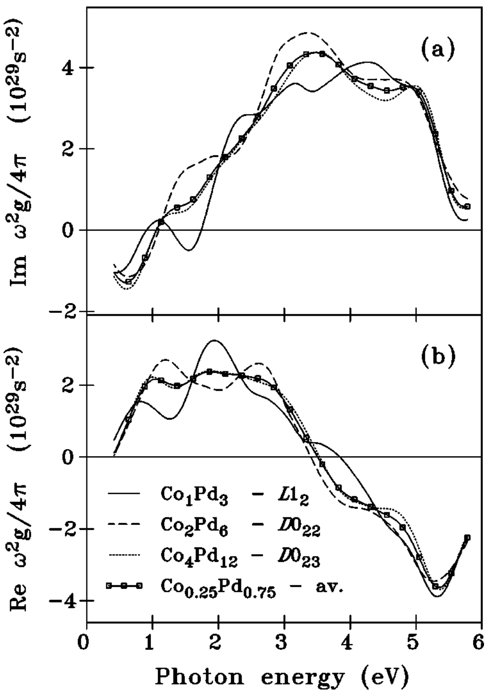

FIG. 3. Effect of the chemical order on the calculated $\omega^2 g(\omega)$ spectra of $Co_{0.25}Pd_{0.75}$ alloy.

location of both ir and uv maxima. The feature at 2.5 eV in the $g_{\|}(\omega)$ spectra disappears completely for $\mathbf{M}\perp c$. For the more diluted tetragonal Co_1Pd_7 alloy the orientation dependence of the $g(\omega)$ spectra was found to be relatively weak and practically disappears for the Co_1Pd_{15} (see Fig. 2). The same tendencies were found also for the $\mathrm{Re}\omega^2 g(\omega)$ spectra.

To take into account the effects of random orientation of microcrystals in the alloys studied, we have to average Eq. (1) over different relative orientations of the crystal axes and magnetization direction, which gives

$$g^{\mathrm{av}}(\omega)=[2g_{\perp}(\omega)+g_{\|}(\omega)]/3. \tag{2}$$

The main effect of the averaging (see Fig. 2) on the Co_1Pd_1 spectrum is a change of the band shape in the uv range as compared to the spectrum calculated with the magnetization directed along the c axis. The position of the main uv maximum shifts to lower energy and in Co_2Pd_6 the band becomes broader. In both alloys the structure at ~1.5 eV becomes less pronounced.

To study the dependence of the MO spectra on chemical ordering, let us consider the results for alloys with 1:3 stoichiometry. The calculations for the cubic $L1_2$ phase together with those for the tetragonal $D0_{22}$, $D0_{23}$, Co_3Pd_9, and second kind of Co_4Pd_{12} supercells have been performed. Tetragonal supercells Co_3Pd_9 and Co_4Pd_{12} consisting of one and two more central planes, respectively, as compared to the $D0_{22}$ structure (see Fig. 1), were used. All the alloys have the same nominal composition of $Co_{0.25}Pd_{0.75}$, but differ in symmetry and local environment of Co and Pd sites. While all these structures have identical first-neighbor coordination (each Co has 12Pd neighbors and each Pd has 8Pd +4Co neighbors), there are differences in the second atomic shell (see Fig. 1). For the $L1_2$, $D0_{22}$, and $D0_{23}$ cells each Co has 6Co, 4Co+2Pd and 5Co+1Pd second neighbors with local site symmetry O_h, D_{4h}, and C_{4v}, respectively. The atomic coordination around Pd in the second shell consists of 6Pd for $L1_2$ structure, while there are two inequivalent Pd sites for $D0_{22}$ (with 6Pd and 2Co+4Pd) and for $D0_{23}$ (with 6Pd and 1Co+5Pd). These three structures will be taken as basic because the other structures studied, like Co_3Pd_9 and Co_4Pd_{12}, are composed of various numbers of the above described inequivalent Co and Pd sites.

In Fig. 3 the spectra of the imaginary (a) and real (b) parts of $\omega^2 g(\omega)$ are shown. For Co_2Pd_6 ($D0_{22}$) and Co_4Pd_{12} ($D0_{23}$) alloys $\omega^2 g^{\mathrm{av}}(\omega)$ is presented. As a result of the change of the atomic arrangement and accompanied lowering of the local site symmetry along the sequence $L1_2$ – $D0_{22}$ – $D0_{23}$, the spectra become less structured and the peaks broaden, as can be seen, for example, comparing the $\mathrm{Re}\omega^2 g(\omega)$ spectra in the spectral range up to 3 eV. This effect apparently arises from the fact that lowering of the local symmetry of atomic sites generally leads to the appearance of an increasing number of nonequivalent interband transitions, with their contributions to the MO spectra spread out over wider energy range. It was found that the resulting $\omega^2 g^{\mathrm{av}}(\omega)$ spectra calculated for Co_3Pd_9 and Co_4Pd_{12} alloys can be well reproduced by averaging the basic $L1_2$, $D0_{22}$, and $D0_{23}$ spectra, weighted by the corresponding numbers of the equivalent sites in these structures.

To estimate the $\omega^2 g^{\mathrm{av}}(\omega)$ spectrum of disordered $Co_{0.25}Pd_{0.75}$ alloy it is necessary to assume a procedure of averaging the spectra over possible atomic arrangements. Here the simplest assumption was made that the basic cells appear with equal probabilities and the contribution of each cell to the spectrum is proportional to its number of atomic sites. On this basis the averaged spectrum was obtained as the weighted average of the $L1_2$, $D0_{22}$, and $D0_{23}$ spectra with the weights of 1, 2, and 4, respectively. As a result of the configuration averaging, the spectrum becomes less structured and is closest to the spectrum of $D0_{23}$ structure (see Fig. 3). It can be concluded that for the alloys studied the local atomic arrangement and sites symmetry controls the MO properties.

To discuss the composition dependence of MO properties of the alloys under consideration, we focus here on the polar Kerr rotation spectra, because they are the quantities measured directly in the experiment. If the optical anisotropy δ_A is small, the complex Kerr rotation angle, neglecting the second-order quantities (δ_A^2, $\delta_A g$, and g^2), has the form

$$\phi_K=\vartheta_K+i\eta_K=ig/[\sqrt{\epsilon}(1-\epsilon)]. \tag{3}$$

Equation (3) is linear with respect to g and this allows us to use for the uniaxial systems g^{av} instead of g. To take into account finite lifetime effects the calculated spectra were convoluted with a Lorentzian of width 1.0 eV. Calculated polar Kerr rotation and ellipticity spectra of the Co-Pd alloys of different composition are shown, together with experimentally measured ones, in Fig. 4. To take into account the decrease of Curie temperature for diluted alloys, all experimental spectra have been scaled with the factor $M_{4.2}/M_{300}$, where $M_{4.2}$ and M_{300} are magnetization at 4.2 K and 300 K, respectively. Both experimental and theoretical data exhibit the same features. As the Co content diminishes, the magnitude of the rotation angle in the energy range 2.5–4.5 eV decreases. The shoulder at $\sim$1.5 eV, clearly seen in the ϑ_K spectrum of the $\mathrm{Co_1Pd_1}$ alloy, disappears completely in diluted alloys and the Kerr rotation changes sign below $\sim$2 eV. In the uv range, the peak centered at 4 eV in $\mathrm{Co_1Pd_1}$ transforms into the broad two-peak structure. Also, a shift of the Kerr ellipticity zero crossing to higher energy with decrease of Co content is observed, as well as a diminishing of the amplitude at $\sim$5 eV. However, the calculated peak positions of both Kerr rotation and ellipticity are slightly shifted to higher energies with respect to the experimental ones. The discrepancy is apparently due to a drawback of the LDA-based calculations that was lately extensively discussed in the literature.[16,24]

It is interesting to note that the calculated MOKE spectra of $\mathrm{Co_2Pd_{10}}$, $\mathrm{Co_1Pd_7}$, $\mathrm{Co_1Pd_{11}}$, and also $\mathrm{Co_1Pd_{15}}$ alloys are close to each other, although the Co content in the alloys decreases from 16.5% to as low value as 6%. From this one can conclude that the MOKE spectra of diluted Co-Pd alloys are determined to a great extent by the MO properties of spin-polarized Pd. This is in line with the relatively large average Pd magnetic moment obtained from the calculations (e.g., $0.24\mu_B$/atom for $\mathrm{Co_1Pd_{15}}$) that depends weakly on Co content in the Co-Pd alloys.

It should be noted that the theoretical spectra shown in Fig. 4, except for the spectrum of $\mathrm{Co_{0.25}Pd_{0.75}}$, have been obtained for one specific atomic arrangement. In Fig. 4 the theoretical spectrum for $\mathrm{Co_{0.25}Pd_{0.75}}$, obtained as the weighted average of the spectra for $L1_2$, $D0_{22}$, and $D0_{23}$ structures according to the procedure described above, is presented. As a result of the averaging over different atomic configurations, the MOKE spectrum of $\mathrm{Co_{0.25}Pd_{0.75}}$ becomes less structured and the minimum in the polar Kerr rotation spectrum, centered at about 4.5 eV in the ordered $\mathrm{Co_1Pd_3}$-$L1_2$, transforms into broad structure as observed in the mea-

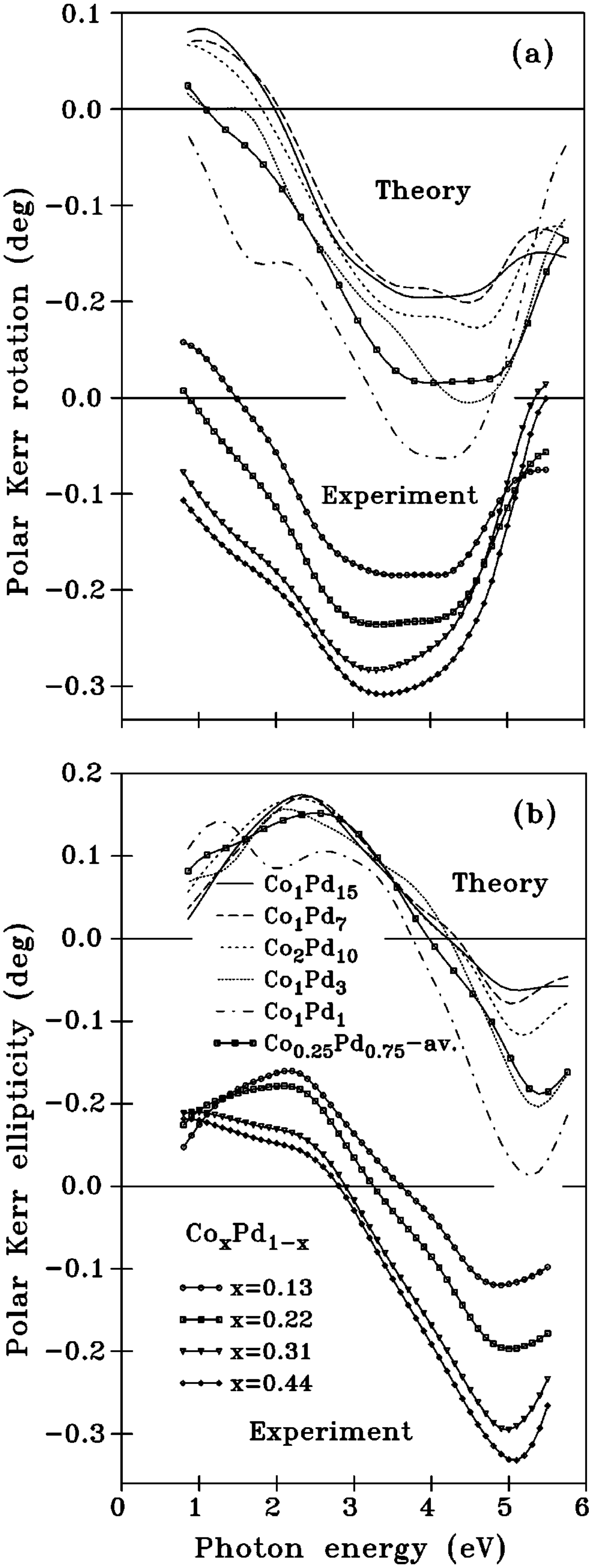

FIG. 4. Comparison of the calculated and experimental polar Kerr rotation (a) and ellipticity (b) spectra for the different composition of the Co-Pd alloys.

sured spectrum of the disordered $Co_{0.22}Pd_{0.78}$ alloy film. It can be concluded that the applied theoretical approach allows us to describe well the main features of the MOKE spectra for Co-Pd disordered alloys.

Of course, a more rigorous procedure of averaging over different atomic arrangements should be elaborated and it would be desirable to compare the results of supercell calculations with available KKR-CPA results. This is, however, beyond the scope of the present paper.

IV. SUMMARY AND CONCLUSIONS

In conclusion, a theoretical approach to calculate the optical and magneto-optical properties of disordered alloys has been proposed and successfully applied to the case of Co-Pd alloys. The optical part of the dielectric tensor of these compounds depends very weakly on both the alloy composition and chemical ordering and does not show noticeable anisotropy for the uniaxial crystals. Moreover, its frequency dependence is structureless and all features in magneto-optical spectra are determined by the gyrotropic part of the dielectric tensor, which exhibits a considerable orientation anisotropy increasing with the Co content. The MOKE calculations in which the effects of random orientation of microcrystals and/or chemical disorder are taken into account reproduce well the experimental spectra and the dependence of the spectra on the composition.

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