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Experimental and *ab initio* theoretical study of optical and magneto-optical properties of Co/Cu multilayers

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Abstract. The polar Kerr rotation and ellipticity spectra were measured in the spectral range 0.8–5.5 eV for a set of Co/Cu multilayered structures (MLS). The magneto-optical (MO) response of Co/Cu MLS being related to the σ_{xy}-part of the optical conductivity tensor was obtained from the Kerr spectra and tensor component σ_{xx}, determined by spectroscopic ellipsometry. The self-consistent spin-polarized fully relativistic linear muffin-tin orbital method within the local spin-density approximation was used to calculate the electronic structure, optical, and MO properties of some model Co/Cu MLS. Good agreement between the measured and calculated optical and MO spectra is observed. The role of the spin–orbit coupling and exchange splitting at the Co and Cu sites, and the hybridization effects are examined and discussed. The spin-polarized Cu interface states appear to give a weak contribution to the σ_{xy}- and Kerr spectra whereas the latter ones are strongly influenced by the Cu-dominated optical spectra structure. The results obtained demonstrate that the MO properties of real large-period multilayered structures can be quantitatively predicted from first-principles band-structure calculations.

1. Introduction

Multilayered structures (MLS), where ferromagnetic transition metals and noble metals are stacked alternately, have become of current interest, since they exhibit unusual properties and have the possibility of new applications. In these systems a number of extraordinary phenomena have been observed such as an oscillatory interlayer indirect magnetic coupling via nonmagnetic metal layers [1, 2], the induced spin polarization of a noble metal spacer layer [3, 4], and a giant-magnetoresistance effect [5, 6]. To clarify the origin of these phenomena a number of theoretical studies have been performed: using RKKY-like theories [7, 8], quantum confinement models [9, 10], and first-principles band calculations [11, 12]. Experiments probing the electronic structure near the Fermi level, like inverse and direct spin-polarized photoemission spectroscopy [13, 14], magnetic circular x-ray dichroism (MCXD) [3, 4], and magnetization-induced second-harmonic-generation [15] studies, provide information about the microscopic origin of these phenomena.

The magneto-optical Kerr effect (MOKE) spectroscopy is a valuable and powerful tool for the study of magnetic properties and electronic structures of magnetic materials. Due to

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their transition energies of up to ∼6 eV, magneto-optical (MO) techniques in general probe a convolution of initial and final states within the valence band of a material. Numerous spectroscopic MOKE discoveries, including the strong resonance effect due to the reduced optical constants in Fe, Co/noble-metal structures [16–21], MOKE enhancement in the uv region in Fe, Co/Pt multilayers [22–25], and quantum confinement effects in ultrathin Fe film [26] have been established.

In the pioneering study of Katayama *et al* (reference [16] and references therein) it was shown that the Kerr rotation in Fe/Cu bilayers and multilayers can be enhanced at the absorption edge energy of Cu. Since then, several investigations of MOKE spectra of Fe, Co/noble-metal bilayers and multilayers have been done [17–21]. The observed features of the MOKE spectra were interpreted as mainly related to the plasma edge in the noble metal and also to magneto-optically active transitions in the noble metal as a consequence of the spin polarization of the noble metal due to the proximity of magnetic layers at the interface.

The aim of the present paper is to clarify this point through a complementary investigation of the electronic structure of Co/Cu MLS by studies of their optical and magneto-optical spectra, both experimentally and theoretically. The measurements of both the optical and MO spectra in Co/Cu MLS are very attractive, because they offer an exceptional opportunity to clearly separate the contribution to the MOKE coming from the diagonal and off-diagonal components of the optical conductivity tensor (σ). This is important, because an appropriate discussion of electronic transitions underlying the MOKE generally requires the analysis of σ. It is well known that the complex Kerr rotation angle is composed of both diagonal and off-diagonal σ-components and that the absorptive part of σ is directly connected with the optical transition between the electronic states, their strength and their energy position.

In this work the complex MOKE spectra and optical properties—refractive index n and extinction coefficient k—have been measured over a wide spectral range for a set of the Co/Cu MLS structures. To clarify the main features in the formation of the MOKE spectra under consideration, *ab initio* theoretical calculations of the band structure and the optical conductivity spectra of some models of Co/Cu compounds have been performed in the framework of the self-consistent spin-polarized fully relativistic LMTO method within the local spin-density approximation. The results obtained show that magneto-optically active transitions coming from the spin-polarized Cu states arising as a result of the hybridization with Co states at the MLS interface give relatively insignificant contributions to the Co/Cu MLS MOKE spectra, whereas these spectra are strongly influenced by the Cu-dominated optical spectra structure.

The paper is organized as follows. In section 2 the experimental details are briefly described, and measured polar Kerr rotation and ellipticity spectra as well as spectra of the optical conductivity tensor components are presented and discussed. In section 3 the results of the band-structure calculations for some model Co/Cu multilayers are presented. The calculated optical and MO properties of the Co/Cu MLS are compared with the measured ones, and the effects of the hybridization, spin–orbit coupling and exchange splitting on the optical conductivity tensor are examined and discussed.

2. Experimental results

For the present study we have prepared the following set of Co/Cu MLS: 14.9 Å Co/10.1 Å Cu, 15.2 Å Co/15.2 Å Cu, 15.1 Å Co/21.7 Å Cu, and 22.3 Å Co/23.7 Å Cu, all with the same number of repetitions of the Co/Cu bilayer—being equal to 40—and with

Co as a top layer. The MLS have been deposited by the face-to-face dc sputtering system described in [27]. All of the samples were deposited on water-cooled glass substrates. The argon pressure during the deposition was about 60 mPa and the deposition rate about 0.5 Å s^{-1}. The chemical composition of the films was determined by x-ray fluorescence analysis with an EDX system from the peak intensities of the characteristic fluorescence radiation. The crystal structure was examined by low- and high-angle x-ray diffraction (XRD) using Co Kα radiation. The layered structure was confirmed by low-angle XRD. The modulation period was deduced from the position of the Bragg peaks. From the high-angle XRD the pronounced (111) texture was inferred.

The complex MOKE spectra were measured by means of the polarization modulation method using a piezobirefringent modulator. More details of the MOKE spectrometer system are given in [25]. The polar Kerr rotation and ellipticity spectra were measured in the photon energy range 0.8–5.5 eV under saturation conditions deduced from the measured hysteresis loops in magnetic fields up to 1.8 T. The angle of light incidence was 3° from the film surface normal. The refractive index n and extinction coefficient k were measured by the spectroscopic ellipsometry technique with the rotating analyser method [25]. The angle of incidence has been set at 67° which is near the average of the principal incidence angles of the metals in the spectral range used.

It is well known that the MOKE properties are determined by both diagonal and off-diagonal components of the 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 $\phi_K = \theta_K + \mathrm{i}\eta_K$ is given by [28, 29]

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

where $\sigma_{\alpha\beta}$ for a multilayer is interpreted as an effective optical conductivity of the modulated medium provided that the modulation period is much smaller than the wavelength used. The conductivity tensor $\sigma_{\alpha\beta}$ is related to the dielectric tensor $\varepsilon_{\alpha\beta} = \delta_{\alpha\beta} + \mathrm{i}(4\pi/\omega)\sigma_{\alpha\beta}$. The complex quantities are defined as $\sigma_{\alpha\beta} = \sigma^{(1)}_{\alpha\beta} + \mathrm{i}\sigma^{(2)}_{\alpha\beta}$, $\varepsilon_{\alpha\beta} = \varepsilon^{(1)}_{\alpha\beta} + \mathrm{i}\varepsilon^{(2)}_{\alpha\beta}$, and the diagonal element $\varepsilon_{\alpha\alpha} = (n + \mathrm{i}k)^2$. From the measured θ_K- and η_K-data, and the diagonal tensor component σ_{xx} determined from the ellipsometric data, the off-diagonal σ_{xy}-component can be evaluated.

In figure 1 the measured polar Kerr rotation θ_K- and ellipticity η_K-spectra of the Co/Cu MLS studied are shown. The measured spectra of θ_K and η_K for thick (∼2000 Å) fcc Co film, discussed in detail in the reference [25], are included for comparison.

The essential points of the experimental MOKE results can be summarized as follows.

(i) The θ_K-spectra of the Co/Cu MLS exhibit a two-peak structure and are strongly reduced compared to pure Co thick film.

(ii) The broad negative θ_K-peak appearing in the uv energy region has an amplitude which is approximately proportional to the MLS Co content. The energy position of this peak remains the same as for the Co thick film.

(iii) The θ_K-spectra in Co/Cu MLS exhibit a peak at $\hbar\omega \sim 2.1$ eV not observed in pure Co film. The amplitude and width of this peak is directly connected with the Cu sublayer thickness. For the MLS with the thinnest Cu sublayer the peak transforms into a broad shallow minimum located between ∼1.5 eV and ∼2.1 eV, the former energy corresponding to the θ_K-peak position of the Co thick film.

(iv) As the amount of Co in the MLS decreases the spectra generally scale down, while there is an increase in the prominence of the θ_K-peak located at the energy of ∼2.1 eV.

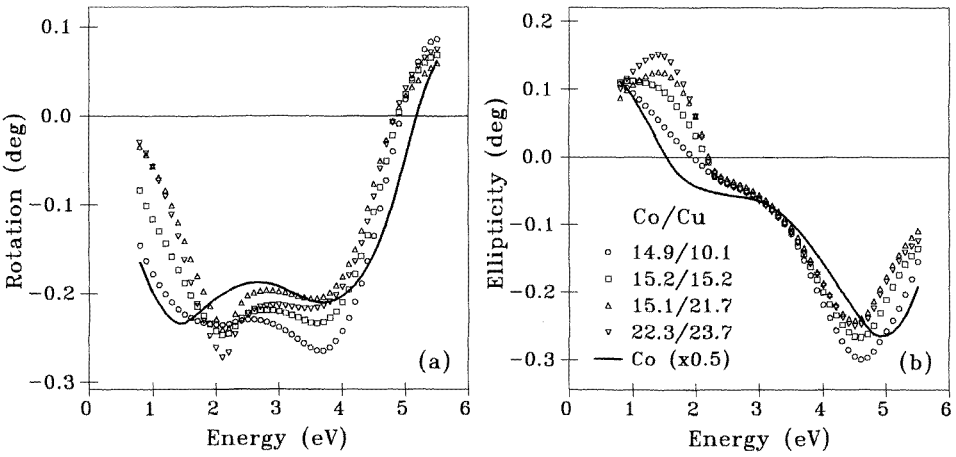

Figure 1. Experimental polar Kerr rotation (a) and ellipticity (b) spectra of Co/Cu MLS and thick fcc Co film (multiplied by a factor of 0.5). Co and Cu sublayer thicknesses are given in the key in units of Å.

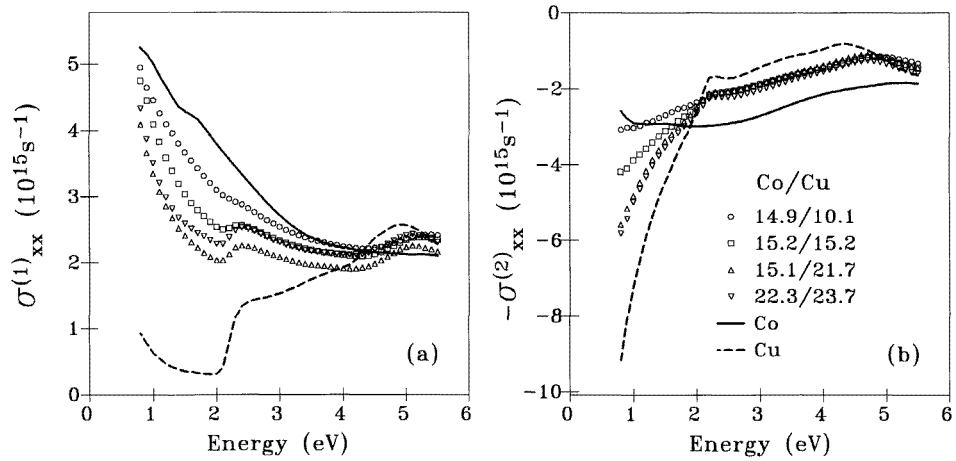

Figure 2. The absorptive (a) and dispersive (b) parts of the conductivity tensor of Co/Cu MLS and thick fcc Co and Cu films determined from the ellipsometric measurements.

(v) At the energies above ∼2.1 eV the Kerr ellipticity spectra are very similar. The uv peak position shifts to lower energy as compared to the Co thick film and its amplitude scales with the amount of Co. In the energy range 0.8–2.1 eV a strong dependence of the spectra on the MLS composition is seen.

The measured optical properties of the MLS as well as of thick fcc Co and Cu films, prepared by the same technique, are shown in the form of spectral dependencies of the absorptive ($\sigma^{(1)}_{xx}$) and dispersive ($\sigma^{(2)}_{xx}$) parts of the diagonal component of the optical conductivity tensor in figures 2(a) and 2(b), respectively. The well known prominent feature at the energy ∼2.1 eV in the Cu optical conductivity tensor, where there is the superposition

of the Drude-like intraband transitions and the interband transition edge, is clearly observed.

As can be seen from figure 2(a), the $\sigma_{xx}^{(1)}$-spectra for the MLS studied are located in between the spectra of pure Co and Cu films. These spectra have two features, a shoulder at the energy of ∼2.1 eV and a broad maximum at about 5 eV. The feature at ∼2.1 eV is related to the Cu plasma edge resonance absorption and its prominence increases with the Cu content. For all Co/Cu MLS studied the $\sigma_{xx}^{(1)}$-spectra have similar shape and their magnitude in the ir range scales with the amount of Co. This can easily be understood by taking into account the weak absorption of Cu metal in the energy region.

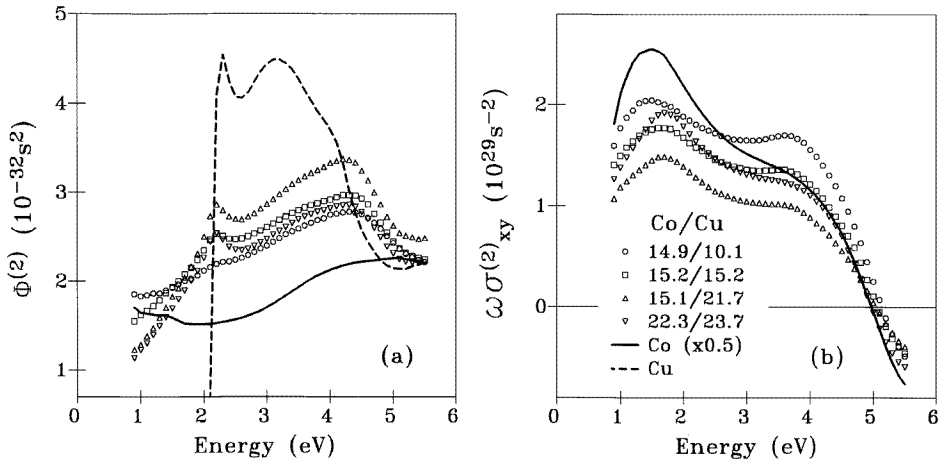

Figure 3. The contribution to the Kerr rotation spectra from the diagonal part of the conductivity tensor (see the text) (a) and the $\omega\sigma_{xy}^{(2)}$-spectra of Co/Cu MLS and the thick fcc Co film determined from the ellipsometric and the MOKE measurements (b). (For $\omega\sigma_{xy}^{(2)}$, Co is multiplied by a factor of 0.5.)

The MOKE depends on the diagonal part of the optical conductivity through the denominator of equation (1). To separate the contribution to the MOKE coming from the diagonal and off-diagonal components of the optical conductivity tensor the function $\Phi(\omega)$

$$\Phi(\omega) = \Phi^{(1)}(\omega) + \mathrm{i}\Phi^{(2)}(\omega) = \frac{1}{\omega\sigma_{xx}\sqrt{1 + \mathrm{i}(4\pi/\omega)\sigma_{xx}}} \tag{2}$$

has been evaluated from the optical measurements. It has been found that for the compounds under consideration the imaginary part of the function multiplied by $\omega\sigma_{xy}^{(2)}$ gives the main contribution to the polar Kerr rotation spectra. (Here and in the following $\sigma_{xy}^{(2)}$ stands for the absorptive part of the off-diagonal conductivity component.) For the Co/Cu MLS the $\Phi^{(2)}(\omega)$ function exhibits a two-peak structure (see figure 3(a)), a sharp peak at the same energy ∼2.1 eV as in the Kerr rotation spectra and a broader one at an energy around 4.5 eV. Thus, it can be clearly seen that diagonal part of the optical conductivity tensor significantly influences the shape of the MOKE spectra.

In figure 3(b) the $\omega\sigma_{xy}^{(2)}$-spectra of the Co/Cu MLS are shown. The shapes of the curves for all MLS are qualitatively similar to the shape of $\omega\sigma_{xy}^{(2)}$ determined for the Co thick film. Inspection of the curves shows that the magnitude of the $\omega\sigma_{xy}^{(2)}$-spectra in the uv spectral region scales with the amount of Co. This is, however, not true for the ir spectral region

and, in particular, the position of the low-energy peak shifts from that for the fcc Co film and depends on the MLS composition. The changes can be explained by a modification of the electronic states involved in the optical transitions due to existence of the Co/Cu interface.

In a preliminary study, the classical multi-reflection method (see, e.g., references [30, 31]) in the formulation as in [32] has been tentatively applied to the Co/Cu MLS investigated. The phenomenological approach reproduces reasonably well the measured spectra, similarly to the case of Fe/Cu multilayers [30, 33]. However, it is impossible in this approach to determine the microscopic origin of the interface influence on the MO properties. Also, the optical and magneto-optical properties of a sublayer used as input data for the procedure are usually taken from measurements of thick metal films. Moreover, the electronic structure of the metal and, consequently, its optical properties can be modified by MLS formation. Such an uncertainty in the input data affects the result of multi-reflection calculations and, for example, leads to erroneous determination of the interface thickness.

It can be concluded that the ir peak observed in the polar Kerr rotation spectra is mainly determined by the corresponding feature in the diagonal part of the conductivity tensor. The position of the θ_K-peak observed in the uv spectral region coincides with the position of the peak in the $\omega\sigma_{xy}^{(2)}$-spectra but its shape is to a great extent affected by the spectral form of the σ_{xx}, just like in the case of pure Co.

3. Theoretical results and discussion

To clarify the microscopic origin of the magneto-optical properties of the Co/Cu multilayers we have performed self-consistent local-spin-density calculations of the electronic structure of some model Co/Cu structures by means of the spin-polarized fully relativistic (SPR) LMTO method [34–36]. To improve the accuracy of the eigenenergies and wave functions the combined correction terms [37, 38] have been taken into account. The core charge densities were evaluated from the solutions of the Dirac equation for atoms. Exchange and correlation contributions to both the atomic and the crystalline potentials have been included through the density functional description in the LSDA of von Barth and Hedin [39]. The Brillouin zone integrations have been performed using the improved tetrahedron method [40].

Starting with eigenfunctions and eigenenergies obtained from the band-structure calculation it is possible to evaluate the frequency dependence of the optical conductivity tensor components [41, 42]. Details of the calculations in the framework of the relativistic LMTO method are given elsewhere [38, 25]. It should be noted that the combined correction term has to be included into expressions for evaluation of the optical transition matrix elements to obtain accurate results [38, 43, 44].

It is currently impossible to carry out *ab initio* calculations of the optical and MO properties of realistic MLS because these are not monocrystalline materials with a well-defined structure. Therefore, we investigate some idealized model structures representing the actual Co/Cu MLS.

Since the experimentally investigated MLS have pronounced fcc (111) texture, in our *ab initio* calculations we have constructed a number of nCo/mCu model MLS periodic along the [111] direction and consisting of n Co and m Cu closely packed atomic planes with an abc stacking sequence. All of the structures possess D_{3d}^3 symmetry. The lattice constant ($a = 3.574$ Å) has been chosen as an average of those of fcc Co and Cu metals. No attempt has been made to optimize the interlayer spacing, which was taken to be a constant corresponding to the ideal-case c/a ratio ($c/a = q\sqrt{2/3}$, where $q = n + m$ is the number

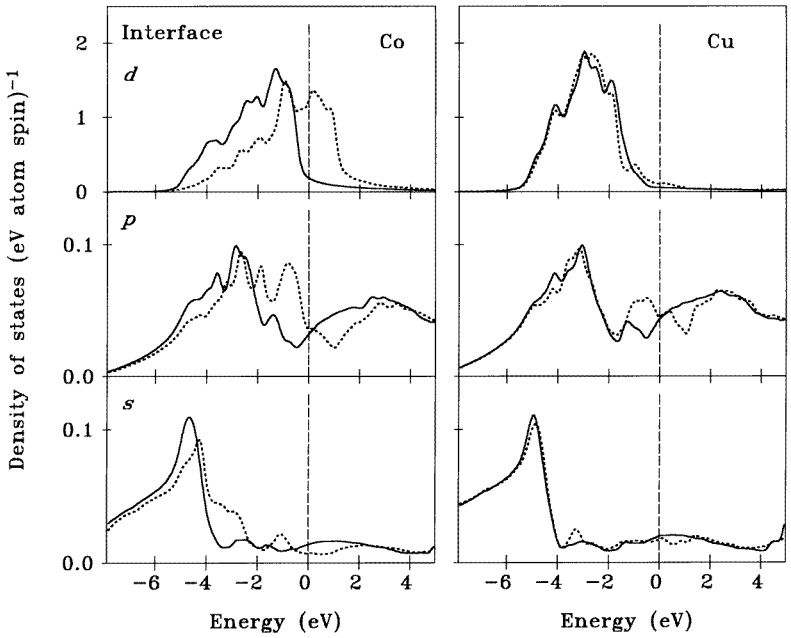

Figure 4. Spin- and l-projected densities of states (states eV^{-1}/(atom spin)) at Co and Cu sites at the interface in a 6Co/6Cu multilayer. Full and dotted lines correspond to majority- and minority-spin states, respectively. The Fermi level is denoted by vertical dashed lines.

of close-packed atomic layers with interlayer spacing equal to 2.063 Å).

The results of the band-structure calculations of the Co/Cu model MLS (density of states, local magnetic moments, etc) agree well with the available theoretical [3, 44, 45] and experimental [3, 4] data. The spin- and l-projected density of the electronic states for Co and Cu atoms in a representative 6Co/6Cu multilayer are shown in figure 4 for the sites at the interface and in figure 5 for the sites located in the middle of the appropriate sublayer. The densities of d states of bulk fcc Co and Cu are shown at the top of figure 5 for comparison. As could be anticipated, the density of Co and Cu d states of the interior atoms are similar to those of bulk metals. At the same time, the Co and Cu states at the interfacial sites are strongly hybridized. It is interesting to note that Cu states at the interface are considerably spin polarized due to the hybridization with Co states. It has been found that the resulting Cu sp and d spin magnetic moments are of opposite sign and almost compensate each other. Recently, the spin polarization of Cu states in Co/Cu MLS has been observed experimentally by means of MCXD [3, 4].

To show the main trends in the formation of the optical and magneto-optical spectra of the Co/Cu MLS and the role of interface electronic structure, the conductivity tensor of 2Co/4Cu, 3Co/3Cu, and 4Co/2Cu model multilayers was calculated. Keeping the fcc *abc* stacking sequence unperturbed, the calculations for 6Co/3Cu, 6Co/6Cu and 6Co/9Cu model multilayers, with the sublayer thicknesses closest to the MLS studied, were also performed for direct comparison with the experiment. In the following, the results for the first group with thin sublayers, together with the representative 6Co/6Cu example of the second group, will be discussed in detail to study the role of interface effects.

The calculated $\sigma_{xx}^{(1)}$-spectra of the selected multilayers are shown in figure 6(a). All

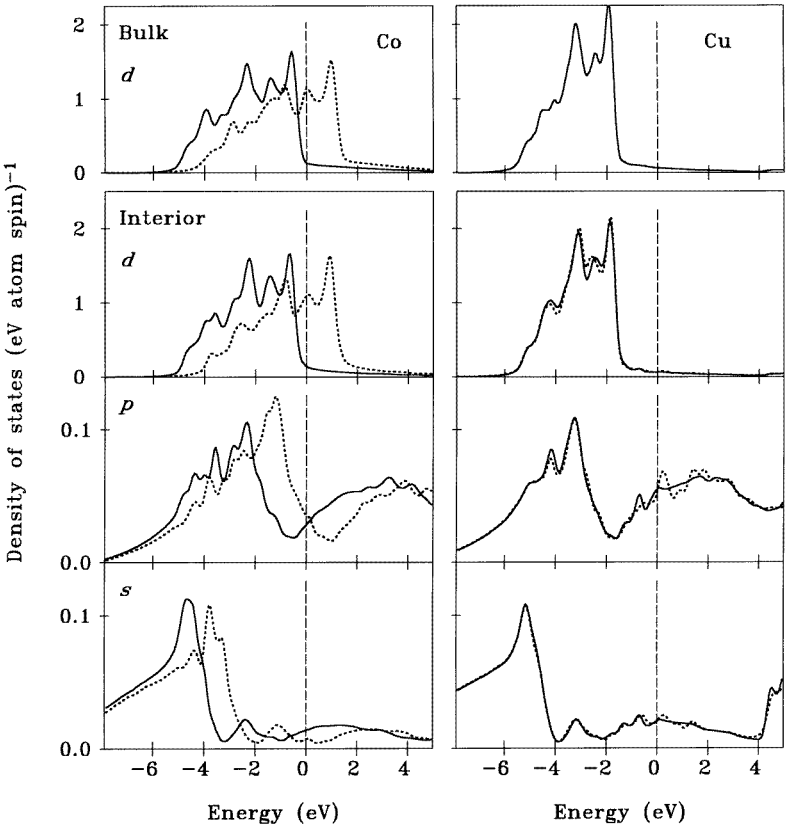

Figure 5. Spin- and l-projected densities of states (states eV^{-1}/atom spin)) at Co and Cu sites located in the middle of the corresponding sublayer in a 6Co/6Cu multilayer. The densities of d states of bulk fcc Co and Cu are also shown. Full and dotted lines correspond to majority- and minority-spin states, respectively. The Fermi level is denoted by vertical dashed lines.

of the spectra, except for that of Cu, were broadened with a Lorentzian of width 1.2 eV to simulate the finite-electron-lifetime effects. For the Cu spectrum a Lorentzian of width 0.4 eV was used. To take into account the intraband contribution, the phenomenological Drude term was also added to the diagonal components of σ with the plasma frequency taken from the calculation. As is seen in figure 6(a) all of the curves lie very close to each other at energies higher than $\sim$4 eV. In the visible and ir spectral regions a noticeable increase of the absorption with an enlargement of Co content can be seen. An interesting feature of these spectra is the minimum at the energy $\hbar\omega \sim 1.5$ eV, which corresponds to a minimum of interband absorption in pure Cu. From the comparison for $\sigma_{xx}^{(1)}$ of 3Co/3Cu and 6Co/6Cu it can be seen that this peculiarity is enhanced as the number of adjacent Cu layers increases. An analysis of the partial density of states shows that such a behaviour of the $\sigma_{xx}^{(1)}$ can be explained by the energy location of Co and Cu d states (see figure 4 and figure 5). A dominant interband contribution to the $\sigma_{xx}^{(1)}$-spectra at photon energy lower than $\sim$4 eV comes from transitions with Co and Cu d states involved. Like in pure Cu metal, the Cu d states in the MLS are located $\sim$1.5 eV below the Fermi level and, consequently, give no contribution to $\sigma_{xx}^{(1)}$ at lower photon energy. In contrast, optical transitions to/from

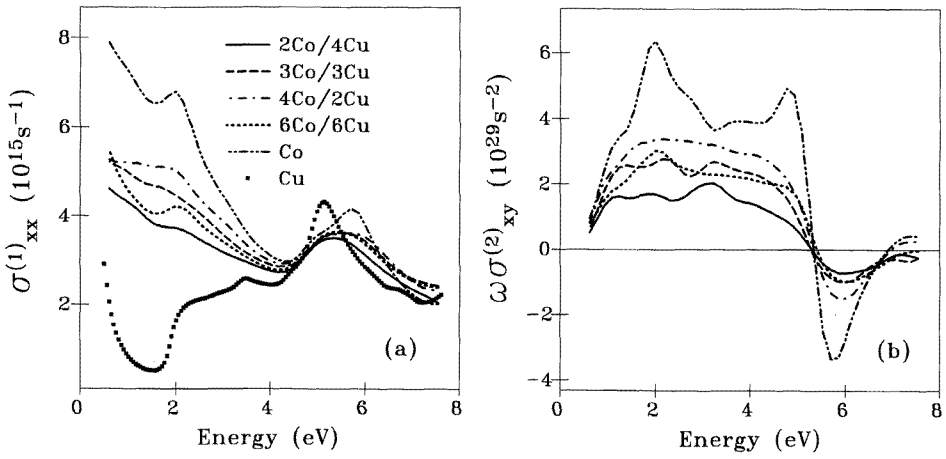

Figure 6. Calculated $\sigma^{(1)}_{xx}$-spectra (a) and (b) $\omega\sigma^{(2)}_{xy}$-spectra of the model nCo/mCu multilayers, where n and m are the numbers of atomic layers.

the Co d states are possible at arbitrary energy because minority-spin Co d states are only partially filled. As a result of a superposition of the absorption in Co and Cu sublayers, the $\sigma^{(1)}_{xx}$-spectra magnitude of the Co/Cu MLS increases with the Co content at $\hbar\omega \leqslant 4$ eV. The peak in $\sigma_{xx}(\omega)$ at $\hbar\omega \sim 5.5$ eV is mainly due to transitions from the s states at the bottom of the valence band to the p states above the Fermi level. The densities of Co and Cu s states are rather similar, except for the spin splitting of the former. As a result, the peak becomes broader with an increase of Co content in the MLS, its magnitude being almost unchanged.

Calculated $\omega\sigma^{(2)}_{xy}$-spectra are shown in figure 6(b). The magnitude of the spectra of the Co/Cu MLS is smaller than that of pure fcc Co, also shown in figure 6(b), and approximately scales with the Co content. As compared to the $\omega\sigma^{(2)}_{xy}$-curve of Co, the spectra of the MLS have less pronounced structure. The peak, centred at $\sim$2 eV in the Co spectrum, for the model multilayers studied is observed most distinctly for the 6Co/6Cu structure where the Co sublayer is sufficiently thick. It is remarkable that all of the spectra cross zero at the same energy of about 5 eV. Considering the $\omega\sigma^{(2)}_{xy}$-spectra leads one to a conclusion that the dominant contribution is provided by optical transitions with Co electronic states involved. The states are modified by the hybridization with Cu states at the interface, the hybridization effects being of greater importance for the MLS with smaller Co sublayer thickness.

Magneto-optical effects originate from a complicated interplay of the spin–orbit coupling and exchange splitting [46–48, 25]. As can be inferred from the perturbation theory (see, e.g., [46] and [49]), the $\sigma^{(2)}_{xy}$-spectrum can be expanded as a sum of contributions proportional to the spin–orbit coupling strength ξ_{tl} of the electronic states with an angular momentum l at a site t. From the results of test calculations with $\xi_{tl} = 0$ for all $l \neq 2$ it has been found that the MO properties of the Co/Cu multilayers are governed mainly by the spin–orbit coupling of d electrons. Then we have accomplished calculations with the only nonzero value of ξ_{tl} for either Co d or Cu d states. The results of the calculations for the 2Co/4Cu multilayer are shown in figure 7. It can be seen that the Cu contribution is, in general, significantly smaller than the contribution from the Co sites. There are two inequivalent Cu atoms in the 2Co/4Cu multilayer, one of them being located at the Co/Cu interface and

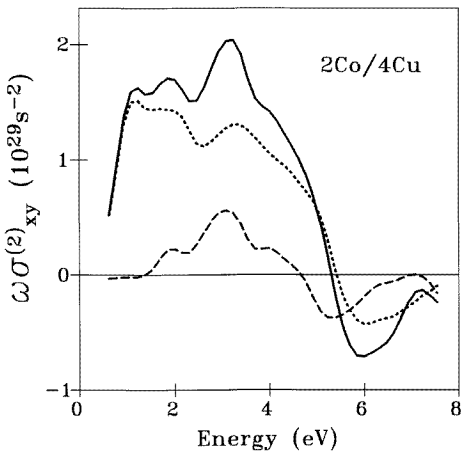

Figure 7. The influence of the Co and Cu spin–orbit coupling strength on the theoretical $\omega\sigma^{(2)}_{xy}$-spectrum of the 2Co/4Cu multilayer. The whole spectrum is shown by the solid line. Dotted and dashed lines denote the spectra calculated with nonzero spin–orbit coupling strength at Co and Cu sites, respectively.

the another one in the interior of the Cu slab. From the test calculations with ξ_{tl} set to zero for the interfacial Cu sites it has been found that the contribution from the interior Cu sites into the $\sigma^{(2)}_{xy}$-spectrum is negligible. The spin–orbit coupling strengths for Co and Cu d states are, however, of the same order of magnitude. The relatively small effect of the Cu sites on the off-diagonal conductivity component can be explained by the fact that the exchange splitting at the Cu sites is much smaller than at the Co ones (see figures 4 and 5). We have performed calculations assuming zero exchange splitting at all Cu sites and have found that the effect of the 'switching-off' of the exchange splitting on $\omega\sigma^{(2)}_{xy}$ for Co/Cu MLS is negligible. The results support the conclusion about the dominant role of the Co contribution as regards MO properties of the Co/Cu MLS.

To investigate the dependence of the $\sigma_{\alpha\beta}$ on the local environment we have performed calculations for 2Co/4Cu MLS with *abc* and *ab* closely packed plane stacking sequences. In both structures the number of Co and Cu atoms among both the first- and the second-nearest neighbours around each site is the same, but the local symmetry is different. The calculated $\omega\sigma^{(2)}_{xy}$-spectra are shown in figure 8. Although the overall shape of the curves is similar the fine structure is rather different. There are, also, differences in the corresponding $\sigma^{(1)}_{xx}$-curves shown in the inset in figure 8. It has been found that the joint densities of states calculated for the MLS with different kinds of stacking sequence are almost identical. Taking into account that the joint density of states is determined only by the band structure of a compound, one can reach a conclusion that the details of the short-range order affect the optical and MO spectra of the Co/Cu MLS mainly via the transition matrix elements.

Calculated polar Kerr rotation and ellipticity spectra for the Co/Cu MLS under discussion are shown in figure 9. The spectra reproduce well the dependence of the magnitude of the uv peak on the Co content observed experimentally for the Co/Cu MLS. However, the calculated spectra are more structured. A well-defined peak at $\sim$2 eV in the polar Kerr rotation, which is a characteristic feature of all the measured spectra, is present only in the theoretical spectrum of a 6Co/6Cu multilayer with sufficiently large Co and Cu sublayer thicknesses. Due to small sublayer thickness in the 2Co/4Cu and 4Co/2Cu MLS

the transitions to/from electronic states modified by the hybridization at the Co/Cu interfaces give relatively large contributions to the MO spectra. The results can, in principle, be used to estimate the effects of the interfacial disorder on the MO spectra. As the roughness at the interface increases the number of Co(Cu) atoms modified by the hybridization with Cu(Co) atoms also increases. This leads, in turn, to broadening of both the Cu absorption edge and the Co-induced peak in the ir range of the off-diagonal optical conductivity. The effects of the Co d–Cu d hybridization at the interface possibly intensified by the interface roughness is supposed to be responsible for the fact that a shallow peak is observed in the measured polar Kerr rotation spectra of 14.9 Å Co/10.1 Å Cu film in the energy range 1.2–2.2 eV.

It can be observed that, according to the experimental data, the low-energy peak in the $\omega\sigma^{(2)}_{xy}$-spectra of the Co film as well as of the Co/Cu MLS is located well below 2 eV. The

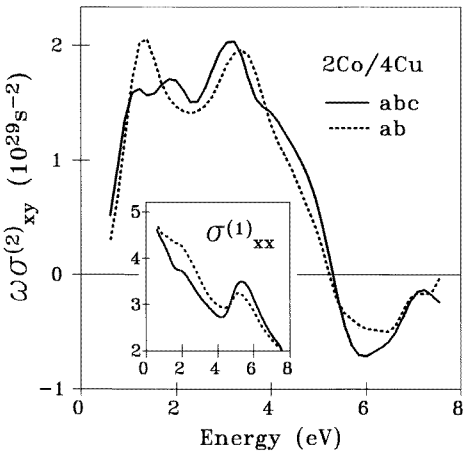

Figure 8. Calculated $\omega\sigma^{(2)}_{xy}$-spectra of 2Co/4Cu multilayers with *abc* and *ab* stacking sequences. In the inset the corresponding $\sigma^{(1)}_{xx}$-spectra are shown.

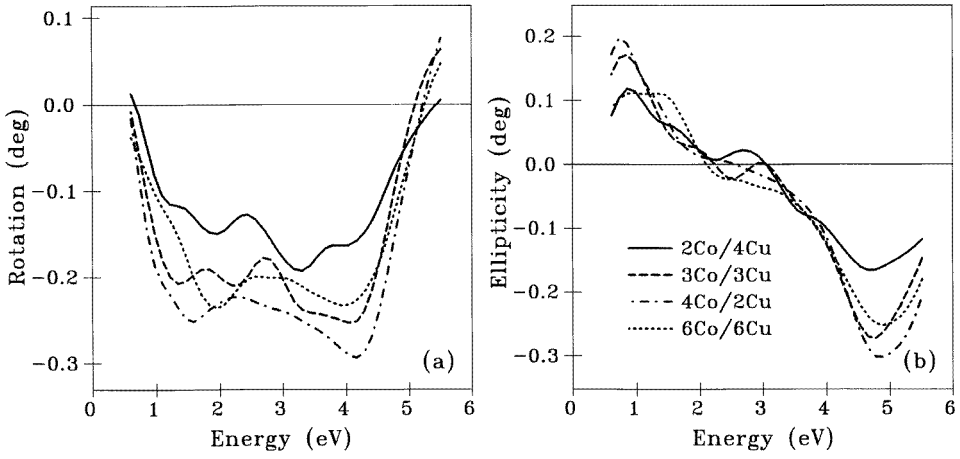

Figure 9. Calculated polar Kerr rotation (a) and ellipticity (b) spectra of the model Co/Cu multilayers.

feature in $\sigma_{xx}^{(1)}$, which is related to Cu plasma edge and manifests itself as a sharp peak in the Kerr rotation spectra, is centred at 2.1 eV. On the other hand, the calculations predict the peak in $\omega\sigma_{xy}^{(2)}$ for Co and the edge of the interband transitions in Cu to be at the same energy of ∼2 eV. The discrepancy may be due to the fact that the LSDA approximation is only moderately successful in the description of the energy band structure and MO properties of ferromagnetic 3d metals, especially Ni and also Co [25, 43, 47].

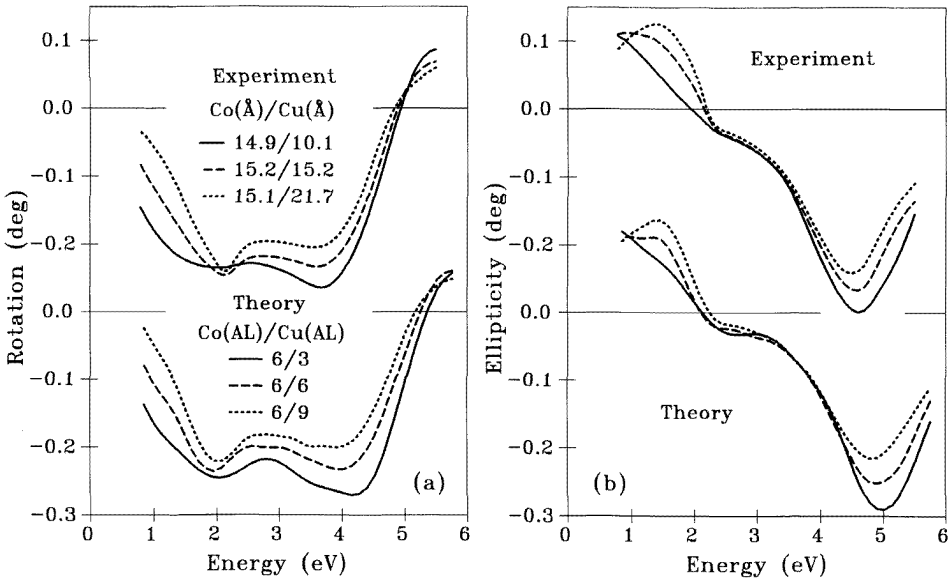

Figure 10. The comparison of the polar Kerr rotation (a) and ellipticity (b) spectra measured for Co/Cu MLS films with the spectra calculated for the model *n*Co/*m*Cu multilayers (the sublayer thicknesses are expressed in the key for the experimental films in (Å) and for the calculated structures in atomic layers (AL)).

Finally, an attempt at a direct comparison between selected groups of the experimental and calculated MO spectra has been undertaken. Such an objective in the case of *ab initio* calculations of the MLS can hardly be achievable because of a number of factors: (i) nonmonocrystalline structure of the sputtered multilayers; (ii) not exactly equal sublayer thicknesses in the real and model MLS; (iii) unknown actual Co/Cu interface structure; (iv) the above-mentioned problem of more general nature with the local density description of the electronic structure. Nevertheless, *ab initio* calculations of polar Kerr rotation and ellipticity spectra were performed for 6Co/3Cu, 6Co/6Cu, and 6Co/9Cu multilayers assuming model multilayer structures with ideal interfaces. The calculated spectra are compared with the experimental ones in figure 10. Despite the idealized multilayer structures, a good agreement between the theory and experiment is observed. Besides the overall similarity in the shape of the measured and calculated spectra, the calculations reproduce well the main trends in amplitudes of the spectra in different energy regions. In particular, the broadening of the peak at 1.2–2.2 eV in the case of the 14.9 Å Co/10.1 Å Cu MLS with the thinnest Cu sublayer is also observed in the theoretical Kerr rotation spectra of the 6Co/3Cu model multilayer. In view of the previous discussion this broadening can be interpreted as an enhanced manifestation of the hybridization effects at the Co/Cu interface. Obviously, a direct modelling of real interface roughness in the *ab initio* calculations would be desirable

to estimate the role of the interface in magnetic multilayers, and this is planned for future work [50].

4. Conclusions

The optical and magneto-optical properties of Co/Cu multilayers have been studied both experimentally and theoretically from first principles using LMTO band-structure calculations. The results show that, despite the spin polarization of Cu d states due to the hybridization with the Co states at the MLS interface, the dominant contribution to the off-diagonal part of the optical conductivity tensor of the Co/Cu MLS comes from the interband transitions with Co electronic states involved. The feature observed at $\hbar\omega \sim 2.1$ eV in the spectra for the optical conductivity tensor component $\sigma_{xx}^{(1)}$ is related to the edge of interband transitions from Cu d states in the Cu sublayers and is responsible for the peak at 2.1 eV in the polar Kerr rotation spectra of the MLS studied. There are some discrepancies between the experimental and theoretical results mainly in the description of the Co-related peaks of the optical and MO spectra of the Co/Cu MLS. Nevertheless, the *ab initio* calculations reproduce well the main trends and features observed in the spectra and provide an explanation of their microscopic origin. The results obtained imply that MO properties of multilayers with various compositions and structures can be quantitatively predicted from first-principles band-structure calculations. Such a possibility is important for basic research as well as applications.

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[50] It should be pointed out that LMTO computational time increases as a third power of the number of atoms in the unit cell. To construct a model of a multilayer with a rough interface, the planar dimensions of the unit cell must be at least doubled, dramatically increasing the computational power requirements.