Theoretical investigation of the orientation dependence of the magneto-optical Kerr effect in Co

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The first-principles spin-polarized, relativistic linear muffin-tin orbital method has been applied to calculate electronic and magneto-optical properties of Co in both fcc and hcp structures. In particular, the magneto-optical properties have been calculated for several magnetization directions in order to study their magnetization orientation dependences. A pronounced anisotropy in the polar Kerr effect is found for hcp Co while the magneto-optical properties of fcc Co are predicted to be insensitive to the magnetization direction. Moreover, this magneto-optical anisotropy in hcp Co is found to be correlated with the presence of a large anisotropy in the orbital magnetic moment and orbital hyperfine field. Changing the structure from fcc to hcp gives rise to even greater changes in the magneto-optical properties of Co. Calculated Kerr angles and ellipticities are in reasonable agreement with the latest experiment.

The magneto-optical Kerr effect (MOKE) is nowadays a widely used probe of magnetic properties of materials. Many recent exciting MOKE discoveries, such as quantum confinement effects[1] and oscillations of the Kerr rotation with magnetic layer thickness,[2] highlight its utility. MOKE not only provides unique information on the electronic structure of magnetic materials but also is technologically important. For example, because of its recent application to the read-out process in magneto-optical storage devices, an enormous amount of experimental work has been devoted to search for materials with high Kerr rotation angles.[3]

Despite the tremendous experimental research, few reports on calculation of MOKE by means of first-principles band-structure method have appeared in the literature (see Refs. 4 and 5 for reviews on former theoretical work). In fact, although the absorptive parts of the optical conductivity tensor that determines the Kerr rotation angle were calculated for transition metals and a few compounds (see Ref. 5 and references therein), the polar Kerr rotation angle itself has been reported for Fe, Co, and Ni only recently.[5] Consequently, the theoretical understanding of MOKE is still rather poor. For example, the appearance of pronounced peaks in the MOKE spectrum has been attributed to various physical origins such as interband transitions and spin-orbit coupling,[6] half-metallic ferromagnetism together with spin-orbit coupling,[7] and free charge carrier plasma resonances.[8] One of the reasons for this situation is undoubtedly that a theoretical treatment of the MOKE has to deal with spin-orbit coupling and spin polarization *simultaneously*. Another reason is perhaps that, to evaluate the Kerr rotation, one needs to obtain an accurate value of both the dispersive and absorptive parts of the off-diagonal elements of the conductivity tensor. These quantities are difficult to calculate because they depend crucially on small changes in the band structure caused by the spin-orbit coupling and exchange splitting.

In this paper, we present theoretical MOKE spectra of fcc and hcp Co and their magnetization orientation dependences. This work is partially motivated by the recent observation of the large anisotropy of MOKE in hcp Co.[9] We used the first-principles spin-polarized, relativistic linear muffin-tin orbital (SPRLMTO) method.[10] The absorptive parts of the conductivity tensor were calculated from the self-consistent relativistic band structures of fcc and hcp Co. The dispersive parts of conductivity tensors were obtained by using the Kramer-Kronig technique.[6] We also present theoretical magnetic moments and hyperfine fields and discuss possible correlations between them and the theoretical MOKE spectra.

MOKE refers to the change in the polarization of light that is reflected from the surface of a magnetic material. As a result, linearly polarized light is turned into elliptically polarized light with ellipticity ε_K and its major axis rotated by the Kerr angle θ_K relative to the polarization axis of the incident beam. Both quantities, which are usually combined into the complex Kerr angle $\phi_K = \theta_K + i\varepsilon_K$, can be related to the optical conductivity tensor σ of the material. For the polar geometry (i.e., the magnetization and the photon propagation direction along the surface normal specifying the z axis), the complex polar Kerr angle ϕ_K is given by[11]

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

For a solid with at least threefold rotational symmetry (e.g., hexagonal and cubic), $\sigma_o = \sigma_{xx} = \sigma_{yy}$ and for orthorhombic systems, $\sigma_o = \frac{1}{2}(\sigma_{xx} + \sigma_{yy})$. The various possibilities to relate the optical conductivity, and by this way MOKE spectra, to the electronic structure of a magnetic solid have been discussed by Erskine and Stern.[12] Here we simply used the relationship between the absorptive parts of the elements of the complex conductivity tensor σ and the corresponding quantum-mechanical photon absorption rates. Calculation of the absorptive parts of σ from the spin-polarized, relativistic band structure of a magnetic solid within the framework of the SPRLMTO method has been described by Ebert.[13] Nevertheless, we have made three improvements in this work. First, the underlying band structures of hcp and fcc Co were

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calculated self-consistently by using the SPRLMTO method. Second, the dispersive parts of σ have also been calculated, from the corresponding absorptive parts by use of the Kramer-Kronig technique.[6] This enabled us to obtain the complex polar Kerr angles ϕ_K via Eq. (1). Our Kramer-Kronig transformation program has been checked by performing double Kramer-Kronig transformations. The absorptive parts of σ recovered by the double Kramer-Kronig transformations agree with those calculated from the band structure to within 2%. Finally, the relativistic photon absorption matrix elements $\langle f|\pi_\lambda|i\rangle = \langle f|mc\,\boldsymbol{\alpha}\cdot\mathbf{a}_\lambda|i\rangle$ were evaluated by taking advantage of the identity[14,15]

$$\langle f|mc\,\boldsymbol{\alpha}\cdot\mathbf{a}_\lambda|i\rangle = \frac{2mc^2}{2mc^2+\varepsilon_i+\varepsilon_f}\left\langle f\left| \mathbf{p}\cdot\mathbf{a}_\lambda + \frac{V}{c}\boldsymbol{\alpha}\cdot\mathbf{a}_\lambda \right.\right.$$
$$\left.\left. - \frac{B}{c}\,i\beta(\boldsymbol{\alpha}\times\mathbf{a}_\lambda)_z \right|i\right\rangle, \qquad (2)$$

where $\mathbf{a}_\lambda$ is the λ photon polarization unit vector, and ε_i and ε_f are the initial- and final-state energies, respectively. Other symbols have their usual meanings.[10,13] The expression on the right is reduced to the nonrelativistic transition matrix elements $\langle f|\mathbf{p}\cdot\mathbf{a}_\lambda|i\rangle$ in the limit of $c\to\infty$. It turned out that the right-hand-side expression gives rise to far more accurate MOKE matrix elements than the simpler expression of $\langle f|mc\,\boldsymbol{\alpha}\cdot\mathbf{a}_\lambda|i\rangle$, though the former is more tedious to implement. More details about this last new feature may be found in Ref. 15.

In the self-consistent electronic structure calculations, the experimental lattice parameters of $a=2.51$ Å and $c=4.07$ Å for hcp Co and $a=3.54$ Å for fcc Co were used. The magnetization $\mathbf{m}$ was assumed to be along the [001] ([0001]) direction (i.e., $\mathbf{m}\|[001]$ ([0001])). The basis functions were s, p, and d MTO's. The number of $\mathbf{k}$ points over the irreducible wedges (IW) of the Brillouin zone (BZ) used, is 720 (over $\frac{3}{48}$ BZ) for fcc and 364 (over $\frac{1}{24}$ BZ) for hcp. The final band structures for the calculation of the magneto-optical properties were generated with a larger basis of s, p, and d as well as f MTO's. The absorptive parts of the conductivity tensor of fcc Co with $\mathbf{m}\|[001]$ and $\mathbf{m}\|[110]$, and of hcp Co with $\mathbf{m}\|[0001]$ and $\mathbf{m}\|[11\bar{2}0]$ were calculated. When the magnetization in fcc (hcp) Co is rotated from [001] ([0001]) to [110] ([11$\bar{2}$0]), the symmetry is reduced to the orthorhombic one.[16] To minimize the numerical uncertainties in the calculation of the magnetization orientation dependences of σ, we used the same IW of the orthorhombic BZ for both magnetizations. The number of $\mathbf{k}$ points used is 4515 (over $\frac{6}{48}$ BZ) for fcc and 6591 (over $\frac{3}{24}$ BZ) for hcp. The calculated absorptive parts of σ were found to converge well within 2% with respect to the number of the $\mathbf{k}$ points over the IWBZ used. The calculated spin (orbital) magnetic moment, m_s (m_o), is 1.594 (0.073) μ_B/Co for fcc Co, 1.581 (0.074) μ_B/Co for hcp Co with $\mathbf{m}\|[0001]$, and 1.581 (0.070) μ_B/Co for $\mathbf{m}\|[11\bar{2}0]$. The calculated total (core) hyperfine field, B_{hf} (B_{hf}^c), is -206.9 (-179.0) kG for fcc Co, -217.3 (-182.0) kG for hcp Co with $\mathbf{m}\|[0001]$, and -223.0 (-182.0) kG for $\mathbf{m}\|[11\bar{2}0]$.

Calculated absorptive parts of the conductivity tensor of Co in both fcc ($\mathbf{m}\|[001]$) and hcp ($\mathbf{m}\|[0001]$, and $\mathbf{m}\|[11\bar{2}0]$) are shown in Fig. 1. These spectra have been
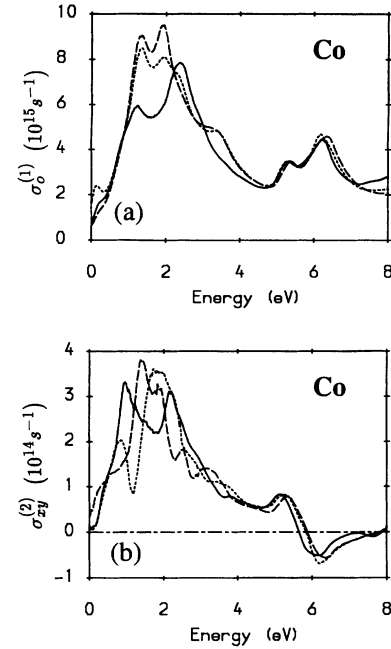

FIG. 1. The absorptive parts of the theoretical conductivity tensor of Co, with (a) diagonal element $\sigma_o^{(1)}$ and (b) off-diagonal element $\sigma_{xy}^{(2)}$. Solid curves are for fcc Co with [001] magnetization ($\sigma_o^{(1)}=\sigma_{xx}^{(1)}$), dashed curves for hcp Co with [0001] magnetization ($\sigma_o^{(1)}=\sigma_{xx}^{(1)}$), and dotted curves for hcp Co with [11$\bar{2}$0] magnetization [$\sigma_o^{(1)}=\frac{1}{2}(\sigma_{xx}^{(1)}+\sigma_{yy}^{(1)})$].

smoothed by convolution with a Lorentzian of 0.1 eV. The results for fcc Co with $\mathbf{m}\|[110]$ are nearly identical to those for $\mathbf{m}\|[001]$, and thus are not plotted in Fig. 1. Broadly speaking, all the calculated spectra look quite similar. For example, for the diagonal elements, they all have a pair of twin peaks between 5 and 7 eV, and a large broad double peak around 2 eV. For the off-diagonal elements, they all exhibit a rather sharp dip just above 6 eV and a broad plateau between 1 and 3 eV. Nevertheless, significant differences exist in σ for different structures and also for different magnetizations in the case of the hcp structure. In particular, the differences between fcc and hcp structures are rather pronounced. There are also considerable differences in $\sigma_{xy}^{(2)}$ of hcp Co for $\mathbf{m}\|[0001]$ and $\mathbf{m}\|[11\bar{2}0]$ between 1 and 4 eV. This clearly demonstrates that the conductivity tensor of Co and consequently, its MOKE spectra, depend not only on its crystalline structure but also on the magnetization direction in the case of the anisotropic hcp structure. In the rest of this paper, we will be mainly concerned with the magnetization orientation dependence.

From the absorptive parts of conductivity tensor, we obtained the dispersive parts by use of the Kramer-Kronig technique.[6] Both dispersive and absorptive parts were then broadened with a Lorentzian of $h/\tau=0.5$ eV to simulate the electron lifetime (τ) effects. To the diagonal components, a phenomenological Drude term of $\sigma_D/(1-i\omega\tau_D)$ was added in an attempt to include the intraband contributions. σ_D of 9×10^{15} s^{-1} and τ_D of 12×10^{-15} s were estimated from experiment.[17] Finally, the complex polar Kerr rotations calculated by using Eq. (1) are shown in Figs. 2 and 3, together
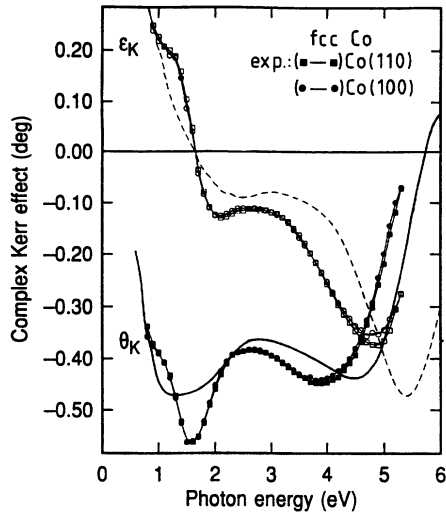
FIG. 2. Theoretical and experimental polar Kerr rotation θ_K and ellipticity ε_K spectra of fcc Co. Thereotical spectra are shown as smooth curves and are for the [001] magnetization. Experimental spectra (Ref. 9) are plotted as dotted lines.

with experimental results of Ref. 9.

Figures 2 and 3 suggest that the main features in the experimental MOKE spectra were reproduced by the present calculations. In particular, good quantitative agreement is found for the polar Kerr angles, θ_K. The strong anisotropy observed in the MOKE spectra of hcp Co (Ref. 9) were essentially reproduced as well (see Fig. 3). The outstanding discrepancies between experiment and theory are in the Kerr ellipticity ε_K spectra of hcp Co between 3 and 5 eV. However, we note that the calculated ε_K spectrum of hcp Co ($\mathbf{m}\|[0001]$) is closer to the experimental spectrum reported by Zeper et al.[18] Also, the local minimum around 4–5 eV in the theoretical θ_K spectra lie about 0.6–1.0 eV too high com-
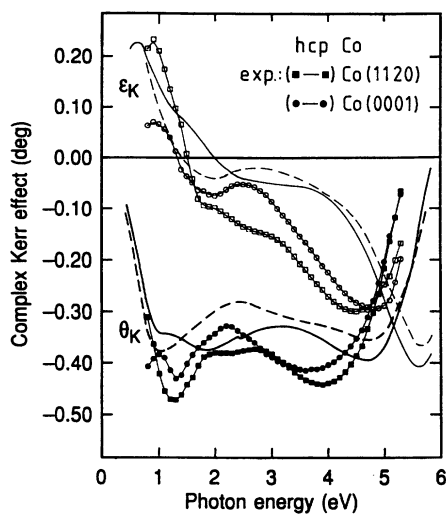

FIG. 3. Theoretical and experimental polar Kerr rotation θ_K and ellipticity ε_K spectra of hcp Co. Theoretical spectra are shown as smooth curves. Solid curves are for the $[11\bar{2}0]$ magnetization and dashed curves for the [0001] magnetization. Experimental spectra (Ref. 9) are plotted as dotted lines.

pared with the corresponding experimental spectra (Figs. 2 and 3). These discrepancies in energy positions may be due to the errors in the local density approximation used in this work. Also discernable is the discrepancy in the θ_K spectrum of hcp Co ($\mathbf{m}\|[11\bar{2}0]$) between 1 and 2 eV. This could be attributed to the neglecting of the intraband contribution to the off-diagonal element of σ. Intraband contributions affect mainly the MOKE spectra below 1–2 eV. Nevertheless, given the complexity and sensitivity of the results of the small band-structure effects, the agreement between the present calculations and experiments is considered to be satisfactory. Oppeneer et al.[5] reported only the θ_K spectrum for hcp Co ($\mathbf{m}\|[0001]$). Gasche[19] calculated the θ_K and ε_K spectra of Co for both the fcc ($\mathbf{m}\|[0001]$) and hcp ($\mathbf{m}\|[0001]$) structures. Unlike the present fully relativistic calculations, the spin-orbit coupling was treated as a (pseudo-) perturbation in these previous works. For hcp Co, the present results are similar to those of Oppeneer et al. but differ significantly from those of Gasche. For fcc Co, the present results also differ noticeably from those of Gasche, but are closer to the experimental spectra.

The results of the present calculations also show that the diagonal elements ($\sigma_{0001}, \sigma_{10\bar{1}0}, \sigma_{11\bar{2}0}$) of hcp Co are independent on the magnetization orientation, and that $\sigma_{10\bar{1}0} = \sigma_{11\bar{2}0}$. For $\mathbf{m}\|[0001]$, $\sigma_{xx} = \sigma_{11\bar{2}0} = \sigma_{yy}$ while for $\mathbf{m}\|[11\bar{2}0]$, $\sigma_{xx} = \sigma_{0001}$, $\sigma_{yy} = \sigma_{11\bar{2}0}$. Therefore, the differences in the $\sigma_o^{(1)}$ spectrum for $\mathbf{m}\|[0001]$ and for $\mathbf{m}\|[11\bar{2}0]$ result entirely from the differences between $\sigma_{11\bar{2}0}^{(1)}$ and $\sigma_{0001}^{(1)}$.

The absorptive part of the off-diagonal element ($\sigma_{xy}^{(2)}$) is directly related to the difference of the absorption rates for left- and right-circularly polarized light.[6] Specifically, $\sigma_{xy}^{(2)} = \frac{1}{2}(\sigma_l^{(1)} - \sigma_r^{(1)})$. According to the recent works on the circular magnetic x-ray dichroism (see, e.g., Refs. 20 and 21 and references therein), this identity suggests that $\sigma_{xy}^{(2)}$ may be related to the orbital magnetic moment (m_o). Indeed, we found that qualitatively, the larger the orbital magnetic moment, the larger the integral of $\sigma_{xy}^{(2)}$ (I^o). For example, in hcp Co, I^o (integrated over up to 8 eV) is 1.350 for $\mathbf{m}\|[0001]$ while I^o is 1.339 for $\mathbf{m}\|[11\bar{2}0]$. In accord with these results, m_o for $\mathbf{m}\|[0001]$ is $0.074\mu_B$/Co, being larger than $0.070\mu_B$/Co for $\mathbf{m}\|[11\bar{2}0]$. Interestingly, the orbital contribution to the hyperfine field, B_{hf}^o (Ref. 22) for $\mathbf{m}\|[0001]$ is 48.1 kG, also being larger than 42.4 kG for $\mathbf{m}\|[11\bar{2}0]$. These findings are not surprising since both m_o and σ_{xy} as well as B_{hf}^o, result from the spin-orbit coupling. Nevertheless, unlike the circular magnetic x-ray dichroism,[20,21] there is no obvious simple relationship between I^o and m_o (B_{hf}^o). For example, I^o for fcc Co is 1.368, being larger than 1.350 for hcp Co ($\mathbf{m}\|[0001]$). Yet, m_o (B_{hf}^o) of $0.073\mu_F$/Co (45.9 kG) for fcc Co, is smaller than that of hcp Co ($\mathbf{m}\|[0001]$). Furthermore, the Kerr angles for hcp Co with $\mathbf{m}\|[11\bar{2}0]$ between 2 and 5 eV are larger than those for $\mathbf{m}\|[0001]$. This appears not to be correlated with the smaller I^o and smaller m_o (B_{hf}^o) for hcp Co ($\mathbf{m}\|[11\bar{2}0]$), suggesting that MOKE is related to σ_{xy} in a nontrivial manner.[8] Note that the spin magnetic moment (m_s), and the contact hyperfine field ($B_{hf}^s = B_{hf} - B_{hf}^c - B_{hf}^o$),[22] is independent of the magnetization direction.

Clearly, the pronounced magneto-optical anisotropy in hcp Co is associated with the large magnetic anisotropy in the orbital magnetic moment ($\Delta m^o = 0.004 \mu_B$/Co) and orbital hyperfine field ($\Delta B_{hf}^o = 5.7$ kG). In fcc Co, the anisotropic orbital magnetic moment and orbital hyperfine field are nearly zero ($\Delta m^o = 0.0001 \mu_B$/Co and $\Delta B_{hf}^o = -0.1$ kG). Weller et al.[9] argued that the observed Kerr anisotropy in hcp Co is correlated with the large magnetocrystalline anisotropy in hcp Co. They noticed that the measured magnetocrystalline anisotropy energy of hcp Co is ten times larger than that of fcc Co. The results of the present calculations further corroborate the argument of Weller et al.[9]

In conclusion, magnetic properties and MOKE spectra of fcc and hcp Co have been calculated by using the first-principles spin-polarized, relativistic band-structure (SPRLMTO) method. The results of these calculations show that the large magneto-optical anisotropy found in hcp Co is strongly correlated with the occurrence of the large anisotropy orbital moment and orbital hyperfine field, which are more than 20 times smaller in fcc Co. Greater changes in the magneto-optical properties of Co are caused by a structural change from hcp to fcc. The present calculations reproduced the main features in the observed MOKE spectra.[9] This suggests that the first-principles SPRLMTO method for the MOKE calculations be applied to important but complex magnetic multilayers. There are no simple rules for fabricating good magneto-optical materials yet. The ongoing first-principles calculations for Fe and Co multilayers[15] will undoubtedly provide not only better understanding of the MOKE spectra in these systems but also guidance in search for large Kerr rotation materials.

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