Fully relativistic description of the magneto-optical properties of arbitrary layered systems

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A fully relativistic formalism is presented that allows us to define the frequency-dependent optical conductivity tensor for arbitrary layered systems in a layer-resolved way. This opens in particular the way to deal with the magneto-optical properties of magnetic surface layer systems and to calculate the corresponding magneto-optical Kerr spectra. The formalism, based on a fully relativistic description of response theory in arbitrary order, is described in some detail. For an implementation the very flexible spin polarized relativistic Korringa-Kohn-Rostoker method of band structure calculation has been used. Results of corresponding applications to the elemental ferromagnets bcc-Fe and fcc-Co treated as homogeneous layer systems are presented. [S0163-1829(99)04141-7]

I. INTRODUCTION

Most magneto-optical effects of magnetic materials known today have already been discovered.[1–3] In spite of this, intensive research on this subject has only been carried out during the last couple of decades. These activities have primarily been triggered by a strongly growing interest in applications in high density magneto-optical data storage based on the magneto-optical Kerr-effect (MOKE).[4,5] In order to achieve a high signal-to-noise ratio, this technology requires first of all materials with large Kerr rotation at optical frequencies. Unfortunately, most bulk compound materials with high Kerr rotations tend to loose this favorable property when they are prepared in the form of thin films.[6,7] A possible way to overcome these difficulties might be to extend material design to an additional degree of freedom, e.g., to use layer systems tailored to optimize their magneto-optical properties.[8,9] Investigations in this direction, especially on metallic layer systems consisting of magnetic and nonmagnetic films and substrates, have brought new exciting discoveries, also from the scientific point of view. The most important of these are quantum confinement effects,[10] oscillations of the Kerr rotation with variation of the thickness of the magnetic layer,[11,12] and an apparent relationship between MOKE and magnetic anisotropies.[13] Another important field for the application of MOKE is destruction-free domain pattern imaging near the surface using optical microscopy. Employed for the first time already in the beginning of the 1950's,[14] there have been some decisive amendments in recent years such as the use of interferometric techniques[15,16] leading to lateral resolutions below 100 nm.

It was already pointed out by Hulme[17] in 1932, that the MOKE is caused by the simultaneous presence of magnetic order and spin-orbit coupling. Because of this complex situation not much theoretical work has been done in the past in spite of the many experimental investigations in the field of magneto-optics. Only about 10 years ago the various technical problems in calculating the magneto-optical Kerr spectra in a parameter-free way were solved for the first time by Oppeneer *et al.*[18] Since then several other groups reported on comparable work (see for example Refs. 19–24), i.e., MOKE spectra can be calculated nowadays in a nearly routine way.

However, all theoretical approaches presented so far to deal with the MOKE are based on a calculation of the underlying electronic structure using a so-called linear band structure method working in $\vec{k}$ space. Accordingly, all of them are restricted in application to ordered bulk materials with three-dimensional translational invariance. As a consequence they do not supply an adequate platform to deal with the interesting magneto-optical properties of surface layered systems mentioned above.

The restriction of the conventional $\vec{k}$ space methods to deal with the optical conductivity only for ordered solids could recently be removed by Banhart.[25] This author extended the application of the Kubo-Greenwood-formalism for the residual resistivity of disordered alloys, as it has been worked out originally by Butler,[26] to finite frequencies. By this way he got access to the optical conductivity of paramagnetic ordered as well as disordered solids. This is achieved by a description of the underlying electronic structure by means of electronic Green's function using the Korringa-Kohn-Rostoker Green's function (KKR-GF) method of band structure calculation in combination with the coherent potential approximation (CPA) alloy theory.

Another important extension of Butler's approach is the concept of the layer-resolved dc conductivity. This was introduced by Butler, Zhang, Nicholson, and MacLaren[27] and by Weinberger, Levy, Banhart, Szunyogh, and Újfalussy[28] to deal with the giant magnetoresistance (GMR) effect in magnetic multilayer systems.

In the following a generalization of the concept to finite frequencies will be presented (this approach as well as first results had already been presented before[54,55]). In contrast to Banhart's approach, however, it gives access to the full complex optical conductivity tensor of arbitrary layered systems. This, in combination with a direct solution of the microscopic Maxwell equations (details on this will be published elsewhere[29]), allows one in particular to deal with the magneto-optical properties of surface layered systems in an adequate way.

In the next two sections our approach that is based on Kubo's linear response formalism will be presented in some detail. For its application use is also made of the KKR-GF

technique to describe the underlying electronic structure. Most important for the investigation of magneto-optical properties, this is done in a spin-polarized relativistic (SPR) way. Further features of the resulting SPR-KKR-GF-scheme relevant for the calculation of optical properties will be discussed below. To demonstrate the feasibility and implications of our approach, results for the elemental ferromagnets bcc-Fe and fcc-Co which have been treated as layered systems, will be presented.

II. THEORETICAL FRAMEWORK AND TECHNICAL DETAILS

A. Fully relativistic description of the nonlocal frequency-dependent optical conductivity

The interaction of an electronic system and external electromagnetic fields in minimal coupling is conventionally described in terms of the time-dependent perturbation Hamiltonian

$$X(t) = -\frac{1}{c} \int d^3 r \vec{j}(\vec{r}) \vec{A}(\vec{r},t),$$

$$\vec{j}(\vec{r}) = -ec\vec{\alpha}\delta(\vec{r}-\vec{r}'). \quad (1)$$

Here the external vector potential $\vec{A}$ which is considered to be adiabatically switched on and coupled to the electronic current density is represented by the operator $\vec{j}(\vec{r})$. To ensure that all possible sources of the MOKE are properly accounted for the following formalism is worked out in a fully relativistic way. Accordingly $\vec{j}(\vec{r})$ is represented by the relativistic velocity operator $c\vec{\alpha}$ with $\vec{\alpha}$ the vector of the standard Dirac matrices.[30]

Employing conventional time-dependent perturbation theory for the density matrix ρ of the system,[31] we find by iteration of the corresponding equation of motion the appropriate relation between $\vec{A}$ and the difference in expectation values of $\vec{j}(\vec{r})$ for the perturbed and the unperturbed system in time t:

$$\langle j_\alpha(\vec{r})\rangle(t) - \langle j_\alpha(\vec{r})\rangle_0$$

$$= \sum_{n=1}^{\infty} \left(\frac{i}{\hbar c}\right)^n \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \cdots$$

$$\times \int_{-\infty}^{t_{n-1}} dt_n \int \cdots \int d^3 r_1 \cdots d^3 r_n$$

$$\times \sum_{\beta_1,\cdots,\beta_n}^{\infty,\ldots,\infty} A_{\beta_1}(\vec{r}_1,t_1) \ldots A_{\beta_n}(\vec{r}_n,t_n)$$

$$\times \text{tr}\{\rho_0[\cdots[j_\alpha^D(\vec{r},t),j_{\beta_1}^D(\vec{r}_1,t_1)]_-,\ldots,j_{\beta_n}^D$$

$$\times (\vec{r}_n,t_n)]_-\}, \quad (2)$$

where 0 represents quantities of the unperturbed system and D denotes Dirac representation of operators. In the following, the quantity on the left hand side will be denoted as the "induced" current density.

Retaining only the linear term in $\vec{A}$, using Coulomb gauge for the representation of $\vec{A}$ and after Fourier transformation with respect to time we arrive at the Kubo formula for linear response of the induced current density $\vec{j}$ to the external electric field $\vec{E}$ with frequency ω:

$$j_\alpha(\vec{r},\omega) = \sum_\beta \int d^3 r' \sigma_{\alpha\beta}(\vec{r},\vec{r}',\omega)E_\beta(\vec{r}',\omega), \quad (3)$$

with α,β the Cartesian components of the involved vectors. The nonlocal optical conductivity $\sigma_{\alpha\beta}$ is defined in terms of the retarded current-current Green's function $G_{j_\alpha j_\beta}^+$ and the ground state $|\eta_0\rangle$ as follows:

$$\sigma_{\alpha\beta}(\vec{r},\vec{r}',\omega) = \frac{i}{(\omega+i0^+)\hbar} \int_{-\infty}^{\infty} dt' G_{j_\alpha j_\beta}^+$$

$$\times (\vec{r},\vec{r}',0,-t')e^{i\omega t'-0^+ t'}, \quad (4)$$

where the limiting value 0^+ stems from the adiabatic switching condition of $\vec{A}$. The quantity $G_{j_\alpha j_\beta}^+$ is given by

$$G_{j_\alpha j_\beta}^+(\vec{r},\vec{r}',t,t') = -i\Theta(t-t')$$

$$\times \langle \eta_0|[j_\alpha^D(\vec{r},t),j_\beta^D(\vec{r}',t')]_-|\eta_0\rangle. \quad (5)$$

Dynamical correlation effects, e.g., electron-hole interaction, will be neglected. On the other hand, inclusion of these effects can be achieved within time-dependent density functional theory. This would imply the solution of a Dyson equation in terms of the optical conductivity given by Eqs. (4) and (5) as the zeroth order approximation.[32] Because magneto-optical properties of metallic surface layer systems within the frequency range considered here are due to delocalised conduction electrons, we expect dynamical correlation effects to be of minor importance.

To evaluate $G_{j_\alpha j_\beta}^+$, we express j_α^D according to second quantization in terms of Fermion construction operators a_i and single particle orthonormal eigenstates $|i\rangle$ with energies E_i of the unperturbed Hamiltonian of the system:

$$j_\alpha^D(\vec{r},t) = \sum_{ij} \langle i|j_\alpha(\vec{r})|j\rangle a_i^+ a_j e^{(E_i-E_j)t/\hbar}. \quad (6)$$

Inserting this to Eq. (5) and using Eq. (4), we finally arrive at

$$\sigma_{\alpha\beta}(\vec{r},\vec{r}',\omega) = \frac{i}{\omega+i0^+}\left\{\sum_{ij} \frac{1}{\hbar\omega+E_i-E_j+i0^+}\langle i|j_\alpha(\vec{r})|j\rangle\right.$$

$$\times \langle j|j_\beta(\vec{r}')|i\rangle$$

$$-\frac{1}{\hbar\omega+E_j-E_i+i0^+}\langle i|j_\beta(\vec{r}')|j\rangle$$

$$\left.\times \langle j|j_\alpha(\vec{r})|i\rangle\right\} \Theta(E_F-E_i)\Theta(E_j-E_F), \quad (7)$$

with E_F the Fermi energy. To simulate finite lifetime effects and other broadening mechanisms due to finite temperature,

indirect transitions and limited experimental resolution, we replace the quantity 0^+ in the energy denominators in Eq. (7) by a lifetime parameter $\hbar/\tau$.

An analogous expression for the optical conductivity in a spatially averaged, nonrelativistic form has been given by Wang and Callaway.[33] However, there is a subtle difference: with the nonrelativistic formalism, the operator for the electrical current density is explicitly dependent on the vector potential $\vec{A}$, while in the relativistic formalism it is not. This originates from the fact that the Schrödinger Hamiltonian is of second order in space coordinates, while the Dirac Hamiltonian is only of first order; therefore, the so-called diamagnetic (Drude-like) contribution to the optical conductivity appearing explicitly in Eq. (5.1) in Ref. 33 is implicitly included in the relativistic form of the electric current density operator.

The formalism described so far could be applied straightforwardly to ordered solids by using a $\vec{k}$-space band structure method and inserting the resulting Bloch states $|\Psi_{n\vec{k}}\rangle$ and associated eigenvalues $E_{n\vec{k}}$ into Eq. (7). A much more general scheme is achieved, however, by representing the manifold of initial and final states by the trace of the corresponding retarded single-particle Green's function operator $G^+(E)$.

For real energies the relation

$$\sum_i |i\rangle\langle i| \delta(E - E_i) = -\frac{1}{\pi} \operatorname{Im} G^+(E) \qquad (8)$$

allows one to rewrite Eq. (7) in the following form:

$$
\sigma_{\alpha\beta}(\vec{r},\vec{r}',\omega) = \frac{i\hbar}{\pi^2} \int_{-\infty}^{E_F} dE \int_{E_F}^{\infty} dE'
$$

$$
\times \frac{\operatorname{tr}\{j_\alpha(\vec{r})\operatorname{Im} G^+(E') j_\beta(\vec{r}')\operatorname{Im} G^+(E)\}}{[E' - E - i(\hbar/\tau)][\hbar\omega + E - E' + i(\hbar/\tau)]}
$$

$$
+ \frac{\operatorname{tr}\{j_\beta(\vec{r}')\operatorname{Im} G^+(E') j_\alpha(\vec{r})\operatorname{Im} G^+(E)\}}{[E' - E + i(\hbar/\tau)][\hbar\omega + E' - E + i(\hbar/\tau)]},
$$

$$(9)$$

where explicit use has been made of the requirement that the optical conductivity is finite in the limit $\omega \to 0$. In the following, the ω-dependence of all quantities will be suppressed in intermediate steps for clarity.

B. Layer-resolved optical conductivity

Equation (9) is still of general validity, but of course tedious to be evaluated directly. Concerning its applications to optical problems, one is in general primarily interested in the reflection and possibly transmission of light with frequency ω and wave vector $\vec{k}$ falling onto a stack of atomic layers with arbitrary mutual distance. In addition, in-plane translational invariance will be assumed in the following; i.e., the atomic positions in all layers can be described by a common basis of lattice vectors $\{\vec{R}_\parallel^i\}$ thus defining a two-dimensional lattice. Because the optical conductivity is to be used later to solve the microscopic Maxwell equations, it is inconvenient to consider a ''layer'' to be set up by atomic Wigner Seitz

cells. In the following, we therefore define a point $\vec{r}$ to belong to a layer I if its distance to the lattice plane I is smaller than its distance to any other lattice plane. To exploit translational invariance, it is convenient to perform a spatial two-dimensional Fourier transform to the optical conductivity and the optical fields with respect to the layer plane. We define the two-dimensional Fourier transform $\vec{E}(\vec{k}_\parallel, \vec{r}_\perp)$ as

$$\vec{E}(\vec{k}_\parallel, \vec{r}_\perp) = \int d^2 r\, \vec{E}(\vec{r}_\parallel + \vec{r}_\perp) e^{-i\vec{k}_\parallel \vec{r}_\parallel} \qquad (10)$$

with $\vec{r} = \vec{r}_\parallel + \vec{r}_\perp$ denoting the separation of $\vec{r}$ into a part parallel ($\vec{r}_\parallel$) and perpendicular ($\vec{r}_\perp$) to the layer plane. Together with the fact that $\sigma_{\alpha\beta}$ possesses translational invariance with respect to any lattice vector $\vec{R}_\parallel^i$:

$$\sigma_{\alpha\beta}(\vec{r} + \vec{R}_\parallel^i, \vec{r}' + \vec{R}_\parallel^i) = \sigma_{\alpha\beta}(\vec{r}, \vec{r}'), \qquad (11)$$

the following relation can be shown to hold by straightforward Fourier analysis:

$$\vec{j}(\vec{k}_\parallel + \vec{G}_\parallel^i, \vec{r}_\perp) = \sum_{\vec{G}_\parallel^j} \int dr_\perp' \, \underline{\sigma}^{ij}(\vec{k}_\parallel, \vec{r}_\perp, \vec{r}_\perp') \vec{E}(\vec{k}_\parallel + \vec{G}_\parallel^j, \vec{r}_\perp'),$$

$$(12)$$

where the quantity $\underline{\sigma}^{ij}$ is given by

$$
\underline{\sigma}^{ij}(\vec{k}_\parallel, \vec{r}_\perp, \vec{r}_\perp') = \frac{1}{A_{WS}^I} \int_{A_{WS}^I} d^2 r
$$

$$
\times \int_{A^J} d^2 r' \, \underline{\sigma}(\vec{r}_\parallel, \vec{r}_\perp, \vec{r}_\parallel', \vec{r}_\perp')
$$

$$
\times e^{-i\vec{k}_\parallel(\vec{r}_\parallel - \vec{r}_\parallel')} e^{i\vec{r}_\parallel'(\vec{G}_\parallel^j - \vec{G}_\parallel^i)}, \qquad (13)
$$

with σ a matrix with respect to Cartesian components. $\vec{G}_\parallel^{i(j)}$ denote reciprocal lattice vectors with respect to the lattice plane and A^J, A_{WS}^I the area of layer J and the area of the two-dimensional Wigner-Seitz cell in layer I, respectively. Furthermore, it is assumed that $\vec{r}_\perp$ ($\vec{r}_\perp'$) is located in layer I (layer J). Equation (13) can be further simplified, if we assume the electric field to vary only slowly on an atomic scale; this is well justified in the optical regime where the typical wave length is by a factor of 1000 larger than the lattice parameters of ordinary solids. Averaging both sides in Eq. (13) with respect to $\vec{r}_\perp$ within layer I—which leads to an additional index I—and integrating the electric field and the optical conductivity independently over each layer J, we arrive at the following nonlocal, however, discrete, relation between the average electric field in layers J and the electrical current density in layer I:

$$\vec{j}^I(\vec{k}_\parallel + \vec{G}_\parallel^i) = \sum_J \sum_{\vec{G}_\parallel^j} \underline{\sigma}_{ij}^{IJ}(\vec{k}_\parallel) \vec{E}^J(\vec{k}_\parallel + \vec{G}_\parallel^j). \qquad (14)$$

We may further neglect all off-diagonal elements of $\underline{\sigma}_{ij}^{IJ}$ with respect to (ij) and drop these indices in all following considerations. Together with Eq. (13), this means that the real

system is replaced by a stack of homogeneous layers of atomic thicknesses in an optical sense. This implies in particular that all local field effects taking place within the dimensions of a single layer are neglected. For the optical regime, this will be justified by an investigation of the corresponding explicit solutions of the Maxwell equations for realistic systems.[29] Relying on this property, it is save to neglect also the remaining $\vec{k}_\parallel$ dependence of σ^{IJ} for optical wavelengths. Using Eq. (9), one obtains finally the following expression for the layer-resolved optical conductivity $\underline{\sigma}^{IJ}$:

$$\sigma^{IJ}_{\alpha\beta}(\omega) = \frac{i\hbar}{\pi^2 A_{WS} d^I} \int_{-\infty}^{E_F} dE \int_{E_F}^{\infty} dE' \int_{A^I_{WS}} d^2 r$$

$$\times \int_{\text{layer } I} dr \int_{\text{layer } J} d^3 r'$$

$$\times \frac{\text{tr}\{j_\alpha(\vec{r})\,\text{Im}\,G^+(E')\,j_\beta(\vec{r}')\,\text{Im}\,G^+(E)\}}{[E'-E-i(\hbar/\tau)][\hbar\omega+E-E'+i(\hbar/\tau)]}$$

$$+ \frac{\text{tr}\{j_\beta(\vec{r}')\,\text{Im}\,G^+(E')\,j_\alpha(\vec{r})\,\text{Im}\,G^+(E)\}}{[E'-E+i(\hbar/\tau)][\hbar\omega+E'-E+i(\hbar/\tau)]},$$

$$(15)$$

with d^I the thickness of layer I.

The explicit solution of the Maxwell equations that is needed to determine the magneto-optical properties of the system is of course greatly simplified if the nonlocal relation in Eq. (14) between the current density in layer I and the electric fields in all layers J can be replaced by an effective local one. In fact, under the assumption that in the direction perpendicular to the layers the electric field varies only slowly compared to the range of the interlayer dependency within $\underline{\sigma}^{IJ}$, we can write with the aid of Eq. (14)

$$\vec{j}^I(\omega) = \underline{\sigma}^I(\omega)\vec{E}^I(\omega) \qquad (16)$$

with $\underline{\sigma}^I = \Sigma_J \underline{\sigma}^{IJ}$ the effective *local* optical conductivity of layer I. For a system with three dimensional translational invariance this local quantity is of course identical with the conventional optical conductivity tensor $\underline{\sigma}$.

C. Optical conductivity within the SPR-KKR-GF

As mentioned above, one has to account in particular for the spin-orbit coupling when one is dealing with magneto-optical properties. This is done here in a fully relativistic way by describing the electronic system on the basis of the Dirac equation for magnetic solids (from now on we will use atomic Rydberg units):

$$\left[\frac{c}{i}\vec{\alpha}\vec{\nabla} + \frac{1}{2}(\beta-1)c^2 + V(\vec{r})\right]\Phi_i(\vec{r}) = E\Phi_i(\vec{r}). \qquad (17)$$

Here it is assumed that the corresponding effective potential V that is spin dependent is set up within the relativistic version of spin density functional theory (SDFT).[34,35] For systems for which orbital magnetism is of appreciable importance appropriate extensions as for example current density functional theory (CDFT) (Ref. 36) can be used as well, without affecting the following scheme.

A very reliable and accurate way to determine the electronic Green's function $G^+(E)$ to be inserted into Eq. (15) on the basis of the above Dirac equation is supplied by relativistic multiple scattering theory or KKR formalism. This allows to write $\text{Im}\,G^+(E)$ in its real-space spin representation in the compact way:

$$\text{Im}\,G^+(\vec{r},\vec{r}',E) = \frac{1}{2i} \sum_{\Lambda\Lambda'} Z^n_\Lambda(\vec{r},E)$$

$$\times [\tau^{nm}_{\Lambda\Lambda'}(E) - \tau^{mn*}_{\Lambda'\Lambda}(E)] Z^{m\times}_{\Lambda'}(\vec{r}',E).$$

$$(18)$$

Here $Z^{n(\times)}_\Lambda$ is the regular (left hand) solution for the single site Dirac equation for the potential well at site n and $\Lambda = (\kappa,\mu)$ stands for the relativistic spin-orbit and magnetic quantum numbers κ and μ, respectively. Finally, $\tau^{nm}_{\Lambda\Lambda'}$ is the so-called scattering path operator that transforms a wave with character Λ' incoming at site m into a wave with character Λ outgoing from site n with all possible multiple scattering events accounted for (for further details see for example Refs. 37 or 38).

Here one should mention that the expression given in Eq. (18) is completely sufficient to evaluate the energy integrals in Eq. (15) for real energies. However, a more efficient scheme is achieved by exploiting the analyticity of the Green's function and deforming the energy integration paths in an appropriate way into the upper complex plane. This has the great advantage that the Green's function is less structured for complex energies allowing to reduce the number of energy points accordingly.[39] However, in this case, also the so-called irregular parts of the Green's function have to be taken into account. This makes the implementation much more tedious and the whole calculation for one single pair of energy points from both paths very time consuming.

An alternative and promising approach seems to use Cauchy's integral theorem to express the integral over the imaginary part of the second Green's function times the energy-dependent prefactor by a single Green's function. This way one gets rid of one of the energy integrations. However, in this case, the photon energy $\hbar\omega$ explicitly appears in the argument of the Green's function. That means that the whole calculation has to be carried out separately for each photon energy, i.e., 100–200 times *in praxi*, depending on the spectral range one is interested in.

Finally it should be mentioned here that a further method to calculate the optical conductivity has been developed recently by Szunyogh and Weinberger.[40] These authors start from the formula given by Wang and Callaway[33] in terms of the single-particle Green's function and also end up with only one single energy integration involved.

For simplicity, it is assumed in the following that the in-layer atomic basis consists only of one atom per primitive unit cell; however, the formalism can be readily extended to the case of complex systems. Furthermore the energy integrals in Eq. (15) will be restricted to the real energy axis. Accordingly Eq. (18) is inserted directly into Eq. (15). Performing a two-dimensional Fourier transformation with respect to the resulting sums over products of τ-matrices and exploiting the fact that the wave functions Z only depend on the respective layer, we can write

$$\int_{A_{\mathrm{WS}}^I} d^2r \int_{\mathrm{layer}\ I} dr \int_{\mathrm{layer}\ J} d^3r'\,\mathrm{tr}\{j_\alpha(\vec{r})$$

$$\times \mathrm{Im}\,G^+(E')j_\beta(\vec{r}')\,\mathrm{Im}\,G^+(E)\}$$

$$= -\sum_{\Lambda\ldots\Lambda'''}\langle Z_{\Lambda'''}^{I\times}(E)|j_\alpha(\vec{r})|Z_\Lambda^I(E')\rangle$$

$$\times \langle Z_{\Lambda'}^{J\times}(E')|j_\beta(\vec{r}')|Z_{\Lambda''}^J(E)\rangle \frac{1}{V_{\mathrm{BZ}}}$$

$$\times \int_{V_{\mathrm{BZ}}} d^2k[\tau_{\Lambda\Lambda'}^{IJ}(\vec{k},E) \quad -\tau_{\Lambda'\Lambda}^{*JI}(\vec{k},E)]$$

$$\times [\tau_{\Lambda''\Lambda'''}^{JI}(\vec{k},E') - \tau_{\Lambda'''\Lambda''}^{*IJ}(\vec{k},E')] \tag{19}$$

with V_{BZ} the corresponding area of the corresponding two-dimensional Brillouin zone. The τ matrices in reciprocal space, $\tau_{\Lambda\Lambda''}^{IJ}(\vec{k},E)$, are obtained by the corresponding host τ matrices and the single-site scattering matrices (t matrices) of the perturbed system via a Dyson equation. Details on this can be found elsewhere.[41]

The quantities in angular brackets in Eq. (19) represent the matrix elements of the current density operator j_α with respect to the regular wave functions $Z_\Lambda(E)$. Technical details concerning the evaluation of these matrix elements can be found for example in Ref. 42. The spatial integration involved here in principle has to be carried out over the regular prism extending according to the layer thickness with the two-dimensional Wigner-Seitz cell of the in-plane lattice as basis. However, the calculations presented in this paper were carried out making use of the atomic sphere approximation (ASA). This means that each atomic cell is represented by a sphere of the volume of the respective atom, which in a first place also represents the integration volume for our matrix elements. The integration volume, i.e., the prism described above is distributed over several adjacent atomic spheres. As an approximation, we therefore neglect the spatial dependence of the current density matrix elements within an atomic cell in order to represent its spatial integral by a weighted sum of integrals over adjacent atomic spheres, according to their volume contributions to the respective right prisms.

D. General transfer matrix T and Kerr rotation angle of magneto-optic layer systems

In general, the transmission, reflection, and coupling of electromagnetic waves for a stack of homogeneous and optically local layers is described in terms of the so-called general transfer matrix T.[43] This approach allows one to calculate for any polarization the amplitude of transmitted light in terms of those of incident and reflected light. We assume the layer stack to be limited by homogeneous, isotropic dielectric media (e.g., air). Then the waves on both sides of the stack can be decomposed into p- and s-polarized contributions:

$$\begin{pmatrix} E_{s,i} \\ E_{p,i} \\ E_{s,r} \\ E_{p,r} \end{pmatrix} = T \begin{pmatrix} E_{s,t} \\ E_{p,t} \\ 0 \\ 0 \end{pmatrix} \tag{20}$$

with $E_{s(p),i(r)(t)}$ the complex amplitudes of s- (p-) polarized components of incident (i), reflected (r), and transmitted waves (t). Light incidence is only permitted from one side of the layer stack, thus there are no reflected waves on the transmission side. As shown by various authors,[44,45] the transfer matrix T can be used in a straightforward way to calculate the reflection coefficients r_{ss}, r_{pp}, r_{ps} and r_{sp} of the layer system. Given that, the following approximate relation between the reflection coefficients and the complex Kerr rotation angle can be used:[46]

$$\theta_s + i\epsilon_s = r_{ps}/r_{ss},$$

$$-\theta_p + i\epsilon_p = r_{sp}/r_{pp}, \tag{21}$$

with $\theta_s(\theta_p)$ the real Kerr rotation angle and $\epsilon_s(\epsilon_p)$ the Kerr ellipticity of the system as a whole.

For the remaining determination of the general transfer matrix T, we employ a formalism based on Eq. (16) and an eigenvector equation for any appropriate wave vector $\vec{k}$ and polarization vector $\vec{\epsilon}^I$ in layer I which follows from simple combination of Maxwell's curl equations:

$$\left(\frac{4\pi i}{\omega}\underline{\sigma}^I + \underline{I} + \frac{c^2}{\omega^2}(\vec{k}\vec{k}^t - \vec{k}^t\vec{k})\right)\vec{\epsilon}^I = 0 \tag{22}$$

with $\underline{I}$ the 3×3 unit matrix and c the velocity of light in vacuum. The procedure can be carried out without any further restrictions to $\underline{\sigma}^I$. We have checked our method by a totally different and far more expensive method employing direct numerical solution of Maxwell's equations in differential form. Details of this, together with a numerical justification of the approximation leading to Eq. (16), may be found in a subsequent publication.[29]

III. APPLICATION TO BCC-FE AND FCC-CO

A. Layer-resolved optical conductivity tensor

The calculation of the layer-resolved optical conductivity tensor σ^{IJ} for the bulk systems bcc-Fe and fcc-Co treated as homogeneous layer systems has been carried out evaluating Eq. (15) together with Eq. (19) by means of the SPR-KKR-GF method. The z axis of the coordinate system as well as the direction of magnetization have been taken in the [001] direction, the x and y axes pointing along the perpendicular crystallographic directions. The first energy path in Eq. (15) was started below the bottom of the conduction band and ended at the Fermi energy. The second energy path extended from the Fermi level over approximately 2 Ryd. To reduce the numerical effort the energy paths have been shifted somewhat (2 mRy) into the complex plane. For the first (second) energy path, 150 (250) energy points have been used, respectively. The number of k points in the irreducible part of the two-dimensional Brillouin zone was fixed to 528. For the relaxation time parameter appearing in Eq. (15), we used 0.4 eV for bcc-Fe as well as for fcc-Co as in Ref. 47 for comparison.

The real part of the diagonal component of the effective *local* conductivity tensor σ_{xx} [see Eq. (16)] for bcc-Fe is shown in Fig. 1, the solid curve being identical to that in Fig. 2. As one would expect, the intralayer part $\sigma_{xx}^{II\,(1)}$ dominates
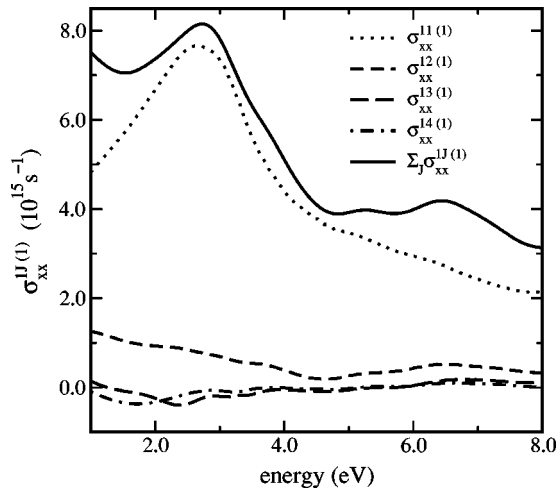
FIG. 1. Real part of the diagonal component σ_{xx}^{IJ} of the layer-resolved local optical conductivity tensor for bcc-Fe. The dotted line gives the layer-diagonal contribution σ_{xx}^{IJ} ($I=J$). Dashed lines give various layer-off-diagonal contributions ($J \neq I$). The solid line stands for the resulting effective local conductivity tensor.

over the whole spectral range. Nevertheless, the coupling to the first neighboring layers still gives a substantial contribution. On the other hand, the layer-off-diagonal optical conductivity $\sigma_{xx}^{IJ\,(1)}$ is seen to fall off quite rapidly with increasing interlayer distance and thus may safely be neglected for the fifth neighboring layer and beyond. This is why one should expect that the approximation made when employing the effective local optical conductivity for the solution of the Maxwell equations should work quite well. Similar results as shown here for $\sigma_{xx}^{IJ\,(1)}$ have been obtained for the off-diagonal tensor elements $\sigma_{xy}^{IJ\,(1)}$. However, these decay somewhat less rapidly with respect to the interlayer distance $(I-J)$.
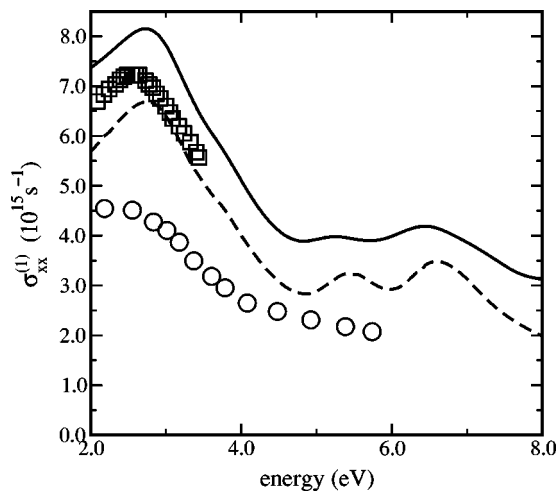

FIG. 2. Real part of the diagonal component σ_{xx} of the effective local optical conductivity tensor for bcc-Fe. Solid line: present results based on the SPR-KKR-GF. Dashed line: results obtained from an SPR-LMTO calculation (Ref. 47). The circles and squares represent the experimental results given in Refs. 48 and 49, respectively.
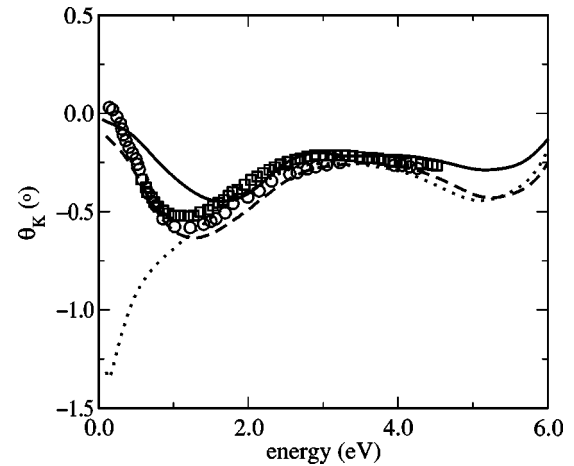

FIG. 3. Kerr rotation angle θ_K for bcc-Fe. Solid line: result based on the SPR-KKR-GF. The dashed and dotted lines give the results of SPR-LMTO calculations (Ref. 47). The latter one has been done without the Drude contribution. Circles and squares represent the experimental results of Refs. 50 and 51, respectively.

B. Kerr rotation spectra

Kerr rotation spectra for bcc-Fe and fcc-Co for light incident along the surface normal (polar Kerr effect) are shown in Figs. 3 and 4 together with theoretical results and measurements from two different sources. The data have been produced using the formalism sketched in Sec II D. Shapes and magnitudes of measured curves are well reproduced, at least in the photon energy range above than 2 eV (see below).

The dotted curve in Fig. 3 is calculated by means of the SPR-LMTO band structure method without making explicit use of the Drude (intraband-) contribution to the optical conductivity tensor. This well-known term is usually added to spectra calculated within the framework of conventional band structure methods where only interband contributions are taken into account in the first place. As one can see, this term is significant for photon energies below 2 eV. To represent the Drude term one is employing two different addi-
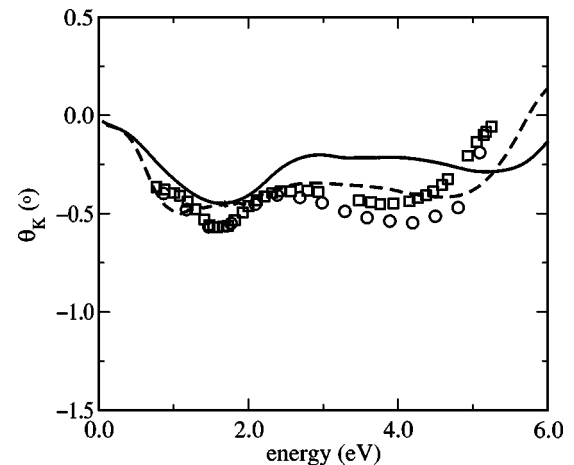

FIG. 4. Kerr rotation angle θ_K for fcc-Co. Solid line: result based on the SPR-KKR-GF. The dashed line gives the result of an SPR-LMTO calculation (Ref. 47). The circles and squares represent the experimental results of Refs. 52 and 53, respectively.
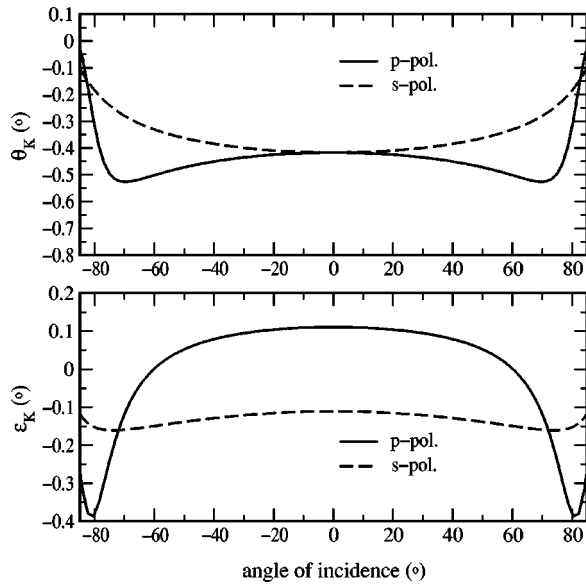
FIG. 5. Kerr rotation angles and ellipticities for bcc-Fe for various angles of incidence for photon energy of 1.96 eV. Solid (dashed) line is for *p*- (*s*-) polarized light.

tional parameters: the zero-frequency conductivity σ_0 and the Drude (free-electron) relaxation time τ_D. In our method, the intraband contributions are intrinsically taken into account with a single relaxation time parameter τ for the whole spectrum as required by Eq. (15). The choice made here for this parameter ($\hbar/\tau = 0.4$ eV) is mainly responsible for the deviation of our Kerr rotation curves from other theoretical as well as experimental data for photon energies below 2 eV.

Finally, Kerr rotation, as well as Kerr ellipticity spectra for various angles of incidence with respect to the surface normal and a fixed photon energy (1.96 eV for common He-Ne-laser light) are presented in Fig. 5. Obviously, the curves depend strongly on the angle of incidence and polarization direction of incoming light. For this reason, ellipsometric magneto-optic measurements for fixed photon energy can be seen as an appropriate tool to check the results of band structure calculations for the initial and final electronic states involved.

IV. SUMMARY

A scheme to calculate the layer-resolved optical conductivity tensor on the basis of the single-particle Green's function has been presented. The most important details of a corresponding implementation within the SPR-KKR-GF have been discussed. The connection of introduced layer-resolved optical conductivity tensor with experimentally accessible magneto-optical quantities such as the Kerr-rotation angle have been outlined.

To demonstrate the feasibility of our approach, first results for the elemental ferromagnets bcc-Fe and fcc-Co treated as a homogeneous layer system were presented. For photon energies above 2 eV, for which the Drude term can be neglected, good agreement was found between our data for the real part of the diagonal component of the effective optical conductivity tensor and Kerr rotation angles for normal incidence for bcc-Fe and corresponding theoretical and experimental data. Discrepancies between the theoretical Kerr spectra for photon energies below 2 eV have been mainly attributed to the distinct treatment of the intraband contributions to the optical conductivity tensor.

On the basis of the very encouraging results for the element systems together with an appropriate solution scheme suggested for the corresponding Maxwell equations, it is expected that the formalism presented here will allow detailed and reliable investigations of the magneto-optical properties of surface-layered systems as well. Work along this line is in progress now.

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