

Limitations of the two-media approach in calculating magneto-optical properties of layered systems

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It is shown that in polar geometry and normal incidence the 2×2 matrix technique—as discussed in detail in a preceding paper [Phys. Rev. B **65**, 144448 (2002)]—accounts correctly for multiple reflections and optical interferences, and reduces only in the case of a periodic sequence of identical layers to the Fresnel formula of reflectivity, which in turn is the theoretical basis of the two-media approach, widely used in the literature to compute magneto-optical Kerr spectra. As a numerical example *ab initio* calculations of the optical constants for an fcc Pt semi-infinite bulk using the spin-polarized relativistic screened Korringa-Kohn-Rostoker method show very good agreement with experimental data.

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INTRODUCTION

In the last few years magneto-optics has started to become of prime interest in dealing with magnetic multilayer systems and magnetic nanostructures (submonolayer coverages of substrates with magnetic material): Kerr measurements not only turned out to be one of the standard experimental tools applied; additional applications, in particular in the context of time-resolved techniques, are presently believed to lead to fast magnetization switching devices. In many cases, however, theoretical descriptions, even for time-integrated magneto-optical effects, are lagging behind experimental efforts, mainly because relevant schemes to deal with semi-infinite systems—not to speak of nanostructured materials—are not commonly used. Bulklike approaches (assumed three-dimensional periodicity) and the so-called two-media model¹ (assumed homogeneity) are still considered to be sufficient to deal with magnetically inhomogeneous layered systems.

In a recent paper,² the authors showed that only by including multiple reflections and optical interferences, e.g., via the 2×2 matrix technique,^{3,4} realistic *ab initio* magneto-optical Kerr spectra for semi-infinite layered systems can be obtained. It is the purpose of the present paper to prove first analytically and then numerically that only in the case of periodic layered systems, i.e., in by definition homogeneous systems, the 2×2 matrix technique reduces to the two-media approach. Formulated oppositely, this implies that the two-media approach is strictly valid only for this kind of systems. This is illustrated for fcc Pt viewed as a periodic layered system, because Pt frequently serves as a substrate for magneto-optically active multilayers.^{5,6}

THEORETICAL APPROACH

Assuming the polar geometry and the magnetization \vec{M}_p in all layers p to point along the z direction, the layer-

resolved permittivity in cubic, hexagonal, or tetragonal systems is given by

$$\tilde{\epsilon}^p = \begin{pmatrix} \tilde{\epsilon}_{xx}^p & \tilde{\epsilon}_{xy}^p & 0 \\ -\tilde{\epsilon}_{xy}^p & \tilde{\epsilon}_{xx}^p & 0 \\ 0 & 0 & \tilde{\epsilon}_{zz}^p \end{pmatrix}, \quad (p = 1, \dots, N), \quad (1)$$

provided that a possible anisotropy in the diagonal elements can be neglected, i.e., $\tilde{\epsilon}_{zz}^p \approx \tilde{\epsilon}_{xx}^p$. In case of normal incidence, $\tilde{n}_{px} = \tilde{n}_{py} = 0$, four electromagnetic beams corresponding to the complex refractive indices

$$\begin{aligned} \tilde{n}_{pz}^{(3)} &= -\tilde{n}_{pz}^{(1)} = \sqrt{\tilde{\epsilon}_-^p} \equiv \tilde{n}_{p-}, \\ \tilde{n}_{pz}^{(4)} &= -\tilde{n}_{pz}^{(2)} = \sqrt{\tilde{\epsilon}_+^p} \equiv \tilde{n}_{p+} \end{aligned}$$

propagate in layer p . By considering harmonic fields

$$\vec{A}(z, t) = \vec{A} \exp[i(\tilde{q}z - \omega t)] \exp(-\delta t) = \vec{A} \exp[i(\tilde{q}z - \tilde{\omega}t)], \quad (2)$$

such that $\delta > 0$ describes the interaction between the layered system and its neighborhood, $\tilde{q} = q_0 \tilde{n}$ is the complex wave vector, with q_0 being the propagation constant in vacuum; and $\tilde{\omega} = \omega - i\delta$, beams 1 and 2 propagate along $-z$, and beams 3 and 4 along $+z$. The surface reflectivity matrix (see in particular the Appendix of Ref. 2),

$$R_{\text{surf}} = \begin{pmatrix} \tilde{r}_{xx} & \tilde{r}_{xy} \\ -\tilde{r}_{xy} & \tilde{r}_{xx} \end{pmatrix}, \quad (3)$$

which relates the incident electric field to the reflected one,

$$R_{\text{surf}} = (B_{\text{vac}} + \mathcal{D}_N \mathcal{A}^{-1})^{-1} (B_{\text{vac}} - \mathcal{D}_N \mathcal{A}^{-1}), \quad (4)$$

is obtained as defined in Eq. (37) of Ref. 2, in terms of reflectivity matrices \mathcal{R}_p ; see Eq. (32) of Ref. 2,

$$\mathcal{R}_p = (\mathcal{B}_p + \mathcal{D}_{p-1})^{-1} (\mathcal{B}_p - \mathcal{D}_{p-1}), \quad p = 1, \dots, N, \quad (5)$$

and propagation matrices \mathcal{C}_p [see Eq. (30) of Ref. 2],

$$\mathcal{C}_p = \begin{pmatrix} \exp(-iq_0 \tilde{n}_p d_p) & 0 \\ 0 & \exp(-iq_0 \tilde{n}_p d_p) \end{pmatrix}, \quad (6)$$

where d_p refers to the thickness of layer p . According to Eqs. (28), (32) and (35) of Ref. 2 the 2×2 matrices occurring in Eq. (4) are defined as follows:

$$\mathcal{D}_p = \mathcal{B}_p [\mathcal{C}_p - (\mathcal{C}_p)^{-1} \mathcal{R}_p] [\mathcal{C}_p + (\mathcal{C}_p)^{-1} \mathcal{R}_p]^{-1}, \quad (7)$$

$$\mathcal{A} = \begin{pmatrix} 1 & i \\ i & 1 \end{pmatrix}, \quad \mathcal{B}_p = \begin{pmatrix} i\tilde{n}_{p-} & \tilde{n}_{p+} \\ -\tilde{n}_{p-} & -i\tilde{n}_{p+} \end{pmatrix},$$

$$\mathcal{B}_{\text{vac}} = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}. \quad (8)$$

\mathcal{R}_N results then recursively starting from the substrate reflectivity matrix $\mathcal{R}_0 = 0$. It has been shown in quite some detail in the appendix of Ref. 2 that the layer-resolved reflectivity matrices \mathcal{R}_p are all diagonal,

$$\mathcal{R}_p = \begin{pmatrix} \tilde{r}_{p-} & 0 \\ 0 & \tilde{r}_{p+} \end{pmatrix},$$

where $\tilde{r}_{p\pm}$ is the complex reflectivity coefficient of the right- and left-handed light in layer p . This implies that the right- and left-handed circularly polarized components of the incident linearly polarized light, once they arrived at the surface layer, propagate independently within the system such that after the first reflection, they become immediately elliptically polarized.

SPECIAL CASE OF HOMOGENEOUS SYSTEMS

In principle in a finite periodic layered system (N can be large but finite; for matters of simplicity a simple parent lattice⁷ is assumed) all layers have identical layer-resolved permittivities. This implies in turn that also all matrices \mathcal{B}_p in Eq. (8) are identical, i.e., are of the form

$$\mathcal{B} = \begin{pmatrix} i\tilde{n}_- & \tilde{n}_+ \\ -\tilde{n}_- & -i\tilde{n}_+ \end{pmatrix}, \quad (9)$$

with $\tilde{n}_\pm = \sqrt{\tilde{\epsilon}_\pm}$. The recursion relation in Eq. (5) therefore reduces to

$$\mathcal{R}_p = (\mathcal{I} + \mathcal{G}_{p-1})^{-1} (\mathcal{I} - \mathcal{G}_{p-1}), \quad p = 1, \dots, N, \quad (10)$$

where \mathcal{I} denotes the 2×2 identity matrix and

$$\mathcal{G}_{p-1} = [\mathcal{C}_{p-1} - (\mathcal{C}_{p-1})^{-1} \mathcal{R}_{p-1}] [\mathcal{C}_{p-1} + (\mathcal{C}_{p-1})^{-1} \mathcal{R}_{p-1}]^{-1}. \quad (11)$$

This finite periodic layered structure has to be properly matched to a semi-infinite system (substrate) of the same material. Inserting $\mathcal{R}_0 = 0$ into Eq. (11) for $p = 1$ yields $\mathcal{G}_0 = \mathcal{I}$, which, substituted into Eq. (10), immediately proves that also $\mathcal{R}_1 = 0$, and so on. In conclusion, for a periodic layered system,

$$\mathcal{R}_p = 0, \quad \forall p = 1, \dots, N. \quad (12)$$

From an optical point of view a periodic layered system behaves like a system with no boundaries. Viewed oppositely, Eq. (12) shows that there is a boundary in-between two adjacent layers, if and only if, the respective layer-resolved permittivities differ.

From $\mathcal{R}_N = 0$ it follows immediately that $\mathcal{D}_N = \mathcal{B}$, with \mathcal{B} defined in Eq. (9); hence Eq. (4) directly yields the surface reflectivity matrix

$$\mathcal{R}_{\text{surf}} = \frac{1}{(\tilde{n}_+ + 1)(\tilde{n}_- + 1)} \begin{pmatrix} 1 - \tilde{n}_+ \tilde{n}_- & -i(\tilde{n}_+ - \tilde{n}_-) \\ i(\tilde{n}_+ - \tilde{n}_-) & 1 - \tilde{n}_+ \tilde{n}_- \end{pmatrix}, \quad (13)$$

i.e., \tilde{r}_{xx} and \tilde{r}_{xy} in Eq. (3) assume the following values

$$\tilde{r}_{xx} = -\frac{\tilde{n}_+ \tilde{n}_- - 1}{(\tilde{n}_+ + 1)(\tilde{n}_- + 1)}, \quad \tilde{r}_{xy} = -i \frac{\tilde{n}_+ - \tilde{n}_-}{(\tilde{n}_+ + 1)(\tilde{n}_- + 1)}.$$

In the case of periodic layered systems the complex reflectivity coefficient of the right- and left-handed polarized light is therefore given by

$$\tilde{r}_\pm = \tilde{r}_{xx} \mp i \tilde{r}_{xy} = -\frac{\tilde{n}_\pm - 1}{\tilde{n}_\pm + 1}, \quad (14)$$

a relation which is known in the literature as the Fresnel formula for s polarization and normal incidence.⁸ Equation (14) then leads directly to the well-known formula for the complex Kerr angle in the two-media approach,¹

$$\theta_K + i\epsilon_K \simeq \frac{\tilde{\sigma}_{xy}}{\tilde{\sigma}_{xx}} \left(1 + \frac{4\pi i}{\tilde{\omega}} \tilde{\sigma}_{xx} \right)^{-1/2},$$

where θ_K is the Kerr rotation angle and ϵ_K the Kerr ellipticity.

Clearly enough in the case of (homogeneous) periodic layered systems the 2×2 matrix technique and the two-media approach provide identical Kerr spectra. However, by using the two-media approach for calculating the Kerr spectra of inhomogeneous layered systems, such spectra are governed almost exclusively by contributions from the surface layer.

APPLICATION TO FCC PT

In Figs. 1–3 the optical constants of fcc Pt bulk as calculated via the 2×2 matrix technique for normal incidence and different surface orientations are compared with available experimental data (Ref. 9, and references cited therein). Because fcc Pt is paramagnetic, the right- and left-handed complex reflectivity coefficients are equal, $\tilde{r}_+(\omega) = \tilde{r}_-(\omega)$

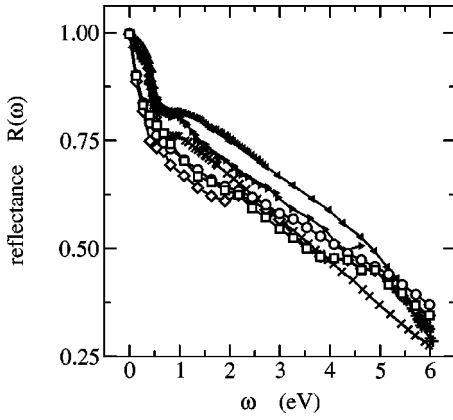


FIG. 1. Reflectance of fcc Pt bulk as calculated for the (100), (110), and (111) surface orientations (diamonds, circles, and squares). The experimental data are taken from Refs. 20 (crosses), 21 (pluses), 22 (stars), 23 (triangle right), 24 (triangle left), and 25 (triangle up).

$\equiv \tilde{r}(\omega)$, and the reflectance $R(\omega)$ given in Figs. 1 is in fact $\tilde{r}(\omega)^2$. In accordance with Eq. (2) the refractive index $n(\omega)$ and the absorption coefficient $k(\omega)$ in Fig. 2 simply correspond to the real and imaginary part of the complex refractive index $\tilde{n}(\omega) = \sqrt{\tilde{\epsilon}(\omega)} \equiv n(\omega) + ik(\omega)$. The total (complex) permittivity $\tilde{\epsilon}(\omega)$ displayed in Fig. 3 is evaluated assuming a periodic layered system consisting of N Pt layers

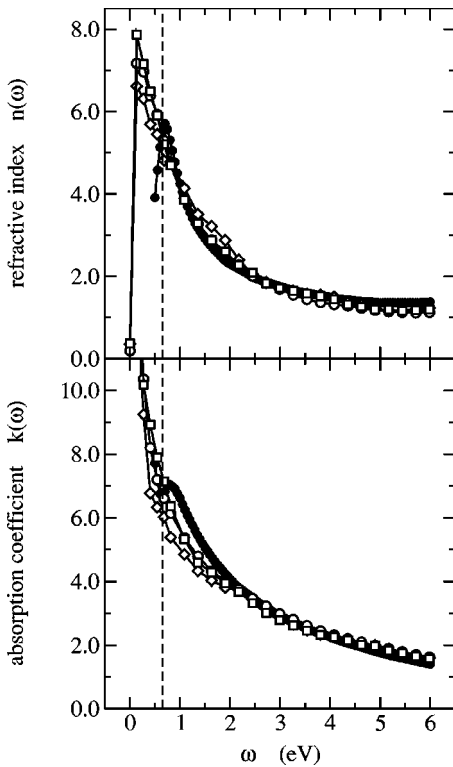


FIG. 2. Optical constants of fcc Pt bulk as calculated for the (100), (110), and (111) surface orientation (diamonds, circles and squares). The experimental data of Ref. 9 are displayed as grey circles. The dotted line marks the photon energy that equals the used lifetime broadening of $\delta = 0.653$ eV.

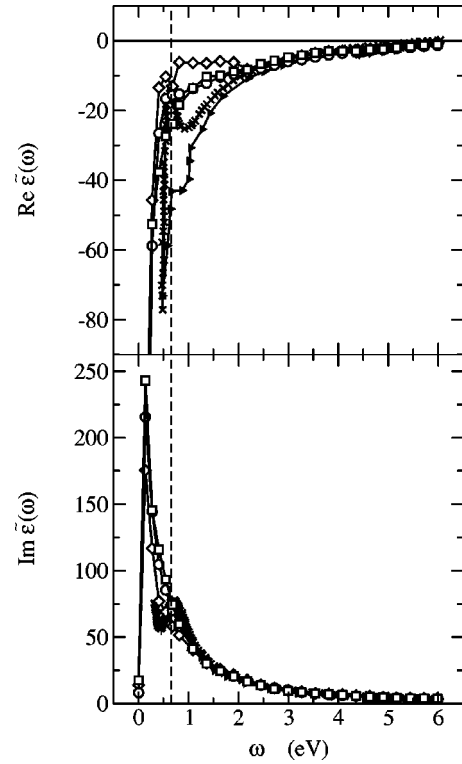


FIG. 3. Permittivity of fcc Pt bulk as calculated for the (100), (110), and (111) surface orientations (diamonds, circles, and squares). The experimental data are taken from Refs. 20 (crosses), 21 (pluses), 22 (stars), 23 (triangle right), and 26 (triangle down). The dotted line marks the photon energy that equals the used lifetime broadening of $\delta = 0.653$ eV.

on top of a semi-infinite Pt substrate, i.e.,

$$\tilde{\epsilon}(\omega) = \frac{1}{N} \sum_{p,q=1}^N \tilde{\epsilon}^{pq}(\omega), \quad (15)$$

where

$$\tilde{\epsilon}^{pq}(\omega) = \delta_{pq} + \frac{4\pi i}{\tilde{\omega}} \tilde{\sigma}^{pq}(\omega), \quad p, q = 1, \dots, N, \quad (16)$$

with $\tilde{\sigma}^{pq}(\omega)$ being the interlayer and intralayer contributions to the optical conductivity and $\tilde{\omega} = \omega - i\delta$. Equations (15) and (16) follow directly from the Fourier-transformed (linear) material equations¹⁰ by assuming that each layer p can be viewed as a homogeneous medium and therefore the layer-resolved optical conductivity $\tilde{\sigma}^p(\omega)$ is related to the layer-resolved permittivity by

$$\tilde{\epsilon}^p(\omega) = 1 + \frac{4\pi i}{\tilde{\omega}} \tilde{\sigma}^p(\omega), \quad p, q = 1, \dots, N. \quad (17)$$

$\tilde{\sigma}^{pq}(\omega)$ in Eq. (16) are calculated in terms of Luttinger's formula¹¹ using the spin-polarized relativistic screened Korringa-Kohn-Rostoker method for layered systems,¹²⁻¹⁵ contour integration techniques,^{16,17} the Konrod quadrature, and the cumulative special points method for the occurring

Brillouin zone integrals.¹⁸ It should be noted that Luttinger's formula uses a vector potential description of the electric field and has several advantages over the well-known Kubo formula:¹⁹ both the absorptive and dissipative parts of the conductivity tensor are included without using Kramers-Kronig relations, and so are all interband and intraband contributions avoiding thus a phenomenological Drude term in order to mimic the latter contributions.

In the present study of fcc Pt bulk (lattice parameter 7.4137 a.u.) all complex energy and \vec{k} -space integrals are performed with an accuracy of 10^{-3} (in atomic units) for all surface orientations considered. The electronic temperature amounts to $T=300$ K. Furthermore, a lifetime broadening of $0.048 \text{ Ryd} \approx 0.653 \text{ eV}$ is used and 2 and 37 Matsubara poles in the lower and upper semi-plane, respectively, are taken into account.

As can be seen from Fig. 1, the reflectance $R(\omega)$ of Pt as calculated for normal incidence and different surface orientations agrees very well with the experimental data. Unfortunately the experimental data⁹ are not specified with respect to surface orientations, i.e., no surface normal is listed. The dependence of all the calculated optical constants on the surface orientation reflects, however, merely the fact that in principle in spectroscopical experiments only physical properties of semi-infinite systems, i.e., of solid systems with a surface, are recorded; see also Figs. 2 and 3.

A close inspection of Fig. 1 in the vicinity of very low photon energies reveals that the fine structure around 0.5 eV seen in the experimental reflectance is missing in the theoretical curves although the maximum in the refractive index as a function of the photon energy is well reproduced. This discrepancy between theory and experiments is caused by an intrinsic feature of the applied approach: the finite lifetime broadening δ that enters Luttinger's formula smears out possible fine structures for $\omega \leq \delta$ (the value of $\omega = \delta$ is indicated in Figs. 2 and 3 by a dotted line).

In Figs. 1–3 the theoretical data refer to a total of $N = 12$ Pt layers, because from calculations for $N=3, \dots, 18$ Pt layers (not shown in here) no significant N dependence of optical constants was found: the optical constants obtained for $N=3$ are almost as good as those obtained with $N=18$ Pt layers in the system.

An extensive analysis of the interlayer and intralayer contributions to the permittivity $\tilde{\epsilon}^{pq}(\omega)$ showed that in the case of periodic layered systems (simple lattices) the pq -like contributions to the permittivity are only functions of the relative position of layers p and q , i.e., $\tilde{\epsilon}^{pq}(\omega) = \tilde{\epsilon}^{|p-q|}(\omega)$; the layer-resolved permittivities $\tilde{\epsilon}^p(\omega)$ are dominated by the corresponding intralayer contribution $\tilde{\epsilon}^{pp}(\omega)$. Thus is not surprising that in the case of periodic layered systems, the following approximative form applies

$$\tilde{\epsilon}(\omega) \approx \frac{1}{N} \sum_{|p-q|=0}^3 \tilde{\epsilon}^{|p-q|}(\omega); \quad (18)$$

see Fig. 4. It was also found that the difference between the exact permittivity and the approximated one defined above, not only is independent of N , but also of ω . Thus a semi-

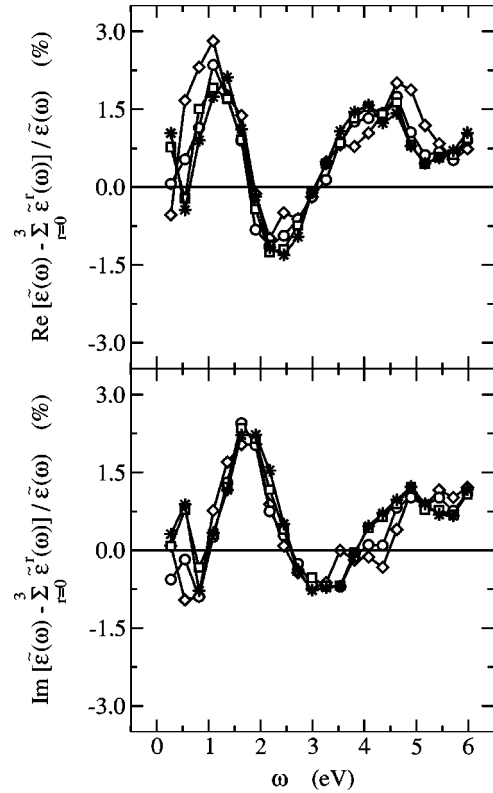


FIG. 4. Relative error made by approximating the permittivity of fcc Pt_N|Pt(111) by the sum over $\tilde{\epsilon}^r(\omega)$ permittivities with $r \equiv |p-q| = 0, \dots, 3$; see the text. Up and down triangles, diamonds, circles, squares, and stars denote data obtained for $N=3, 6, 9, 12, 15$, and 18 .

infinite periodic layered system can be simply modeled as a sequence of seven layers (a central one plus three layers above and below this) in a constant dispersionless permittivity background, the contribution of which to the total permittivity is negligible. Because of the fast convergence in $|p-q|$, calculations for periodic layered systems restricted to $N=3$ layers can already yield reasonably accurate optical constants.

SUMMARY

It was shown analytically and numerically that in polar geometry and normal incidence the complex reflectivity coefficient of the right- and left-handed polarized light for periodic (layered) systems is given by the Fresnel formula: only in the case of periodic (layered) systems the 2×2 matrix technique and the two-media approach become equivalent, i.e., they lead to identical Kerr spectra. This in turn clearly marks the limitations of the two-media approach, since applied to inhomogeneous layered systems, the corresponding Kerr spectra are dominated entirely by the optical activity of the surface layer.

Interlayer and intralayer contributions to the optical conductivity for fcc Pt_N/Pt layered systems as calculated within the framework of the spin-polarized relativistic screened Korringa-Kohn-Rostoker (SKKR) method by means of a contour integration technique lead to optical constants,

which are in very good agreement with available experimental data for photon energies $\omega > \delta$ (δ being the finite lifetime broadening). The calculated optical constants for fcc Pt_N/Pt do depend on the surface orientation; for $N \geq 3$ (number of Pt layers) they become virtually independent of N .

The conclusion to be drawn for magnetic multilayer systems is rather simple: only the 2×2 matrix technique that includes all multiple reflections and optical interferences can produce reliable spectra for this kind of systems. This in turn implies that a computational scheme such as the SKKR method, designed especially for systems with only two-dimensional translational symmetry, has to be applied exclu-

sively in order to evaluate the elements of the optical conductivity tensor.

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