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Linear optical responses beyond the electric dipole approximation on reflection and transmission: a perturbation treatment

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Abstract: There exist in a material a wide range of linear optical responses to external electromagnetic fields beyond the electric dipole process. These responses reveal more detailed information on properties of the material through their corrections to the zeroth-order dielectric tensor. These corrections introduce small yet distinguishable modifications to reflection and transmission. I here describe a perturbation method for computing these modifications. The method simplifies the computation of optical reflection and transmission that include first-order contributions from processes such as magneto-optic effects, electro-optic effects, surface and ultrathin films, electric quadrupole effects, photoelastic effects, and effects of meta-materials.

1. Introduction

Under the illumination of an external electromagnetic field, electrons in a solid material respond and produce time-varying electric dipoles. The latter in turn radiate and alter the spatial distribution of the electromagnetic field. Such an electric dipole process is the leading optical response from the material. Its effect on the propagation of the electromagnetic wave is prescribed by a zeroth-order linear dielectric tensor. In a crystalline material, electrons within a unit cell experience different local fields and their responses differ accordingly. Such a "non-local" effect is taken into consideration by adding a time-varying polarization vector in response to a quadrupole interaction, $H_{int,\,Q} = -\overleftrightarrow{Q} : \nabla\vec{E}$ [1,2,3]. The magnetic field associated with the electromagnetic wave yields another contribution through the magnetic dipole interaction, $H_{int,\,M} = -\vec{m}\cdot\vec{B}$ [1]. If there exist other fields in a material such as a low frequency magnetic field (or a magnetization) or a low frequency electric field or an elastic strain field, we expect additional linear optical responses such as magneto-optic effects [4–8], electro-optic effects and photo-elastic effects [9]. Furthermore, the surface region of a material more often than not has different chemical composition and symmetry from those in the interior of the material. Extrinsic ultrathin films on top of a solid further expand and complicate the surface region. Such a distinct surface layer on an otherwise homogeneous material modifies optical reflection and transmission perturbatively [10,11]. If the material is opaque, these effects can be observed only by the changes they cause to reflection.

Corrections to reflection and transmission due to optical processes beyond the linear electric dipole approximation have been explored for material characterization [3,10–19]. These corrections reveal more detailed information on structures and phases of materials that are easily absent in the zeroth-order dielectric tensor. To find effects on reflection and transmission, one usually starts from the full dielectric tensor that includes corrections from optical processes of interest and solves for eigen modes of the electromagnetic wave. By matching boundary conditions for the *full* electric field and the *full* magnetic field at each applicable surface, one finds reflected and transmitted fields in terms of the incident field. Calculations of this kind on magneto-optic effects [20–23] and those on surface and thin film effects [10,11,24] are examples.

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Yet the *full* expressions are rarely useful for insights. Instead, to relate experimental observation to structural and compositional ingredients responsible for changes to reflection and transmission, one more often than not resorts to Taylor expansions so that only terms varying linearly with the corrections to the dielectric tensor are kept for analysis [10–12,20,22,24].

This calls for a perturbation method to directly compute changes to reflection and transmission to the first order of corrections to the dielectric tensor. In this paper, I present such a method. The results are applicable to any linear optical process beyond the electric dipole approximation in the bulk.

2. Corrections to zeroth-order optical dielectric tensors:

A zeroth-order optical dielectric tensor $\overleftrightarrow{\epsilon}$ relates the electric field $\vec{E}$ to the displacement vector $\vec{D}$ when only the electric dipole response of electrons in a material is considered:

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} + \begin{pmatrix} P_x^{(0)} \\ P_y^{(0)} \\ P_z^{(0)} \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \epsilon_{xx}^{(0)} & \epsilon_{xy}^{(0)} & \epsilon_{xz}^{(0)} \\ \epsilon_{yx}^{(0)} & \epsilon_{yy}^{(0)} & \epsilon_{yz}^{(0)} \\ \epsilon_{zx}^{(0)} & \epsilon_{zy}^{(0)} & \epsilon_{zz}^{(0)} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \varepsilon_0 \overleftrightarrow{\epsilon}^{(0)} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} \tag{1}$$

If the material has a crystalline symmetry, a suitable principal coordinate frame (crystalline frame) exists in which off-diagonal elements vanish and some of the diagonal elements have same values [25,26]. Given the phase propagation direction, the dielectric tensor determines two eigen modes of the electromagnetic wave and in turn reflection and transmission when the wave is incident on such a material [25].

Beyond the electric dipole approximation, other linear optical responses abound. They include distinct electric dipole responses from the surface region or an ultrathin film [10], magneto-optic responses [4,5,13,14], electro-optic response [9], photoelastic response [9], electric quadrupole response [3], and magnetic dipole response [1]. Their effects are typically small as they modify the reflected electric field by only a few percent or less. Nonetheless interests in these other responses are motivated by the fact that they uncover further information on structures, phases, and other properties of a material. Efforts have been made and reported to understand and detect these weaker and yet more informative optical processes for materials characterization. Examples include ellipsometry studies of surfaces and thin films on solids [10,24], magneto-optic Kerr effect (MOKE) studies of materials with broken time-reversal symmetry [12–15], and the electric quadrupole response study of crystalline solids [3].

I start with polarizations arising from these additional optical processes and in turn their modification to the dielectric tensor as follows,

$$\begin{pmatrix} \Delta P_x \\ \Delta P_y \\ \Delta P_z \end{pmatrix} = \varepsilon_0 \Delta\overleftrightarrow{\epsilon} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \Delta\chi_{xx} & \Delta\chi_{xy} & \Delta\chi_{xz} \\ \Delta\chi_{yx} & \Delta\chi_{yy} & \Delta\chi_{yz} \\ \Delta\chi_{zx} & \Delta\chi_{zy} & \Delta\chi_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} \tag{2}$$

The full dielectric tensor can then be written as

$$\overleftrightarrow{\epsilon} \equiv \overleftrightarrow{\epsilon}^{(0)} + \Delta\overleftrightarrow{\epsilon} = \begin{pmatrix} \epsilon_{xx}^{(0)} + \Delta\chi_{xx} & \epsilon_{xy}^{(0)} + \Delta\chi_{xy} & \epsilon_{xz}^{(0)} + \Delta\chi_{xz} \\ \epsilon_{yx}^{(0)} + \Delta\chi_{yx} & \epsilon_{yy}^{(0)} + \Delta\chi_{yy} & \epsilon_{yz}^{(0)} + \Delta\chi_{yz} \\ \epsilon_{zx}^{(0)} + \Delta\chi_{zx} & \epsilon_{zy}^{(0)} + \Delta\chi_{zy} & \epsilon_{zz}^{(0)} + \Delta\chi_{zz} \end{pmatrix} \tag{3}$$

I now describe examples of Eq. (3) in special cases.

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Magneto-optic effects Consider materials belonging to orthorhombic (biaxial), tetragonal/trigonal/ hexagonal (uniaxial), and cubic (isotropic) crystal systems. The zero-order dielectric tensors in the principal coordinate frame only have diagonal elements. For isotropic materials including those belonging to the cubic crystal system, the diagonal elements are equal $\epsilon_{xx}^{(0)} = \epsilon_{yy}^{(0)} = \epsilon_{zz}^{(0)} \equiv \epsilon$. For uniaxial materials, diagonal elements associated with axes perpendicular to the optic axis (chosen as the z-axis by convention) are equal, $\epsilon_{xx}^{(0)} = \epsilon_{yy}^{(0)} \equiv \epsilon_{\perp}$, and $\epsilon_{zz}^{(0)} \equiv \epsilon_{\parallel} \neq \epsilon_{\perp}$. For biaxial materials, three diagonal elements are different. A low frequency magnetization $\vec{m} = (m_x, m_y, m_z)$ modifies the dielectric tensor as follows,

$$\Delta\overleftrightarrow{\epsilon}^{(MOKE)} = \begin{pmatrix} 0 & -iQ_{xy}m_z & iQ_{xz}m_y \\ iQ_{xy}m_z & 0 & -iQ_{yz}m_x \\ -iQ_{xz}m_y & iQ_{yz}m_x & 0 \end{pmatrix} \tag{4}$$

For isotropic materials, $Q_{xy} = Q_{yz} = Q_{zx} \equiv \epsilon Q$ where Q is the magneto-optic parameter or the magnitude of the Voigt vector $\vec{Q}$ [20]; for uniaxial materials, $Q_{yz} = Q_{zx} \equiv Q_{\perp} \neq Q_{xy} \equiv Q_{\parallel}$ [27]; for biaxial materials, Q_{xy}, Q_{yz} and Q_{zx} are different from one another. The magnitude of $\Delta\overleftrightarrow{\epsilon}^{(MOKE)}$ is typically 1/100 of $\overleftrightarrow{\epsilon}^{(0)}$ and thus the bulk magneto-optic effect on reflection is proportionally small.

If the magneto-optic effect is confined to a surface region over a thickness d, modifications to the dielectric tensor becomes

$$\Delta\overleftrightarrow{\epsilon}^{(SMOKE)} = d\delta(z)\begin{pmatrix} 0 & -iQ_{xy}m_z & iQ_{xz}m_y \\ iQ_{xy}m_z & 0 & -iQ_{yz}m_x \\ -iQ_{xz}m_y & iQ_{yz}m_x & 0 \end{pmatrix} \tag{5}$$

If the thickness d is small compared to optical wavelengths λ, such a surface magneto-optic effect on reflection is reduced from the bulk magneto-optic effect by another factor of d/λ. Equation (5) can be extended to a stack of ultrathin layers, each having its respective thickness, $Q_{\alpha\beta}$ values, and magnetization [12] and their effects on optical reflection is additive.

Electric dipole response from a surface layer The surface region of a crystalline material is usually different in composition, structure and crystalline symmetry from those in the bulk and thus has a distinct electric dipole response. The dielectric tensor for the surface region is generally expressed as

$$\overleftrightarrow{\epsilon}^{(surf)} = \begin{pmatrix} \epsilon_{xx}^{(s)} & \epsilon_{xy}^{(s)} & \epsilon_{xz}^{(s)} \\ \epsilon_{yx}^{(s)} & \epsilon_{yy}^{(s)} & \epsilon_{yz}^{(s)} \\ \epsilon_{zx}^{(s)} & \epsilon_{zy}^{(s)} & \epsilon_{zz}^{(s)} \end{pmatrix} \tag{6}$$

To treat the surface effect as a perturbation (the theme of this work), I replace the surface region of thickness d with a bulk material of same thickness. The surface effect on reflection and transmission is incorporated by adding a correction to the dielectric tensor in this region as follows,

$$\Delta\overleftrightarrow{\epsilon}^{(surf)}(z) = d\delta(z)\Delta\overleftrightarrow{\epsilon}^{(surf)} = d\delta(z)\begin{pmatrix} \epsilon_{xx}^{(s)} - \epsilon_{xx}^{(0)} & \epsilon_{xy}^{(s)} & \epsilon_{xz}^{(s)} \\ \epsilon_{yx}^{(s)} & \epsilon_{yy}^{(s)} - \epsilon_{yy}^{(0)} & \epsilon_{yz}^{(s)} \\ \epsilon_{zx}^{(s)} & \epsilon_{zy}^{(s)} & \epsilon_{zz}^{(s)} - \epsilon_{zz}^{(0)} \end{pmatrix} \tag{7}$$

I should note here that the magnitude of $\Delta\overleftrightarrow{\epsilon}^{(surf)}$ needs not to be small compared to that of $\overleftrightarrow{\epsilon}^{(0)}$. The effect on reflection and transmission though is small by the factor of d/λ.

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Electric quadrupole effects Beyond the electric dipole approximation, electric quadrupole responses to the gradient of the electromagnetic field produce effects comparable to that from a surface layer [3]. The quadruple response is prescribed by a polarization vector as follows [1],

$$\Delta\vec{P}^{(EQ)} = \varepsilon_0 \overleftrightarrow{\chi}_{EQ} : \nabla\vec{E} = \varepsilon_0 \left(i\overleftrightarrow{\chi}_{EQ} : \vec{k} \right) : \vec{E} \tag{8}$$

This introduces a correction to the zeroth order dielectric tensor,

$$\Delta\overleftrightarrow{\epsilon}^{(EQ)} = i\overleftrightarrow{\chi}_{EQ} : \vec{k} \tag{9}$$

$\overleftrightarrow{\chi}_{EQ}$ is a second-rank tensor and vanishes if the material has an inversion center. $\vec{k}$ is the wave vector of the electromagnetic field. For example, GaAs belongs to the cubic crystal system and has the T_d symmetry. In the principal coordinate frame, the electric quadrupole correction to the dielectric tensor is [3,28]

$$\Delta\overleftrightarrow{\epsilon}^{(EQ)} = i\chi_{EQ,xyz} \begin{pmatrix} 0 & k_z & k_y \\ k_z & 0 & k_x \\ k_y & k_x & 0 \end{pmatrix} = id_{14} \begin{pmatrix} 0 & k_z & k_y \\ k_z & 0 & k_x \\ k_y & k_x & 0 \end{pmatrix} \tag{10}$$

The magnitude of $\overleftrightarrow{\chi}_{EQ}$ is of the order of $\overleftrightarrow{\epsilon}^{(0)}$ multiplied by the Bohr radius a_B. This means $\Delta\overleftrightarrow{\epsilon}^{(EQ)} \sim (2\pi a_B/\lambda)\overleftrightarrow{\epsilon}^{(0)} < 0.01\ \overleftrightarrow{\epsilon}^{(0)}$ or smaller. For more detailed estimates, I note that d_{14} defined in Eq. (10) has the unit of meter and is roughly equal to the second-order nonlinear susceptibility $d_{14}^{(2)}$ in unit of m/V multiplied by $\hbar\omega/e$. At a wavelength $\lambda = 0.6\ \mu m$ or $\hbar\omega = 2\ eV$, $d_{14} \cong 7.4 \times 10^{-10}\ m$ for GaAs [28].

Corrections to the dielectric tensors due to electro-optic effect and photo-elastic effect have been described in the literature [9]. In Table 1, I list effects on optical reflection from various linear optical processes beyond the leading order electric dipole response.

Table 1. Magnitudes of effects on optical reflection from various linear optical processes in materials beyond the bulk electric dipole response.

	Change in dielectric tensor	Strength relative to bulk electric dipole response	Estimate of $\Delta r/r^{(0)}$
Magneto-optic Kerr effect (MOKE)	$\Delta\overleftrightarrow{\epsilon}^{(MOKE)}$	Q (Voigt vector)	$\mathbf{10^{-2} \sim 10^{-5}}$
Surface magneto-optic Kerr effect (SMOKE)	$\Delta\overleftrightarrow{\epsilon}^{(MOKE)} d\delta(z)$	$\left(\frac{2\pi d}{\lambda}\right) Q$	$\mathbf{10^{-4} \sim 10^{-8}}$
Surface electric dipole effect	$\Delta\overleftrightarrow{\epsilon}^{(surf)} d\delta(z)$	$\frac{2\pi d}{\lambda}$	$\mathbf{10^{-2} \sim 10^{-5}}$
Electric quadrupole effect (EQ)	$\Delta\overleftrightarrow{\epsilon}^{(EQ)}$	$\frac{2\pi a_B}{\lambda}$	$\mathbf{10^{-3} \sim 10^{-5}}$
Magnetic dipole effect (MD)	$\Delta\overleftrightarrow{\epsilon}^{(MD)}$	$\alpha = \frac{v_e}{c}$	$\mathbf{10^{-2} \sim 10^{-3}}$
Photo-elastic effect	$\Delta\overleftrightarrow{\epsilon}^{(ER)}$	Strain-optical constant	$\mathbf{10^{-2} \sim 10^{-3}}$

3. First-order modifications to reflection and transmission

Since modifications to reflection and transmission are expected to be small from optical processes beyond the electric dipole process in the bulk, a perturbation calculation of such modifications to the first-order is sufficient. I describe such a perturbation calculation, following the work of Heinz on optical second-harmonic generation [29].

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The key is to compute radiation fields produced by an extra polarization associated with an optical process of interest to the first order and then add them to reflection and transmission. Since additional radiation fields are small, I only consider the extra polarization induced by the zero-the order electric fields. Zeroth order fields are easily found for biaxial/uniaxial/isotropic materials in which dielectric tensors are diagonal in the principal frame. Furthermore, only boundary conditions up to the "first-order" electromagnetic fields need to be matched. This leaves out the second and higher order corrections and thus greatly simplifies the calculation.

To calculate the radiation fields from an extra polarization, I adopt the method that Heinz used to compute radiation fields from a time-varying second-order nonlinear polarization sheet [29]. Only in second-harmonic generation, there are essentially no leading order reflection and transmission [28,30,31]. This method enables computing corrections to reflection and transmission to the first order and the corrections satisfy the boundary condition to the first-order.

I start with an isotropic material or a materials of cubic symmetry group. It has dielectric constant ϵ_2 and occupies the semi-infinite space $z>0$. The semi-infinite space with $z<0$ is filled with an isotropic ambient with dielectric constant ϵ_1. **Fig. 1** shows the laboratory coordinate frame and choices of s-polarization and p-polarization for electric fields in both media. Linear optical processes under consideration in this work are confined to the material of ϵ_2. A monochromatic light beam with $\vec{E}_1^{(+)} exp[ik_{1x}^{(+)}x + ik_{1z}^{(+)}z - i\omega t]$ is incident on the surface from ϵ_1 (i.e., $z<0$). The zeroth order electric fields in reflection and transmission are $\vec{E}_1^{(-)} exp[ik_{1x}^{(-)}x + ik_{1z}^{(-)}z - i\omega t]$ and $\vec{E}_2^{(+)} exp[ik_{2x}^{(+)}x + ik_{2z}^{(+)}z - i\omega t]$, respectively. They can be found easily through Fresnel equations. "+" and "-" in superscripts indicate signs of z-components of relevant wavevectors [20]. They are defined as follows, along with the Snell's law,

$$k_{1x}^{(+)} = k_{2x}^{(+)} = k_{1x}^{(-)} = (2\pi\sqrt{\epsilon_1}/\lambda)\sin\theta_1 \tag{11}$$

$$k_{1z}^{(+)} = +\sqrt{(2\pi/\lambda)^2\epsilon_1 - (k_{1x}^{(+)})^2} \tag{12a}$$

$$k_{1z}^{(-)} = -\sqrt{(2\pi/\lambda)^2\epsilon_1 - (k_{1x}^{(+)})^2} \tag{12b}$$

$$k_{2z}^{(+)} = +\sqrt{(2\pi/\lambda)^2\epsilon_2 - (k_{1x}^{(+)})^2} \tag{12c}$$

Unit vectors for s-polarized and p-polarized components of the incident, reflected and transmitted beams in two media are chosen as follows,

$$\hat{e}_{1p}^{(+)} = (k_{1z}^{(+)}/k_1)\hat{x} - (k_{1x}^{(+)}/k_1)\hat{z} \tag{13a}$$

$$\hat{e}_{1p}^{(-)} = (k_{1z}^{(+)}/k_1)\hat{x} + (k_{1x}^{(+)}/k_1)\hat{z} \tag{13b}$$

$$\hat{e}_{2p}^{(+)} = (k_{2z}^{(+)}/k_2)\hat{x} - (k_{1x}^{(+)}/k_2)\hat{z} \tag{13c}$$

$$\hat{e}_{2p}^{(-)} = (k_{2z}^{(+)}/k_2)\hat{x} + (k_{1x}^{(+)}/k_2)\hat{z} \tag{13d}$$

$$\hat{e}_{1s}^{(+)} = \hat{e}_{1s}^{(-)} = \hat{e}_{2s}^{(+)} = \hat{e}_{2s}^{(-)} = \hat{y} \tag{13e}$$

In **Fig. 2**, I consider an infinitesimal layer of thickness dz' situated at $z'>0$. A linear polarization (not included in the zeroth order dielectric tensor) is induced in the layer by $\vec{E}_2^{(+)}$,

$$\Delta\vec{P}(z') = \varepsilon_0\Delta\overleftrightarrow{\epsilon} : \vec{E}_2^{(+)} exp[ik_{1x}^{(+)}x + ik_{2z}^{(+)}z' - i\omega t] \tag{14}$$

Such a polarization layer (or polarization sheet as it is sometimes called) produces two outward radiative beams. Since $\Delta\overleftrightarrow{\epsilon}$ is small compared to ϵ_2 in magnitude, the dielectric tensor of the

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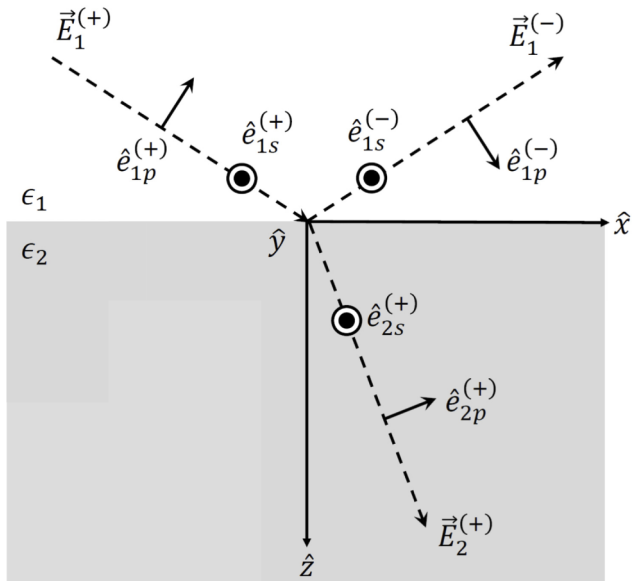

Fig. 1. Sketch of the incident, reflected, and transmitted electric fields in the laboratory coordinate frame. The incident electric field is in medium of ϵ_1. Choices of unit vectors for s-polarization and p-polarization are indicated.

layer for the purpose of the beam propagation is taken to be $\epsilon_2 \overleftrightarrow{I}$, consistent with dropping the second-order and higher-order terms. One beam, $\delta\vec{E}_2^{(-)}(\vec{r},t)$, propagates towards the surface [29],

$$\begin{aligned}\hat{e}_2^{(-)} \cdot \delta\vec{E}_2^{(-)}(\vec{r},t) &= i\frac{2\pi(\omega/c)^2}{4\pi\varepsilon_0 k_{2z}^{(+)}}(\hat{e}_2^{(-)} \cdot \Delta\vec{P}(z'))dz' exp[ik_{2z}^{(-)}z - ik_{2z}^{(-)}z'] \\ &= i\frac{2\pi(\omega/c)^2}{4\pi k_{2z}^{(+)}}\left(\hat{e}_2^{(-)} \cdot \Delta\overleftrightarrow{\epsilon} : \vec{E}_2^{(+)}\right) dz' exp[ik_{1x}^{(+)}x + ik_{2z}^{(-)}z - i\omega t + i(k_{2z}^{(+)} - k_{2z}^{(-)})z']\end{aligned} \tag{15a}$$

The other beam, $\delta\vec{E}_2^{(+)}(\vec{r},t)$, propagates away from the surface,

$$\begin{aligned}\hat{e}_2^{(+)} \cdot \delta\vec{E}_2^{(+)}(\vec{r},t) &= i\frac{2\pi(\omega/c)^2}{4\pi\varepsilon_0 k_{2z}^{(+)}}(\hat{e}_2^{(+)} \cdot \Delta\vec{P}(z'))dz' exp[ik_{2z}^{(+)}z - ik_{2z}^{(+)}z']\, dz' \\ &= i\frac{2\pi(\omega/c)^2}{4\pi k_{2z}^{(+)}}\left(\hat{e}_2^{(+)} \cdot \Delta\overleftrightarrow{\epsilon} : \vec{E}_2^{(+)}\right) dz' exp[ik_{1x}^{(+)}x + ik_{2z}^{(+)}z - i\omega t]\end{aligned} \tag{15b}$$

The first beam contributes to a correction to reflection and transmission. The second beam contributes to a correction to transmission. In a magnetic material, the second beam yields the Faraday effect while the first yields the Kerr effect. I will focus on the correction to reflection.

Extra electric field in reflection produced by bulk polarizations If $\Delta\overleftrightarrow{\epsilon}$ exists throughout the medium ϵ_2, the total electric field propagating toward the surface in response to $\Delta\overleftrightarrow{\epsilon}$ is obtained

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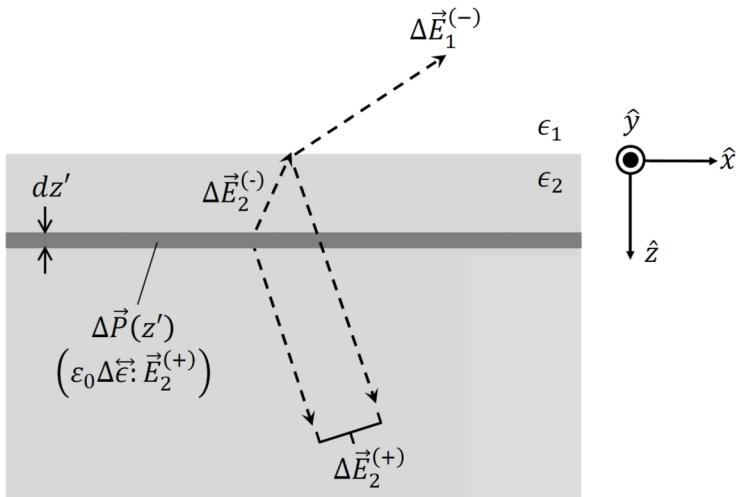

Fig. 2. Electric fields produced by a thin sheet of polarization $\Delta\vec{P}(z')$ with thickness dz' in medium ϵ_2. These fields yield corrections to the reflected field back in medium ϵ_1, $\Delta\vec{E}_1^{(-)}(\vec{r},t)$, and the transmitted field in medium ϵ_2, $\Delta\vec{E}_2^{(+)}(\vec{r},t)$, away from the surface.

by integrating Eq. (15a) over $z'>0$, recalling $k_{2z}^{(-)} = -k_{2z}^{(+)}$,

$$\begin{aligned}\hat{e}_2^{(-)}\cdot\Delta\vec{E}_2^{(-)}(\vec{r},t) &= \int_0^{+\infty} i\frac{2\pi(\omega/c)^2}{4\pi\varepsilon_0 k_{2z}^{(+)}}\hat{e}_2^{(-)}\cdot\Delta\vec{P}(z')dz' exp[ik_{2z}^{(-)}z - ik_{2z}^{(-)}z'] \\ &= (-)\left[\frac{(\omega/c)^2}{2k_{2z}^{(+)}}\right]\left[\frac{\hat{e}_2^{(-)}\cdot\left(\Delta\overleftrightarrow{\epsilon}:\vec{E}_2^{(+)}\right)}{2k_{2z}^{(+)}}\right] exp[ik_{1x}^{(+)}x + ik_{2z}^{(-)}z - i\omega t]\end{aligned} \tag{16}$$

After transmission into the ambient of ϵ_1, the extra electric field $\Delta\vec{E}_1^{(-)}(\vec{r},t)$ propagating in the direction of specular reflection is the correction to reflection. $\Delta\vec{E}_1^{(-)}(\vec{r},t)$ is found from $\Delta\vec{E}_2^{(-)}(\vec{r},t)$, using the method and notation of Heinz [29], as follows,

$$\hat{e}_1^{(-)}\cdot\Delta\vec{E}_1^{(-)}(\vec{r},t) = (-)\left[\frac{(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_1^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)\cdot\left(\Delta\overleftrightarrow{\epsilon}:\vec{E}_2^{(+)}\right)}{2k_{2z}^{(+)}}\right] exp[ik_{1x}^{(+)}x + ik_{1z}^{(-)}z - i\omega t] \tag{17}$$

$\overleftrightarrow{F}^{(1\to2)}$ is the local-field factor tensor. It consists of transmission coefficients for cartesian components of the electric field going from medium ϵ_1 into medium ϵ_2,

$$\overleftrightarrow{F}^{(1\to2)} = \sum F_{jj}^{(1\to2)}\hat{j}\hat{j} = \hat{x}\hat{x}\frac{2\epsilon_1 k_{2z}^{(+)}}{\epsilon_1 k_{2z}^{(+)} + \epsilon_2 k_{1z}^{(+)}} + \hat{y}\hat{y}\frac{2k_{1z}^{(+)}}{k_{2z}^{(+)} + k_{1z}^{(+)}} + \hat{z}\hat{z}\frac{2\epsilon_1 k_{1z}^{(+)}}{\epsilon_1 k_{2z}^{(+)} + \epsilon_2 k_{1z}^{(+)}} \tag{18}$$

so that

$$\vec{E}_{2p}^{(+)} = \hat{e}_{2p}^{(+)}E_{2p}^{(+)} = \overleftrightarrow{F}^{(1\to2)} : (\hat{e}_{1p}^{(+)}E_{1p}^{(+)}) \tag{19}$$

$$\vec{E}_{2s}^{(+)} = \hat{e}_{2s}^{(+)}E_{2s}^{(+)} = \overleftrightarrow{F}^{(1\to2)} : (\hat{e}_{1s}^{(+)}E_{1s}^{(+)}) \tag{20}$$

For biaxial and uniaxial materials, Eq. (18) is slightly modified (see **Appendix A**).

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Equation (17) (dropping the phase factor now) can be specialized to yield the correction to the s-polarized electric field in reflection, using Eq. (13e),

$$\Delta E_{1s}^{(-)} = \left[\frac{(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{-1}{2k_{2z}^{(+)}}\right](F_{yy}^{(1\to 2)}\hat{y})\cdot\left(\Delta\overleftrightarrow{\epsilon}:\vec{E}_2^{(+)}\right) \tag{21}$$

and the correction to the p-polarized electric field in reflection, by using Eq. (13b),

$$\Delta E_{1p}^{(-)} = \left[\frac{(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{-1}{2k_{2z}^{(+)}}\right]\left(\left(\frac{k_{1z}^{(+)}}{k_1}\right)F_{xx}^{(1\to 2)}\hat{x}+\left(\frac{k_{1x}^{(+)}}{k_1}\right)F_{zz}^{(1\to 2)}\hat{z}\right)\cdot\left(\Delta\overleftrightarrow{\epsilon}:\vec{E}_2^{(+)}\right) \tag{22}$$

Extra electric field in reflection produced by a distinct surface layer Unlike optical processes in the bulk of material ϵ_2, the magnitude of $\Delta\overleftrightarrow{\epsilon}^{(surf)}$ in the surface layer can be comparable to or even larger than ϵ_2. For the purpose of finding the extra radiation from the surface layer, I first replace the layer with a bulk layer of same thickness and then treat the effect of the surface layer through $\Delta\overleftrightarrow{\epsilon}^{(surf)}(z) = d\delta(z')\Delta\overleftrightarrow{\epsilon}^{(surf)}$. Using Eq. (7), an extra polarization is induced by $\vec{E}_2^{(+)}$,

$$\Delta\vec{P}^{(surf)}(z') = \varepsilon_0 d\delta(z')\Delta\overleftrightarrow{\epsilon}^{(surf)}:\vec{E}_2^{(+)}exp[ik_{1x}^{(+)}x + ik_{2z}^{(+)}z' - i\omega t] \tag{23}$$

The two beams produced by $\Delta\vec{P}^{(surf)}(z')$ are initially inside the surface layer characterized by dielectric tensor $\overleftrightarrow{\epsilon}^{(surf)}$. For the purpose of beam propagation, I can no longer ignore the reflection at boundaries that separate the surface layer and the bulk of ϵ_2. The effect of such reflection on the electric fields emerging from two sides of the surface layer into ϵ_2 is easily calculated [29]. Let the surface layer be an isotropic material,

$$\overleftrightarrow{\epsilon}^{(surf)} = \begin{pmatrix} \epsilon_s & 0 & 0 \\ 0 & \epsilon_s & 0 \\ 0 & 0 & \epsilon_s \end{pmatrix} \tag{24}$$

$$\Delta\overleftrightarrow{\epsilon}^{(surf)} = \begin{pmatrix} \epsilon_s-\epsilon_2 & 0 & 0 \\ 0 & \epsilon_s-\epsilon_2 & 0 \\ 0 & 0 & \epsilon_s-\epsilon_2 \end{pmatrix}. \tag{25}$$

Produced by an infinitesimal layer of thickness dz' in the surface layer, a beam emerges from the surface layer and propagates in ϵ_2 towards the surface with an electric field

$$\hat{e}_2^{(-)}\cdot\delta\vec{E}_2^{(-)}(\vec{r},t) = i\frac{2\pi(\omega/c)^2[e_{2x}^{(-)}\hat{x}+e_{2y}^{(-)}\hat{y}+(\epsilon_2/\epsilon_s)e_{2z}^{(-)}\hat{z}]\cdot\Delta\vec{P}(z')dz'}{4\pi\varepsilon_0 k_{2z}^{(+)}}exp[ik_{2z}^{(-)}z - ik_{2z}^{(-)}z'] \tag{26}$$

Integrating Eq. (26) yields the total electric field produced by the surface layer that propagates in ϵ_2 toward the surface,

$$\hat{e}_2^{(-)}\cdot\Delta\vec{E}_2^{(-)} = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{2z}^{(+)}}\right][(e_{2x}^{(-)}\hat{x}+e_{2y}^{(-)}\hat{y}+({}^{\epsilon_2}/_{\epsilon_s})e_{2z}^{(-)}\hat{z})\cdot\vec{E}_2^{(+)}] \tag{27}$$

After transmission into the ambient ϵ_1, I arrive at the correction to the reflected electric field from the surface layer as follows,

$$\hat{e}_1^{(-)}\cdot\Delta\vec{E}_1^{(-)} = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right]\left[\left(\overleftrightarrow{F}^{(1\to 2)}\cdot\left(e_{1x}^{(-)}\hat{x}+e_{1y}^{(-)}\hat{y}+\left(\frac{\epsilon_2}{\epsilon_s}\right)e_{1z}^{(-)}\hat{z}\right)\right)\cdot\vec{E}_2^{(+)}\right] \tag{28}$$

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The correction to the s-polarized reflected electric field is

$$\Delta E_{1s}^{(-)} = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right](F_{yy}^{(1\to 2)}E_{2s}^{(+)}) = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right](F_{yy}^{(1\to 2)})^2E_{1s}^{(+)} \tag{29}$$

The correction to the p-polarized reflected electric field is

$$\begin{aligned}\Delta E_{1p}^{(-)} &= i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right]\left[\left(\frac{k_{1z}^{(+)}}{k_1}\right)F_{xx}^{(1\to 2)}\hat{x}+\left(\frac{k_{1x}^{(+)}\epsilon_2}{k_1\epsilon_s}\right)F_{zz}^{(1\to 2)}\hat{z}\right]\cdot\vec{E}_{2p}^{(+)}\\ &= i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right]\left[\left(\frac{k_{1z}^{(+)}}{k_1}\right)\left(\frac{k_{1z}^{(+)}}{k_1}\right)(F_{xx}^{(1\to 2)})^2-\left(\frac{\epsilon_2}{\epsilon_s}\right)\left(\frac{k_{1x}^{(+)}}{k_1}\right)^2(F_{zz}^{(1\to 2)})^2\right]E_{1p}^{(+)}\end{aligned} \tag{30}$$

The last step is done with the help of Eq. (13), (19) and (20).

Before going forward, I check some of the results obtained so far with the exact calculation [11]. Wong and Zhu calculated $\Delta_s \equiv \Delta E_{1s}^{(-)}/E_{1s}^{(-)}$ and $\Delta_p \equiv \Delta E_{1p}^{(-)}/E_{1p}^{(-)}$ from a surface layer by finding the full expression of s-polarized and p-polarized electric fields in reflection using a three-layer model. By keeping only terms that vary linearly with the thickness d, they found

$$\Delta_s \equiv \frac{\Delta E_{1s}^{(-)}}{E_{1s}^{(-)}} = i\left(\frac{2d\omega}{c}\right)\left(\frac{k_{1z}^{(+)}}{k_1}\right)\frac{(\epsilon_s-\epsilon_2)}{(\epsilon_s-\epsilon_1)} \tag{31}$$

$$\Delta_p \equiv \frac{\Delta E_{1p}^{(-)}}{E_{1p}^{(-)}} = i\left(\frac{2d\omega}{c}\right)\left(\frac{k_{1z}^{(+)}}{k_1}\right)\left[\frac{\epsilon_2^2(\epsilon_s-(k_{1x}^{(+)}/k_1)^2)-\epsilon_s^2(\epsilon_2-(k_{1x}^{(+)}/k_1)^2)}{\epsilon_s(\epsilon_2^2(k_{1z}^{(+)}/k_1)^2-\epsilon_2+(k_{1x}^{(+)}/k_1)^2)}\right] \tag{32}$$

Since $\Delta_s \equiv \Delta E_{1s}^{(-)}/E_{1s}^{(-)} = (\Delta E_{1s}^{(-)}/E_{1s}^{(+)})/r_{ss}^{(0)}$ and $\Delta_p \equiv \Delta E_{1p}^{(-)}/E_{1p}^{(-)} = (\Delta E_{1p}^{(-)}/E_{1p}^{(+)})/r_{pp}^{(0)}$, Eq. (29) and (30) reproduce Eq. (31) and (32) with the help of the zeroth-order reflection coefficients,

$$r_{ss}^{(0)} \equiv \frac{E_{1s}^{(-)}}{E_{1s}^{(+)}} = \frac{k_{1z}^{(+)}-k_{2z}^{(+)}}{k_{1z}^{(+)}+k_{2z}^{(+)}} \tag{33}$$

$$r_{pp}^{(0)} \equiv \frac{E_{1p}^{(-)}}{E_{1p}^{(+)}} = \frac{\epsilon_2k_{1z}^{(+)}-\epsilon_1k_{2z}^{(+)}}{\epsilon_2k_{1z}^{(+)}+\epsilon_1k_{2z}^{(+)}} \tag{34}$$

Combining Eq. (31) with (32), Zhu *et al.* further arrived at the oblique-incidence reflectivity difference defined as [16,17,24]

$$\Delta_p-\Delta_s = (-i)\frac{(2dk_{1z}^{(+)})(k_{1x}^{(+)})^2\epsilon_2(\epsilon_s-\epsilon_2)(\epsilon_s-\epsilon_1)}{[\epsilon_2(k_{1z}^{(+)})^2-\epsilon_1(k_{1x}^{(+)})^2]\epsilon_s(\epsilon_2-\epsilon_1)} \tag{35}$$

4. Modifications to reflection matrices

Extra radiative electric fields in reflection modify the reflection matrix [4]. The latter relates p-polarized and s-polarized components of the reflected electric field to the incident electric field,

$$\begin{pmatrix} E_{1p}^{(-)} \\ E_{1s}^{(-)} \end{pmatrix} = R\begin{pmatrix} E_{1p}^{(+)} \\ E_{1s}^{(+)} \end{pmatrix} = \begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix}\begin{pmatrix} E_{1p}^{(+)} \\ E_{1s}^{(+)} \end{pmatrix} \tag{36}$$

Equation (36) is suitable as long as the ambient in z < 0 is isotropic. By choosing the laboratory coordinate frame to overlap with the principal coordinate frame of medium $\overleftrightarrow{\epsilon}_2$, s-polarized and

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p-polarized electric fields are also eigen modes in $\overleftrightarrow{\epsilon}_2$ to the zeroth order and thus the zeroth order reflection matrix is diagonal,

$$\begin{pmatrix} E_{1p}^{(-)} \\ E_{1s}^{(-)} \end{pmatrix} = R^{(0)} \begin{pmatrix} E_{1p}^{(+)} \\ E_{1s}^{(+)} \end{pmatrix} = \begin{pmatrix} r_{pp}^{(0)} & 0 \\ 0 & r_{ss}^{(0)} \end{pmatrix} \begin{pmatrix} E_{1p}^{(+)} \\ E_{1s}^{(+)} \end{pmatrix} \tag{37}$$

Electric fields produced by additional optical responses, $\Delta E_{1p}^{(-)}$ and $\Delta E_{1s}^{(-)}$, add corrections to the reflection matrix

$$R = R^{(0)} + \Delta R = \begin{pmatrix} r_{pp}^{(0)} + \Delta r_{pp} & \Delta r_{ps} \\ \Delta r_{sp} & r_{ss}^{(0)} + \Delta r_{ss} \end{pmatrix} \tag{38}$$

The key advantages of the present perturbation calculation are that (a) corrections due to extra optical processes in the bulk can be generally found from Eq. (21) and (22), and (b) corrections due to optical processes from an isotropic surface layer can be extracted from Eq. (29) and (30). Specifically, with the help of Eq. (19) and (20), Eq. (21) and (22) become

$$\Delta E_{1p}^{(-)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1p}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1p}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)E_{1p}^{(+)} + \left(\hat{e}_{1p}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1s}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)E_{1s}^{(+)}}{2k_{2z}^{(+)}}\right] \tag{39}$$

$$\Delta E_{1s}^{(-)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1s}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1p}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)E_{1p}^{(+)} + \left(\hat{e}_{1s}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1s}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)E_{1s}^{(+)}}{2k_{2z}^{(+)}}\right] \tag{40}$$

I arrive at corrections to the reflection matrix due to extra optical processes in the bulk as follows,

$$\Delta r_{pp} \equiv \frac{E_{1p}^{(-)}}{E_{1p}^{(+)}} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1p}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1p}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)}{2k_{2z}^{(+)}}\right] \tag{41}$$

$$\Delta r_{ss} = \frac{E_{1s}^{(-)}}{E_{1s}^{(+)}} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1s}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1s}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)}{2k_{2z}^{(+)}}\right] \tag{42}$$

$$\Delta r_{ps} = \frac{E_{1p}^{(-)}}{E_{1s}^{(+)}} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1p}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1s}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)}{2k_{2z}^{(+)}}\right] \tag{43}$$

$$\Delta r_{sp} = \frac{E_{1s}^{(-)}}{E_{1p}^{(+)}} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{\left(\hat{e}_{1s}^{(-)}\cdot\overleftrightarrow{F}^{(1\to2)}\right):\Delta\overleftrightarrow{\epsilon}:\left(\hat{e}_{1p}^{(+)}\cdot\overleftrightarrow{F}^{(1\to2)}\right)}{2k_{2z}^{(+)}}\right] \tag{44}$$

The corrections due to optical processes from an isotropic surface layer from Eq. (29) and (30) are as follows,

$$\Delta r_{pp}^{(surf)} = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right]\left[\left(\frac{k_{1z}^{(+)}}{k_1}\right)\left(\frac{k_{1z}^{(+)}}{k_1}\right)\left(F_{xx}^{(1\to2)}\right)^2 - \left(\frac{\epsilon_2}{\epsilon_s}\right)\left(\frac{k_{1x}^{(+)}}{k_1}\right)^2\left(F_{zz}^{(1\to2)}\right)^2\right] \tag{45}$$

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$$\Delta r_{ss}^{(surf)} = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right](F_{yy}^{(1\to2)}E_{2s}^{(+)}) = i\left[\frac{(\omega/c)^2(\epsilon_s-\epsilon_2)d}{2k_{1z}^{(+)}}\right](F_{yy}^{(1\to2)})^2 \tag{46}$$

Equation (41)–(46) are general results of this work. They can be used to analyze a wide range of linear optical processes. I now apply these results to a few specific optical processes.

Surface/Thin film effect due to a distinct surface layer or a stack of ultrathin films I showed in the preceding section that Eq. (45) and (46) reproduce the Taylor expansion of an exact calculation given by Eq. (31) and (32). In terms of modification to the reflection matrix, I have [17,24]

$$R^{(surf)} = \begin{pmatrix} r_{pp}^{(0)} + \Delta r_{pp}^{(surf)} & 0 \\ 0 & r_{ss}^{(0)} + \Delta r_{ss}^{(surf)} \end{pmatrix} \equiv \begin{pmatrix} r_{pp}^{(0)}(1+\Delta_p) & 0 \\ 0 & r_{ss}^{(0)}(1+\Delta_s) \end{pmatrix} \tag{47}$$

For a stack of ultrathin films, as long as the total thickness of the stack is less than the optical wavelength, effects from the films are simply additive. If the surface layer is anisotropic and its principal coordinate frame does not overlap with the laboratory frame, there can be off-diagonal terms.

Magneto-optic Kerr effects (MOKE) from the bulk $\Delta\overleftrightarrow{\epsilon}^{(MOKE)}$ due to a magnetization present in material of $\overleftrightarrow{\epsilon}_2$ is given by Eq. (4). Since I have chosen the laboratory x-z plane as the incidence plane, x and z components of the magnetization, m_x and m_z, couple the s-polarized components of the electric fields with the p-polarized components. Thus Δr_{ps} and Δr_{sp} have the longitudinal Kerr effect (from m_x) and the polar Kerr effect (from m_z). The y component of the magnetization, m_y, only couples the p-polarized components of the electric fields and therefore only Δr_{pp} has the transverse Kerr effect (from m_y). From Eq. (41) through (44) and Eq. (4), I arrive at

$$\Delta r_{ss}^{(MOKE)} = 0 \tag{48}$$

$$\Delta r_{pp}^{(MOKE)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{(e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1p,z}^{(+)}F_{zz}^{(1\to2)} - e_{1p,z}^{(-)}F_{zz}^{(1\to2)}e_{1p,x}^{(+)}F_{xx}^{(1\to2)})(iQ_{zx}m_y)}{2k_{2z}^{(+)}}\right] \tag{49}$$

$$\Delta r_{ps}^{(MOKE)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(-iQ_{xy}m_z) + e_{1p,z}^{(-)}F_{zz}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(iQ_{zy}m_x)}{2k_{2z}^{(+)}}\right] \tag{50}$$

$$\Delta r_{sp}^{(MOKE)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,x}^{(+)}F_{xx}^{(1\to2)}(iQ_{xy}m_z) + e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,z}^{(+)}F_{zz}^{(1\to2)}(-iQ_{yz}m_x)}{2k_{2z}^{(+)}}\right] \tag{51}$$

By writing corrections to the reflection matrix in form of

$$\Delta r_{pp}^{(MOKE)} = \alpha_y m_y \tag{52}$$

$$\Delta r_{ps}^{(MOKE)} = \alpha_x m_x + \alpha_z m_z \tag{53}$$

$$\Delta r_{sp}^{(MOKE)} = \alpha_x m_x - \alpha_z m_z \tag{54}$$

I arrive at a familiar form of reflection matrix for MOKE [20,32,33],

$$R^{(MOKE)} = \begin{pmatrix} r_{pp}^{(0)} + \alpha_y m_y & \alpha_x m_x + \alpha_z m_z \\ \alpha_x m_x - \alpha_z m_z & r_{ss}^{(0)} \end{pmatrix} \tag{55}$$

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Coefficients in the matrix are as follows,

$$\alpha_y = \left[\frac{(-iQ_{zx})(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{2e_{1p,x}^{(-)}e_{1p,z}^{(+)}F_{xx}^{(1\to2)}F_{zz}^{(1\to2)}}{2k_{2z}^{(+)}}\right] \tag{56}$$

$$\alpha_x = \left[\frac{(-iQ_{yz})(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,z}^{(-)}F_{zz}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}}{2k_{2z}^{(+)}}\right] \tag{57}$$

$$\alpha_z = \left[\frac{(-iQ_{xy})(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{-e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}}{2k_{2z}^{(+)}}\right] \tag{58}$$

For magnetic materials that are isotropic or belong to cubic symmetry group, $Q_{xy} = Q_{yz} = Q_{zx} \equiv \epsilon_2 Q$ as is customarily done in the literature [12,20], Eq. (56) – (58) become

$$\alpha_y = +\left[\frac{(iQ)\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1x}^{(+)}k_{1z}^{(+)}F_{xx}^{(1\to2)}F_{zz}^{(1\to2)}}{k_{2z}^{(+)}k_1^2}\right] \tag{59}$$

$$\alpha_x = -\left[\frac{(iQ)\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1x}^{(+)}F_{yy}^{(1\to2)}F_{zz}^{(1\to2)}}{2k_{2z}^{(+)}k_1}\right] \tag{60}$$

$$\alpha_z = +\left[\frac{(iQ)\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1z}^{(+)}F_{yy}^{(1\to2)}F_{xx}^{(1\to2)}}{2k_{2z}^{(+)}k_1}\right] \tag{61}$$

These equations reproduce the findings of Hunt [20] who performed the exact calculation and the Taylor expansion to arrive at his reflection matrix. In **Appendix B**, I show how Eq. (55)–(61) are converted to results in the literatures [20,32,33,34]. For completeness, I show in **Appendix C** that the perturbation treatment of the magneto-optic effect on optical transmission yields the Faraday rotation.

Surface magneto-optic Kerr effects (SMOKE) When the magneto-optic effect is confined to a thin surface layer such that $\Delta\overleftrightarrow{\epsilon}^{(SMOKE)} = d\delta(z)\Delta\overleftrightarrow{\epsilon}^{(MOKE)}$, the reflection matrix after modification has the same form as Eq. (55),

$$R^{(SMOKE)} = \begin{pmatrix} r_{pp}^{(0)} + \alpha_y^{(surf)}m_y & \alpha_x^{(surf)}m_x + \alpha_z^{(surf)}m_z \\ \alpha_x^{(surf)}m_x - \alpha_z^{(surf)}m_z & r_{ss}^{(0)} \end{pmatrix} \tag{62}$$

except that coefficients are now given by

$$\alpha_y^{(surf)} = (-i2dk_{2z}^{(+)})\alpha_y = +\left[\frac{2Qd\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1x}^{(+)}k_{1z}^{(+)}F_{xx}^{(1\to2)}F_{zz}^{(1\to2)}}{k_1^2}\right] \tag{63}$$

$$\alpha_x^{(surf)} = (-i2dk_{2z}^{(+)})\alpha_x = -\left[\frac{Qd\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1x}^{(+)}F_{yy}^{(1\to2)}F_{zz}^{(1\to2)}}{k_1}\right] \tag{64}$$

$$\alpha_z^{(surf)} = (-i2dk_{2z}^{(+)})\alpha_z = +\left[\frac{Qd\epsilon_2(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{k_{1z}^{(+)}F_{yy}^{(1\to2)}F_{xx}^{(1\to2)}}{k_1}\right] \tag{65}$$

For a stack of magnetic layers, as long as the total thickness is less than the optical wavelength, effects are additive such that Eq. (63) - (65) are replaced with sums of coefficients from each layer [12].

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Electric quadrupole effects (EQ) in the bulk For materials that lack inversion centers, the electric quadrupole response exists and produces $\Delta\overleftrightarrow{\epsilon}^{(EQ)} = i\overleftrightarrow{\chi}_{EQ} : \vec{k}$ where $\overleftrightarrow{\chi}_{EQ}$ is a second-rank tensor. In the principal coordinate frame, non-vanishing elements of $\overleftrightarrow{\chi}_{EQ}$ depend on the crystalline symmetry. For GaAs having T_d symmetry, only 6 of 27 elements in $\overleftrightarrow{\chi}_{EQ}$ are non-zero [28] and they are equal in magnitude. $\Delta\overleftrightarrow{\epsilon}^{(EQ)}$ is given by Eq. (10) in the principal frame,

$$\Delta\overleftrightarrow{\epsilon}^{(EQ)} = i\chi_{EQ,xyz}\begin{pmatrix} 0 & k_{2z}^{(+)} & k_{2y}^{(+)} \\ k_{2z}^{(+)} & 0 & k_{2x}^{(+)} \\ k_{2y}^{(+)} & k_{2x}^{(+)} & 0 \end{pmatrix} = id_{14}\begin{pmatrix} 0 & k_{2z}^{(+)} & k_{2y}^{(+)} \\ k_{2z}^{(+)} & 0 & k_{2x}^{(+)} \\ k_{2y}^{(+)} & k_{2x}^{(+)} & 0 \end{pmatrix} \tag{66}$$

Corrections to the reflection matrix is similar to magneto-optic effects. By overlapping the principal coordinate frame with the laboratory frame (see **Fig. 1**), namely on a GaAs(001) surface with the crystalline axes parallel to the x-axis and y-axis, $k_{2y}^{(+)} = 0$, and I arrive at

$$\Delta r_{ss}^{(EQ)} = 0 \tag{67}$$

$$\Delta r_{pp}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{(e_{1p,x}^{(-)}e_{1p,z}^{(+)} + e_{1p,z}^{(-)}e_{1p,x}^{(+)})F_{xx}^{(1\to2)}F_{zz}^{(1\to2)}(id_{14}k_{2y}^{(+)})}{2k_{2z}^{(+)}}\right] = 0 \tag{68}$$

$$\Delta r_{ps}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(id_{14}k_{2z}^{(+)}) + e_{1p,z}^{(-)}F_{zz}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(id_{14}k_{2x}^{(+)})}{2k_{2z}^{(+)}}\right] \tag{69}$$

$$\Delta r_{sp}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,x}^{(+)}F_{xx}^{(1\to2)}(id_{14}k_{2z}^{(+)}) + e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,z}^{(+)}F_{zz}^{(1\to2)}(id_{14}k_{2x}^{(+)})}{2k_{2z}^{(+)}}\right] \tag{70}$$

When the GaAs crystalline sample rotates counter-clockwise about the z-axis of both coordinate frame by an angle ϕ, the correction to the dielectric tensor becomes

$$\Delta\overleftrightarrow{\epsilon}^{(EQ)}(\phi) = id_{14}\begin{pmatrix} k_{2z}^{(+)}\sin 2\phi & k_{2z}^{(+)}\cos 2\phi & k_{2x}^{(+)}\sin 2\phi \\ k_{2z}^{(+)}\cos 2\phi & -k_{2z}^{(+)}\sin 2\phi & k_{2x}^{(+)}\cos 2\phi \\ k_{2x}^{(+)}\sin 2\phi & k_{2x}^{(+)}\cos 2\phi & 0 \end{pmatrix} \tag{71}$$

This yields corrections in the reflection matric as follows

$$\Delta r_{ss}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{F_{yy}^{(1\to2)}F_{yy}^{(1\to2)}(-id_{14}\,k_{2z}^{(+)})}{2k_{2z}^{(+)}}\right]\sin 2\phi \tag{72}$$

$$\Delta r_{pp}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{F_{xx}^{(1\to2)}e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1p,x}^{(+)}(id_{14}k_{2z}^{(+)})}{k_{2z}^{(+)}}\right]\sin 2\phi \tag{73}$$

$$\Delta r_{ps}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,x}^{(-)}F_{xx}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(id_{14}k_{2z}^{(+)}) + e_{1p,z}^{(-)}F_{zz}^{(1\to2)}e_{1s}^{(+)}F_{yy}^{(1\to2)}(id_{14}k_{2x}^{(+)})}{2k_{2z}^{(+)}}\right]\cos 2\phi \tag{74}$$

$$\Delta r_{sp}^{(EQ)} = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,x}^{(+)}F_{xx}^{(1\to2)}(id_{14}k_{2z}^{(+)}) + e_{1s}^{(-)}F_{yy}^{(1\to2)}e_{1p,z}^{(+)}F_{zz}^{(1\to2)}(id_{14}k_{2x}^{(+)})}{2k_{2z}^{(+)}}\right]\cos 2\phi \tag{75}$$

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At normal incidence, $k_{2x}^{(+)} = k_{1x}^{(+)} = 0$ and $k_{2z}^{(+)} = k_2^{(+)}$, $e_{1p,x}^{(-)} e_{1p,x}^{(+)} = e_{1p,x}^{(-)} e_{1s}^{(+)} = e_{1s}^{(-)} e_{1p,x}^{(+)} = 1$, $(F_{xx}^{(1\to 2)})^2 = (F_{yy}^{(1\to 2)})^2 = F_{xx}^{(1\to 2)} F_{yy}^{(1\to 2)}$, Eq. (72) - (75) become

$$\Delta r_{ss}^{(EQ)}(\phi) = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1s}^{(-)} e_{1s}^{(+)} (F_{yy}^{(1\to 2)})^2 (-id_{14}\, k_{2z}^{(+)})}{2k_{2z}^{(+)}}\right] \sin 2\phi \equiv a \sin 2\phi \tag{72a}$$

$$\Delta r_{pp}^{(EQ)}(\phi) = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,x}^{(-)} e_{1p,x}^{(+)} (F_{xx}^{(1\to 2)})^2 (id_{14} k_{2z}^{(+)})}{k_{2z}^{(+)}}\right] \sin 2\phi \equiv -a \sin 2\phi \tag{73a}$$

$$\Delta r_{ps}^{(EQ)}(\phi) = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1p,x}^{(-)} e_{1s}^{(+)} F_{xx}^{(1\to 2)} F_{yy}^{(1\to 2)} (id_{14} k_{2z}^{(+)})}{2k_{2z}^{(+)}}\right] \cos 2\phi \equiv -a \cos 2\phi \tag{74a}$$

$$\Delta r_{sp}^{(EQ)}(\phi) = \left[\frac{(-)(\omega/c)^2}{2k_{1z}^{(+)}}\right]\left[\frac{e_{1s}^{(-)} e_{1p,x}^{(+)} F_{yy}^{(1\to 2)} F_{xx}^{(1\to 2)} (id_{14} k_{2z}^{(+)})}{2k_{2z}^{(+)}}\right] \cos 2\phi \equiv -a \cos 2\phi \tag{75a}$$

and $a = \left[\frac{id_{14}\,(\omega/c)^2 F_{yy}^{(1\to 2)} F_{yy}^{(1\to 2)}}{4k_1^{(+)}}\right] = \frac{id_{14}\,(\omega/c) n_1}{(n_1+n_2)^2} = \frac{id_{14}\,(2\pi/\lambda) n_1}{(n_1+n_2)^2}$. These results led to Eq. (6) in the work reported by Zhu and Zhang [3] with $r_{pp}^{(0)}(\theta_1 = 0) \equiv r_0$.

5. Discussion and conclusion

Putting aside for the moment the algebra and multitude of equations, the essential message is that first-order effects from optical processes beyond the electric dipole response in the bulk of a material can be treated as a perturbation so that one really does not need to find exact electric fields in transmission and reflection. The exact approach can be tedious and often needs to be repeated when a new optical process is under consideration. As I have shown here, given corrections to the zeroth order dielectric tensor $\Delta\overleftrightarrow{\epsilon}$ from an optical process, the corresponding modification to the reflection matrix can be found generally from Eq. (41) through (44). If the optical process is confined to a surface layer, the modification is given by Eq. (45) and (46). By dropping off higher order contributions as is usually justified, it is appealing to see clearly how the perturbation to optical reflection is originated, how the resultant electric field emerges in the direction of specular reflection, and the parameters that determine the eventual magnitude of the effect. The simplicity of such an approach is appealing. It is noteworthy that Eq. (41) - (46) are mostly properties of zeroth-order electric fields except for $\Delta\overleftrightarrow{\epsilon}$, and these properties determine how an optical process changes the reflection in details. For example, a thin film sample and a bulk sample of an otherwise same magnetic material can produce different Kerr effects that are experimentally relevant. Consider the magneto-optic Kerr effect (MOKE) from a transparent ferromagnetic magnetic material such as yttrium iron garnet (YIG) at $\lambda = 780$ nm with a real magneto-optic parameter Q. From the surface of a bulk YIG crystal (with the thickness much larger than the wavelength), the measurable Kerr effect for an s-polarized light is given by Eq. (59),

$$\phi_s \equiv \frac{\alpha_y m_y}{r_{ss}^{(0)}} = \phi_s' + i\phi_s'' \tag{76}$$

The real part (ϕ_s') is the Kerr rotation while the imaginary part is the Kerr ellipticity (ϕ_s''). From Eq. (59) and the fact that Q is a real quantity, a magnetized YIG bulk crystal only yields a Kerr ellipticity ϕ_s'' but no Kerr rotation effect. If instead we have a thin YIG crystalline film

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with a thickness much less than the optical wavelength on a transparent non-magnetic substrate such as gadolinium gallium garnet (GGG), the Kerr effect is given by Eq. (63),

$$\phi_s^{(Surf)} \equiv \frac{\alpha_y^{(Surf)} m_y}{r_{ss}^{(0)}} = \phi_s^{(Surf)\prime} + i\phi_s^{(Surf)\prime\prime} \tag{77}$$

In this case, a magnetized YIG thin film produces a Kerr rotation $\phi_s^{(Surf)\prime}$ and exhibits no Kerr ellipticity effect. We have experimentally confirmed these different Kerr effects from YIG bulk crystals and a 150-nm YIG film on GGG.

The perturbation method also significantly simplifies treatments of the Faraday effect, magnetic circular dichroism, and the Voigt effect (i.e., quadratic magneto-optic effect). In **Appendix C**, I present a detailed perturbation calculation of the Faraday effect in a transparent magnetic material on optical transmission. Furthermore, I generalize the perturbation treatment to materials in which both magnetic circular birefringence and magnetic circular dichroism are present. These two effects can be separately measured using a suitable form of polarization-modulated transmission ellipsometry. Finally, I briefly discuss the treatment of the Voigt effect (the leading order non-linear magneto-optic effect) on optical transmission and how it can be detected experimentally.

The preceding examples illustrate the utility of explicitly detailed equations as I have done in this work. These equations can be directly used in the analysis of experimental measurements and in treating other optical processes. Clearly, the present approach can be applied to the treatment of photoelastic effects, excitation and detection of localized electromagnetic waves (e.g., surface plasmon-polariton and wave-guide modes) [35,36]. It can also be used to analyze optical reflection from metamaterials to the first-order. For metamaterials designed for cloaking application, one expects the leading order correction to the optical reflection to vanish. The perturbation method offers a simple way for computing such a correction.

As a final note, the present perturbation method is readily extended to treat linear optical responses in 2D materials beyond the electric dipole approximation. One starts by treating a 2D material as having a finite thickness d, albeit much smaller than the optical wavelength λ, and having a zeroth order dielectric tensor due to the electric dipole response. One needs to find the zeroth order electric fields in directions of reflection and transmission and in turn the zeroth order reflection and transmission matrices. One only needs to keep terms up to the first order in d/λ. Afterward, corrections to the zeroth order dielectric tensor in the 2D material produce extra electric fields in reflection and transmission. The latter can be calculated using the method as outlined in Section III. They lead to modifications to the reflection matrix and the transmission matrix. These modifications can be measured experimentally just as their counterparts for 3D materials and serve as sources of information on properties of the 2D material.

Appendix A: Local-field factors for uniaxial materials and biaxial materials

The following local-field factor tensor given by Eq. (18) applies when both the ambient of $\overleftrightarrow{\epsilon}_1$ and a material of $\overleftrightarrow{\epsilon}_2$ are isotropic [29].

$$\overleftrightarrow{F}^{(1\to 2)} = \sum F_{jj}^{(1\to 2)} \hat{j}\hat{j} = \hat{x}\hat{x}\frac{2\epsilon_1 k_{2z}^{(+)}}{\epsilon_1 k_{2z}^{(+)} + \epsilon_2 k_{1z}^{(+)}} + \hat{y}\hat{y}\frac{2k_{1z}^{(+)}}{k_{2z}^{(+)} + k_{1z}^{(+)}} + \hat{z}\hat{z}\frac{2\epsilon_1 k_{1z}^{(+)}}{\epsilon_1 k_{2z}^{(+)} + \epsilon_2 k_{1z}^{(+)}} \tag{78}$$

Equation (78) needs to be modified when the material of $\overleftrightarrow{\epsilon}_2$ is uniaxial. If one chooses the optic axis to be along the z-axis of the laboratory coordinate frame into material $\overleftrightarrow{\epsilon}_2$, s-polarization

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inside material $\overleftrightarrow{\epsilon}_2$ is an o-ray while the p-polarization is an e-ray [25,26]. One has

$$k_{1x}^{(+)} = k_{1x}^{(-)} = k_{2x,o}^{(+)} = k_{2x,e}^{(+)} \tag{79}$$

$$k_{1x}^{(-)} = (2\pi/\lambda)n_1 \sin\theta_1 \tag{80a}$$

$$k_{2x,o}^{(+)} = (2\pi/\lambda)n_{2o} \sin\theta_{2o} \tag{80b}$$

$$k_{2x,e}^{(+)} = (2\pi/\lambda)n_{2e}(\theta_{2e}) \sin\theta_{2e} \tag{80c}$$

$$\frac{1}{(n_{2e}(\theta_{2e}))^2} = \frac{(\cos\theta_{2e})^2}{n_{2o}^2} + \frac{(\sin\theta_{2e})^2}{n_{2e}^2} \tag{81}$$

$$k_{1z}^{(+)} = +\sqrt{(2\pi/\lambda)^2\epsilon_1 - (k_{1x}^{(+)})^2} \tag{82a}$$

$$k_{1z}^{(-)} = -\sqrt{(2\pi/\lambda)^2\epsilon_1 - (k_{1x}^{(+)})^2} \tag{82b}$$

$$k_{2z,o}^{(+)} = +\sqrt{(2\pi/\lambda)^2 n_{2o}^2 - (k_{1x}^{(+)})^2} \tag{82c}$$

$$k_{2z,e}^{(+)} = +\sqrt{(2\pi/\lambda)^2 (n_{2e}(\theta_{2e}))^2 - (k_{1x}^{(+)})^2} \tag{82d}$$

Let $\epsilon_{2e}(\theta_{2e}) = (n_{2e}(\theta_{2e}))^2$, the local-field factor tensor is now changed to

$$\overleftrightarrow{F}^{(1\to 2)} = \sum F_{jj}^{(1\to 2)}\hat{j}\hat{j} = \hat{x}\hat{x}\frac{2\epsilon_1 k_{2z,e}^{(+)}}{\epsilon_1 k_{2z,e}^{(+)} + \epsilon_{2e}(\theta_{2e})k_{1z}^{(+)}} + \hat{y}\hat{y}\frac{2k_{1z,o}^{(+)}}{k_{2z,o}^{(+)} + k_{1z}^{(+)}} + \hat{z}\hat{z}\frac{2\epsilon_1 k_{1z}^{(+)}}{\epsilon_1 k_{2z,e}^{(+)} + \epsilon_{2e}(\theta_{2e})k_{1z}^{(+)}} \tag{83}$$

If one chooses the optic axis to be along the x-axis of the laboratory coordinate frame, the s-polarized electric field inside material $\overleftrightarrow{\epsilon}_2$ remains an o-ray while the p-polarized electric field is still an e-ray except that $n_{2e}(\theta_{2e})$ and θ_{2e} are determined from the following equation and the Snell's law

$$\frac{1}{(n_{2e}(\theta_{2e}))^2} = \frac{(\sin\theta_{2e})^2}{n_{2o}^2} + \frac{(\cos\theta_{2e})^2}{n_{2e}^2} \tag{84}$$

The local-field factor tensor is still given by Eq. (83).

If one has the optic axis along the y-axis of the laboratory coordinate frame and thus perpendicular to the incidence plane, the s-polarized electric field inside material $\overleftrightarrow{\epsilon}_2$ now is an e-ray while the p-polarized electric field is an o-ray. One has

$$k_{2x,o}^{(+)} = (2\pi/\lambda)n_{2o} \sin\theta_{2o} \tag{85a}$$

$$k_{2x,e}^{(+)} = (2\pi/\lambda)n_{2e} \sin\theta_{2e} \tag{85b}$$

$$k_{2z,o}^{(+)} = +\sqrt{(2\pi/\lambda)^2 n_{2o}^2 - (k_{1x}^{(+)})^2} \tag{86a}$$

$$k_{2z,e}^{(+)} = +\sqrt{(2\pi/\lambda)^2 n_{2e}^2 - (k_{1x}^{(+)})^2} \tag{86b}$$

The local-field factor tensor becomes

$$\overleftrightarrow{F}^{(1\to 2)} = \sum F_{jj}^{(1\to 2)}\hat{j}\hat{j} = \hat{x}\hat{x}\frac{2\epsilon_1 k_{2z,o}^{(+)}}{\epsilon_1 k_{2z,o}^{(+)} + n_{2o}^2 k_{1z}^{(+)}} + \hat{y}\hat{y}\frac{2k_{1z}^{(+)}}{k_{2z,e}^{(+)} + k_{1z}^{(+)}} + \hat{z}\hat{z}\frac{2\epsilon_1 k_{1z}^{(+)}}{\epsilon_1 k_{2z,o}^{(+)} + n_{2o}^2 k_{1z}^{(+)}} \tag{87}$$

It is easy to extend Eq. (83) to biaxial materials, as long as the laboratory coordinate frame overlaps with the principal coordinate frame.

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Appendix B: Conversion of magneto-optic corrections to reflection matrix between different choices of laboratory coordinate frames and unit vectors for *s*-polarization and *p*-polarization

In the original work of Hunt on Kerr effects [20], he chose

$$\hat{e}^{(-)}_{1p,H} = +(k^{(+)}_{1z}/k_1)\hat{x}_H + (k^{(+)}_{1x}/k_1)\hat{z}_H = sin\theta_1\hat{x}_H + cos\theta_1\hat{z}_H \tag{88a}$$

$$\hat{e}^{(+)}_{1p,H} = -(k^{(+)}_{1z}/k_1)\hat{x}_H + (k^{(+)}_{1x}/k_1)\hat{z}_H = -sin\theta_1\hat{x}_H + cos\theta_1\hat{z}_H \tag{88b}$$

$$\hat{e}^{(-)}_{1s,H} = \hat{e}^{(+)}_{1s,H} = \hat{y}_H \tag{88c}$$

There are two consequences. The first is that his zeroth order reflection coefficient $r^{(0)}_{pp,H}$ for the p-polarized component has the opposite sign while the coefficient $r^{(0)}_{ss,H}$ for the s-polarized component remains unchanged when compared with Eq. (33) and Eq. (34) in the main text:

$$r^{(0)}_{pp,H} = -r^{(0)}_{pp} \tag{89a}$$

$$r^{(0)}_{ss,H} = r^{(0)}_{ss} \tag{89b}$$

The second is that in his expression for corrections to the reflection matrix,

$$\Delta r_{pp}(MOKE) = \alpha_{y,H} m_y \tag{90a}$$

$$\Delta r_{ps}(MOKE) = +\alpha_{x,H} m_x + \alpha_{z,H} m_z \tag{90b}$$

$$\Delta r_{sp}(MOKE) = -\alpha_{x,H} m_x + \alpha_{z,H} m_z \tag{90c}$$

the coefficients are related to those in Eq. (56,57,58) as follows,

$$\alpha_{y,H} = \left[\frac{(-iQ_{zx})(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,H,x}e^{(+)}_{1p,H,z}F^{(1\to2)}_{xx}F^{(1\to2)}_{zz}}{k^{(+)}_{2z}}\right] = -\alpha_y \tag{91a}$$

$$\alpha_{x,H} = \left[\frac{(-iQ_{yz})(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,H,z}e^{(+)}_{1s,H}F^{(1\to2)}_{yy}F^{(1\to2)}_{zz}}{2k^{(+)}_{2z}}\right] = +\alpha_x \tag{91b}$$

$$\alpha_{z,H} = \left[\frac{(+iQ_{xy})(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,H,x}e^{(+)}_{1s,H}F^{(1\to2)}_{yy}F^{(1\to2)}_{xx}}{2k^{(+)}_{2z}}\right] = +\alpha_z \tag{91c}$$

Furthermore, Hunt chose a coordinate frame such that his y-z plane is the incidence plane. As a result, $m_{x,H} = -m_y$, $m_{y,H} = m_x$, $m_{z,H} = m_z$. Sine the corrections to reflection matrix remain,

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the final conversion between the result of this work and the work of Hunt is as follows,

$$\Delta r_{pp}(MOKE) = \alpha'_{x,H} m_{x.H} \tag{92a}$$

$$\Delta r_{ps}(MOKE) = +\alpha'_{y,H} m_{y,H} + \alpha'_{z,H} m_{z,H} \tag{92b}$$

$$\Delta r_{sp}(MOKE) = -\alpha'_{y,H} m_{y,H} + \alpha'_{z,H} m_{z,H} \tag{92c}$$

$$r^{(0)}_{pp,H} = -r^{(0)}_{pp} \tag{93a}$$

$$r^{(0)}_{ss,H} = +r^{(0)}_{ss} \tag{93b}$$

$$\alpha'_{x,H} = \alpha_y \tag{93c}$$

$$\alpha'_{y,H} = \alpha_x \tag{93d}$$

$$\alpha'_{z,H} = \alpha_z \tag{93e}$$

Eq. (92) and (93) reproduce the findings reported by Hunt [20]. In Table II of Ref. 20, Hunt reported $r_{ps} = -r_{sp} = \alpha'_{y,H} m_{y,H}$ for longitudinal Kerr effect and $r_{ps} = r_{sp} = \alpha'_{z,H} m_{z,H}$ for polar Kerr effect.

In the work reported by Kapitulnik and coworkers [32], these authors chose

$$\hat{e}^{(-)}_{1p,K} = -(k^{(+)}_{1z}/k_1)\hat{x}_H - (k^{(+)}_{1x}/k_1)\hat{z}_H = -sin\theta_1 \hat{x}_H - cos\theta_1 \hat{z}_H \tag{94a}$$

$$\hat{e}^{(+)}_{1p,K} = (k^{(+)}_{1z}/k_1)\hat{x}_H - (k^{(+)}_{1x}/k_1)\hat{z}_H = sin\theta_1 \hat{x}_H - cos\theta_1 \hat{z}_H \tag{94b}$$

$$\hat{e}^{(-)}_{1s,} = \hat{e}^{(+)}_{1s,K} = \hat{y}_K \tag{94c}$$

Again, there are two consequences. The first is that the zeroth order reflection coefficient $r^{(0)}_{pp,K}$ for the p-polarized component has the opposite sign while the coefficient for the s-polarized component $r^{(0)}_{ss,K}$ remains unchanged when compared with Eq. (33) and Eq. (34):

$$r^{(0)}_{pp,K} = -r^{(0)}_{pp} \tag{95a}$$

$$r^{(0)}_{ss,K} = r^{(0)}_{ss} \tag{95b}$$

The second is that in the Eq. (4) of Ref. 32,

$$\Delta r_{pp}(MOKE) = \alpha_{y,K} m_y \tag{96a}$$

$$\Delta r_{ps}(MOKE) = \alpha_{x,K} m_x + \alpha_{z,K} m_z \tag{96b}$$

$$\Delta r_{sp}(MOKE) = \alpha_{x,K} m_x - \alpha_{z,K} m_z \tag{96c}$$

their coefficients are related to Eq. (56,57,58) as follows,

$$\alpha_{y,K} = \left[\frac{(-iQ)\epsilon_2(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,K,x} e^{(+)}_{1p,K,z} F^{(1\to2)}_{xx} F^{(1\to2)}_{zz}}{k^{(+)}_{2z}}\right] = -\alpha_y \tag{97a}$$

$$\alpha_{x,K} = \left[\frac{(-iQ)\epsilon_2(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,K,z} e^{(+)}_{1s,K} F^{(1\to2)}_{yy} F^{(1\to2)}_{zz}}{2k^{(+)}_{2z}}\right] = -\alpha_x \tag{97b}$$

$$\alpha_{z,K} = \left[\frac{(+iQ)\epsilon_2(\omega/c)^2}{2k^{(+)}_{1z}}\right]\left[\frac{e^{(-)}_{1p,K,x} e^{(+)}_{1s,K} F^{(1\to2)}_{yy} F^{(1\to2)}_{xx}}{2k^{(+)}_{2z}}\right] = -\alpha_z \tag{97c}$$

In fact, Kapitulnik *et al.* further chose a coordinate frame such that their y-z plane is the incidence plane in a different way such that $m_{x,K} = m_y$, $m_{y,K} = -m_x$, $m_{z,K} = m_z$. Sine the

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corrections to reflection matrix remain, the final conversion between the present work and the work reported by Kapitulnik *et al.* [32] is as follows,

$$\Delta r_{pp}(MOKE) = \alpha'_{x,K} m_{x,K} \tag{98a}$$

$$\Delta r_{ps}(MOKE) = \alpha'_{y,K} m_{y,K} + \alpha'_{z,K} m_{z,K} \tag{98b}$$

$$\Delta r_{sp}(MOKE) = \alpha'_{y,K} m_{y,K} - \alpha'_{z,K} m_{z,K} \tag{98c}$$

$$r^{(0)}_{pp,K} = -r^{(0)}_{pp} \tag{99a}$$

$$r^{(0)}_{ss,K} = +r^{(0)}_{ss} \tag{99b}$$

$$\alpha'_{x,K} = -\alpha_y \tag{99c}$$

$$\alpha'_{y,K} = +\alpha_x \tag{99d}$$

$$\alpha'_{z,K} = -\alpha_z \tag{99e}$$

In the work of Kapitulnik *et al.* [32], they used $a = \alpha'_{x,K}$, $b = \alpha'_{y,K}$, and $c = \alpha'_{z,K}$.

Appendix C: Perturbation treatment of magneto-optic effects on optical transmission, - revisiting the Faraday rotation, magnetic circular dichroism, and the Voigt effect

A thin sheet of polarization $\Delta\vec{P}(z')$ (see Eq. (14) in the main text) of thickness dz' due to $\Delta\overleftrightarrow{\epsilon}$ produces an electric field, $\delta\vec{E}^{(+)}_2(\vec{r},t)$, in the direction of transmission. $\delta\vec{E}^{(+)}_2(\vec{r},t)$ is given by Eq. (15b). Let the thickness of the material $\overleftrightarrow{\epsilon}_2$ be L. The integration of z' over the thickness L yields the extra electric field from

$$\begin{aligned}\hat{e}^{(+)}_2 \cdot \Delta\vec{E}^{(+)}_2(\vec{r},t) &= \int_0^L i\frac{2\pi(\omega/c)^2}{4\pi\varepsilon_0 k^{(+)}_{2z}}\hat{e}^{(+)}_2 \cdot \Delta\vec{P}(z')dz' exp[ik^{(+)}_{2z}z - ik^{(+)}_{2z}z'] \\ &= i\left[\frac{(\omega/c)^2 L}{2k^{(+)}_{2z}}\right]\left[\hat{e}^{(+)}_2 \cdot \left(\Delta\overleftrightarrow{\epsilon} : \vec{E}^{(+)}_2\right)\right] exp[ik^{(+)}_{1x}x + ik^{(+)}_{2z}z - i\omega t]\end{aligned} \tag{100}$$

Specifically, extra s-polarized and p-polarized electric fields are as follows (dropping the phase factor for now),

$$\Delta E^{(+)}_{2s} = i\left[\frac{(\omega/c)^2 L}{2k^{(+)}_{2z}}\right]\left[\left(\hat{e}^{(+)}_{2s} : \Delta\overleftrightarrow{\epsilon}\right) \cdot \vec{E}^{(+)}_2\right] \tag{101}$$

$$\Delta E^{(+)}_{2p} = i\left[\frac{(\omega/c)^2 L}{2k^{(+)}_{2z}}\right]\left[\left(\hat{e}^{(+)}_{2p} : \Delta\overleftrightarrow{\epsilon}\right) \cdot \vec{E}^{(+)}_2\right] \tag{102}$$

We define a transmission matrix T to relate the s-polarized and p-polarized components of the transmitted electric field at $z = 0$ to the components of the field at $z = L$,

$$\begin{pmatrix} E^{(+)}_{2p}(z=L) \\ E^{(+)}_{2s}(z=L) \end{pmatrix} = T\begin{pmatrix} E^{(+)}_{2p}(z=0) \\ E^{(+)}_{2s}(z=0) \end{pmatrix} = \begin{pmatrix} 1+\Delta t_{pp} & \Delta t_{ps} \\ \Delta t_{sp} & 1+\Delta t_{ss} \end{pmatrix}\begin{pmatrix} E^{(+)}_{2p}(z=0) \\ E^{(+)}_{2s}(z=0) \end{pmatrix} \tag{103}$$

From Eq. (101) and Eq. (102), we have

$$\Delta t_{ss} = i\left[\frac{(\omega/c)^2 L}{2k^{(+)}_{2z}}\right]\Delta\epsilon_{yy} \tag{104a}$$

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$$\Delta t_{pp} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right](\Delta\epsilon_{xx}e_{2p,x}^{(+)}e_{2p,x}^{(+)} + \Delta\epsilon_{zx}e_{2p,z}^{(+)}e_{2p,x}^{(+)} + \Delta\epsilon_{xz}e_{2p,x}^{(+)}e_{2p,z}^{(+)} + \Delta\epsilon_{zz}e_{2p,z}^{(+)}e_{2p,z}^{(+)}) \tag{104b}$$

$$\Delta t_{ps} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right](\Delta\epsilon_{xy}e_{2p,x}^{(+)} + \Delta\epsilon_{zy}e_{2p,z}^{(+)}) \tag{104c}$$

$$\Delta t_{sp} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right](\Delta\epsilon_{yx}e_{2p,x}^{(+)} + \Delta\epsilon_{yz}e_{2p,z}^{(+)}) \tag{104d}$$

We apply these results to the magneto-optic effect on optical transmission at normal incidence. In this case, the corrections to the dielectric tensor are as follows

$$\Delta\overleftrightarrow{\epsilon}^{(MOKE)} = \begin{pmatrix} 0 & -i\epsilon_2 Q & 0 \\ i\epsilon_2 Q & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{105}$$

By substituting Eq. (105) into Eq. (104) and noting that $e_{2p,x}^{(+)} = 1$ and $e_{2p,z}^{(+)} = 0$,

$$\Delta t_{ss} = 0 \tag{106a}$$

$$\Delta t_{pp} = 0 \tag{106b}$$

$$\Delta t_{ps} = +\left[\frac{(\omega/c)^2 L\epsilon_2 Q}{2k_2^{(+)}}\right] \tag{106c}$$

$$\Delta t_{sp} = -\left[\frac{(\omega/c)^2 L\epsilon_2 Q}{2k_2^{(+)}}\right] \tag{106d}$$

The transmission matrix becomes

$$T = \begin{pmatrix} 1 & \left[\frac{(\omega/c)^2 L\epsilon_2 Q}{2k_2^{(+)}}\right] \\ -\left[\frac{(\omega/c)^2 L\epsilon_2 Q}{2k_2^{(+)}}\right] & 1 \end{pmatrix} \tag{107}$$

Equation (107) is a rotation matrix in the limit when $\left|\frac{(\omega/c)^2 L\epsilon_2 Q}{2k_2^{(+)}}\right| \ll 1$. By expressing Eq. (107) in the form of a standard rotation matrix with angle θ_F (viewed as the beam travels towards the viewer),

$$T = \begin{pmatrix} cos\theta_F & -sin\theta_F \\ sin\theta_F & cos\theta_F \end{pmatrix} \tag{108}$$

we have the rotation angle due to the magneto-optic effect on transmission,

$$\theta_F = -\frac{\pi\sqrt{\epsilon_2}L}{\lambda}Q \tag{109}$$

This is precisely the Faraday rotation through a magnetic material of thickness L and having a dielectric tensor [12]

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \epsilon_2 & -i\epsilon_2 Q & 0 \\ i\epsilon_2 Q & \epsilon_2 & 0 \\ 0 & 0 & \epsilon_2 \end{pmatrix} \tag{110}$$

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This validates the perturbation approach of dealing with linear optical responses beyond the leading order electric dipole response on optical transmission, as long as the overall effect is small.

This discussion can be generalized to materials that exhibit both magnetic circular birefringence (Faraday Effect) and magnetic circular dichroism (MCD). In these cases, corrections to the zeroth order dielectric tensor has the following form [37],

$$\Delta\overleftrightarrow{\epsilon}^{(MC)} = \begin{pmatrix} 0 & -(\beta + i\alpha) & 0 \\ \beta + i\alpha & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{111}$$

Here α prescribes the magnetic circular birefringence (Faraday effect) while β gives rise to the magnetic circular dichroism (MCD). By substituting Eq. (111) into Eqs.(104) and with the optical beam normally incident on the sample so that $e^{(+)}_{2p,x} = 1$ and $e^{(+)}_{2p,z} = 0$,

$$\Delta t_{ss} = 0 \tag{112a}$$

$$\Delta t_{pp} = 0 \tag{112b}$$

$$\Delta t_{ps} = -i\left[\frac{(\omega/c)^2 L(\beta + i\alpha)}{2k_2^{(+)}}\right] \tag{112c}$$

$$\Delta t_{sp} = +i\left[\frac{(\omega/c)^2 L(\beta + i\alpha)}{2k_2^{(+)}}\right] \tag{112d}$$

As a result, the transmission matrix becomes

$$T = \begin{pmatrix} 1 & -i\left[\frac{(\omega/c)^2 L(\beta+i\alpha)}{2k_2^{(+)}}\right] \\ i\left[\frac{(\omega/c)^2 L(\beta+i\alpha)}{2k_2^{(+)}}\right] & 1 \end{pmatrix} \tag{113}$$

In the absence of the MCD effect so that $\beta = 0$ and $\alpha = \epsilon_2 Q$, Eq. (113) is reduced to Eq. (107),

$$T = \begin{pmatrix} 1 & \left[\frac{(\omega/c)^2 L\alpha}{2k_2^{(+)}}\right] \\ -\left[\frac{(\omega/c)^2 L\alpha}{2k_2^{(+)}}\right] & 1 \end{pmatrix}. \tag{114}$$

It again prescribes the Faraday effect in the limit of $\left|\frac{(\omega/c)^2 L\alpha}{2k_2^{(+)}}\right| \ll 1$.

In the absence of the magnetic circular birefringence so that $\alpha = 0$, Eq. (113) becomes

$$T = \begin{pmatrix} 1 & -i\left[\frac{(\omega/c)^2 L\beta}{2k_2^{(+)}}\right] \\ i\left[\frac{(\omega/c)^2 L\beta}{2k_2^{(+)}}\right] & 1 \end{pmatrix} \tag{115}$$

It yields MCD in the limit when $\left|\frac{(\omega/c)^2 L\beta}{2k_2^{(+)}}\right| \ll 1$, i.e.,

$$\frac{T_L - T_R}{T_L + T_R} = 2 \times \left[\frac{(\omega/c)^2 L\beta}{2k_2^{(+)}}\right] = \left(\frac{(\omega/c)^2 L}{k_2^{(+)}}\right)\beta \tag{116}$$

T_L and T_R are the transmittance for left- and right-circularly polarized components of the light beam.

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When both magnetic circular birefringence and magnetic circular dichroism are present, Eq. (113) can be used, along with a suitable form of polarization-modulated transmission ellipsometry, to determine β and α, i.e., both Faraday rotation and MCD.

Finally, I briefly discuss the treatment of the quadratic magneto-optic (MO) effect, - the Voight effect. Corrections to the zeroth order dielectric tensor up to the second order in an isotropic material are as follows [38],

$$\Delta\overleftrightarrow{\epsilon}^{(MO)} = \begin{pmatrix} 0 & -i\epsilon_2 Q m_z & i\epsilon_2 Q m_y \\ i\epsilon_2 Q m_z & 0 & -i\epsilon_2 Q m_x \\ -i\epsilon_2 Q m_y & i\epsilon_2 Q m_x & 0 \end{pmatrix} + \begin{pmatrix} A_1 m_x^2 & A_1 m_x m_y & A_1 m_x m_z \\ A_1 m_x m_y & A_1 m_y^2 & A_1 m_y m_z \\ A_1 m_x m_z & A_1 m_y m_z & A_1 m_z^2 \end{pmatrix} \tag{117}$$

The second matrix prescribes the Voigt effect. As an example. I examine its effect on the optical transmission when the magnetization vector is perpendicular to the direction of the light propagation (the positive z axis) so that $m_z = 0$. Equation (117) is then simplified as,

$$\Delta\overleftrightarrow{\epsilon}^{(MO)} = \begin{pmatrix} A_1 m_x^2 & A_1 m_x m_y & i\epsilon_2 Q m_y \\ A_1 m_x m_y & A_1 m_y^2 & -i\epsilon_2 Q m_x \\ -i\epsilon_2 Q m_y & i\epsilon_2 Q m_x & 0 \end{pmatrix} \tag{118}$$

By inserting matrix elements in Eq. (118) into Eq. (104a)–(104d) and recalling that $e_{2p,x}^{(+)} = 1$ and $e_{2p,z}^{(+)} = 0$, I find

$$\Delta t_{ss} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_y^2 \tag{119a}$$

$$\Delta t_{pp} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x^2 \tag{119b}$$

$$\Delta t_{ps} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x m_y \tag{119c}$$

$$\Delta t_{sp} = i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x m_y \tag{119d}$$

The resultant transmission matrix is as follows,

$$T = \begin{pmatrix} 1 + i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x^2 & i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x m_y \\ i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_x m_y & 1 + i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 m_y^2 \end{pmatrix} \tag{120}$$

To measure A_1, one can polarize the sample so that $m_y = 1$ and $m_x = 0$. Equation (120) is simplified to

$$T = \begin{pmatrix} 1 & 0 \\ 0 & 1 + i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1 \end{pmatrix} \cong \begin{pmatrix} 1 & 0 \\ 0 & exp\left(i\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1\right) \end{pmatrix} \tag{121}$$

$\left[\frac{(\omega/c)^2 L}{2k_{2z}^{(+)}}\right] A_1$ appears as an extra phase for the y-component of the light beam and can be detected with a transmission difference technique, similar to normal-incidence reflectivity difference techniques [39].

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References

1. E. Alder, "Nonlinear Optical Frequency Polarization in a Dielectric," Phys. Rev. **134**(3A), A728–A733 (1964).
2. N. Bloembergen, R. K. Chang, S. S. Jha, and C. H. Lee, "Optical second-harmonic generation in reflection from media with inversion symmetry," Phys. Rev. **174**(3), 813–822 (1968).; Erratum 178, 1528 (1969).
3. X. D. Zhu and H. Zhang, "Hidden Linear Optical Response Reveal Crystalline Symmetry," Opt. Lett. **45**(8), 2439 (2020).
4. W. Voigt, *Magneto- und Electro-Optik* (Teuner, Leipzig, 1908).
5. H. R. Hulme, "The Faraday Effect in Ferromagnetics," Proc. R. Soc. London, Ser. A **135**(826), 237–257 (1932).
6. P. Argyes, "Theory of the Faraday and Kerr Effects in Ferromagnetics," Phys. Rev. **97**(2), 334–345 (1955).
7. P. M. Oppeneer, T. Maurer, J. Sticht, and J. Kuebler, "Ab initio calculated magneto-optical Kerr effect of ferromagnetic metals: Fe and Ni," Phys. Rev. B **45**(19), 10924–10933 (1992).
8. T. Gasche, M. S. S. Brooks, and B. Johnasson, "Calculated magneto-optical Kerr effect in Fe, Co, and Ni," Phys. Rev. B **53**(1), 296–301 (1996).
9. A. Yariv, *Quantum Electronics*, 2nd Ed. (John Wiley & Sons, New York, 1975).
10. J. D. E. McIntyre and D. E. Aspnes, "Differential reflection spectroscopy of very thin surface films," Surf. Sci. **24**(2), 417–434 (1971).
11. A. Wong and X. D. Zhu, "An optical differential reflectance study of adsorption and desorption of xenon and deuterium on Ni(111)," Appl. Phys. A **63**(1), 1–8 (1996).
12. Z. Q. Qiu and S. D. Bader, "Surface magneto-optic Kerr effects," Rev. Sci. Instrum. **71**(3), 1243–1255 (2000).
13. Y. K. Kato, R. C. Myers, A. C. Gossard, and D. D. Awshalom, "Observation of the spin Hall effects in semiconductors," Science **306**(5703), 1910–1913 (2004).
14. S. Spielman, J. S. Dodge, L. W. Lombardo, C. B. Eom, M. M. Fejer, T. H. Geballe, and A. Kapitulnik, "Measurement of the spontaneous polar Kerr effect in YBa2Cu3O7 and Bi2Sr2CaCu2O8," Phys. Rev. Lett. **68**(23), 3472–3475 (1992).
15. E. R. Schemm, W. J. Gannon, C. M. Wishne, W. P. Halperin, and A. Kapitulnik, "Observation of broken time-reversal symmetry in the heavy-fermion superconductor UPt3," Science **345**(6193), 190–193 (2014).
16. W. Schwarzacher, J. Gray, and X. D. Zhu, "Oblique-incidence reflectivity difference as an in-situ probe of electrodeposition: Co on Au," Electrochem. Solid-State Lett. **6**(5), C73 (2003).
17. P. Thomas, E. Nabighian, M. C. Bartelt, C. Y. Fong, and X. D. Zhu, "An oblique-incidence optical reflectivity difference) OI-RD) and LEED study of rare gas growth on a lattice-mismatched metal substrate," Appl. Phys. A **79**(1), 131–137 (2004).
18. Y. Y. Fei, L. F. Liu, H. B. Lu, Z. H. Chen, G. Z. Yang, and X. D. Zhu, "Oscillations in oblique-incidence optical reflection from a growth surface during layer-by-layer epitaxy," Phys. Rev. B **69**(23), 233405 (2004).
19. Y. Y. Fei, J. P. Landry, Y. S. Sun, X. D. Zhu, J. T. Luo, X. B. Wang, and K. S. Lam, "A novel high-throughput scanning microscope for label-free detection of protein and small-molecule chemical microarrays," Rev. Sci. Instrum. **79**(11), 113708 (2008).
20. R. P. Hunt, "Magneto-optic scattering from thin solid films," J. Appl. Phys. **38**(4), 1652–1671 (1967).
21. T. Yoshino and S. Tanaka, "Longitudinal Magneto-Optic Effect in Ferromagnetic Thin Films. I," Jpn. J. Appl. Phys. **5**(11), 989–993 (1966).
22. J. Zak, E. R. Moog, C. Liu, and S. D. Bader, "Magneto-Optics of Multilayers with Arbitrary Magnetization Directions," Phys. Rev. B **43**(8), 6423–6429 (1991).
23. J. Zak, E. R. Moog, C. Liu, and S. D. Bader, "Universal Approach to Magneti-Optics," J. Magn. Magn. Mater. **89**(1-2), 107–123 (1990).
24. X. D. Zhu, Y. Y. Fei, X. Wang, H. B. Lu, and G. Z. Yang, "General theory of optical reflection from a thin film on a solid and its application to heteroepitaxy," Phys. Rev. B **75**(24), 245434 (2007).
25. M. Born and E. Wolf, *Principles of Optics* 6th Ed. (Pergamon Press, New York, 1980), p 665.
26. G. R. Fowler, *Introduction to Modern Optics* (Holt, Rinehart and Winston, New York, 1968), p 173.
27. Y. A. Uspenskii, E. T. Kulatov, and S. V. Halilov, "Effect of anisotropy on magneto-optical properties of uniaxial crystals: Application to CrO2," Phys. Rev. B **54**(1), 474–482 (1996).
28. Y. R. Shen, *The Principles of Nonlinear Optics* (John Wiley & Sons, New York, 1984).
29. T. F. Heinz, "Second-order nonlinear optical effects at surfaces and interfaces," in *Nonlinear Surface Electromagnetic Phenomena*, H. E. Ponath and G. I. Stegeman, eds. (Elsevier Science Publisher, B.V., 1991), p. 353.
30. H. W. K. Tom, T. F. Heinz, and Y. R. Shen, "Second-harmonic reflection from silicon surfaces and its relation to structural symmetry," Phys. Rev. B **51**(21), 1983–1986 (1983).
31. H. W. K. Tom and G. D. Aumiller, "Observation of rotational anisotropy in the second-harmonic generation from a metal surface," Phys. Rev. B **33**(12), 8818–8821 (1986).
32. A. Kapitulnik, J. S. Dodge, and M. M. Fejer, "High-Resolution Magneto-Optic Measurements with a Sagnac Interferometer," J. Appl. Phys. **75**(10), 6872–6877 (1994).

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33. X. D. Zhu, "Symmetry Consideration in Zero Loop-Area Sagnac Interferometry at Oblique-Incidence for Detecting Magneto-Optic Kerr Effect," Rev. Sci. Instrum. **88**(8), 083112 (2017).
34. X. D. Zhu and G. Malovichko, "Zero Loop-Area Sagnac Interferometer at Oblique-Incidence for Detecting In-Plane Magneto-Optic Kerr Effect," AIP Adv. **7**(5), 055008 (2017).
35. M. Neviere, R. Petit, and M. Cadilhac, "About the Theory of Optical Grating Coupler-Waveguide Systems," Opt. Commun. **8**(2), 113–117 (1973).
36. Y. Y. Fei, X. Wang, and X. D. Zhu, "Novel Excitation and Detection of Surface-Plasmon Polariton Wave on Cu(111) with Gratings of Rare Gas Monolayers," Opt. Lett. **33**(16), 1914 (2008).
37. D. Zhao, H. Hochst, and D. L. Huber, "Dielectric tensor formulation of magnetic dichroism sum rules," J. Appl. Phys. **84**(5), 2858–2860 (1998).
38. J. McCord, "Progress in magnetic domain observed by advanced magneto-optical microscopy," J. Phys. D: Appl. Phys. **48**(33), 333001 (2015).
39. X. D. Zhu, "Measuring linear electric quadrupole susceptibility of non-centrosymmetric crystals with reflectivity difference," Rev. Sci. Instrum. **92**(2), 025103 (2021).