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Macroscopic theory of reflection from antiferromagnetic Cr_2O_3

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Abstract. Reflection matrices with the correct space–time symmetry properties may be obtained from the standard Maxwell boundary conditions only when the constitutive relations for the $\boldsymbol{D}$ and $\boldsymbol{H}$ fields satisfy covariance requirements. In this paper we present the first application of covariant multipole forms for $\boldsymbol{D}$ and $\boldsymbol{H}$, to the order of electric quadrupole and magnetic dipole, to a magnetic crystal in order to compare the predictions obtained from this theory with recent experimental results. In particular, the theory is used to determine the Fresnel reflection amplitudes for antiferromagnetic Cr_2O_3 when a monochromatic plane light wave is incident normally from a vacuum on a crystal face perpendicular first to the C_3 axis and then to the C_2 axis. The theory agrees well with experiment. In addition, it reveals a novel relationship between certain components of the frequency-dependent magnetoelectric tensor, which, with the aid of transmission and reflection data, would enable the surface and bulk contributions in reflection to be distinguished.

1. Introduction

Theories of reflection from spatially dispersive media have been shown to be sensitive both to the form assumed for the constitutive relations, which are often of an empirical or phenomenological nature, and to the boundary conditions that are used when matching the light wave fields at the interface [1–6]. However, an acid test for distinguishing between the conflicting results of the various theories is that the reflection matrix for a particular system should exhibit the form that is prescribed by space–time symmetry [7, 8]. It has been shown for non-magnetic media [6, 9] that reflection matrices which meet this requirement can be obtained from the standard Maxwell boundary conditions [10] only when the $\boldsymbol{D}$ and $\boldsymbol{H}$ fields are taken in covariant form. This problem is discussed in a recent article [11] where the covariant multipole forms for $\boldsymbol{D}$ and $\boldsymbol{H}$ that apply to non-magnetic and magnetic media are derived in the electric quadrupole-magnetic dipole approximation in terms of the macroscopic property tensors of the medium.

In this paper use is made for the first time of the magnetic forms of the covariant $\boldsymbol{D}$ and $\boldsymbol{H}$ fields to produce a theory of reflection from antiferromagnetic Cr_2O_3, so providing a quantitative basis for describing the experimental observations of Krichevtsov *et al* [12], which these authors, in the absence of such a theory, interpreted phenomenologically. Although our theory is macroscopic and as such does not allow for microscopic surface effects [13], it yields a relationship between the components of the magnetoelectric tensor which has not previously been noted and which, with the aid of reflection and transmission data, would permit the surface and bulk contributions in reflection to be distinguished. In addition, our theory provides an important test of the validity of the covariant multipole

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forms for the $\boldsymbol{D}$ and $\boldsymbol{H}$ fields for a magnetic medium, which have only recently been derived [11].

We begin in section 2 by considering wave propagation in Cr_2O_3, since the eigenpolarizations that the medium supports must necessarily be determined before boundary conditions can be applied. In order to compare our theoretical results with the experimental data of Krichevtsov *et al* for a single-domain crystal of antiferromagnetic Cr_2O_3 [12], we derive in section 3 the reflection matrices for the propagation directions used in [12]. This is done by applying the standard Maxwell boundary conditions at the vacuum–crystal interface and by taking the $\boldsymbol{D}$ and $\boldsymbol{H}$ fields in the medium in covariant form. In section 4 the predictions based on our theory are shown to be in agreement with experiment [12]. The discussion follows in section 5.

2. Wave propagation in Cr_2O_3

The equation that describes the propagation of a plane electromagnetic wave with an electric field of the form

$$\boldsymbol{E} = \boldsymbol{E}^{(0)} \exp\{\mathrm{i}\omega(\tilde{n}\boldsymbol{\sigma}\cdot\boldsymbol{r}/c - t)\} \tag{1}$$

in an optically inactive antiferromagnet such as Cr_2O_3 is [14]

$$\left[\tilde{n}^2(\sigma_\alpha\sigma_\beta - \delta_{\alpha\beta}) + \delta_{\alpha\beta} + \epsilon_0^{-1}\tilde{\alpha}_{\alpha\beta} + \mu_0 c\tilde{n}\sigma_\gamma\tilde{A}_{\alpha\beta\gamma}\right]E_\beta^{(0)} = 0 \tag{2}$$

where $\tilde{n}$ is the complex refractive index for the polarization state described by the amplitude $\boldsymbol{E}^{(0)}$ when propagation is in the direction of the unit wave-normal $\boldsymbol{\sigma}$, $\epsilon_0^{-1}\tilde{\alpha}_{\alpha\beta}$ is the electric susceptibility and

$$\tilde{A}_{\alpha\beta\gamma} = -\epsilon_{\alpha\gamma\delta}\tilde{G}_{\beta\delta} - \epsilon_{\beta\gamma\delta}\tilde{G}_{\alpha\delta} + \tfrac{1}{2}\omega\big(\tilde{a}'_{\alpha\beta\gamma} + \tilde{a}'_{\beta\alpha\gamma}\big) = \tilde{A}_{\beta\alpha\gamma}. \tag{3}$$

In (2) and (3), $\tilde{\alpha}_{\alpha\beta}$, $\tilde{G}_{\alpha\beta}$ and $\tilde{a}'_{\alpha\beta\gamma}$ are macroscopic property tensors of the medium and are complex (denoted by a tilde) to allow for absorption. They describe the induction of multipole polarization densities as follows [14]:

$$P_\alpha = \tilde{\alpha}_{\alpha\beta}E_\beta + \tfrac{1}{2}\omega^{-1}\tilde{a}'_{\alpha\beta\gamma}\nabla_\gamma\dot{E}_\beta + \tilde{G}_{\alpha\beta}B_\beta + \ldots \tag{4}$$

$$Q_{\alpha\beta} = \omega^{-1}\tilde{a}'_{\gamma\alpha\beta}\dot{E}_\gamma + \ldots \tag{5}$$

$$M_\alpha = \tilde{G}_{\beta\alpha}E_\beta + \ldots\ . \tag{6}$$

An earlier theory of wave propagation in Cr_2O_3 has been presented by Hornreich and Shtrikman [15], who were the first to recognize the necessity for including electric quadrupole contributions in addition to those of magnetic dipoles. However, their theory yields expressions for the polarization states and refractive indices of the characteristic waves (eigenpolarizations) propagating along a two-fold rotation axis in Cr_2O_3 which differ from those that are obtained from (2) [14]. The problem arises because they assume that their electric quadrupole tensor γ''_{ijl}, which is symmetric in the interchange of the subscripts i and j, is also symmetric in the interchange of the subscripts j and l (see equation (26) in [15]). It is readily verified from the quantum-mechanical expression for γ''_{ijl} in their equation (86*a*) that this assumption is erroneous. We therefore make use of (2) to determine the refractive indices and corresponding eigenpolarizations that propagate in Cr_2O_3 when deriving the reflection matrices in the following section.

3. Normal-incidence reflection from Cr_2O_3

In order to apply our theory to the experimental results of Krichevtsov *et al* [12] we consider reflection at normal incidence from a single-domain crystal of Cr_2O_3 in a vacuum when the light path is parallel first to the optic axis (C_3 axis) and then to the C_2 axis. The crystal properties are specified relative to Cartesian crystallographic axes x, y, z where, following Birss [16], we take $C_3 \parallel z$ and $C_2 \parallel x$. A laboratory reference frame which coincides with the crystallographic system is used to describe the incident, reflected and transmitted waves, for which the electric fields have the form in (1). Standard Maxwell boundary conditions are applied to the light wave fields at the vacuum–crystal interface. Although these boundary conditions place restrictions on the normal components of $\boldsymbol{D}$ and $\boldsymbol{B}$ and also on the tangential components of $\boldsymbol{E}$ and $\boldsymbol{H}$, they yield at most four independent relationships which can be obtained from the conditions on the $\boldsymbol{E}$ and $\boldsymbol{H}$ fields [17]. These relationships may be rewritten in the form [18]

$$\left(E_r^{(0)}\right)_j = R_{jk}\left(E_i^{(0)}\right)_k \tag{7}$$

where R_{jk} is the 2×2 reflection matrix that relates the Cartesian components of the reflected electric field amplitude $\boldsymbol{E}_r^{(0)}$ to those of the incident electric field amplitude $\boldsymbol{E}_i^{(0)}$. Various forms can be obtained for R_{jk} which depend on the choice that is made for the $\boldsymbol{D}$ and $\boldsymbol{H}$ fields. However, time-reversal symmetry (reciprocity) imposes the following condition on the matrices that describe normal-incidence reflection from a sample in two time-conjugated equilibrium states (t) and $(-t)$ [7]:

$$R_{jk}(t) = R_{kj}(-t). \tag{8}$$

We have found that the relationship in (8) is satisfied only when covariant $\boldsymbol{D}$ and $\boldsymbol{H}$ fields are used in the standard Maxwell boundary conditions. For an optically inactive antiferromagnet such as Cr_2O_3 [16] the appropriate forms are [11]

$$D_\alpha = \left[\epsilon_0\delta_{\alpha\beta} + \tilde{\alpha}_{\alpha\beta} - \tfrac{1}{3}\mathrm{i}\left(\tilde{a}'_{\alpha\beta\gamma} + \tilde{a}'_{\beta\alpha\gamma} + \tilde{a}'_{\gamma\alpha\beta}\right)\nabla_\gamma\right]E_\beta + \tilde{T}_{\alpha\beta}B_\beta \tag{9}$$

$$H_\alpha = -\tilde{T}_{\beta\alpha}E_\beta + \mu_0^{-1}\delta_{\alpha\beta}B_\beta \tag{10}$$

where

$$\tilde{T}_{\alpha\beta} = \tilde{G}_{\alpha\beta} - \tfrac{1}{3}\delta_{\alpha\beta}\tilde{G}_{\gamma\gamma} - \tfrac{1}{6}\omega\epsilon_{\beta\gamma\delta}\tilde{a}'_{\gamma\delta\alpha}. \tag{11}$$

As antiferromagnetic Cr_2O_3 has magnetic point group symmetry $\underline{\bar{3}}\,\underline{m}$ [19], we find from Birss's tables [16] and the symmetry property [14]

$$\tilde{a}'_{\alpha\beta\gamma} = \tilde{a}'_{\alpha\gamma\beta} \tag{12}$$

that the non-vanishing components of the polarizability tensors $\tilde{\alpha}_{\alpha\beta}$, $\tilde{G}_{\alpha\beta}$, and $\tilde{a}'_{\alpha\beta\gamma}$ are

$$\begin{aligned} &\tilde{\alpha}_{xx} = \tilde{\alpha}_{yy}, \quad \tilde{\alpha}_{zz} \\ &\tilde{G}_{xx} = \tilde{G}_{yy}, \quad \tilde{G}_{zz} \\ &\tilde{a}'_{xyz} = \tilde{a}'_{xzy} = -\tilde{a}'_{yxz} = -\tilde{a}'_{yzx} \\ &\tilde{a}'_{xxx} = -\tilde{a}'_{yyx} = -\tilde{a}'_{yxy} = -\tilde{a}'_{xyy}. \end{aligned} \tag{13}$$

Although $\tilde{G}_{\alpha\beta}$ and $\tilde{a}'_{\alpha\beta\gamma}$ are origin-dependent tensors, the combinations in (3) and (11), which enter expressions for observables in transmission [14] and reflection [11] respectively, can be shown to be origin-independent (see [11] for details), as is required of a physical property.

3.1. Light path parallel to z axis (optic axis)

For propagation parallel to the optic axis, the unit wave normal $\boldsymbol{\sigma} = (0, 0, 1)$. We then find from (3) and (13) that the propagation equation (2) can be written in component form as

$$\begin{bmatrix} -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_x & 0 & 0 \\ 0 & -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_x & 0 \\ 0 & 0 & \epsilon_0^{-1}\tilde{\epsilon}_z \end{bmatrix} \begin{bmatrix} E_x^{(0)} \\ E_y^{(0)} \\ E_z^{(0)} \end{bmatrix} = 0 \tag{14}$$

where $\tilde{\epsilon}_k = \epsilon_0 + \tilde{\alpha}_{kk}$. Equation (14) shows that the characteristic waves travelling along the z axis in the crystal are polarized parallel to the x and y axes and have corresponding refractive indices

$$\tilde{n}_x = \tilde{n}_y = (\epsilon_0^{-1}\tilde{\epsilon}_x)^{\frac{1}{2}}. \tag{15}$$

As the tensors $\tilde{G}_{\alpha\beta}$ and $\tilde{a}'_{\alpha\beta\gamma}$ do not enter (14), the magnetoelectric properties of Cr_2O_3 cannot be determined in transmission when the light path is along the optic axis [14].

The $\boldsymbol{H}$ fields that are associated with the two characteristic waves in the crystal may be determined from (10), (11) and (13) and have the following amplitude components:

$$\boldsymbol{E} \parallel x : H_x^{(0)} = \tilde{T}_{xx} E_x^{(0)}, \qquad H_y^{(0)} = (\mu_0 c)^{-1} \tilde{n}_x E_y^{(0)} \tag{16}$$

$$\boldsymbol{E} \parallel y : H_x^{(0)} = -(\mu_0 c)^{-1} \tilde{n}_x E_y^{(0)}, \qquad H_y^{(0)} = \tilde{T}_{yy} E_y^{(0)} \tag{17}$$

where

$$\tilde{T}_{xx} = \tilde{T}_{yy} = \tfrac{1}{3}\big(\tilde{G}_{zz} - \tilde{G}_{xx} - \tfrac{1}{2}\omega \tilde{a}'_{xyz}\big) \tag{18}$$

and use has also been made of (15) and the Maxwell equation

$$\nabla \times \boldsymbol{E} = -\dot{\boldsymbol{B}}. \tag{19}$$

Application of the Maxwell condition of continuity to the tangential components of the $\boldsymbol{E}$ and $\boldsymbol{H}$ fields of the incident, reflected and transmitted waves at the vacuum–crystal interface then yields the reflection matrix

$$R(z) = \begin{bmatrix} (1 - \tilde{n}_x)/(1 + \tilde{n}_x) & -2\mu_0 c \tilde{T}_{xx}/(1 + \tilde{n}_x)^2 \\ 2\mu_0 c \tilde{T}_{xx}/(1 + \tilde{n}_x)^2 & (1 - \tilde{n}_x)/(1 + \tilde{n}_x) \end{bmatrix} \tag{20}$$

in which terms that are quadratic in $\tilde{T}_{xx}$ have been neglected. Because $\tilde{T}_{xx}$ is time-odd, the matrix in (20) satisfies the reciprocity condition in (8).

3.2. Light path parallel to x axis

We have previously shown [14] that for propagation along the C_2 axis in Cr_2O_3 the refractive indices and polarization states of the two characteristic waves are

wave 1:

$$\tilde{n}_1 = (\epsilon_0^{-1}\tilde{\epsilon}_y)^{\frac{1}{2}}\big[1 + \tfrac{1}{2}\mu_0 \tilde{C}^2/(\tilde{\epsilon}_y - \tilde{\epsilon}_z)\big] \mp \tfrac{1}{2}\mu_0 c \omega \tilde{a}'_{xxx} \tag{21}$$

$$E_z^{(0)}/E_y^{(0)} = \tilde{C}(\mu_0\tilde{\epsilon}_y)^{\frac{1}{2}}/(\tilde{\epsilon}_y - \tilde{\epsilon}_z) \tag{22}$$

wave 2:

$$\tilde{n}_2 = (\epsilon_0^{-1}\tilde{\epsilon}_z)^{\frac{1}{2}}\big[1 - \tfrac{1}{2}\mu_0 \tilde{C}^2/(\tilde{\epsilon}_y - \tilde{\epsilon}_z)\big] \tag{23}$$

$$E_y^{(0)}/E_z^{(0)} = -\tilde{C}(\mu_0\tilde{\epsilon}_z)^{\frac{1}{2}}/(\tilde{\epsilon}_y - \tilde{\epsilon}_z) \tag{24}$$

where

$$\tilde{\epsilon}_k = \epsilon_0 + \tilde{\alpha}_{kk} \tag{25}$$

$$\tilde{C} = \tilde{G}_{zz} - \tilde{G}_{xx} - \tfrac{1}{2}\omega\tilde{a}'_{xyz} = 3\tilde{T}_{xx}. \tag{26}$$

From (10), (11), (13) and (19) it follows that for propagation in the x direction the amplitude components of the $\boldsymbol{H}$ field are

$$H_y^{(0)} = -\tilde{n}(\mu_0 c)^{-1}E_z^{(0)} + \tilde{T}_{yy}E_y^{(0)} \tag{27}$$

$$H_z^{(0)} = \tilde{n}(\mu_0 c)^{-1}E_y^{(0)} + \tilde{T}_{zz}E_z^{(0)} \tag{28}$$

where

$$\tilde{T}_{zz} = \tfrac{2}{3}\big(\tilde{G}_{xx} - \tilde{G}_{zz} + \tfrac{1}{2}\omega\tilde{a}'_{xyz}\big) = -2\tilde{T}_{yy} = -2\tilde{T}_{xx}. \tag{29}$$

The appropriate forms of (27) and (28) for the characteristic waves 1 and 2 may be determined with the aid of (21)–(26).

The boundary conditions on the $\boldsymbol{E}$ and $\boldsymbol{H}$ fields at the interface then yield, to first order in $\tilde{T}_{xx}$, the reflection matrix

$$R(x) = \begin{bmatrix} (1-\tilde{n}_y)/(1+\tilde{n}_y) & \tilde{R}_{yz} \\ \tilde{R}_{zy} & (1-\tilde{n}_z)/(1+\tilde{n}_z) \end{bmatrix} \tag{30}$$

where

$$\tilde{n}_y = \tilde{n}_x = (\epsilon_0^{-1}\tilde{\epsilon}_x)^{\frac{1}{2}} \tag{31}$$

$$\tilde{n}_z = (\epsilon_0^{-1}\tilde{\epsilon}_z)^{\frac{1}{2}} \tag{32}$$

$$\tilde{R}_{yz} = -\tilde{R}_{zy} = 2\mu_0 c\tilde{T}_{xx}(2\tilde{n}_x - \tilde{n}_z)/(\tilde{n}_x + \tilde{n}_z)(1+\tilde{n}_x)(1+\tilde{n}_z). \tag{33}$$

It follows from (33) that $\tilde{R}_{yz}$ and $\tilde{R}_{zy}$ change sign under time reversal because $\tilde{T}_{xx}$ is time-odd and hence that the matrix in (30) satisfies the reciprocity requirement in (8).

4. Comparison with experiment

The properties predicted by our theory for reflection from Cr_2O_3 are most easily compared with experiment [12] if we assume $\tilde{n}_x = \tilde{n}_z$. (This is a reasonable approximation since the measured value for the birefringence [12] is $n_z - n_x = 5.8 \times 10^{-2}$ and also since $n_z - n_x$ does not enter our results in (20) or (30)–(33).) We then find from (30), (31) and (33) that

$$R(x) = \begin{bmatrix} (1-\tilde{n}_x)/(1+\tilde{n}_x) & \mu_0 c\tilde{T}_{xx}/(1+\tilde{n}_x)^2 \\ -\mu_0 c\tilde{T}_{xx}/(1+\tilde{n}_x)^2 & (1-\tilde{n}_x)/(1+\tilde{n}_x) \end{bmatrix}. \tag{34}$$

As the off-diagonal elements in the reflection matrices in (20) and (34) are linear in the time-odd property $\tilde{T}_{xx}$, the rotation and circular dichroism in the reflected beam are non-reciprocal. This has been confirmed experimentally [12].

Inspection of (20) and (34) shows that for incident linearly polarized light of azimuth $\phi_i = 0^\circ$ and $\phi_i = 90^\circ$, the rotation $\Delta\phi$ for $\boldsymbol{\sigma} \parallel \boldsymbol{z}$ is twice that for $\boldsymbol{\sigma} \parallel \boldsymbol{x}$. Experiment [12], however, yields the ratio $(\Delta\phi)_z/(\Delta\phi)_x \simeq 1.6$. The difference between this value and the predicted value of two is probably due to the presence of microscopic surface effects [13] which are not accounted for in our macroscopic theory.

Because the off-diagonal elements in (20) are of opposite sign to those in (34), the rotations when $\phi_i = 0^\circ,\ 90^\circ$ and the circular dichroism for $\boldsymbol{\sigma} \parallel \boldsymbol{z}$ are of opposite sign, for a given spin form of Cr_2O_3, to the corresponding properties when $\boldsymbol{\sigma} \parallel \boldsymbol{x}$. This is observed in practice [12].

It can also be shown from (20) and (34), in agreement with experiment [12], that for $\boldsymbol{\sigma} \parallel \boldsymbol{x}$ and an azimuth $\phi_i = 45°$, the difference in the rotations for the two spin forms of Cr_2O_3 is equal in magnitude to the rotation for either time form when $\boldsymbol{\sigma} \parallel \boldsymbol{z}$.

5. Discussion

Several interesting observations follow from the multipole theory presented in this paper. As shown in [11], the frequency-dependent magnetoelectric tensor $\tilde{T}_{\alpha\beta}$ is an origin-independent combination of magnetic dipole and electric quadrupole tensors. The latter contribution vanishes in the low-frequency limit ($\omega \to 0$) where the magnetoelectric effect is described by the magnetic dipole tensor $\tilde{G}_{\alpha\beta}$ alone [20].

A novel result, which has not previously been noted, is the relationship in (29) between the components of the frequency-dependent magnetoelectric tensor for Cr_2O_3, namely

$$\tilde{T}_{zz} = -2\tilde{T}_{xx} = -2\tilde{T}_{yy} = \tfrac{2}{3}\left(\tilde{G}_{xx} - \tilde{G}_{zz} + \tfrac{1}{2}\omega\tilde{a}'_{xyz}\right)$$

which shows that for $\tilde{T}_{\alpha\beta}$ in the form

$$\tilde{T}_{\alpha\beta} = t'_{\alpha\beta} + \mathrm{i}t''_{\alpha\beta} \tag{35}$$

the expected ratio of components for the real and the imaginary parts ($t'_{\alpha\beta}$ and $t''_{\alpha\beta}$, respectively) is

$$t'_{zz}/t'_{xx} = t''_{zz}/t''_{xx} = -2. \tag{36}$$

The components of $t'_{\alpha\beta}$ and $t''_{\alpha\beta}$ have been evaluated for Cr_2O_3 from measurements in reflection of the change in azimuth and the circular dichroism, respectively [12], and these values yield

$$t'_{zz}/t'_{xx} = t''_{zz}/t''_{xx} \simeq -2.4. \tag{37}$$

Again a possible explanation for the difference between the theoretical and observed values for these ratios is the existence of surface effects which are of a microscopic nature and may be attributed to the rotating power of the first ferromagnetic layer in an antiferromagnetic crystal [13]. However, these effects are absent in transmission, and it follows from (22), (24) and (26) that the bulk value for $\tilde{T}_{xx}$, evaluated from the rotation of the optical indicatrix when propagation is along the C_2 axis in Cr_2O_3, could be used to obtain an estimate of the surface contributions to $\tilde{T}_{xx}$ and $\tilde{T}_{zz}$ in reflection.

It can be shown from the covariant $\boldsymbol{D}$ and $\boldsymbol{H}$ fields in (9)–(11) and from Birss's tables [16] that cubic antiferromagnets belonging to the symmetry classes $m3$, 432, $\underline{4}3\underline{m}$, $m3m$, $m3\underline{m}$ and $\underline{m}3\underline{m}$ may not exhibit bulk magnetic effects induced by a time-harmonic wave. Consequently these crystals may be suitable for the direct observation of surface effects in reflection [13].

Finally, by using the new covariant multipole forms for the constitutive relations for a magnetic medium [11], we have produced a quantitative theory of reflection from Cr_2O_3 which satisfies the reciprocity condition in (8) and which also yields results that agree well with experiment [12].

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