# Extensions to the Maxwell boundary conditions: Reflection from uniaxial and cubic antiferromagnets

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Standard Maxwell boundary conditions do not always apply at the surface of an anisotropic magnetic medium in a vacuum, even when the **D** and **H** fields that describe the properties of the medium are taken in covariant form. It is shown that discontinuities may arise in these fields due to bound surface current and electric-dipole moment, and the leading multipole forms of these discontinuities are identified. Reflection matrices for normal incidence are derived for all uniaxial and cubic antiferromagnetic crystals. These matrices satisfy reciprocity (time-reversal symmetry) only when the extended boundary conditions are used. Various magnetic point groups are identified that exhibit nonreciprocal effects in reflection, but not in transmission. It is also shown that when nonreciprocal birefringence exists in transmission, nonreciprocal effects are absent in reflection at normal incidence. The magnetic point-group symmetry assigned to $Nd_2CuO_4$ is questioned on the basis of the theory. [S0163-1829(99)00106-X]

## I. INTRODUCTION

Theories of reflection that satisfy the requirements of space-time symmetry[1,2] have recently been derived for various anisotropic nonmagnetic media[3,4] with the aid of standard Maxwell boundary conditions[5] and covariant constitutive relations to the order of electric quadrupole and magnetic dipole.[6] A similar procedure applied to a uniaxial magnetic crystal, namely, antiferromagnetic chromium oxide ($Cr_2O_3$),[7] has also been successful in producing a theory of reflection for normal incidence with the correct symmetry properties which, in addition, agrees well with experiment.[8] In this paper the theory is extended to all antiferromagnetic crystals of the uniaxial and cubic systems. An interesting point that arises in this treatment is that, even for normal incidence, there are magnetic media to which the standard boundary conditions do not apply in the electric-quadrupole–magnetic-dipole approximation. Examples of such media are cubic crystals with the magnetic point-group symmetries $\underline{m}3m$, $\underline{m}3$, $\bar{4}3\underline{m}$, $4\underline{3}\underline{2}$, and 23 and uniaxial crystals belonging to the magnetic point groups listed in III–VI in Table I. A characteristic feature of these point groups is the existence of a third-rank time-odd polar tensor that is totally symmetric in its three subscripts. As shown in Sec. II, this tensor is associated with discontinuities due to bound surface current and electric dipole moment that may occur in the light-wave fields at an interface. The validity of boundary conditions that do not include such a tensor has previously and presciently been questioned.[8]

In this paper we make use of covariant multipole forms for the **D** and **H** fields and work to the order of induced electric quadrupoles and magnetic dipoles. The boundary conditions on the **D** and **H** fields at the surface of a source-free, anisotropic, magnetic medium in a vacuum are determined in Sec. II to this multipole order. These conditions are shown to reduce to the standard Maxwell forms[5] for all nonmagnetic media and also for certain magnetic media.

Since reflection matrices have been derived to the order of electric quadrupole and magnetic dipole for all nonmagnetic crystals of the uniaxial[3] and cubic[9] systems, using the standard Maxwell boundary conditions, the concern of this paper is to demonstrate the need to extend these boundary conditions when reflection from magnetic crystals is described. To this end only the magnetic contributions of electric-quadrupole–magnetic-dipole order are included in the theory of reflection from antiferromagnetic crystals that is developed in Sec. III.

In Sec. IV the theory is applied to uniaxial antiferromagnets for normal incidence on a plane face perpendicular to the optic axis. Various point groups are identified for which the nonreciprocal properties in reflection turn out to be the same as those displayed by $Cr_2O_3$ (symmetry $\bar{3}\underline{m}$), namely, a change in azimuth of an incident linearly polarized light beam and circular dichroism.[7,8] (These effects are termed nonreciprocal because they have opposite signs for the two states of a crystal that are related to each other by time reversal.) Another prediction, somewhat unexpected, is the absence in reflection of nonreciprocal effects for classes of tetragonal magnetoelectrics that have been shown to exhibit nonreciprocal linear birefringence for propagation along their optic axis.[10–12] Magnetic cubic crystals are considered in Sec. V where reflection matrices are derived for normal incidence on a cube face. Concluding remarks follow in Sec. VI.

## II. COVARIANT CONSTITUTIVE RELATIONS AND BOUNDARY CONDITIONS

In any theory of reflection two aspects of the problem require careful consideration, namely, the choice that is made for the **D** and **H** fields that describe the response properties of the medium to the fields of the light wave, and the boundary conditions that apply to these fields at an interface.

### A. Covariant constitutive relations

The importance of using covariant forms for **D** and **H** to ensure the spatial and temporal invariance of the equations of

 

TABLE I. Components of $\alpha_{\alpha\beta}$, $A_{\alpha\beta\gamma}$, $t_{\alpha\beta}$, and $S_{\alpha\beta\gamma}$ relative to Cartesian crystallographic axes 1,2,3 for uniaxial antiferromagnets of the magnetic point groups shown, when the 3 axis (optic axis) is parallel to the $z$ axis of a laboratory reference frame $0(x,y,z)$ lying along the normal to the reflecting surface in which the 1 and 2 axes are at an angle $\theta$ to the $x$ and $y$ axes, respectively. The noncentrosymmetric point groups are shown in parentheses. The remaining point groups possess a geometric center of symmetry. For I–IV: $\alpha_{xx}=\alpha_{yy}$, $A_{xxz}=A_{yyz}$, $t_{xx}=t_{yy}$, $t_{xy}=-t_{yx}$, $S_{xxz}=S_{yyz}$ For V and VI: $\alpha_{xx}=\alpha_{yy}$, $A_{xxz}=-A_{yyz}$, $t_{xx}=-t_{yy}$, $t_{xy}=t_{yx}$, $S_{xxz}=-S_{yyz}$, $A_1=\frac{1}{3}(G_{33}-G_{11}-\frac{1}{2}\omega a'_{123})$, $A_2=2G_{12}+\omega a'_{113}$, $A_3=\omega a'_{333}$, $A_4=G_{12}+\frac{1}{6}\omega(a'_{113}-a'_{311})$, $A_5=\frac{1}{3}\omega(2a'_{113}+a'_{311})$, $A_6=-2G_{11}+\omega a'_{123}$, $A_7=G_{11}-\frac{1}{6}\omega(a'_{123}-a'_{312})$, $A_8=\frac{1}{3}\omega(2a'_{123}+a'_{312})$, $S=\sin 2\theta$, $C=\cos 2\theta$.

| | Magnetic point group | $\alpha_{xx}$ | $\alpha_{zz}$ | $A_{xxz}$ | $A_{xyz}$ | $A_{zzz}$ | $t_{xx}$ | $t_{xy}$ | $S_{xxz}$ | $S_{xyz}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| I | $\underline{6}/mm\underline{m},\underline{6}/m(\bar{6}m2,\underline{6}m\underline{m},\underline{6}2\underline{2},\underline{6})$ | $\alpha_{11}$ | $\alpha_{33}$ | | | | | | | |
| II | $6/\underline{mmm},\underline{\bar{3}m},4/\underline{mmm}$ $(\underline{\bar{6}m}2,622,32,\underline{\bar{4}}2\underline{m},422)$ | $\alpha_{11}$ | $\alpha_{33}$ | | | | $-A_1$ | | | |
| III | $6/\underline{m}mm,\underline{\bar{3}}m,4/\underline{m}mm$ $(\underline{\bar{6}}m\underline{2},6mm,3m,\underline{\bar{4}2}m,4mm)$ | $\alpha_{11}$ | $\alpha_{33}$ | $A_2$ | | $A_3$ | | $A_4$ | $A_5$ | |
| IV | $6/\underline{m},\underline{\bar{3}},4/\underline{m},(\underline{\bar{6}},\underline{\bar{4}})$ | $\alpha_{11}$ | $\alpha_{33}$ | $A_2$ | | $A_3$ | $-A_1$ | $A_4$ | $A_5$ | |
| V | $\underline{4}/\underline{m}m\underline{m},(\bar{4}2m,\underline{4}m\underline{m},\underline{4}22)$ | $\alpha_{11}$ | $\alpha_{33}$ | $SA_6$ | $CA_6$ | | $CA_7$ | $-SA_7$ | $CA_5$ | $-SA_5$ |
| VI | $\underline{4}/\underline{m},(\underline{4})$ | $\alpha_{11}$ | $\alpha_{33}$ | $SA_6+CA_2$ | $CA_6-SA_2$ | | $CA_7+SA_4$ | $CA_4-SA_7$ | $CA_5+SA_8$ | $CA_8-SA_5$ |

continuity at an interface has been discussed in a number of articles.[6,13,14] Also reported in these articles are the appropriate forms for **D** and **H** in the electric-quadrupole–magnetic-dipole approximation for a magnetic, anisotropic, chiral medium exposed to a light wave with time dependence $\exp\{-i\omega t\}$. Although the expressions for **D** and **H** were previously displayed in the format adopted by Post,[15] it is desirable when discussing boundary conditions to rewrite them in multipole form. To the order of electric quadrupoles and magnetic dipoles the multipole expansions for **D** and **H** are[16,17]

$$D_\alpha=\epsilon_0 E_\alpha+P_\alpha-\frac{1}{2}\nabla_\beta Q_{\alpha\beta}+\cdots, \tag{1}$$

$$H_\alpha=\mu_0^{-1}B_\alpha-M_\alpha+\cdots, \tag{2}$$

where $P_\alpha$, $Q_{\alpha\beta}$, and $M_\alpha$ are, in Cartesian tensor notation, macroscopic densities of electric-dipole moment, electric-quadrupole moment, and magnetic-dipole moment, respectively. We then find by inspection from the covariant constitutive relations in Refs. 6, 13, and 14 the following expressions for the polarization densities in Eqs. (1) and (2):

$$P_\alpha=F_{\alpha\beta}E_\beta-\frac{1}{6}iS_{\alpha\beta\gamma}\nabla_\gamma E_\beta+(t_{\alpha\beta}-it'_{\alpha\beta})B_\beta+\cdots, \tag{3}$$

$$Q_{\alpha\beta}=\frac{1}{3}iS_{\alpha\beta\gamma}E_\gamma+\cdots, \tag{4}$$

$$M_\alpha=(t_{\beta\alpha}+it'_{\beta\alpha})E_\beta+\cdots, \tag{5}$$

where

$$F_{\alpha\beta}=\alpha_{\alpha\beta}-i\alpha'_{\alpha\beta}, \tag{6}$$

$$S_{\alpha\beta\gamma}=a'_{\alpha\beta\gamma}+a'_{\beta\gamma\alpha}+a'_{\gamma\alpha\beta}, \tag{7}$$

$$t_{\alpha\beta}=G_{\alpha\beta}-\frac{1}{3}\delta_{\alpha\beta}G_{\gamma\gamma}-\frac{1}{6}\omega\epsilon_{\beta\gamma\delta}a'_{\gamma\delta\alpha}, \tag{8}$$

$$t'_{\alpha\beta}=G'_{\alpha\beta}-\frac{1}{2}\omega\epsilon_{\beta\gamma\delta}a_{\gamma\delta\alpha}. \tag{9}$$

In Eqs. (8) and (9), $\delta_{\alpha\beta}$ is the Kronecker delta and $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita tensor. The remaining tensors on the right-hand sides of Eqs. (6)–(9) are macroscopic property tensors of the medium to which symmetry considerations may be properly applied.[18] These tensors describe the induction of multipole moments in a macroscopic volume element of the material by the light-wave fields and their space and time derivatives.[14] Certain of these tensors exhibit permutation symmetry in their subscripts[14] as shown below:

$$\alpha_{\alpha\beta}=\alpha_{\beta\alpha}, \quad \alpha'_{\alpha\beta}=-\alpha'_{\beta\alpha}, \tag{10}$$

$$a_{\alpha\beta\gamma}=a_{\alpha\gamma\beta}, \quad a'_{\alpha\beta\gamma}=a'_{\alpha\gamma\beta}. \tag{11}$$

It also follows from the second equality in Eq. (11) that $S_{\alpha\beta\gamma}$ in Eq. (7) is symmetric in the permutation of all three subscripts $\alpha$, $\beta$, and $\gamma$. It has previously been shown that the tensors $\alpha_{\alpha\beta}, G'_{\alpha\beta}$, and $a_{\alpha\beta\gamma}$ are time even and $\alpha'_{\alpha\beta}$, $G_{\alpha\beta}$, and $a'_{\alpha\beta\gamma}$ time odd,[12] so that from Eq. (7) $S_{\alpha\beta\gamma}$ is time odd. (Time-even property tensors may exist for both nonmagnetic and magnetic media, while time-odd ones belong only to magnetic materials.[18])

### B. Boundary conditions

In order to determine the matching condition on the **H** fields at an interface between a medium and a vacuum, we consider the total bound current density at a macroscopic volume element of the material. It follows from a multipole expansion of the vector potential[17] that in the electric-quadrupole–magnetic-dipole approximation there are two contributions: a volume density of bound current $\mathbf{J}_b$ as well as a surface density of bound current $\mathbf{K}_b$ where

$$J_{b_\alpha}=\dot{P}_\alpha-\frac{1}{2}\nabla_\beta\dot{Q}_{\alpha\beta}+\epsilon_{\alpha\beta\gamma}\nabla_\beta M_\gamma, \tag{12}$$

$$K_{b_\alpha}=\left(\frac{1}{2}\dot{Q}_{\alpha\beta}-\epsilon_{\alpha\beta\gamma}M_\gamma\right)\hat{n}_\beta; \tag{13}$$

an overdot denotes partial differentiation with respect to time, and $\hat{\mathbf{n}}$ is the outward unit normal to the surface.

For time-dependent light-wave fields **E** and **B** in matter, Ampère's law takes the form[19]

$$\nabla\times\mathbf{B}=\mu_0(\epsilon_0\dot{\mathbf{E}}+\mathbf{J}), \tag{14}$$

where **J** is the total current density, both free and bound, in the medium. Application of standard procedures[5] to the integral form of Eq. (14) yields the following condition on the tangential components of **B** at a vacuum-dielectric interface:

$$\hat{\mathbf{n}}\times(\mathbf{B}_2-\mathbf{B}_1)=\mu_0\mathbf{K}, \tag{15}$$

where medium 1 is the dielectric, medium 2 is the vacuum, $\hat{\mathbf{n}}$ is here the unit normal at the interface that points from the first to the second medium, and **K** is the total surface current density. In the absence of free current the only contribution to **K** is that in Eq. (13). Substitution of Eq. (13) into Eq. (15) followed by rearrangement, in which use is made of Eq. (2), yields the following boundary condition on the **H** fields:

$$\epsilon_{\alpha\beta\gamma}\hat{n}_\beta(H_{2\gamma}-H_{1\gamma})=\frac{1}{2}\dot{Q}_{\alpha\beta}\hat{n}_\beta. \tag{16}$$

Although not used here the matching condition on the **D** fields is included for completeness. To the order of electric quadrupoles and magnetic dipoles it is

$$\hat{n}_\alpha(D_{2\alpha}-D_{1\alpha})=-\frac{1}{2}\nabla_\beta Q_{\alpha\beta}\hat{n}_\alpha \quad (\alpha\neq\beta). \tag{17}$$

The above equation is obtained when the multipole expression for the surface density of electric-dipole moment **p**, namely,[20]

$$p_\beta=-\frac{1}{2}Q_{\alpha\beta}\hat{n}_\alpha, \tag{18}$$

is substituted into the S.I. version of Langreth's result for the discontinuity in the normal component of **D**.[21]

When $Q_{\alpha\beta}=0$ in Eqs. (16) and (17), the standard Maxwell boundary conditions on the **D** and **H** fields are recovered. It follows from Eq. (4) that these conditions apply to all nonmagnetic media because for them the time-odd tensor $S_{\alpha\beta\gamma}$ in Eq. (7) does not exist.[18] Certain magnetic media also have $Q_{\alpha\beta}=0$ because $S_{\alpha\beta\gamma}$ vanishes for symmetry reasons, as Table I shows.

We find for normal incidence that there are no discontinuities at the interface in the normal component of **B** or, as shown by Langreth,[21] in the tangential components of **E**.

## III. REFLECTION FROM ANTIFERROMAGNETIC CRYSTALS

In this paper we consider antiferromagnets of the uniaxial and cubic systems and present details of reflection only for normal incidence.

The properties of a crystal are traditionally specified relative to its crystallographic Cartesian axes, here denoted 1,2,3 with the 3 axis that of highest symmetry,[18] which in a uniaxial crystal is also the optic axis. The crystal is oriented with respect to a laboratory system of Cartesian axes $x,y,z$ such that $3\|z$ and the 1 and 2 crystallographic axes are at an arbitrary angle $\theta$ to the $x$ and $y$ axes, respectively, which lie in the reflecting surface. Tensor components in the laboratory frame $0(x,y,z)$ can be expressed in terms of those in the crystallographic system 0(1,2,3) by means of the transformation for a rotation

$$X_{\alpha\beta\ldots}=X_{ij\ldots}a_i^\alpha a_j^\beta\ldots, \tag{19}$$

where $a_i^\alpha$ is the direction cosine between the $\alpha$ axis in $0(x,y,z)$ and the $i$ axis in 0(1,2,3).

For antiferromagnets $\alpha'_{\alpha\beta}$ in Eq. (6) is zero.[12] Also, as explained in Sec. I, the time-even tensors $G'_{\alpha\beta}$ and $a_{\alpha\beta\gamma}$ in Eq. (9) are not included here. Thus the appropriate forms of the constitutive relations in Eqs. (1) and (2) are from Eqs. (3)–(6)

$$D_\alpha=\left(\epsilon_0\delta_{\alpha\beta}+\alpha_{\alpha\beta}-\frac{1}{3}iS_{\alpha\beta\gamma}\nabla_\gamma\right)E_\beta+t_{\alpha\beta}B_\beta, \tag{20}$$

$$H_\alpha=-t_{\beta\alpha}E_\beta+\mu_0^{-1}B_\alpha, \tag{21}$$

where $S_{\alpha\beta\gamma}$ and $t_{\alpha\beta}$ are defined, respectively, in Eqs. (7) and (8).

We then find from Eqs. (20) and (21) and the two inhomogeneous Maxwell equations that, for a plane monochromatic light wave of angular frequency $\omega$ with an electric field of the form

$$\mathbf{E}=\mathbf{E}^{(0)}\exp\{i\omega(n\boldsymbol{\sigma}\cdot\mathbf{r}/c-t)\}, \tag{22}$$

the propagation equation is[12]

$$[n^2(\sigma_\alpha\sigma_\beta-\delta_{\alpha\beta})+\delta_{\alpha\beta}+\epsilon_0^{-1}\alpha_{\alpha\beta}+n\mu_0c\sigma_\gamma A_{\alpha\beta\gamma}]E_\beta^{(0)}=0, \tag{23}$$

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

$$A_{\alpha\beta\gamma}=-\epsilon_{\alpha\gamma\delta}G_{\beta\delta}-\epsilon_{\beta\gamma\delta}G_{\alpha\delta}+\frac{1}{2}\omega(a'_{\alpha\beta\gamma}+a'_{\beta\alpha\gamma})=A_{\beta\alpha\gamma}. \tag{24}$$

Nontrivial solutions for the components $\mathbf{E}^{(0)}$ in Eq. (23) may be found by setting the determinant of the coefficients equal to zero. The roots of the determinantal equation are the refractive indices of the propagating waves which, when substituted into Eq. (23), yield the corresponding eigenpolarizations in terms of the components of $\mathbf{E}^{(0)}$.

It follows from Eqs. (21) and (22) and the Maxwell equation

$$\nabla\times\mathbf{E}=-\dot{\mathbf{B}}, \tag{25}$$

that the associated **H** fields are given by

$$H_\alpha=(\mu_0c)^{-1}n\epsilon_{\alpha\beta\gamma}\sigma_\beta E_\gamma-t_{\beta\alpha}E_\beta. \tag{26}$$

At the interface $(z=0)$ the tangential components of the electric field are continuous,[21] i.e.,

$$\epsilon_{\alpha z\gamma}(E_{2\gamma}-E_{1\gamma})=0, \tag{27}$$

while from Eqs. (16), (4), and (22) the condition on the **H** fields is

$$\epsilon_{\alpha z\gamma}(H_{2\gamma}-H_{1\gamma})=\frac{1}{6}\omega S_{\alpha\gamma z}E_{1\gamma}. \tag{28}$$

The boundary conditions in Eqs. (27) and (28) yield four independent relationships that can be rewritten in the form[22]

$$(E_r^{(0)})_j=R_{jk}(E_i^{(0)})_k, \tag{29}$$

where $R_{jk}$ is the $2\times2$ reflection matrix that relates the Cartesian components of the reflected electric-field amplitude $\mathbf{E}_r^{(0)}$ to those of the incident electric-field amplitude $\mathbf{E}_i^{(0)}$.

Equations (19)–(29) are applied to uniaxial antiferromagnets in Sec. IV and to cubic antiferromagnets in Sec. V.

## IV. UNIAXIAL ANTIFERROMAGNETS

In this section we consider reflection at normal incidence from uniaxial antiferromagnets when the light path is parallel to the optic axis. The magnetic point groups to which this class of crystal belongs have been identified from Birss's tables[18] and are listed in Table I. Here, the noncentrosymmetric crystal classes have been placed in parentheses to distinguish them from the remaining classes that possess a geometric center of symmetry.

Also shown in Table I are the nonvanishing components of the polarizability tensor $\alpha_{\alpha\beta}$ and of the time-odd tensors $A_{\alpha\beta\gamma}$, $t_{\alpha\beta}$, and $S_{\alpha\beta\gamma}$ [defined, respectively, in Eqs. (24), (8), and (7)] for the selected experimental arrangement. The various entries were obtained with the aid of Eq. (19) and the tabulations of tensor components for different crystal symmetries by Birss.[18] It is interesting that, with the exception of the point groups listed in V and VI in Table I, the crystal properties are independent of the relative orientation $\theta$ of the crystallographic and laboratory axes in the $xy$ plane.

In the electric-quadrupole–magnetic-dipole approximation, crystals with a geometric center of symmetry do not possess properties other than those shown in Table I for the experimental arrangement under consideration. On the other hand, additional contributions due to the time-even tensors $G'_{\alpha\beta}$ and $a_{\alpha\beta\gamma}$ may exist for the noncentrosymmetric classes in parentheses in Table I. These contributions have previously been determined for the relevant nonmagnetic uniaxial classes,[3] and also apply to the associated magnetic subgroups in each case. However, we have excluded such contributions from the theory in Sec. III, and hence the results that follow apply strictly only to centrosymmetric antiferromagnets, the point groups of which do not appear in parentheses in Table I.

For normal incidence $\boldsymbol{\sigma}=(0,0,1)$, and then the propagation equation (23) may be written as

$$\begin{bmatrix} -n^2+\mu_0c^2\epsilon_x+n\mu_0cA_{xxz} & n\mu_0cA_{xyz} & 0\\ n\mu_0cA_{xyz} & -n^2+\mu_0c^2\epsilon_x+n\mu_0cA_{yyz} & 0\\ 0 & 0 & \mu_0c^2\epsilon_z+n\mu_0A_{zzz}\end{bmatrix}\begin{bmatrix}E_x^{(0)}\\ E_y^{(0)}\\ E_z^{(0)}\end{bmatrix}=0, \tag{30}$$

where $\epsilon_k=\epsilon_0+\alpha_{kk}$, $\epsilon_x=\epsilon_y$, and use has been made of the symmetry property in Eq. (24).

Because the off-diagonal components of $A_{\alpha\beta\gamma}$ in Eq. (30) vanish for the magnetic point groups in I–IV of Table I, but not for those in V and VI, it is necessary to treat these two groups of crystal separately. The results that follow from Table I and Eqs. (26)–(30) are presented below.

### A. Magnetic point groups listed in I–IV in Table I

Reference to Table I shows that $A_{xyz}=0$ for the point groups in I–IV, while $A_{xxz}=A_{yyz}$ exists only for the classes in III and IV. It therefore follows from Eq. (30) that the characteristic waves are polarized parallel to the $x$ and $y$ axes with corresponding refractive indices

$$n_x=n_y=\frac{1}{2}\mu_0cA_{xxz}+N, \tag{31}$$

where

$$N=\frac{1}{2}c(\mu_0^2A_{xxz}^2+4\mu_0\epsilon_x)^{1/2}. \tag{32}$$

Because $n_x=n_y$ there is no birefringence for propagation along the optic axis. However, as shown previously,[12] a reversal of the light path changes the sign of $A_{xxz}$ in Eq. (31) and hence crystals belonging to the magnetic point groups listed in III and IV of Table I may exhibit directional birefringence, i.e., a difference in the refractive indices for forward and reverse propagation along the same path.

From Eqs. (26)–(28), (31), and the entries in Table I we find the following expressions for the elements of the reflection matrix $R_{jk}$ in Eq. (29):

$$R_{xx}=R_{yy}=(1-N)/(1+N), \tag{33}$$

$$R_{xy}=-R_{yx}=-2\mu_0ct_{xx}/(1+N)^2. \tag{34}$$

Since time reversal leaves $N$ unchanged, as Eq. (32) shows, but reverses the sign of $t_{xx}$ in Eq. (34), the reciprocity condition[1]

$$R_{jk}(t)=R_{kj}(-t) \tag{35}$$

is clearly satisfied by Eqs. (33) and (34).

It follows from Eq. (34) that the off-diagonal elements of the reflection matrix vanish when $t_{xx}=0$, and hence, from

Table I, that nonreciprocal effects in reflection may not exist for the magnetic point groups listed in I and III when the light path is parallel to the optic axis.

### B. Magnetic point groups listed in V and VI in Table I

For the magnetic point groups in V and VI in Table I, Eq. (30) yields the following eigenpolarizations and associated refractive indices:

$$(E_y^{(0)}/E_x^{(0)})_\pm = [\pm(A_{xxz}^2 + A_{xyz}^2)^{1/2} - A_{xxz}]/A_{xyz}, \quad (36)$$

$$n_\pm = \pm \frac{1}{2}\mu_0 c (A_{xxz}^2 + A_{xyz}^2)^{1/2} + V, \quad (37)$$

where

$$V = \frac{1}{2} c [\mu_0^2 (A_{xxz}^2 + A_{xyz}^2) + 4\mu_0 \epsilon_x]^{\frac{1}{2}}. \quad (38)$$

When Eqs. (36) and (37) are used in Eqs. (26)–(28), together with the relevant entries in Table I, the elements of the reflection matrix $R_{jk}$ in Eq. (29) are found to be

$$R_{xx} = R_{yy} = (1-V)/(1+V) \quad (39)$$

and

$$R_{xy} = R_{yx} = 0, \quad (40)$$

where

$$V = \frac{1}{2} c [\mu_0^2 (A_2^2 + A_6^2) + 4\mu_0 \epsilon_x]^{1/2} \quad (41)$$

for V and

$$V = \frac{1}{2} c [\mu_0^2 A_6^2 + 4\mu_0 \epsilon_x]^{1/2} \quad (42)$$

for VI. These results clearly satisfy the reciprocity condition in Eq. (35).

It is evident from Eqs. (39), (41), and (42) that despite the angular dependence of the entries for $A_{\alpha\beta\gamma}$, $t_{\alpha\beta}$, and $S_{\alpha\beta\gamma}$ in Table I, effects in reflection at normal incidence are independent of the orientation of the crystallographic 1 and 2 axes in the reflecting plane. Furthermore, nonreciprocal effects are absent in reflection because there are no contributions to Eqs. (39)–(42) that are linear in $G_{\alpha\beta}$ and $a'_{\alpha\beta\gamma}$. However it has previously been noted,[10–12] and is readily seen from Eq. (37), that nonreciprocal birefringence may exist in transmission.

## V. CUBIC ANTIFERROMAGNETS

The constitutive relations in Eqs. (20) and (21) have simpler forms for all magnetic point groups of the cubic system because $t_{\alpha\beta} = t_{\beta\alpha} = 0$, as Birss's tables[18] show. Thus magnetic effects in transmission and reflection are described solely by the symmetric tensor $S_{\alpha\beta\gamma}$ in Eq. (7). In this section we consider reflection from cubic crystals for which $S_{\alpha\beta\gamma}$ may exist. Reference to tables[18] shows that the relevant magnetic point-group symmetries are

$$\underline{m}3m,\ \underline{m}3,\ (\bar{4}3m,\ \underline{4}3\underline{2},\ 23), \quad (43)$$

where, once again, the noncentrosymmetric classes are shown in parentheses.

When propagation is along the normal to a cube face in crystals belonging to the point groups listed in Eq. (43), we find from tables[18] and Eq. (19) that the independent components of $S_{\alpha\beta\gamma}$ in Eq. (7) are

$$S_{xxz} = -S_{yyz} = a'_{123} \sin 2\theta, \quad (44)$$

$$S_{xyz} = a'_{123} \cos 2\theta, \quad (45)$$

where $\theta$ is the angle between a crystallographic axis and the corresponding laboratory axis in the reflecting surface. It also follows from the tables[18] and Eqs. (19), (23), and (24) that the propagation equation has the form in Eq. (30) with

$$\epsilon_x = \epsilon_z, \quad (46)$$

$$A_{xxz} = -A_{yyz} = \omega a'_{123} \sin 2\theta, \quad (47)$$

$$A_{xyz} = \omega a'_{123} \cos 2\theta, \quad (48)$$

$$A_{zzz} = 0. \quad (49)$$

Then, from Eqs. (30) and (46)–(49), the eigenpolarizations and associated refractive indices of the waves in the crystal are

$$(E_y^{(0)}/E_x^{(0)})_\pm = (\pm 1 - \cos 2\theta)/\sin 2\theta \quad (50)$$

and

$$n_\pm = \pm \frac{1}{2}\mu_0 c \omega a'_{123} + W, \quad (51)$$

where

$$W = \frac{1}{2} c [(\mu_0 \omega a'_{123})^2 + 4\mu_0 \epsilon_x]^{1/2}. \quad (52)$$

Subsequent use of Eq. (26), in which $t_{\beta\alpha} = 0$, and also of Eqs. (27), (28), (44), (45), (50), and (51) yields for the elements of $R_{jk}$ in Eq. (29),

$$R_{xx} = R_{yy} = (1-W)/(1+W), \quad (53)$$

$$R_{xy} = R_{yx} = 0, \quad (54)$$

where the expression for $W$ is given in Eq. (52). It is evident from Eqs. (52)–(54) that here also the reciprocity condition in Eq. (35) is satisfied. In addition, because there are no contributions linear in $a'_{123}$ to Eqs. (52)–(54), nonreciprocal effects may not occur in reflection at normal incidence from crystals with the point-group symmetries in Eq. (43), even though these crystals may exhibit linear birefringence in transmission that is proportional to $a'_{123}$,[12] as Eq. (51) shows.

## VI. CONCLUSION

The theory described in this paper is based on a consistent multipole treatment, to the order of electric quadrupoles and magnetic dipoles, of both the covariant **D** and **H** fields for an anisotropic, magnetic medium, and also of the discontinuities in these fields at a vacuum-medium interface. Several interesting results emerge from this treatment.

In the first instance it is shown that the bound surface densities of electric-dipole moment and current, which give rise, respectively, to the discontinuities in the **D** and **H** fields in a source-free medium, are both related to a third-rank, time-odd, polar tensor $S_{\alpha\beta\gamma}$ with full permutation symmetry in its subscripts. Because this tensor does not exist for nonmagnetic media and also vanishes due to symmetry for certain magnetic crystals, it is possible to explain why the standard Maxwell boundary conditions should apply to such media in the electric-quadrupole–magnetic-dipole approximation. Furthermore, when $S_{\alpha\beta\gamma}$ exists, it is readily shown that, even for normal incidence, its omission from the boundary conditions on the **D** and **H** fields leads to reflection matrices that do not satisfy the reciprocity condition in Eq. (35).

In Sec. IV A the theory is applied to crystals belonging to the magnetic point groups listed in I–IV in Table I. Although these crystals do not exhibit birefringence for propagation along the optic axis, those in groups II and IV nevertheless display nonreciprocal effects in reflection. These are a rotation $\Delta\phi$ of an incident linearly polarized beam and circular dichroism.[7,8] Reflection measurements at normal incidence have been performed on two antiferromagnetic crystals belonging to group II,[8] namely, $Cr_2O_3$ (symmetry $\bar{3}m$) and $Nd_2CuO_4$ which has been assigned the magnetic point-group symmetry $4/mmm$, in the temperature range 70–245 K.[23] Whereas a rotation $\Delta\phi \sim 10^{-4}$ was measured in $Cr_2O_3$,[8] as confirmed by our present and earlier theories,[7] null results were obtained for $Nd_2CuO_4$, even though the sensitivity of the apparatus was $\sim 10^{-6}$.[8] In view of this uncharacteristic behavior of a crystal with symmetry $4/mmm$, the question arises whether the appropriate magnetic point group is not perhaps $4/mmm$, or even $4/mmm$, for both of which the theory in this paper predicts a null effect in reflection at normal incidence, as shown in Sec. IV. Inasmuch as the properties of different materials provide a basis for determining their relevant point-group symmetries, birefringence measurements would distinguish between the three groups $4/mmm$, $4/mmm$, and $4/mmm$. For propagation along the optic axis, it follows from Eqs. (31) and (37) that birefringence would exist only for a crystal with magnetic point group symmetry $4/mmm$. The remaining two point groups may be distinguished by means of a path reversal since, as explained in Sec. IV A, directional birefringence may exist for $4/mmm$ but not for $4/mmm$.

The application of the theory in Sec. IV B to crystals belonging to the point-group symmetries listed in V and VI in Table I reveals the somewhat surprising result that nonreciprocal effects are absent in reflection at normal incidence from a face perpendicular to the optic axis, despite the existence in these crystals of nonreciprocal linear birefringence along their optic axis.[10–12]

It follows from an earlier theory,[3] in which we consider reflection from nonmagnetic uniaxial crystals, that contributions to the off-diagonal elements of the reflection matrix, due to the time-even tensors $G'_{\alpha\beta}$ and $a_{\alpha\beta\gamma}$, would exist only for those magnetic subgroups associated with the noncentrosymmetric classes $\bar{4}$ and $\bar{4}2m$. Thus reflection matrices for the remaining noncentrosymmetric magnetic point groups, listed in I–VI in Table I, have the same general forms as those derived in Secs. IV A and IV B for the corresponding centrosymmetric classes.

In Sec. V we consider cubic antiferromagnets belonging to the magnetic point groups $m3m$, $m3$, $\bar{4}3m$, $432$, and 23. Whereas these classes may exhibit nonreciprocal linear birefringence for propagation along the normal to a cube face,[12] we find from Eqs. (52)–(54) that nonreciprocal effects are once again absent in reflection. It has previously been shown[9] that even for chiral cubic crystals the matrix elements have the general forms in Eqs. (53) and (54) (i.e., $R_{xx}=R_{yy}, \quad R_{xy}=R_{yx}=0$), which are also those for an isotropic, optically inactive dielectric.[24] Thus, to the order of electric quadrupoles and magnetic dipoles, all cubic crystals, irrespective of their magnetic or chiral properties, exhibit the same response in reflection at normal incidence from a cube face as an achiral, nonmagnetic, cubic crystal. To distinguish between the different crystal properties it is therefore necessary to make use of transmission measurements or to work at oblique incidence in reflection.


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