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The optical properties of antiferromagnetic chromium oxide in transmission

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Abstract. A multipole theory of wave propagation in a magnetic medium is applied to a single-domain crystal of Cr_2O_3 in the electric quadrupole–magnetic dipole approximation. It is shown that the optical response of the crystal may be regarded as the net effect of three independent linear birefringences and their respective dichroisms, of which two are non-reciprocal and change sign when either the light path or spins are reversed; the third is reciprocal and insensitive to these influences. The six properties are contained in the Jones \mathbf{M} -matrix, which describes the effect of an arbitrary length of the crystal on an incident polarized light beam. This matrix is used to express transmitted intensities, for different incident and analysed polarization states, in terms of crystal properties. An experiment is suggested for determining the six optical properties of Cr_2O_3 from suitable measurements of the transmitted intensities.

1. Introduction

In 1963 Brown, Shtrikman and Treves [1] noted that chromium oxide (Cr_2O_3) should exhibit a spontaneous optical effect which they termed gyrotropic or non-reciprocal birefringence. The theory of this effect in Cr_2O_3 was subsequently given by Hornreich and Shtrikman [2], who were the first to offer an explanation which included the contribution of electric quadrupoles. However, a recent multipole theory of optical effects in antiferromagnetics [3], in which allowance was made for the consistent inclusion of all contributions to the order of electric quadrupoles and magnetic dipoles, has shown that certain electric quadrupole terms were omitted in the earlier theory.

The first values of components of the magnetoelectric tensor at an optical frequency have recently been determined by Krichevtsov and co-workers from reflection measurements [4]. They quote results of a similar magnitude to the corresponding low-frequency data, namely $\sim 10^{-4}$ (cgs), which also agree with an estimation they made [4], assuming electronic transitions in the optical region. Their values are several orders of magnitude higher than those predicted by Hornreich and Shtrikman [2], but support a later calculation [3], based on the quantum-mechanical expression for the magnetoelectric tensor, which showed that the optical and low-frequency values should be comparable in the near-infrared and visible range. The first attempt to measure gyrotropic birefringence in Cr_2O_3 has recently been made at an optical frequency by Pisarev and co-workers [5]. Although they were unable to detect a difference in the birefringence for the two types of antiferromagnetic domain, the respective angular shifts of the optical indicatrix were of opposite sign and much larger than originally anticipated [2], namely $\Delta\theta \simeq 1.2 \times 10^{-3}$ at 1156 nm.

In this paper we show that the gyrotropic birefringence in Cr_2O_3 may be regarded as the net effect of three independent linear birefringences and their respective dichroisms, and

we describe a method, based on the measurement of intensity differentials, for determining these six properties. In order to express transmitted intensities in terms of crystal properties use is made of the Jones \mathbf{M} -matrix [6] which, through its four complex elements, is related to the independent birefringences and dichroisms that a general dielectric medium may possess. This matrix describes the effect of an arbitrary length of the dielectric on an incident polarized light beam and enables the transverse components of the electric field of the emergent beam to be expressed in terms of the matrix elements. If each of the six basic polarization states identified by Stokes [7] is used in turn for the incident beam, and the emergent beam is analysed for the same six states, a total of 36 intensity measurements can be made. A table has been produced [8] which lists the various combinations of matrix elements for the 36 transmitted intensities. With the aid of this table and the \mathbf{M} -matrix for propagation along a twofold rotation axis in Cr_2O_3 , we have identified a suitable set of intensity measurements for determining the three independent linear birefringences and their respective dichroisms exhibited by this uniaxial magnetic crystal.

2. Wave propagation in Cr_2O_3

A multipole theory of wave propagation in a non-absorbing magnetic medium has previously been used to describe the gyrotropic birefringence which occurs in a single-domain crystal of Cr_2O_3 when the light path is along a twofold rotation axis [3]. If the crystal is oriented with respect to a laboratory system of Cartesian axes $O(x, y, z)$ such that $C_2 \parallel x$ and $C_3 \parallel z$, it follows from the theory that the eigenvectors propagating along the x axis are described by the matrix equation [3]

$$\begin{bmatrix} -\tilde{n}^2 + \varepsilon_0^{-1}\tilde{\varepsilon}_x - \tilde{n}\tilde{a} & -\tilde{n}\tilde{u} \\ -\tilde{n}\tilde{u} & -\tilde{n}^2 + \varepsilon_0^{-1}\tilde{\varepsilon}_z \end{bmatrix} \begin{bmatrix} E_{0y} \\ E_{0z} \end{bmatrix} = 0 \quad (1)$$

where

$$\tilde{\varepsilon}_i = \varepsilon_0 + \tilde{\alpha}_{ii} \quad (2)$$

$$\tilde{a} = \omega \tilde{a}'_{xxx} / \varepsilon_0 c \quad (3)$$

$$\tilde{u} = (\tilde{G}_{xx} - \tilde{G}_{zz} + \frac{1}{2}\omega \tilde{a}'_{xyz}) / \varepsilon_0 c. \quad (4)$$

The polarizability tensors $\tilde{\alpha}_{\alpha\beta}$, $\tilde{a}'_{\alpha\beta\gamma}$ and $\tilde{G}_{\alpha\beta}$ in (2)–(4) are complex (denoted by a tilde) to allow for absorption and are related to the polarization densities induced in the medium by the light wave fields as follows:

$$P_\alpha = \tilde{\alpha}_{\alpha\beta} E_\beta + (1/2\omega) \tilde{a}'_{\alpha\beta\gamma} \nabla_\gamma \dot{E}_\beta + \dots + \tilde{G}_{\alpha\beta} B_\beta + \dots \quad (5)$$

$$Q_{\alpha\beta} = -(1/\omega) \tilde{a}'_{\gamma\alpha\beta} \dot{E}_\gamma + \dots \quad (6)$$

$$M_\alpha = \tilde{G}_{\beta\alpha} E_\beta + \dots \quad (7)$$

where P_α , $Q_{\alpha\beta}$ and M_α are respectively the electric dipole, electric quadrupole and magnetic dipole moments per unit macroscopic volume. The relative multipole orders of the polarizability tensors $\tilde{\alpha}_{\alpha\beta}$, $\tilde{a}'_{\alpha\beta\gamma}$ and $\tilde{G}_{\alpha\beta}$ are, respectively, electric dipole, electric quadrupole

and magnetic dipole. Theories of optical activity [9, 10] have shown the necessity for the consistent inclusion of electric quadrupoles and magnetic dipoles as in (5)–(7).

In this paper our starting point is the 2 × 2 determinantal equation which follows from (1), namely

$$\begin{vmatrix} -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_x - \tilde{n}\tilde{a} & -\tilde{n}\tilde{u} \\ -\tilde{n}\tilde{u} & -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_z \end{vmatrix} = 0. \tag{8}$$

However, instead of solving (8) directly for the two refractive indices that are associated with gyrotropic birefringence, as was done in the earlier theory [3], we apply a different procedure [11]. In this the determinant is replaced by a sum of determinants, each representing an independent birefringence and dichroism that Jones [6] identified in his optical calculus.

Equation (8) may be rewritten exactly as

$$\begin{vmatrix} -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_x & 0 \\ 0 & -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_z \end{vmatrix} + \begin{vmatrix} \tilde{n}^2 - \tilde{n}\tilde{a} - \epsilon_0^{-1}\tilde{\epsilon}_z & 0 \\ 0 & -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_z \end{vmatrix} + \begin{vmatrix} -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_z & -\tilde{n}\tilde{u} \\ -\tilde{n}\tilde{u} & -\tilde{n}^2 + \epsilon_0^{-1}\tilde{\epsilon}_z \end{vmatrix} = 0. \tag{9}$$

For independent effects each determinant may be set equal to zero. From the first we find

$$\tilde{n}_y = n_y + ik_y = (\epsilon_0^{-1}\tilde{\epsilon}_x)^{1/2} \tag{10}$$

and a similar form for \tilde{n}_z . Thus the determinant is associated with the well known linear birefringence for propagation at right-angles to the optic axis in a uniaxial crystal like Cr₂O₃

$$n_y - n_z = \text{Re}\{\tilde{\epsilon}_x^{1/2} - \tilde{\epsilon}_z^{1/2}\}/\epsilon_0^{1/2} \tag{11}$$

and its associated dichroism

$$k_y - k_z = \text{Im}\{\tilde{\epsilon}_x^{1/2} - \tilde{\epsilon}_z^{1/2}\}/\epsilon_0^{1/2} \tag{12}$$

for which the fast and slow axes are along the crystallographic y and z axes. It follows from (2) and (5) that the properties in (11) and (12) are electric dipole effects and are time-even.

As the second determinant in (9) yields, to first order in \tilde{a} ,

$$\tilde{n}_y - \tilde{n}_z = \frac{1}{2}\tilde{a} = \omega\tilde{a}'_{xxx}/2\epsilon_0c \tag{13}$$

it also describes linear birefringence and linear dichroism relative to the crystallographic y and z axes. However, the two properties are independent of those in (11) and (12) because they are of electric quadrupole origin and are time-odd, as (6) shows.

The remaining determinant in (9) has exact solutions

$$\tilde{n} = \frac{1}{2}[\mp\tilde{u} + (\tilde{u}^2 + 4\epsilon_0^{-1}\tilde{\epsilon}_z)^{1/2}]. \tag{14}$$

When these are substituted into the equations implicit in the determinant we find

$$E_{0y}/E_{0z} = \pm 1 \tag{15}$$

corresponding to the \mp signs in (14). Thus the two roots in (14) are associated with eigenvectors that are linearly polarized at $+45^\circ$ and -45° to the crystallographic y axis in the yz plane, and the difference in the complex refractive indices for propagation along the x axis is

$$\bar{n}_{-45} - \bar{n}_{45} = \bar{u} = (\tilde{G}_{xx} - \tilde{G}_{zz} + \frac{1}{2}\omega\tilde{a}'_{xyz})/\varepsilon_0 c. \quad (16)$$

The determinant therefore describes a third independent linear birefringence and linear dichroism, namely Jones birefringence and Jones dichroism [6, 12], which, as (6) and (7) show, are due to time-odd tensors of electric quadrupole and magnetic dipole order. (Jones birefringence was first predicted by Jones in the formulation of the optical calculus that bears his name [6]. It is a linear birefringence which may, in certain systems, co-exist with the normal linear birefringence, but its fast and slow axes bisect those of the normal birefringence. Jones gave no indication of the systems which would exhibit this birefringence and measurement of it has yet to be reported. However, a systematic analysis has now identified the directions of propagation in crystals of the 32 non-magnetic symmetry point groups, in which it would exist [11].)

A reversal of the light path or of the spins in a single-domain crystal of Cr_2O_3 changes the sign of the non-reciprocal properties in (13) and (16). As shown previously [3], the combination of tensors in (16), which includes an electric quadrupole contribution \tilde{a}'_{xyz} , is proportional to the angular shift in the principal axes.

The combined effect of the various birefringences and dichroisms in (11)–(13) and (16) on an incident polarized light beam, propagating along a twofold rotation axis in a Cr_2O_3 crystal of length l , is described by the Jones \mathbf{M} -matrix [6]

$$\mathbf{M} = \exp(i\omega\bar{n}l/c) \begin{bmatrix} \cosh(Ql) - i\tilde{g}_0 Q^{-1} \sinh(Ql) & -i\tilde{g}_{45} Q^{-1} \sinh(Ql) \\ -i\tilde{g}_{45} Q^{-1} \sinh(Ql) & \cosh(Ql) + i\tilde{g}_0 Q^{-1} \sinh(Ql) \end{bmatrix} \quad (17)$$

where \bar{n} is the average complex refractive index, and [6]

$$\tilde{g}_0 = g_0 + ip_0 = \frac{1}{2}\omega c^{-1}(\bar{n}_y - \bar{n}_z) = \frac{1}{2}\omega\mu_0[ce_0^{1/2}(\bar{\varepsilon}_x^{1/2} - \bar{\varepsilon}_z^{1/2}) + \frac{1}{2}\omega\tilde{a}'_{xxx}] \quad (18)$$

$$\tilde{g}_{45} = g_{45} + ip_{45} = \frac{1}{2}\omega c^{-1}(\bar{n}_{-45} - \bar{n}_{45}) = \frac{1}{2}\omega\mu_0(\tilde{G}_{xx} - \tilde{G}_{zz} + \frac{1}{2}\omega\tilde{a}'_{xyz}) \quad (19)$$

$$Q^2 = -(\tilde{g}_0^2 + \tilde{g}_{45}^2). \quad (20)$$

Then, if \mathcal{E} is the Jones vector of the incident light beam, the corresponding quantity \mathcal{E}' for the emergent beam is given by

$$\mathcal{E}' = \mathbf{M}\mathcal{E}. \quad (21)$$

Thus the polarization of the emergent beam is a function of all the independent properties of the crystal. Clearly these cannot be separated by a single simple operation such as a path reversal. The measurement of intensity differentials, however, provides a possible solution to the problem [8]. In the following section we consider how these may be applied to determine the optical properties of Cr_2O_3 .

3. Experimental considerations

In the proposed experiment a magnetoelectrically annealed crystal of Cr_2O_3 , with plane and parallel faces perpendicular to the twofold rotation axis (the crystallographic x axis in our theory), is arranged such that its optic axis (z axis) is in the horizontal plane. A polarized light beam of intensity I_0 propagating in a vacuum is incident normally on one of the plane faces. The beam that emerges from the crystal is then passed through an analyser and the transmitted intensity is measured.

The polarization of the incident beam is taken to be in one of the six basic states identified by Stokes [7]. They are, as judged by an observer into whose eye the light is travelling, linearly polarized horizontally \leftrightarrow and vertically \updownarrow , linearly polarized at $+45^\circ$ \nearrow and -45° \nwarrow to the horizontal, and right \odot and left \ominus circularly polarized. The same six states are used for the analyser, thus yielding a total of 36 analysed intensities. Expressions for these intensities in terms of the elements of the Jones \mathbf{M} -matrix have previously been tabulated [8]. To distinguish the various analysed intensities a notation is used in which the symbols for the incident and analysed polarization states are displayed. For example, $\nearrow I \odot$ represents the intensity transmitted by a right circular analyser when the incident beam is linearly polarized at $+45^\circ$ to the horizontal. A difference between two analysed intensities when either the incident or analysed polarization states are in common, such as $\nearrow I \odot - \nwarrow I \ominus$, is termed an intensity differential.

The independent properties to be measured in Cr_2O_3 are g_0 and p_0 which, as (18) shows, are proportional to the linear birefringence and the linear dichroism relative to the crystallographic y and z axes, and g_{45} and p_{45} which are respectively proportional to the Jones birefringence and Jones dichroism described by (19). It follows from the table by Raab [8] and the form of the \mathbf{M} -matrix in (17) that the four properties can be determined from just four intensity differentials. Each intensity differential is measured using an incident polarized beam of the same intensity I_0 , and is normalized by dividing by the transmitted intensity I when an unpolarized beam, also of intensity I_0 , is incident on the sample [8]. The four normalized intensity differentials shown below have been selected on the basis of experimental considerations, as they allow in each case for switching between orthogonal linear polarizations of the incident beam for a fixed setting of the analyser:

$$I_1 = \nearrow I \odot - \nwarrow I \ominus \quad (22)$$

$$I_2 = \nearrow I \ominus - \nwarrow I \odot \quad (23)$$

$$I_3 = \leftrightarrow I \ominus - \updownarrow I \odot \quad (24)$$

$$I_4 = \leftrightarrow I \odot - \updownarrow I \ominus \quad (25)$$

Combinations of the intensity differentials in (22)–(25) are related to the properties of the medium as follows:

$$d_1 = I_1 + I_2 = K[pg_{45} + gp_{45} \sin(2gl) - (gg_{45} - pp_{45}) \sinh(2pl)] \quad (26)$$

$$d_2 = I_3 - I_4 = K[(gg_{45} - pp_{45}) \sin(2gl) + (pg_{45} + gp_{45}) \sinh(2pl)] \quad (27)$$

$$d_3 = I_3 + I_4 = K[(pg_0 + gp_0) \sin(2gl) - (gg_0 - pp_0) \sinh(2pl)] \quad (28)$$

$$d_4 = I_2 - I_1 = K[(gg_0 - pp_0) \sin(2gl) + (pg_0 + gp_0) \sinh(2pl)] \quad (29)$$

where $K = 2(QQ^*)^{-1}$ and $Q = p + ig$.

From suitable combinations of (26)–(29), namely

$$\frac{1}{4}(d_2^2 - d_1^2 + d_4^2 - d_3^2) = -(I_1 I_2 + I_3 I_4) = \sin^2(2gl) - \sinh^2(2pl) \quad (30)$$

and

$$\frac{1}{2}(d_1 d_2 + d_3 d_4) = (I_2 I_3 - I_1 I_4) = -2 \sin(2gl) \sinh(2pl) \quad (31)$$

it can be shown that

$$g = \sin^{-1} G/2l \quad G = (F - I_1 I_2 - I_3 I_4)^{1/2}/\sqrt{2} \quad (32)$$

$$p = \sin^{-1} P/2l \quad P = (F + I_1 I_2 + I_3 I_4)^{1/2}/\sqrt{2} \quad (33)$$

where

$$F = [(I_1^2 + I_3^2)(I_2^2 + I_4^2)]^{1/2}. \quad (34)$$

Then (26)–(29) can be solved to yield the following expressions for the four properties in a length l of the crystal

$$g_{45} = [d_1(pG - gP) + d_2(gG + pP)]/2F \quad (35)$$

$$p_{45} = [d_1(gG + pP) - d_2(pG - gP)]/2F \quad (36)$$

$$g_0 = [d_3(pG - gP) + d_4(gG + pP)]/2F \quad (37)$$

$$p_0 = [d_3(gG + pP) - d_4(pG - gP)]/2F. \quad (38)$$

However, it follows from (11)–(13) and (18) that g_0 and p_0 represent respectively a composite linear birefringence and linear dichroism composed of two parts, a reciprocal part due to electric dipoles and a non-reciprocal part due to electric quadrupoles. It is possible to separate the two independent contributions to this birefringence and dichroism, which are due to different multipole orders, by repeating the measurements in (22)–(25) with the light path reversed. Reciprocity considerations [13] have shown the necessity for performing a second set of measurements of differing geometry, both in reflection and transmission, in order to isolate a time-odd quantity. Because the arcsine function is many-valued, and g and p in (32) and (33) respectively enter (35)–(38), it may also be necessary to repeat the measurements on a sample of different thickness to determine g_0 , p_0 , g_{45} and p_{45} unambiguously.

In the absence of absorption $p = p_0 = p_{45} = 0$ and, as (26) and (28) show that $I_1 = -I_2$ and $I_3 = -I_4$, only two intensity differentials need be measured to determine g_0 and g_{45} .

4. Conclusion

The procedure described in section 3 provides a new method for determining the independent birefringences and dichroisms in a magnetoelectrically annealed crystal of Cr_2O_3 . It follows from the theory that, for propagation along a twofold rotation axis in this crystal, the four intensity measurements in (22)–(25) are sufficient for evaluating the linear birefringence

g_0 and its respective dichroism p_0 , which are relative to orthogonal crystallographic axes at right angles to the light path, and also the Jones birefringence g_{45} and Jones dichroism p_{45} along the bisectors of these axes. The time-odd contributions to g_0 and p_0 , which are of electric quadrupole order, account for the difference in these properties for the two types of antiferromagnetic domain that are exhibited by Cr_2O_3 [5]. Pisarev and co-workers [5], however, have been unable to detect a difference in the birefringence for the two types of domain in the near-infrared. Equation (5) shows that the tensor \tilde{a}'_{xxx} , which is associated with the non-reciprocal contributions to g_0 and p_0 , describes the electric dipole moment induced in the medium by the electric field-gradient of the light wave. This implies that the non-reciprocal behaviour of g_0 and p_0 is more likely to be observed at shorter wavelengths in the visible region of the spectrum. The frequency factor ω in (18) confirms this interpretation. Repeating the intensity measurements in (22)–(25) with the light path reversed in principle allows the time-odd contributions to g_0 and p_0 to be separated from the time-even electric dipole contributions.

Inspection of (16) shows that the magnitude of the electric quadrupole contribution relative to that of one of the magnetic dipole terms is given by the ratio

$$R = \frac{1}{2}\omega\tilde{a}'_{xyz}/\tilde{G}_{xx}. \quad (39)$$

From the quantum mechanical expressions for the \tilde{G} and \tilde{a}' tensors [3] we then find

$$R \simeq \frac{\omega^2}{2\omega_{jn}} \frac{\langle j|Q_{yz}|n\rangle}{\langle j|M_x|n\rangle}. \quad (40)$$

Taking the ratio of matrix elements of the order

$$\frac{\langle j|Q_{yz}|n\rangle}{\langle j|M_x|n\rangle} \simeq \frac{\text{electronic charge} \times (\text{cell dimension})^2}{\text{Bohr magneton}}$$

and assuming an electronic transition wavelength of 500 nm for incident radiation of 600 nm, we find from (40) that

$$R \simeq 8.$$

Thus the electric quadrupole contribution is of a similar order of magnitude to that of the magnetic dipole.

A suitable method for determining the various transmitted intensities in (22)–(25) would be to modulate the polarization of the incident beam between two orthogonal linear states, and to use a phase-sensitive detector to measure the corresponding intensity differential after passage of the beam through the sample and the selected circular analyser. A practical aspect which may need consideration in the design of a suitable measuring system is the magnitude of the transmitted intensities. As the intensities are measured for non-orthogonal polarization settings of the polarizer and analyser, they would be quite large, particularly when the crystal is transparent. However, this may not be a problem in an absorbing medium such as Cr_2O_3 as the light intensity could be controlled by a suitable choice for the sample length.

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