On the existence of linear non-reciprocal bi-isotropic (NRBI) media

1997 J. Phys. A: Math. Gen. 30 1335

(http://iopscience.iop.org/0305-4470/30/4/032)

J. Phys. A: Math. Gen. **30** (1997) 1335–1344. Printed in the UK PII: S0305-4470(97)69692-4

On the existence of linear non-reciprocal bi-isotropic (NRBI) media

R E Raab† and A H Sihvola‡

† Physics Department, University of Natal, P/Bag X01, Scottsville, South Africa 3209
‡ Electromagnetics Laboratory, Helsinki University of Technology, Otakaari 5A, FIN-02150, Espoo, Finland

Received 17 November 1995, in final form 6 September 1996

Abstract. A recent letter in this journal and a number of related publications have purported to prove that Maxwell's equations forbid the existence of linear non-reciprocal bi-isotropic (NRBI) media. The basis of such proofs is shown to be unfounded, with the constraint on matter being due not to electromagnetism but to symmetry. It is also shown that linear NRBI media may exist in some situations in the form of certain magnetic cubic crystals. In addition, in their response to an electromagnetic wave there are also magnetic cubics in which propagation, although not perfectly isotropic, deviates in practice to an extent which is probably undetectable. Finally, the existence of a Tellegen medium, as an example of a NRBI substance, is shown to be possible in principle, in contrast to the findings of recent papers.

1. Introduction

In a recent publication [1] Lakhtakia and Weiglhofer presented a proof, based on a covariance requirement deduced by Post [2], that a bi-isotropic medium must be reciprocal. Their proof limited consideration to a linear and homogeneous substance. That non-reciprocal bi-isotropic (NRBI) substances might occur has been tacitly assumed in the past [3], without, however, any examples being identified. By contrast, the existence of non-reciprocal bi-anisotropic media is well accepted [4, 5], of which antiferromagnetic chromium oxide is a familiar example.

Following their paper in [1] Lakhtakia and Weiglhofer published, either jointly or singly, numerous other papers [6, 7, 8, 9, 10, 11, 12, 13, 14] in which the implications of the Post covariance requirement were investigated more fully, particularly with regard to electromagnetic constraints on the existence of certain materials. The claim is made in a number of these papers that the non-existence of NRBI media is a consequence of electromagnetic theory [6, 7, 8, 11]. This claim and the possible existence of NRBI substances are investigated in the present paper, using a different approach from that in [1]. Because of this, certain terms and notation require definition or explanation.

We begin by stating that the fundamental macroscopic fields in matter are taken in this paper to be the electric field $\boldsymbol{E}$ and the magnetic field $\boldsymbol{B}$ (rather than $\boldsymbol{H}$ which, like $\boldsymbol{D}$, is regarded as a response field). A variety of reasons for this choice appears in the literature [15, 16, 17, 18].

The traditional definition of a bi-isotropic substance is one for which the constitutive

0305-4470/97/041335+10$19.50

relations have the form [1, 3, 4]

$$\begin{aligned} \boldsymbol{D} &= \epsilon \boldsymbol{E} + \alpha \boldsymbol{B} \\ \boldsymbol{H} &= \beta \boldsymbol{E} + k \boldsymbol{B} \end{aligned} \tag{1}$$

where $\boldsymbol{E}$ and $\boldsymbol{B}$ are frequency-dependent fields and ϵ, α, β, k are macroscopic scalar properties of the substance, which, for a linear response, are independent of the fields.

The term 'non-reciprocal' has been used for a material whose measured response to a source changes when the source and observer are interchanged [2, 3, 4]. This meaning is closely related to the time-antisymmetric behaviour of the material, and, in this sense, accords with the use in the literature of 'non-reciprocal' to describe an effect which changes sign under time reversal [2, 19]. When it comes to describing the behaviour under time reversal of the medium itself or of its properties, the terms 'time-even' and 'time-odd' will be used in this paper in preference to 'reciprocal' and 'non-reciprocal'. This choice conforms to established usage [20]. Time-odd substances are also termed 'magnetic' and time-even ones 'non-magnetic'.

A further difference in approach is that we are concerned primarily with the electric and magnetic multipole moments per unit macroscopic volume (rather than with the response fields $\boldsymbol{D}$ and $\boldsymbol{H}$) that are induced by the fields $\boldsymbol{E}$ and $\boldsymbol{B}$ and, in certain situations, also by their space and time derivatives. The expressions for such multipole moment densities serve to define valid property tensors of the medium [21], to which symmetry considerations may be applied in order to determine the conditions for their existence and also the nature and number of their components. As an example, the magnetoelectric effect may be defined in terms of the electric dipole moment density (the polarization $\boldsymbol{P}$) induced by a uniform magnetic field $\boldsymbol{B}$ and by the magnetic dipole moment density (the magnetization $\boldsymbol{M}$) induced by a uniform electric field $\boldsymbol{E}$ [16, 22]. Once the moment densities are precisely specified, the constitutive relations for $\boldsymbol{D}$ and $\boldsymbol{H}$ may be obtained when required.

To determine the extent of the constraints, if any, imposed on the structure of matter by electromagnetic theory, the material tensors entering the constitutive relations for a non-absorbing linear and homogeneous bi-anisotropic medium in the field of a plane time-harmonic electromagnetic wave are analysed with regard to their time-inversion behaviour. This analysis is presented in section 2, where it is shown that an isotropic medium is necessarily reciprocal as a result of its symmetry and not of any constraint arising from electromagnetic theory.

When they exist, the second-rank property tensors of magnetic cubic crystals are isotropic in form [21]. The implication of this for possible bi-isotropic constitutive relations is investigated in section 3 for uniform electric and magnetic fields and in section 4 for the fields of a plane time-harmonic wave.

A Tellegen medium [23] is a postulated isotropic substance comprising macroscopically small magnetoelectric particles, and would therefore exhibit a NRBI response to uniform electric and magnetic fields. Despite recent arguments against the existence of such a medium [6, 10, 12], the matter is considered again using a different approach. This is presented in section 5.

2. Effect of symmetry on constitutive relations

The proof in [1] that a linear bi-isotropic medium must be reciprocal begins with the constitutive relations describing the response of a linear and homogeneous bi-anisotropic medium to a plane time-harmonic wave, namely

$$\begin{aligned} D_\alpha &= \epsilon_{\alpha\beta} E_\beta + \alpha_{\alpha\beta} B_\beta \\ H_\alpha &= \beta_{\alpha\beta} E_\beta + k_{\alpha\beta} B_\beta \, . \end{aligned} \tag{2}$$

We consider such a wave propagating through the medium with an angular frequency ω, which is far from any absorption band. The wave fields $\boldsymbol{E}$ and $\boldsymbol{B}$ that enter (2) are complex. We take $\boldsymbol{E}$ in the form

$$\boldsymbol{E} = \boldsymbol{E}_0 \exp\{-\mathrm{i}\omega(t - n\boldsymbol{r} \cdot \boldsymbol{\sigma}/c)\} \tag{3}$$

where c is the speed of light in a vacuum, $\boldsymbol{\sigma}$ the unit vector perpendicular to the plane wavefront, and n the refractive index of the medium for the propagation direction $\boldsymbol{\sigma}$ and the polarization state described by the amplitude $\boldsymbol{E}_0$, which may be complex.

For complex fields the four material parameters in (2) are in general complex. As the time-even and time-odd parts of these parameters are separately required for symmetry considerations, each is written in the form

$$U_{\alpha\beta} = U^r_{\alpha\beta} - \mathrm{i}U^i_{\alpha\beta} \tag{4}$$

where $U^r_{\alpha\beta}$ and $U^i_{\alpha\beta}$ are real in the absence of absorption. Then equation (2) is readily shown to be, for the form of the field in (3),

$$\begin{aligned} D_\alpha &= \epsilon^r_{\alpha\beta} E_\beta + \omega^{-1}\epsilon^i_{\alpha\beta} \dot{E}_\beta + \alpha^r_{\alpha\beta} B_\beta + \omega^{-1}\alpha^i_{\alpha\beta} \dot{B}_\beta \\ H_\alpha &= \beta^r_{\alpha\beta} E_\beta + \omega^{-1}\beta^i_{\alpha\beta} \dot{E}_\beta + k^r_{\alpha\beta} B_\beta + \omega^{-1}k^i_{\alpha\beta} \dot{B}_\beta \end{aligned} \tag{5}$$

in which $\dot{\boldsymbol{E}} = \partial \boldsymbol{E}/\partial t$ and $\dot{\boldsymbol{B}} = \partial \boldsymbol{B}/\partial t$. Although the complex form in (3) was used to obtain (5), the fields $\boldsymbol{E}$, $\dot{\boldsymbol{E}}$, $\boldsymbol{B}$ and $\dot{\boldsymbol{B}}$ in it may nevertheless be understood to be real (see pp 76, 77 of [20]). It has been shown for negligible absorption [20, 24, 25] that

$$\alpha^r_{\alpha\beta} = -\beta^r_{\beta\alpha} \qquad \alpha^i_{\alpha\beta} = \beta^i_{\beta\alpha} \, . \tag{6}$$

We now determine the time-inversion behaviour of the eight property tensors in (5). To do this we note that for Maxwell's equations to be invariant under time inversion, $\boldsymbol{D}$ must be time-even and $\boldsymbol{H}$ time-odd. From their definitions $\boldsymbol{E}$ is time-even, while $\boldsymbol{B}$ is time-odd. By inspection of (5), with the understanding that all quantities are real, the following classification is obtained:

$$\begin{aligned} &\text{Time-even:} \qquad \epsilon^r_{\alpha\beta}, \quad \alpha^i_{\alpha\beta}, \quad \beta^i_{\alpha\beta}, \quad k^r_{\alpha\beta} \\ &\text{Time-odd:} \qquad \epsilon^i_{\alpha\beta}, \quad \alpha^r_{\alpha\beta}, \quad \beta^r_{\alpha\beta}, \quad k^i_{\alpha\beta} \, . \end{aligned} \tag{7}$$

By Neumann's principle [21] a magnetic medium may possess both time-even and time-odd property tensors, while only the former may belong to a non-magnetic substance. Thus equation (5) describes a linear and homogeneous bi-anisotropic magnetic medium, and to this point no limitation has been placed on the structure of the medium.

We now introduce such a constraint by assuming that the medium has isotropic symmetry. Only such a medium has all its property tensors isotropic, including any time-odd ones should they exist. However, Van Vleck [26] and Buckingham *et al* [27] have shown that an isotropic medium may not possess any time-odd macroscopic property tensors, even when its constituent molecules are time-odd, by having, for example, a magnetic dipole. The proof of this statement rests on the effect of time reversal at the microscopic level (see section 5) and is quite independent of electromagnetic theory. Thus it is isotropic symmetry and not Maxwell's equations that limit bi-isotropic media to being time-even or reciprocal.

The arguments in [26] and [27] apply only if the medium is isotropic. Could there be media which have bi-isotropic constitutive relations but which are not isotropic? Cubic crystals suggest themselves, since all their second-rank property tensors are isotropic, while in general their higher-rank ones are not [21]. For them the proof by Lakhtakia and Weiglhofer based on Post's covariance requirement, might then, if correct, be applicable. However, magnetic cubic crystals exist and are examples of non-reciprocal or time-odd systems [21].

For an isotropic medium only the time-even tensors in (7) survive and are now isotropic. Then from (5) and (6)

$$\begin{aligned} \boldsymbol{D} &= \epsilon^r \boldsymbol{E} - \mathrm{i}\alpha^i \boldsymbol{B} = \epsilon^r \boldsymbol{E} + \omega^{-1}\alpha^i \dot{\boldsymbol{B}} \\ \boldsymbol{H} &= -\mathrm{i}\alpha^i \boldsymbol{E} + k^r \boldsymbol{B} = \omega^{-1}\alpha^i \dot{\boldsymbol{E}} + k^r \boldsymbol{B}\,. \end{aligned} \tag{8}$$

These equations now have a time-even (reciprocal) bi-isotropic form, which is characteristic of a chiral fluid.

In a recent paper [6] entitled 'On a medium constraint arising directly from Maxwell's equations', Weiglhofer uses constitutive relations in the bi-isotropic form

$$\begin{aligned} \boldsymbol{D} &= \epsilon \boldsymbol{E} + (\alpha + \beta)\boldsymbol{B} \\ \boldsymbol{H} &= (-\alpha + \beta)\boldsymbol{E} + k\boldsymbol{B} \end{aligned} \tag{9}$$

and shows that α, termed the 'non-reciprocity parameter', does not enter Maxwell's equations when (9) are substituted for $\boldsymbol{D}$ and $\boldsymbol{H}$. The author then argues that α may be set equal to zero for a bi-isotropic medium; that is, Maxwell's equations constrain such a medium to be reciprocal. However, Weiglhofer's α is the isotropic form of the dynamic magnetoelectric tensor $\alpha^r_{\alpha\beta} = -\beta^r_{\beta\alpha}$ in (5) and (6), and, as shown above, this time-odd tensor vanishes for an isotropic system. The constraint on such a medium arises from symmetry, not from Maxwell's equations.

3. A magnetic medium in uniform $\boldsymbol{E}$ and $\boldsymbol{B}$ fields

Uniform fields are constant in time and thus independent. They induce in a linear and homogeneous medium a polarization and magnetization according [16] to

$$\begin{aligned} P_\alpha &= \epsilon_0 \chi^{(\mathrm{e})}_{\alpha\beta} E_\beta + G_{\alpha\beta} B_\beta \\ M_\alpha &= \mu_0^{-1} \chi^{(\mathrm{m})}_{\alpha\beta} B_\beta + \mathcal{G}_{\alpha\beta} E_\beta \end{aligned} \tag{10}$$

where the quantities $\chi^{(\mathrm{e})}_{\alpha\beta}$, $G_{\alpha\beta}$, $\chi^{(\mathrm{m})}_{\alpha\beta}$, $\mathcal{G}_{\alpha\beta}$ are valid property tensors of the medium [21]. To determine their behaviour under space and time inversion, we begin by noting that $\boldsymbol{P}$ is a polar time-even vector, while $\boldsymbol{M}$ is an axial time-odd one. This follows from their definitions:

$$\boldsymbol{P} = \sum_i q_i \boldsymbol{r}_i / \Delta V \tag{11}$$

$$\boldsymbol{M} = \sum_i (q_i/2m_i)(\boldsymbol{r}_i \times \boldsymbol{p}_i + g_i \boldsymbol{s}_i)/\Delta V\,. \tag{12}$$

In these $\boldsymbol{r}_i$ is the displacement from an arbitrary origin inside the macroscopic volume element ΔV of a particle with charge q_i, mass m_i, linear momentum $\boldsymbol{p}_i$, spin $\boldsymbol{s}_i$, and g-factor g_i.

Inspection of (10) then yields the classification:

$$\begin{aligned} &\text{Polar time-even:} \qquad \chi^{(\mathrm{e})}_{\alpha\beta}, \quad \chi^{(\mathrm{m})}_{\alpha\beta} \\ &\text{Axial time-odd:} \qquad G_{\alpha\beta}, \quad \mathcal{G}_{\alpha\beta}\,. \end{aligned} \tag{13}$$

Thus equations (10) describe the induction of $\boldsymbol{P}$ and $\boldsymbol{M}$ by uniform $\boldsymbol{E}$ and $\boldsymbol{B}$ in a general anisotropic magnetic medium. The tensors $G_{\alpha\beta}$ and $\mathcal{G}_{\alpha\beta}$ have been shown to have non-vanishing components for 58 of the 90 magnetic symmetry point groups [28, 29], so that it is in these that the magnetoelectric effect is exhibited. Symmetry considerations also indicate that the electric and magnetic susceptibility tensors $\chi^{(e)}_{\alpha\beta}$ and $\chi^{(m)}_{\alpha\beta}$, respectively, exist for all magnetic and non-magnetic crystals [21].

The static magnetoelectric tensors $G_{\alpha\beta}$ and $\mathcal{G}_{\alpha\beta}$ can be simply shown by quantum-mechanical perturbation theory to be related [25] by

$$G_{\alpha\beta} = \mathcal{G}_{\beta\alpha} \tag{14}$$

although other approaches have been used to prove this [16].

We now consider the possible isotropy of the three independent property tensors in (13). Use of tables 7 and 4d in Birss [21] shows that for all 11 cubic magnetic classes $\chi^{(e)}_{\alpha\beta}$ and $\chi^{(m)}_{\alpha\beta}$ exist and are isotropic. Of these 11 classes $G_{\alpha\beta}$ exists and is isotropic for five, namely

$$23, \quad \underline{m}3, \quad 432, \quad \bar{4}3\underline{m}, \quad \underline{m}3\underline{m}\,. \tag{15}$$

Thus it is for these five magnetic point groups that the medium responds isotropically to both uniform $\boldsymbol{E}$ and $\boldsymbol{B}$ fields.

To express this conclusion in the form of constitutive relations we use

$$\begin{aligned} \boldsymbol{D} &= \epsilon_0 \boldsymbol{E} + \boldsymbol{P} \\ \boldsymbol{H} &= \mu_0^{-1} \boldsymbol{B} - \boldsymbol{M} \end{aligned} \tag{16}$$

which apply when the fields are uniform. Then from (16), (10), and (14)

$$\begin{aligned} \boldsymbol{D} &= \epsilon_0 (1 + \chi^{(e)}) \boldsymbol{E} + G\boldsymbol{B} \\ \boldsymbol{H} &= \mu_0^{-1} (1 - \chi^{(m)}) \boldsymbol{B} - G\boldsymbol{E}\,. \end{aligned} \tag{17}$$

As equation (17) is identical in form to (1), it is evident that there exist magnetic (or non-reciprocal) media whose constitutive relations for uniform electric and magnetic fields are bi-isotropic, namely the magnetic crystals belonging to the five cubic point groups in (15).

4. A plane time-harmonic wave in a magnetic crystal

Since no crystal is perfectly isotropic, it is evident that a plane time-harmonic wave will not propagate isotropically through it. Nevertheless, in view of the controversy regarding the existence of NRBI media, it is of interest to determine the extent to which wave propagation in magnetic cubic crystals, in particular, deviates from isotropic. This might allow one to identify media which, to a very good approximation, behave as NRBI for wave propagation through them.

To investigate this matter we begin by recognizing that the wave possesses not only electric and magnetic fields but also, because of its finite wavelength, field gradients of all orders. In addition, it has time-derivative fields. However, because of the harmonic condition

$$\ddot{\boldsymbol{E}} = -\omega^2 \boldsymbol{E}, \qquad \dddot{\boldsymbol{E}} = -\omega^2 \dot{\boldsymbol{E}}, \qquad \ldots$$

only two of the time-derivative fields are independent, which we take to be the zeroth and the first. Thus the wave interacts with matter [20, 24, 25] through the fields

$$E_\alpha,\ \dot{E}_\alpha; \quad \nabla_\beta E_\alpha,\ \nabla_\beta \dot{E}_\alpha; \quad \ldots; \qquad B_\alpha,\ \dot{B}_\alpha; \quad \nabla_\beta B_\alpha,\ \nabla_\beta \dot{B}_\alpha; \quad \ldots\,. \tag{18}$$

The existence of all these fields is confirmed by a quantum-mechanical treatment [30, 31].

For wavelengths very much larger than the dimensions of a macroscopic volume element, the fields in (18) may be considered to induce multipole moments in each macroscopic volume element. The relative magnitudes of the multipole contributions to a physical effect due to electromagnetic radiation [32] are

$$\text{Electric dipole} \gg \left\{ \begin{array}{l} \text{Electric quadrupole} \\ \text{Magnetic dipole} \end{array} \right. \gg \left\{ \begin{array}{l} \text{Electric octopole} \\ \text{Magnetic quadrupole} \end{array} \right. \gg \ldots\,. \tag{19}$$

Against this background we now investigate the behaviour of a magnetic crystal exposed to a plane time-harmonic wave. In the approximation where multipole contributions beyond those of electric quadrupole and magnetic dipole are omitted, the induced multipole moment densities linear in the fields of the wave [25], in a modified form of the notation of Buckingham [24] and Barron [20], are

$$P_\alpha = \epsilon_0 \chi_{\alpha\beta} E_\beta + \omega^{-1}\epsilon_0 \chi'_{\alpha\beta}\dot{E}_\beta + \tfrac{1}{2}a_{\alpha\beta\gamma}\nabla_\gamma E_\beta + \tfrac{1}{2}\omega^{-1}a'_{\alpha\beta\gamma}\nabla_\gamma \dot{E}_\beta$$
$$+ G_{\alpha\beta}B_\beta + \omega^{-1}G'_{\alpha\beta}\dot{B}_\beta + \cdots \tag{20}$$

$$Q_{\alpha\beta} = \mathrm{a}_{\alpha\beta\gamma}E_\gamma + \omega^{-1}\mathrm{a}'_{\alpha\beta\gamma}\dot{E}_\gamma + \cdots \tag{21}$$

$$M_\alpha = \mathcal{G}_{\alpha\beta}E_\beta + \omega^{-1}\mathcal{G}'_{\alpha\beta}\dot{E}_\beta + \cdots\,. \tag{22}$$

In equation (21) $Q_{\alpha\beta}$ is the electric quadrupole moment density, defined by

$$Q_{\alpha\beta} = \sum_i q_i r_{\alpha i} r_{\beta i}/\Delta V\,. \tag{23}$$

Equations (20)–(22) are consistent in their multipole order, as may be seen from the quantum-mechanical expressions for the property tensors in these equations [25], or may be suggested intuitively by the well known associations [33]

Electric field $\sim$ Electric dipole

First gradient of $\boldsymbol{E}$ $\sim$ Electric quadrupole

Magnetic field $\sim$ Magnetic dipole.

It is of interest to note that equation (22) contains no magnetic susceptibility term, as appeared in (10). This is because it is of magnetic quadrupole order [32], since its quantum-mechanical expression, as derived by Van Vleck [26], contains the product of two magnetic dipole moment matrix elements. Consequently it falls outside our multipole approximation.

The property tensors in (20)–(22) depend on frequency, satisfy the Kramers–Kronig relations [20], and in the absence of absorption are real, as confirmed by their quantum-mechanical expressions, which also show [25] that

$$\begin{array}{llll} \chi_{\alpha\beta} = \chi_{\beta\alpha} & \chi'_{\alpha\beta} = -\chi'_{\beta\alpha} & a_{\alpha\beta\gamma} = a_{\alpha\gamma\beta} & a'_{\alpha\beta\gamma} = a'_{\alpha\gamma\beta} \\ \mathrm{a}_{\alpha\beta\gamma} = a_{\gamma\alpha\beta} & \mathrm{a}'_{\alpha\beta\gamma} = -a'_{\gamma\alpha\beta} & \mathcal{G}_{\alpha\beta} = G_{\beta\alpha} & \mathcal{G}'_{\alpha\beta} = -G'_{\beta\alpha}\,. \end{array} \tag{24}$$

Thus of the ten property tensors in (20)–(22) six are independent. Their space–time classification can be readily determined by a similar procedure to that in section 2, and is presented in table 1.

To derive the wave equation we use $\boldsymbol{E}$ in (3), together with the Maxwell equations

$$\nabla\times\boldsymbol{E} = -\dot{\boldsymbol{B}} \qquad \nabla\times\boldsymbol{B} = \mu_0(\epsilon_0\dot{\boldsymbol{E}} + \boldsymbol{J} + \boldsymbol{J}_\mathrm{b}) \tag{25}$$

Table 1. Space–time classification of the independent multipole property tensors in (20)–(22).

	Time-even		Time-odd	
Relative multipole order	Polar	Axial	Polar	Axial
Electric dipole	$\chi_{\alpha\beta}$		$\chi'_{\alpha\beta}$	
Electric quadrupole Magnetic dipole	$a_{\alpha\beta\gamma}$	$G'_{\alpha\beta}$	$a'_{\alpha\beta\gamma}$	$G_{\alpha\beta}$

where $\boldsymbol{E}$ and $\boldsymbol{B}$ are macroscopic fields and $\boldsymbol{J}$ and $\boldsymbol{J}_{\mathrm{b}}$ are the free and bound current densities respectively [17, 34]. In the electric quadrupole–magnetic dipole approximation [34]

$$J_{\mathrm{b}_\alpha} = \dot{P}_\alpha - \tfrac{1}{2}\nabla_\beta \dot{Q}_{\alpha\beta} + (\nabla \times \boldsymbol{M})_\alpha + \cdots . \tag{26}$$

The wave equation for a source-free medium follows from (3), (25), (26), (20)–(22) and to the order of electric quadrupoles and magnetic dipoles is

$$[n^2\sigma_\alpha\sigma_\beta - (n^2-1)\delta_{\alpha\beta} + \chi_{\alpha\beta} - \mathrm{i}\chi'_{\alpha\beta} + \epsilon_0^{-1}c^{-1}n(U^s_{\alpha\beta} - \mathrm{i}U^a_{\alpha\beta})]E_{0\beta} = 0 . \tag{27}$$

In this

$$U^s_{\alpha\beta} = \sigma_\gamma[-\epsilon_{\beta\gamma\delta}G_{\alpha\delta} - \epsilon_{\alpha\gamma\delta}G_{\beta\delta} + \tfrac{1}{2}\omega(a'_{\alpha\beta\gamma} + a'_{\beta\alpha\gamma})] = U^s_{\beta\alpha} \tag{28}$$

$$U^a_{\alpha\beta} = \sigma_\gamma[-\epsilon_{\beta\gamma\delta}G'_{\alpha\delta} + \epsilon_{\alpha\gamma\delta}G'_{\beta\delta} - \tfrac{1}{2}\omega(a_{\alpha\beta\gamma} - a_{\beta\alpha\gamma})] = -U^a_{\beta\alpha} . \tag{29}$$

Equation (27) is the fundamental equation, in the electric quadrupole–magnetic dipole approximation, which describes the propagation of a plane time-harmonic wave through a linear source-free anisotropic magnetic medium in a direction specified by the wavefront normal $\boldsymbol{\sigma}$.

The tensor $U^a_{\alpha\beta}$ in (29) is time-even, as table 1 shows, and it accounts for any reciprocal optical activity which is manifest in magnetic and non-magnetic crystals [25, 35]. For those cubic crystals for which $U^a_{\alpha\beta}$ exists tables 7, 4a, 4d, and 4e in Birss [21] show that it is isotropic. By contrast, the tensor $U^s_{\alpha\beta}$ in (28) is time-odd and, because of $a'_{\alpha\beta\gamma}$ in it, is not isotropic for the magnetic cubics for which it exists, as follows from the tables in Birss [21]. Accordingly, within the electric quadrupole–magnetic dipole approximation, wave propagation in magnetic cubics is isotropic only for those symmetry classes for which $a'_{\alpha\beta\gamma}$ in $U^s_{\alpha\beta}$ vanishes, namely

$$m3, \quad 432, \quad \underline{\bar{4}}3\underline{m}, \quad m3m, \quad m3\underline{m}, \quad \underline{m}3\underline{m} . \tag{30}$$

In the remaining five magnetic cubic classes the propagation anisotropy is manifest in linear birefringence, an expression for which has been derived in terms of a component of $a'_{\alpha\beta\gamma}$ when propagation is along a cube edge [25].

If the wave equation were derived to the next multipole order, would the six magnetic cubic symmetries in (30) still permit isotropic propagation? It is known that anisotropic propagation, as manifest in linear birefringence, occurs in non-magnetic cubic crystals. This was predicted over a century ago by Lorentz [36] and later measured by him and others [37, 38, 39]. Subsequently, a multipole theory to the order of electric octopoles and magnetic quadrupoles has shown that all non-magnetic cubic symmetries exhibit linear birefringence [35]. As magnetic crystals possess the same time-even tensors as their non-magnetic counterparts [21], the magnetic cubic classes in (30) are therefore anisotropic to wave propagation, when this is described in the electric octopole–magnetic quadrupole approximation.

Thus magnetic cubics belonging to the six classes in (30) are the only non-reciprocal substances that will transmit a plane time-harmonic wave isotropically when described within both the electric dipole and the electric quadrupole–magnetic dipole approximations, although not to higher multipole orders. However, in practice the non-isotropy of propagation, being of electric octopole–magnetic quadrupole order, will probably be too small to detect. For instance, the linear birefringence due to such induced multipoles is typically 10^{-6} at optical frequencies [37], roughly two orders of magnitude smaller than circular birefringence, which is explained in the electric quadrupole–magnetic dipole approximation [40]. (These relative magnitudes provide a partial justification of (19).)

It is of interest to note that three of the magnetic classes in (15) that respond bi-isotropically to uniform $\boldsymbol{E}$ and $\boldsymbol{B}$ fields also appear in (30), that is, they transmit a plane time-harmonic wave isotropically when described in the electric quadrupole–magnetic dipole approximation but not to higher multipole orders. These common classes are

$$432, \quad \underline{\bar{4}}3\underline{m}, \quad \underline{m}3\underline{m} \tag{31}$$

of which the first is chiral.

5. The Tellegen medium

A particular example of a magnetoelectric medium was conceptualized by Tellegen in 1948 [23], namely one comprising very small macroscopic particles, randomly oriented as in a liquid suspension, with each particle containing parallel electric and magnetic dipoles (they could equally well be antiparallel). A very large number of ferromagnetic particles, each glued in this way to a microcrystal of an electret and then suspended in a colloidal solution, would in principle constitute a Tellegen medium. (Colloids of ferromagnetic particles are already known, being examples of a magnetic fluid [41].)

Were a uniform electric field $\boldsymbol{E}$ to be applied to a Tellegen medium, the partial alignment of its electric dipoles would produce macroscopic electric and magnetic dipoles parallel to $\boldsymbol{E}$, and similarly if a uniform magnetic field were applied. Thus the four property tensors in (10) would be isotropic and the Tellegen medium would be NRBI in its response to uniform fields.

A real Tellegen medium has never been produced, while its equivalent on the molecular scale is disallowed [26, 27]. So may such a medium really exist? An emphatic negative has been given in two recent papers by Lakhtakia: *'The Tellegen medium is "A boojum, you see"'* [10] and *'Tellegen media: a fecund but incorrect speculation'* [12]. Two main arguments are used in these papers. One is that the magnetoelectric tensor of an isotropic medium is zero (this result being claimed [10] to follow from a covariance condition [1], whereas it is due to the constraint of isotropic symmetry, as discussed in section 2). The other argument is based on Weiglhofer's demonstration in [6] that the magnetoelectric tensor does not enter Maxwell's equations when the bi-isotropic constitutive relations in (9) are substituted into them. Accordingly, if a Tellegen medium is to be shown to exist in principle, it becomes necessary to address these two arguments.

We begin by identifying the basis of the proof by Van Vleck [26] that the molecular equivalent of a Tellegen medium is forbidden. In effect, it is that the time-reversal operator T is a symmetry operator of the molecular Hamiltonian, so doubling for a paramagnetic molecule any other degeneracies and giving rise to states

$$\psi \qquad \text{and} \qquad T\psi = \psi^*$$

which have equal probability and opposite time behaviour [42]. Thus in a fluid of paramagnetic molecules, which also possess an electric dipole (a time-even property), there are necessarily as many with the magnetic dipole (a time-odd property) pointing in one direction, relative to the electric dipole, as in the opposite direction. By contrast, this situation does not occur for the macroscopic 'molecules' of parallel dipoles in a Tellegen medium, simply because they have all been rigidly constructed in this way, with the implication that none exists in the opposite time state with antiparallel dipoles. Furthermore, from symmetry considerations a system with spherical rotational symmetry may possess second-rank axial tensors [42], which can be shown to include time-odd ones like the magnetoelectric tensor in (13). Thus far, then, there seems to be no reason to exclude the existence in principle of a Tellegen medium.

In regard to the second argument, Weiglhofer has correctly established that in its response to a radiation field a Tellegen medium behaves as if its magnetoelectric tensor were zero. However, as shown earlier in this section, such a medium would respond magnetoelectrically to uniform fields, so it would manifest itself physically in this way.

Accordingly, not only may a Tellegen medium exist but it would be experimentally distinguishable from non-magnetoelectric fluids.

6. Conclusion

Various claims have been made [1, 6] that electromagnetic theory places a constraint on the structure of matter, in particular that a linear bi-isotropic medium is necessarily non-magnetic (or reciprocal). In contrast to these claims it is shown in section 2 that symmetry and not electromagnetic theory is responsible for this constraint. To this end we have analysed the behaviour under time reversal of the material properties which enter the constitutive relations for a linear and homogeneous anisotropic medium in the field of a plane time-harmonic electromagnetic wave. For the special case of an isotropic medium we use a result deduced by Van Vleck [26] and Buckingham *et al* [27] to show that its constitutive relations reduce to the bi-isotropic form characteristic of a non-magnetic or reciprocal medium.

Although cubic crystals have isotropic second-rank property tensors, they do not possess isotropic symmetry [21], so that the result in [26] and [27] does not apply to them. Accordingly, it is of relevance to the main issue of this paper to determine whether magnetic cubic crystals, being time-odd or non-reciprocal, are described by bi-isotropic constitutive relations. It is shown in section 3 that magnetic cubics with the point group symmetries in (15) respond bi-isotropically to uniform electric and magnetic fields. In this sense such crystals are NRBI media.

In section 4 a multipole theory is used to derive an equation for the propagation of a plane time-harmonic electromagnetic wave through a magnetic cubic crystal, since this has the highest symmetry of any non-isotropic system. This equation allows the extent of anisotropy of propagation to be determined. It is found that to the order of electric quadrupoles and magnetic dipoles only the magnetic cubics with the point group symmetries in (30) will support isotropic propagation. However, when allowance is made in the theory for higher-order multipoles, propagation in these crystals is seen to be anisotropic, but to an extent which is probably undetectable in practice. Accordingly, the magnetic cubics in (30) would appear to be NRBI to a very good approximation.

Recent electromagnetic theories have claimed to prove that a Tellegen medium is forbidden [6, 10, 12], this being an example of a NRBI substance with very small macroscopic 'molecules' randomly oriented. In section 5 it is shown that, despite these theories, such a medium may exist in principle. Direct experimental evidence for this would be the isotropic polarization and magnetization induced in it by either a uniform electric or magnetic field.

Acknowledgments

We thank Dr Elizabeth Graham and Professor Clive Graham for stimulating and helpful discussions.

References

[1] Lakhtakia A and Weiglhofer W S 1994 *IEEE Trans.* **MTT-42** 1715
[2] Post E J 1962 *Formal Structure of Electromagnetics* (Amsterdam: North-Holland)
[3] Lindell I V, Sihvola A H, Tretyakov S A and Viitanen A J 1994 *Electromagnetic Waves in Chiral and Bi-Isotropic Media* (Norwood, MA: Artech House)
[4] Kong J A 1972 *Proc. IEEE* **60** 1036
[5] Cheng D K and Kong J A 1968 *Proc. IEEE* **56** 248
[6] Weiglhofer W S 1994 *J. Phys. A: Math. Gen.* **27** L871
[7] Lakhtakia A and Weiglhofer W S 1994 *Phys. Rev.* E **50** 5017
[8] Weiglhofer W S and Lakhtakia A 1994 *Electron. Lett.* **30** 1656
[9] Lakhtakia A 1994 *Int. J. Infrared Millim. Wave* **15** 1363
[10] Lakhtakia A 1994 *Int. J. Infrared Millim. Wave* **15** 1625
[11] Weiglhofer W S and Lakhtakia A 1995 *IEEE Ant. & Prop. Mag.* **37** (3) 32
Weiglhofer W S and Lakhtakia A 1995 *IEEE Ant. & Prop. Mag.* **37** (5) 96
Weiglhofer W S and Lakhtakia A 1995 *IEEE Ant. & Prop. Mag.* **37** (6) 111
Weiglhofer W S and Lakhtakia A 1995 *Radio Science Bulletin* No. 237 4
Weiglhofer W S and Lakhtakia A 1996 *IEEE Ant. & Prop. Mag.* **38** (2) 85
Weiglhofer W S and Lakhtakia A 1996 *IEEE Ant. & Prop. Mag.* **38** (4) 94
[12] Lakhtakia A 1995 *Speculations Sci. Technol.* **18** 1
[13] Lakhtakia A and Weiglhofer W S *IMA J. Appl. Math.* **54** 301
[14] Lakhtakia A and Weiglhofer W S 1996 *Phys. Lett.* **213A** 107
[15] Feynman R P, Leighton R B and Sands M 1964 *The Feynman Lectures on Physics* vol II (Reading MA: Addison-Wesley) pp 32-4, 32-5, 36-12
[16] O'Dell T H 1970 *The Electrodynamics of Magneto-electric Media* (Amsterdam: North-Holland) pp 21–5
[17] Purcell E M 1985 *Electricity and Magnetism* (New York: McGraw-Hill) ch 11
[18] Raab R E and Cloete J H 1994 *J. Electromagn. Waves Applic.* **8** 1073
[19] Hornreich R M and Shtrikman S 1968 *Phys. Rev.* **171** 1065
[20] Barron L D 1982 *Molecular Light Scattering and Optical Activity* (Cambridge: Cambridge University Press)
[21] Birss R R 1966 *Symmetry and Magnetism* (Amsterdam: North-Holland)
[22] Folen V J, Rado G T and Stalder E W 1961 *Phys. Rev. Lett.* **6** 607
[23] Tellegen B D H, 1948 *Phillips Res. Rep.* **3** 81
[24] Buckingham A D 1967 *Adv. Chem. Phys.* **12** 107
[25] Graham E B and Raab R E 1992 *Phil. Mag.* B **66** 269
[26] Van Vleck J H 1932 *The Theory of Electric and Magnetic Susceptibilities* (Oxford: Clarendon) p 279
[27] Buckingham A D, Graham C and Raab R E 1971 *Chem. Phys. Lett.* **8** 622
[28] Indenbom V L 1960 *Sov. Phys. Crystallogr.* **5** 493
[29] Birss R R 1963 *Rep. Prog. Phys.* **26** 307
[30] Eyring H, Walter J and Kimball G E 1967 *Quantum Chemistry* (New York: Wiley) p 335
[31] Raab R E 1975 *Molec. Phys.* **29** 1323
[32] de Figueiredo I M B and Raab R E 1981 *Proc. R. Soc.* A **375** 425
[33] Jackson J D 1975 *Classical Electrodynamics* (New York: Wiley) pp 142, 186
[34] Graham E B, Pierrus J and Raab R E 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 4673
[35] Graham E B and Raab R E 1990 *Proc. R. Soc.* A **430** 593
[36] Lorentz H A 1878 *Verh. K. Akad. Wet. Amsterd.* **18**
[37] Lorentz H A 1922 *Proc. K. Akad. Wet. Amsterd.* **24** 333
[38] Pastrnak J and Cross L E 1971 *Phys. Status Solidi* B **44** 313
[39] Pastrnak J and Vedam K 1971 *Phys. Rev.* B **3** 2567
[40] Buckingham A D and Dunn M B 1971 *J. Chem. Soc.* A 1988
[41] Fertman V E 1990 *Magnetic Fluids Guidebook: Properties and Applications* (New York: Hemisphere)
[42] Tinkham M 1964 *Group Theory and Quantum Mechanics* (New York: McGraw-Hill) ch 5