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Homogeneous models for bianisotropic crystals

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Abstract

We extend to bianisotropic structures a formalism already developed, based on the Bloch method for defining the effective dielectric tensor of anisotropic crystals in the long-wavelength approximation. More precisely, we provide a homogenization scheme which yields a wavevector-dependent effective medium for any 3D, 2D, or 1D bianisotropic crystal. We illustrate our procedure by applying this to a 1D magneto-electric smectic C^*-type structure. The resulting equations confirm that the presence of dielectric and magnetic susceptibilities in the periodic structures generates magneto-electric pseudo-tensors for the effective medium. Their contribution to the optical activity of structurally chiral media can be of the same order of magnitude as the one present in dielectric helix-shaped crystals. Simple analytical expressions are found for the most important optical properties of smectic C^*-type structures which are simultaneously dielectric and magnetic.

1. Introduction

Natural and artificial complex and bianisotropic media have received a great deal of attention during the last few decades. The reasons for this are their many potential applications and a desire to gain a physical understanding of them. The structures studied theoretically and experimentally in the applications are diverse. We can mention for instance chiral media (Lindell *et al* 1994, Kharina *et al* 1998), multi-component composites, and artificial dielectrics (Ziolkowski 1997). The peculiarities of various optical aspects of these media, such as the propagation of waves, reflectance properties (Khaliullin and Tretyakov 1998, Semchenko *et al* 1998), anomalous dispersion (Lakhtakia 1998), and biased absorption (Venugopal and Lakhtakia 1998, 2000) of waves, have been widely treated. It should be mentioned that in this paper we shall restrict our consideration to those bianisotropic media whose properties vary over spatial scales much smaller ($\lesssim 10^{-8}$ m) than the wavelength of the electromagnetic signals used to test them. In this sense, the use of an effective medium theory valid in the long-wavelength limit and leading to a macroscopic description is feasible and useful.

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The fundamentals for supporting the macroscopic description in the quasistatic or long-wavelength limit established long ago for dielectric materials (Born and Huang 1954) are discussed for bianisotropic materials in the book of Lakhtakia *et al* (1989) and in the controversial paper of Kamenetskii (1998a). There, it is stated that the separation between the macroscopic and microscopic electromagnetic descriptions is not quite as sharp in these media as it is in pure dielectrics due to the fact that Tellegen pseudo-tensors vanish in the long-wavelength limit. In the literature, there are also dynamical microscopic models for studying the phenomenon of electromagnetic activity in bianisotropic composites for the microwave regime of frequency (Mackay *et al* 2000, Kamenetskii 1998b).

Some direct approaches have been developed to calculate the optical properties of periodic media. However, the determination of such properties requires elaborate numerical calculations. For this reason it is useful to develop homogeneous models approximating the effective properties by means of simple and analytical expressions. The simplest approximation is obtained by spatially averaging the inhomogeneous dielectric and magnetic tensors. This approximation is very rough and does not allow one to account for any effect related to the wavevector dispersion. A better approximation consists in dividing the periodic sample into many homogeneous layers (the thin-plate approximation given by Reese and Lakhtakia (1990) each one providing a transfer matrix. The homogeneous effective transfer matrix is obtained by averaging over all these matrices. The effective dielectric tensor calculated by such a procedure yields a better approximation but still does not account for the spatial dispersion (Becchi *et al* 1999).

By using the Tellegen constitutive relations, we generalize to bianisotropic media a previously developed theory valid only for dielectric materials (Galatola 1997, Ponti *et al* 2001) where the Landau constitutive relations were used. The theory is based on the Bloch wave method which is suitable for exactly describing the wave propagation in periodic media. Our procedure can be used to optically define a homogeneous model, by means of effective material tensors, which globally take into account the non-locality and the multiple scattering of the electromagnetic interaction caused by the periodic inhomogeneities of the medium. This allows us to provide a homogenization scheme which yields a wavevector-dependent effective medium for any 3D, 2D, or 1D periodic structure, in the long-wavelength approximation. The wavevector dispersion of the effective medium is originated by the multiple scattering of the electromagnetic field in the whole crystal.

The outline of this work is as follows. In section 2 the general formalism is derived from the Tellegen constitutive relations for perfectly periodic media—that is to say, crystals without defects and for which the thermal fluctuations of the lattices can be neglected. In section 3 we illustrate our formalism by giving the exact analytical expression up to two-photon scattering events for the effective material tensor of a medium periodic in only one direction, having the typical helical structure of chiral smectic C^* liquid crystals but with non-vanishing magnetic polarizability. In section 4 we use the effective material tensor previously obtained to calculate the plane-wave solutions for the Maxwell equations and derive the expression for the optical activity of the effective medium. Finally, in section 5 we summarize and discuss our results.

2. Basic equations

Maxwell's equations can be written for monochromatic fields in matrix form as

$$\mathbf{R}\vec{\psi} = \mathrm{i}\frac{\omega}{c}\mathbf{M}\vec{\phi}, \tag{1}$$

where $\mathbf{R}$ is

$$\mathbf{R} = \begin{pmatrix} \mathbf{rot} & \mathbf{0} \\ \mathbf{0} & \mathbf{rot} \end{pmatrix}, \tag{2}$$

$$\mathbf{rot} = \begin{pmatrix} 0 & -\partial_3 & \partial_2 \\ \partial_3 & 0 & \partial_1 \\ -\partial_2 & -\partial_1 & 0 \end{pmatrix}, \tag{3}$$

where 1, 2, and 3 are the Cartesian coordinates. The vectors $\vec{\psi}$ and $\vec{\phi}$ are defined as

$$\vec{\psi} \equiv \begin{pmatrix} \Xi_0^{1/2}\vec{e} \\ \Xi_0^{-1/2}\vec{h} \end{pmatrix}, \qquad \vec{\phi} \equiv \begin{pmatrix} \Xi_0^{1/2}\varepsilon_0^{-1}\vec{d} \\ \Xi_0^{-1/2}\mu_0^{-1}\vec{b} \end{pmatrix}, \tag{4}$$

where $\Xi_0 \equiv \sqrt{\mu_0/\varepsilon_0}$ is the vacuum impedance, $\vec{e}$, $\vec{h}$, $\vec{d}$, and $\vec{b}$ are dimensionless electromagnetic fields, and $\mathbf{M}$ is an interchange 6×6 tensor defined by

$$\mathbf{M} \equiv \begin{pmatrix} \mathbf{0} & \mathbf{I} \\ -\mathbf{I} & \mathbf{0} \end{pmatrix}, \tag{5}$$

where $\mathbf{I}$ is the 3×3 identity tensor. For linear media, the most general constitutive relations are those of Tellegen (1948) corresponding to bianisotropic media which in the frequency domain can be written as

$$\vec{\phi} = \gamma\vec{\psi}, \qquad \gamma(\vec{r}) \equiv \begin{pmatrix} \varepsilon(\vec{r}) & \chi(\vec{r}) \\ \zeta(\vec{r}) & \mu(\vec{r}) \end{pmatrix}, \tag{6}$$

where ε and μ are the relative electric permittivity and magnetic permeability tensors, respectively. Since χ and ζ relate polar and axial vectors, these quantities are just dimensionless pseudo-tensors accounting for the fact that in such media the electric field can additionally induce a magnetic polarization and the magnetic field may generate electric polarization. For lossless media, the material tensor γ is Hermitian (Kong 1986).

Now, we generalize the Bloch wave method developed in the paper of Galatola (1997), to define the homogeneous model for bianisotropic media. According to this procedure, the tensor γ defining the material properties of the system, which for a crystal satisfies the periodicity condition $\gamma(\vec{r} + \vec{p}) = \gamma(\vec{r})$, is expanded in a Fourier series as

$$\gamma(\vec{r}) = \sum_{\vec{q}} \gamma(\vec{q}) \exp[\mathrm{i}k_0\vec{q} \cdot \vec{r}], \tag{7}$$

where $k_0\vec{q}$ are the lattice vectors of the reciprocal space. The electromagnetic field can be written in terms of normal modes defined as

$$\vec{\psi}(\vec{r}) = \sum_{\vec{q}} \vec{\psi}(\vec{q}) \exp[\mathrm{i}k_0(\vec{n} + \vec{q}) \cdot \vec{r}], \tag{8}$$

where $k_0\vec{n}$ is the Bloch vector and $\vec{\psi}(\vec{q})$ are the Fourier amplitudes of the Bloch waves. Inserting equations (7) and (8) in equation (6), we find

$$\vec{\phi}(\vec{q}) = \sum_{\vec{q}_a} \gamma(\vec{q} - \vec{q}_a)\vec{\psi}(\vec{q}_a) \tag{9}$$

and

$$\mathbf{S}\vec{\psi}(\vec{q}) = \sum_{\vec{q}_a} \mathbf{M}\gamma(\vec{q} - \vec{q}_a)\vec{\psi}(\vec{q}_a), \tag{10}$$

where $\mathbf{S}$ is the tensor defined as

$$\mathbf{S} = \begin{pmatrix} \alpha & \mathbf{0} \\ \mathbf{0} & \alpha \end{pmatrix} \tag{11}$$

and $\boldsymbol{\alpha}$ is the antisymmetric tensor given by

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & -(\vec{n}+\vec{q})_3 & (\vec{n}+\vec{q})_2 \\ (\vec{n}+\vec{q})_3 & 0 & -(\vec{n}+\vec{q})_1 \\ -(\vec{n}+\vec{q})_2 & (\vec{n}+\vec{q})_1 & 0 \end{pmatrix}. \tag{12}$$

Here also the subscripts 1, 2, 3 correspond to the Cartesian coordinates of the vector $\vec{n}+\vec{q}$. By separating the terms $\vec{q}_a = \vec{q}$ and $\vec{0}$ in equation (10), and solving for $\vec{\psi}(\vec{q})$, we obtain

$$\vec{\psi}(\vec{q}) = \mathbf{G}(\vec{q})\gamma(\vec{q})\vec{\psi}(\vec{0}) + \mathbf{G}(\vec{q}) \sum_{\vec{q}_a \neq \vec{q},\vec{0}} \gamma(\vec{q}-\vec{q}_a)\vec{\psi}(\vec{q}_a) \tag{13}$$

where

$$\mathbf{G}(\vec{q}) = -\begin{pmatrix} \varepsilon(\vec{0}) & \chi(\vec{0}) - \boldsymbol{\alpha}(\vec{q}) \\ \boldsymbol{\alpha}(\vec{q}) + \zeta(\vec{0}) & \boldsymbol{\mu}(\vec{0}) \end{pmatrix}^{-1}. \tag{14}$$

The effective macroscopic model is implicitly defined by the long-wavelength component of the Bloch wave (Born and Huang 1954). According to the first of equations (6), the effective material tensor $\tilde{\gamma}$ is therefore defined by the relation $\vec{\phi}(\vec{0}) = \tilde{\gamma}\vec{\psi}(\vec{0})$. Thus, taking into account equation (9), one obtains

$$\tilde{\gamma}\vec{\psi}(\vec{0}) \equiv \sum_{\vec{q}} \gamma(-\vec{q})\vec{\psi}(\vec{q}). \tag{15}$$

Up to zero order, $\tilde{\gamma}$ is defined by setting $\vec{q} = \vec{0}$ in equation (15) and is given by the zeroth-order Fourier component of $\gamma(\vec{r})$, which coincides with its spatial average $\bar{\gamma}$.

The first-order approximation is obtained by taking into account the components $\vec{q} \neq 0$ through equation (13), where the terms of the sum over $\vec{q}_a$ are neglected. It yields

$$\tilde{\gamma} = \bar{\gamma} + \sum_{\vec{q}\neq 0} \gamma(-\vec{q})\mathbf{G}(\vec{q})\gamma(\vec{q}), \tag{16}$$

which only accounts for two-photon scattering events.

The next iteration is obtained by keeping the summation over $\vec{q}_a$ in equation (13) and approximating $\vec{\psi}(\vec{q}_a)$ by $\mathbf{G}(\vec{q}_a)\gamma(\vec{q}_a)\vec{\psi}(\vec{0})$. The tensor $\tilde{\gamma}$ turns out to be

$$\tilde{\gamma} = \bar{\gamma} + \sum_{\vec{q}\neq 0} \gamma(-\vec{q})\mathbf{G}(\vec{q})\gamma(\vec{q}) + \sum_{\vec{q}\neq 0} \sum_{\vec{q}_a \neq \vec{q},\vec{0}} \gamma(-\vec{q})\mathbf{G}(\vec{q})\gamma(\vec{q}-\vec{q}_a)\mathbf{G}(\vec{q}_a)\gamma(-\vec{q}_a). \tag{17}$$

Iteration of this procedure gives

$$\tilde{\gamma} = \bar{\gamma} + \sum_{N=2}^{\infty} \sum_{\vec{q}_1,\ldots,\vec{q}_{N-1}} \gamma(\vec{q}_1)\mathbf{G}(-\vec{q}_1)\gamma(\vec{q}_2)\mathbf{G}(-\vec{q}_1-\vec{q}_2)\gamma(\vec{q}_3) \times \cdots \times \mathbf{G}(-\vec{q}_1-\vec{q}_2-\vec{q}_{N-1})\gamma(\vec{q}_N), \tag{18}$$

where N defines the multiplicity of the scattering, $\vec{q}_1 \neq \vec{0}, \ldots, \vec{q}_{N-1} \neq \vec{0}$, and

$$\begin{aligned} &\vec{q}_1 + \vec{q}_2 \neq \vec{0}, \ldots, \qquad \vec{q}_1 + \vec{q}_2 + \cdots + \vec{q}_{N-1} \neq \vec{0}, \\ &\vec{q}_1 + \vec{q}_2 + \cdots + \vec{q}_N = \vec{0}. \end{aligned} \tag{19}$$

The latter relation states that only the forward scattering contributes to $\tilde{\gamma}$.

For most periodic structures the two-photon approximation given by equation (16) is good enough for any practical purpose. The role of the multiple scattering is discussed in the paper by Ponti *et al* (2001). As shown by equations (12) and (14), the effective tensor $\tilde{\gamma}$ depends strongly on the wavevector $k_0\vec{n}$ of the plane waves propagating in the effective homogeneous medium (wavevector dispersion).

Before closing this section it is important to stress that our formalism is useful for modelling any 3D, 2D, or 1D periodic structure, as was done in the work by Ponti *et al* (2001) for periodic dielectric structures. Next, to illustrate our formalism, we will analyse a 1D dielectric–magnetic periodic medium with the typical chiral structure of smectic C* (de Gennes and Prost 1994).

3. Magnetic and dielectric media

Let us consider an anisotropic medium ($\chi = \zeta = 0$), whose magnetic and dielectric tensors $\varepsilon(\vec{r})$ and $\boldsymbol{\mu}(\vec{r})$ are periodic along the x_3-axis, and such that their space averages $\bar{\varepsilon}$ and $\bar{\mu}$ show uniaxial symmetry:

$$\bar{\varepsilon} = \begin{pmatrix} \varepsilon_\perp & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_\parallel \end{pmatrix}, \tag{20}$$

$$\bar{\boldsymbol{\mu}} = \begin{pmatrix} \mu_\perp & 0 & 0 \\ 0 & \mu_\perp & 0 \\ 0 & 0 & \mu_\parallel \end{pmatrix}. \tag{21}$$

The dimensionless Bloch vector $\vec{n} = \vec{k}/k_0$ can be chosen without loss of generality as $(n_1, 0, n_3)$. The *exact expression* for $\mathbf{G}$ at any order in $\vec{n}$ and $\vec{q} = (0, 0, q)$ is

$$\mathbf{G} = -\begin{pmatrix} \mathbf{G}_{ee} & \mathbf{G}_{em} \\ \mathbf{G}_{me} & \mathbf{G}_{mm} \end{pmatrix} \tag{22}$$

where

$$\mathbf{G}_{ee} = \begin{pmatrix} \frac{\varepsilon_\parallel \mu_\perp - n_1^2}{g_1} & 0 & \frac{-n_1(n_3+q)}{g_1} \\ 0 & \frac{\mu_\parallel \mu_\perp}{g_2} & 0 \\ -\frac{n_1(n_3+q)}{g_1} & 0 & \frac{\varepsilon_\perp \mu_\perp - (n_3+q)^2}{g_1} \end{pmatrix}, \tag{23}$$

$$\mathbf{G}_{em} = \begin{pmatrix} 0 & \frac{\varepsilon_\parallel (n_3+q)}{g_1} & 0 \\ -\frac{\mu_\parallel (n_3+q)}{g_2} & 0 & \frac{\mu_\perp n_1}{g_2} \\ 0 & -\frac{\varepsilon_\perp n_1}{g_1} & 0 \end{pmatrix}, \tag{24}$$

$$\mathbf{G}_{mm} = \begin{pmatrix} \frac{\varepsilon_\perp \mu_\parallel - n_1^2}{g_2} & 0 & -\frac{n_1(n_3+q)}{g_2} \\ 0 & \frac{\varepsilon_\parallel \varepsilon_\perp}{g_1} & 0 \\ -\frac{n_1(n_3+q)}{g_2} & 0 & \frac{\varepsilon_\perp \mu_\perp - (n_3+q)^2}{g_2} \end{pmatrix}, \tag{25}$$

$\mathbf{G}_{me}$ is the transpose of $\mathbf{G}_{em}$, and

$$\begin{aligned} g_1 &\equiv \varepsilon_\parallel \varepsilon_\perp \mu_\perp - \varepsilon_\perp n_1^2 - \varepsilon_\parallel (n_3+q)^2, \\ g_2 &\equiv \mu_\parallel \mu_\perp \varepsilon_\perp - \mu_\perp n_1^2 - \mu_\parallel (n_3+q)^2. \end{aligned} \tag{26}$$

Notice that $\mathbf{G}_{mm}$ can be obtained from $\mathbf{G}_{ee}$ by swapping ε_i with μ_i where $i = \perp, \parallel$. Even though the medium considered has vanishing magneto-electric tensors (i.e. $\chi = \zeta = 0$), the tensor $\mathbf{G}$ contains non-zero elements which might couple the magnetic and electric fields, since the 3×3 subtensors $\mathbf{G}_{em}$ and $\mathbf{G}_{me}$ are non-vanishing. The elements G_{ij} of the tensor $\mathbf{G}$ vanish if the indices i and j have different parity (for instance when i is even and j is odd). The others are fractions with denominators g_1 and g_2 for odd or even indices, respectively. These last quantities depend quadratically on the parameter $q = \lambda/p$, where p is the period of the medium and λ is the light wavelength. In the limit $\lambda \to \infty$ (static fields) the matrices $\mathbf{G}_{em}$ and $\mathbf{G}_{me}$ go to zero. We should mention that this result is in agreement with some of the discussion addressed by Lakhtakia *et al* (1989) and Kamenetskii (1998a).

We analyse now the simple case of a periodic medium having the same structure as a chiral smectic C* liquid crystal. The local dielectric and magnetic tensors rotate along z in such a way that their principal axes 2 and 3 maintain constant angles $\pi/2 - \theta$ and θ, respectively, with z, where θ is the tilt angle of the smectic structure.

The dielectric and magnetic tensors of the periodic medium are

$$\begin{aligned} \varepsilon(z) &= \overline{\varepsilon} + \varepsilon_{-1} \exp(-\mathrm{i}k_0 q z) + \varepsilon_1 \exp(\mathrm{i}k_0 q z) + \varepsilon_{-2} \exp(-\mathrm{i}\, 2k_0 q z) + \varepsilon_2 \exp(\mathrm{i}\, 2k_0 q z), \\ \boldsymbol{\mu}(z) &= \overline{\boldsymbol{\mu}} + \boldsymbol{\mu}_{-1} \exp(-\mathrm{i}k_0 q z) + \boldsymbol{\mu}_1 \exp(\mathrm{i}k_0 q z) + \boldsymbol{\mu}_{-2} \exp(-\mathrm{i}\, 2k_0 q z) + \boldsymbol{\mu}_2 \exp(\mathrm{i}\, 2k_0 q z). \end{aligned} \tag{27}$$

The tensors $\overline{\varepsilon}$, $\varepsilon_{\pm1}$, $\varepsilon_{\pm2}$ are given by equation (20) and by

$$\varepsilon_{\pm1} = \frac{\varepsilon_{23}}{2}\begin{pmatrix} 0 & 0 & \pm\mathrm{i} \\ 0 & 0 & 1 \\ \pm\mathrm{i} & 1 & 0 \end{pmatrix}, \tag{28}$$

$$\varepsilon_{\pm2} = \frac{\varepsilon_1 - \varepsilon_{22}}{4}\begin{pmatrix} 1 & \mp\mathrm{i} & 0 \\ \mp\mathrm{i} & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{29}$$

respectively, where

$$\begin{aligned} \varepsilon_\perp &= (\varepsilon_1 + \varepsilon_{22})/2, \\ \varepsilon_\parallel &= \varepsilon_2 \sin^2\theta + \varepsilon_3 \cos^2\theta, \\ \varepsilon_{22} &= \varepsilon_2 \cos^2\theta + \varepsilon_3 \sin^2\theta, \\ \varepsilon_{23} &= \tfrac{1}{2}(\varepsilon_2 - \varepsilon_3)\sin 2\theta. \end{aligned} \tag{30}$$

Here ε_1, ε_2, ε_3 are the principal values of the rotating tensor ε. The tensors $\overline{\boldsymbol{\mu}}$, $\boldsymbol{\mu}_{\pm1}$, and $\boldsymbol{\mu}_{\pm2}$ are similarly defined, with μ_{ij} instead of ε_{ij}.

In the two-photon scattering approximation and at any order in the small parameter $q^{-1} \equiv p/\lambda$, the four tensors appearing in the defining equation

$$\tilde{\gamma} = \begin{pmatrix} \tilde{\varepsilon} & \tilde{\boldsymbol{\chi}} \\ \tilde{\boldsymbol{\zeta}} & \tilde{\boldsymbol{\mu}} \end{pmatrix}, \tag{31}$$

are given by

$$\tilde{\varepsilon} = \overline{\varepsilon} - \begin{pmatrix} \varepsilon'_{11} & -\mathrm{i}\varepsilon'_{12} & 0 \\ \mathrm{i}\varepsilon'_{12} & \varepsilon'_{11} & \mathrm{i}\varepsilon'_{23} \\ 0 & -\mathrm{i}\varepsilon'_{23} & \varepsilon'_{33} \end{pmatrix}, \tag{32}$$

where

$$\varepsilon'_{11} = \frac{1}{2}\left[\frac{\varepsilon_{23}^2}{\varepsilon_{33}} - \frac{(\varepsilon_1 - \varepsilon_{22})^2(\mu_1 + \mu_{22})}{16q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22})}\right], \tag{33}$$

$$\varepsilon'_{33} = -\frac{2\varepsilon_{23}^2(\mu_1 + \mu_{22})}{4q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22})}, \tag{34}$$

$$\varepsilon'_{12} = \frac{8n_3 q(\varepsilon_1 - \varepsilon_{22})^2(\mu_1 + \mu_{22})}{(16q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))^2}, \tag{35}$$

$$\varepsilon'_{23} = \frac{2n_1 q \varepsilon_{23}{}^2}{\varepsilon_{33}(4q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))}; \tag{36}$$

the tensor $\tilde{\boldsymbol{\mu}}$ is obtained from $\tilde{\varepsilon}$ by exchanging ε_{ij} with μ_{ij}; the pseudo-tensors are

$$\tilde{\boldsymbol{\zeta}}^\dagger = \tilde{\boldsymbol{\chi}} = -\begin{pmatrix} \mathrm{i}\chi'_{11} & -\chi'_{12} & 0 \\ \chi'_{12} & \mathrm{i}\chi'_{11} & -\chi'_{23} \\ 0 & \chi'_{32} & \mathrm{i}\chi'_{33} \end{pmatrix}, \tag{37}$$

where the superscript † means complex conjugation and transposition and

$$\chi'_{12} = \frac{n_3(\varepsilon_1 - \varepsilon_{22})(\mu_1 - \mu_{22})(16q^2 + (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))}{(-16q^2 + (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))^2}, \tag{38}$$

$$\chi'_{23} = \frac{n_1\varepsilon_{23}(\varepsilon_1 + \varepsilon_{22})\mu_{23}}{\varepsilon_{33}(-4q^2 + (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))}, \tag{39}$$

$$\chi'_{32} = \frac{n_1\varepsilon_{23}(\mu_1 + \mu_{22})\mu_{23}}{\mu_{33}(-4q^2 + (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22}))}, \tag{40}$$

$$\chi'_{11} = \frac{2q(\varepsilon_1 - \varepsilon_{22})(\mu_1 - \mu_{22})}{16q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22})}, \tag{41}$$

$$\chi'_{33} = \frac{4q\varepsilon_{23}\mu_{23}}{4q^2 - (\varepsilon_1 + \varepsilon_{22})(\mu_1 + \mu_{22})}. \tag{42}$$

One may note that γ is Hermitian if $\overline{\varepsilon}(\vec{r})$ and $\bar{\mu}(\vec{r})$ are real. It is worth mentioning that the above expressions have been obtained up to first order in $\vec{n}$, and are valid at any order in the parameter $q^{-1} \equiv p/\lambda$.

4. Plane-wave solutions and optical activity

Since the most interesting optical property of chiral media is their optical activity, in what follows we derive the contributions associated with $\tilde{\varepsilon}$, $\tilde{\boldsymbol{\mu}}$, $\tilde{\boldsymbol{\zeta}}$, and $\tilde{\boldsymbol{\chi}}$ by considering the cases of wave propagation parallel and orthogonal to the helix axis. The integration of Maxwell's equations is not an easy problem since the material tensor $\tilde{\gamma}$ depends on the wavevector $\vec{k} = k_0\vec{n}$. They give an infinite number of plane-wave solutions for any direction of $\vec{k}$, but in our case only four waves are physically meaningful. To find such solutions we expand $\tilde{\gamma}$ given by equation (31) in power series in the small parameter $q^{-1} \equiv p/\lambda$.

For a wave propagating along the x_1-axis and at first order in p/λ we have

$$\tilde{\varepsilon} = \begin{pmatrix} \tilde{\varepsilon}_\perp & 0 & 0 \\ 0 & \tilde{\varepsilon}_\perp & 0 \\ 0 & 0 & \varepsilon_\parallel \end{pmatrix} + \mathrm{i}\frac{p}{\lambda}\frac{\varepsilon_{23}^2 n_1}{2\varepsilon_{33}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, \tag{43}$$

$$\tilde{\boldsymbol{\zeta}}^\dagger = \tilde{\boldsymbol{\chi}} = -\mathrm{i}\frac{p}{\lambda} \begin{pmatrix} \frac{(\varepsilon_1-\varepsilon_{22})(\mu_1-\mu_{22})}{8} & 0 & 0 \\ 0 & \frac{(\varepsilon_1-\varepsilon_{22})(\mu_1-\mu_{22})}{8} & 0 \\ 0 & 0 & \varepsilon_{23}\mu_{23} \end{pmatrix}, \tag{44}$$

$$\tilde{\varepsilon}_\perp = \varepsilon_\perp - \frac{\varepsilon_{23}^2}{2\varepsilon_\parallel}. \tag{45}$$

The tensor $\tilde{\boldsymbol{\mu}}$ is obtained from $\tilde{\varepsilon}$ by exchanging ε_{ij} with μ_{ij}. In order to find the plane-wave solutions from Maxwell's equations, we consider the elements of $\tilde{\gamma}$ depending on p/λ as perturbing terms. At zero order, the wavevector is $\vec{k} = k_0 n_0 \hat{x}_1$ where n_0 satisfies the biquadratic equation

$$n_0^4 - n_0^2(\tilde{\varepsilon}_\perp\tilde{\mu}_\parallel + \tilde{\varepsilon}_\parallel\tilde{\mu}_\perp) + \tilde{\varepsilon}_\parallel\tilde{\varepsilon}_\perp\tilde{\mu}_\parallel\tilde{\mu}_\perp = 0, \tag{46}$$

whose solutions are

$$n_0^\perp = \sqrt{\tilde{\varepsilon}_\perp\tilde{\mu}_\parallel} \qquad \text{and} \qquad n_0^\parallel = \sqrt{\tilde{\varepsilon}_\parallel\tilde{\mu}_\perp}. \tag{47}$$

The waves are linearly polarized. The superscripts $\perp$ and $\parallel$ refer to the direction of the electric field with respect to the plane $(\vec{k}, \hat{x}_3)$.

Each one of the first-order solutions is found by the following iteration procedure. The expression for n_0 given by equation (47) is inserted into the expression for $\tilde{\gamma}$ to obtain a new value of n. The iteration of this procedure converges rapidly for small values of p/λ. The plane waves obtained are in general elliptically polarized.

To show explicitly the presence of the optical rotation, it is convenient to consider the case where the unperturbed solutions are degenerate, i.e. $n_0^\perp = n_0^\parallel$. The first-order solutions are circularly polarized, and the circular birefringence $\Delta n = n^{left} - n^{right}$ is

$$\Delta n = \frac{p}{\lambda}\left[\frac{\varepsilon_{23}^2}{2\varepsilon_{33}}\sqrt{\tilde{\mu}_\parallel\tilde{\mu}_\perp} + \frac{\mu_{23}^2}{2\mu_{33}}\sqrt{\tilde{\varepsilon}_\parallel\tilde{\varepsilon}_\perp} + \rho\varepsilon_{23}\mu_{23} + (\varepsilon_1 - \varepsilon_{22})\frac{(\mu_1 - \mu_{22})}{8\rho}\right], \tag{48}$$

where $\rho = (\tilde{\mu}_\perp \tilde{\varepsilon}_\perp / \tilde{\mu}_\| \tilde{\varepsilon}_\|)^{1/4}$. The first term represents the well known contribution to the rotatory power given by the tensors $\tilde{\varepsilon}$ for $\tilde{\mu}_\perp = \tilde{\mu}_\| = 1$ (Hubert *et al* 1998, Etxebarria and Folcia 2001, Oldano and Rajteri 1997). The second term is new, has the same form as the latter one, and is related to μ, as expected. Finally, the third and fourth terms represent the rotatory power coming from the magneto-electric pseudo-tensors $\tilde{\boldsymbol{\chi}}$ and $\tilde{\boldsymbol{\zeta}}$. The presence of such terms was not obvious *a priori*.

Let us now consider a wave propagation along the x_3-axis (parallel propagation). Up to the third order in p/λ the equations (18) and (38) give

$$\tilde{\varepsilon} = \begin{pmatrix} \tilde{\varepsilon}_\perp & 0 & 0 \\ 0 & \tilde{\varepsilon}_\perp & 0 \\ 0 & 0 & \varepsilon_\| \end{pmatrix} + \mathrm{i}\left(\frac{p}{\lambda}\right)^3 \frac{(\varepsilon_1 - \varepsilon_{22})^2(\mu_1 + \mu_{22})n_3}{32} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{49}$$

$$\begin{aligned} \tilde{\boldsymbol{\zeta}}^\dagger = \tilde{\boldsymbol{\chi}} = -\mathrm{i}\frac{p}{\lambda} & \begin{pmatrix} \frac{(\varepsilon_1-\varepsilon_{22})(\mu_1-\mu_{22})}{8} & 0 & 0 \\ 0 & \frac{(\varepsilon_1-\varepsilon_{22})(\mu_1-\mu_{22})}{8} & 0 \\ 0 & 0 & \varepsilon_{23}\mu_{23} \end{pmatrix} \\ & + \left(\frac{p}{\lambda}\right)^2 \frac{(\varepsilon_1 - \varepsilon_{22})(\mu_1 - \mu_{22})n_3}{16} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \\ & - \mathrm{i}\left(\frac{p}{\lambda}\right)^3 \begin{pmatrix} \frac{(\varepsilon_1^2-\varepsilon_{22}^2)(\mu_1^2-\mu_{22}^2)}{128} & 0 & 0 \\ 0 & \frac{(\varepsilon_1^2-\varepsilon_{22}^2)(\mu_1^2-\mu_{22}^2)}{128} & 0 \\ 0 & 0 & \frac{\varepsilon_{23}\mu_{23}(\varepsilon_1+\varepsilon_{22})(\mu_1+\mu_{22})}{4} \end{pmatrix}. \end{aligned} \tag{50}$$

As before, the tensor $\tilde{\boldsymbol{\mu}}$ is obtained from $\tilde{\varepsilon}$ by switching ε_{ij} and μ_{ij}.

At zero order of the perturbative approach, the wavevector is $\vec{k} = k_0 n_0 \hat{x}_3$, where

$$n_0 = \pm\sqrt{\varepsilon_\perp \mu_\perp}. \tag{51}$$

The two solutions for the forward (and for the backward) propagating modes are coincident, as expected for parallel propagation in uniaxial media. As before, the contribution to the optical activity is given by terms depending on p/λ. The term linear in p/λ is different from zero and is given by

$$\Delta n = \frac{1}{4}\frac{p}{\lambda}(\varepsilon_1 - \varepsilon_{22})(\mu_1 - \mu_{22}). \tag{52}$$

This result is new and unexpected. In fact, it has been shown in the paper written by Ponti *et al* (2001) that such terms are absent for light propagating along the periodicity direction of any 1D dielectric crystal, whose optical activity scales as $(p/\lambda)^3$.

The next contribution comes from the term related to $(p/\lambda)^3$ and reads

$$\Delta n = \frac{1}{32}\left(\frac{p}{\lambda}\right)^3 \left[(\varepsilon_1 - \varepsilon_{22})^2(\mu_1 + \mu_{22})\mu_\perp + \frac{1}{2}(\varepsilon_1^2 - \varepsilon_{22}^2)(\mu_1^2 - \mu_{22}^2)\right]. \tag{53}$$

For non-magnetic media, it reduces to the already known contribution (Ponti *et al* 2001) coming from the dielectric anisotropy (Becchi *et al* 1999).

We finally observe that Maxwell's equations admit exact solutions for axial propagation in the helical media considered here, as shown by de Vries (1951) for cholesteric liquid crystals, by Lakhtakia and Weiglhofer (1994, 1995a, 1995b, 1999) for media simultaneously dielectric and magnetic, and by Lakhtakia and Weiglhofer (2000) for the general case of bianisotropic media. Equations (52) and (53) are in agreement with the exact solutions. The results given in this section, valid in the long-wavelength limit, allow us to find analytical but approximate solutions for any light direction in cholesteric-like and smectic-like structures.

5. Summary and concluding remarks

We generalized to bianisotropic materials the theory based on the Bloch wave method developed in the works by Galatola (1997) and Ponti *et al* (2001) for crystals optically defined by a local permittivity tensor $\varepsilon(\vec{r})$.

We provided exact expressions for the tensors $\tilde{\varepsilon}$, $\tilde{\mu}$, $\tilde{\chi}$, and $\tilde{\zeta}$ defining the effective medium and appearing in the Tellegen constitutive relations. These effective tensors are expressed as the sums of space averages of $\varepsilon(\vec{r})$, $\mu(\vec{r})$, $\chi(\vec{r})$, and $\zeta(\vec{r})$, and of terms taking into account the multiple scattering due to the inhomogeneities of the periodic structure on a mesoscopic scale. Such terms depend explicitly on the wavevector $\vec{k}$ of the plane waves propagating in the effective medium, that as a consequence exhibits a strong wavevector dispersion. The multiple scattering involves all the Fourier components of the functions $\varepsilon(\vec{r})$, $\mu(\vec{r})$, $\chi(\vec{r})$, and $\zeta(\vec{r})$, in such a way as to give a contribution to any one of the four effective tensors even when two of the quantities $\varepsilon(\vec{r})$, $\mu(\vec{r})$, $\chi(\vec{r})$, and $\zeta(\vec{r})$ are zero. If $\chi(\vec{r}) \equiv \zeta(\vec{r}) \equiv 0$, the dominant terms appearing in the expressions for $\tilde{\chi}$ and $\tilde{\zeta}$ scale as the ratio of the structure period and the light wavelength, p/λ. Such terms tend to zero in the limit $\lambda \to \infty$ (static fields) as argued by Lakhtakia *et al* (1989) and Kamenetskii (1998b).

The equations obtained have been applied to periodic media having the typical structure of cholesteric and chiral smectic C* liquid crystals, which are made of molecules whose long axis rotates uniformly along a given direction (the helix axis of the periodic structure). Such crystals, when they are purely dielectric media, have been the object of intensive research because of their huge optical activity, so their optical properties are well known. We showed that the simultaneous presence of dielectric and magnetic susceptibilities gives an additional contribution to the optical activity. More precisely, we showed that the circular birefringence for light propagating along the helix axis, which for $p/\lambda \ll 1$ is practically absent in purely dielectric media, acquires a term scaling as p/λ and proportional to the product of the magnetic and dielectric anisotropies. In the other directions, four terms appear in the expression for the circular birefringence. One of those terms is the magnetic equivalent of the well known term associated with the dielectric anisotropy. The other two are related to the presence of the magneto-electric pseudo-tensors. All these terms are of the same order of magnitude, scale as p/λ, and are proportional to the dielectric and magnetic anisotropies.

For oblique propagation the circular birefringence has small macroscopic effects in purely dielectric media because of the presence of a strong linear birefringence. In the presence of magnetization the linear birefringence can be absent, a fact that gives a new interest to such helical structures.

Finally, we address the following observations. Many sets of bulk and boundary material equations are found in the literature for media displaying wavevector dispersion (Lakhtakia and Weiglhofer 1994). The theory developed in section 3, in which the macroscopic properties are derived from a mesoscopic model defined by the periodic functions $\varepsilon(\vec{r})$ and $\mu(\vec{r})$, is particularly suitable for discerning the limits of validity of different approaches. Indeed, the optical properties of a periodic medium can be easily calculated by simple numerical analysis and used as a test for our equations defining the macroscopic model. In this sense the results given here can be considered as a first and very partial step towards a more complete analysis. In fact, we have studied only the bulk properties of perfect crystals, corresponding to homogeneous and unlimited effective media, whereas important discrepancies in the different macroscopic equations are expected at the boundaries of bounded media.

In our approach all the terms appearing in the Tellegen equations contain terms depending on p/λ. Particular emphasis was given to the linear terms which are related to the optical activity of the medium. The formulation of Born and Wolf (1999) and Landau and Lifshitz

(1985) emphasizes the non-locality of this property because it is given by terms explicitly containing the wavevector $\vec{k}$ and coming from the polarization induced by the space derivatives $\partial E_j/\partial x_i$. In contrast, the optical activity appears, at least formally, in the Tellegen formulation as a local property, in spite of the fact that for homogeneous media the two formulations are equivalent (Peterson 1975). In our approach the optical activity is given by terms contained in $\tilde{\varepsilon}$ and $\tilde{\mu}$, depending explicitly on $\vec{k}$, and by terms contained in $\tilde{\chi}$ and $\tilde{\zeta}$ which are independent of $\vec{k}$. The contributions of all these terms to the optical activity exhibit the same linear dependence on p/λ and are of the same order of magnitude, a fact that is in agreement with the analysis given by Peterson (1975).

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