Bloch Wave Approach to the Optics of Crystals: The Role of Spatial Dispersion

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Abstract

In this work we give new results in the framework of a research on the macroscopic optical properties of crystals whose period p is shorter than the light wavelength but large on the molecular scale, so that they can be treated in the framework of a continuum theory. More precisely, we assume that the optical properties of the crystal are fully defined by a perfectly periodic function $\varepsilon(\vec{r})$, representing the local permittivity of the medium (assumption of locality). To define the effective permittivity tensor $\widetilde{\varepsilon}$ we use a method based on the analysis of the normal modes for the electromagnetic field propagating within of the periodic medium (Bloch waves).

1. Introduction

A perfect crystal is generally treated in optics as a homogeneous medium, despite the fact that in crystals the time harmonic solutions of Maxwell equations (*i.e.* the normal modes of the electromagnetic field) are Bloch waves, whereas in homogeneous media they are plane waves. We recall that a Bloch wave can be considered as a plane wave whose amplitude is a periodic function of $\vec{r}$, or equivalently as a superposition of plane waves, obtained by expanding the amplitude in a Fourier series. The dielectric tensor $\widetilde{\varepsilon}$ of the homogeneous (*macroscopic*) model for crystals is defined by considering as a starting point the Bloch wave, and neglecting the space dependence of its amplitude. This means that only the plane wave component with $\vec{q} = \vec{0}$, that defines the *macroscopic field* [1, 2, 3], is taken into account, and considered as the best 'plane wave approximation' of the actual Bloch wave. The homogeneous medium, where this plane wave can freely propagate, is the *effective macroscopic* medium. Its tensor $\widetilde{\varepsilon}$ is therefore implicitly defined by the equation

$$\vec{D}_{\vec{0}} = \widetilde{\varepsilon}\left(\epsilon_0 \vec{E}_{\vec{0}}\right), \tag{1}$$

where $\vec{D}_{\vec{0}}$, $\vec{E}_{\vec{0}}$ define the zeroth order Fourier components of the Bloch wave and ϵ_0 is the vacuum permittivity.

Here we discuss the application of the Bloch wave method to the study of a short period crystal whose optical properties are assumed as perfectly known and fully defined by a periodic tensor field $\varepsilon(\vec{r})$. This field represents a *mesoscopic model* for the actual crystal, where its molecular (*microscopic*) structure is neglected, and the assumption of *locality* is explicitly made. Despite this last assumption, the spatial dispersion plays a main role in our theory, since $\widetilde{\varepsilon}$ explicitly depends on the wave vector $\vec{k}$ of the internal plane wave, *i.e.* $\widetilde{\varepsilon} = \widetilde{\varepsilon}(\vec{k})$. The tensor field $\varepsilon(\vec{r})$ is well known for many periodic liquid crystal (LC) phases and for artificially made structures (as for instance Reusch piles). Our problem reduces therefore to the search of equations relating $\tilde{\varepsilon}(\vec{k})$ to $\varepsilon(\vec{r})$. Fully analytic expressions for $\tilde{\varepsilon}(\vec{k})$ will be given, that allow for a quantitative analysis of the approximations required to define homogeneous models for non-homogeneous and periodic media. This is the main motivation of our research. The found equations will be applied to the particularly interesting and simple case of cholesteric liquid crystals.

2. Theory

The effective dielectric tensor $\tilde{\varepsilon}$ of the macroscopic model is expressed as a function of the Fourier components $\varepsilon_{\vec{q}}$ of the tensor field $\varepsilon(\vec{r})$:

$$\varepsilon(\vec{r}) = \sum_{\vec{q}} \varepsilon_{\vec{q}} \exp{(i\vec{q}\vec{r})}. \tag{2}$$

The component of order zero is the space average of $\varepsilon(\vec{r})$, *i.e.* $\varepsilon_{\vec{0}} = \bar{\varepsilon}$, and constitutes a first rough approximation for $\tilde{\varepsilon}$. The other approximations are obtained by adding to $\bar{\varepsilon}$ terms having the general structure:

$$\varepsilon_{\vec{q}_1} G_{-\vec{q}_1} \varepsilon_{\vec{q}_2} G_{-(\vec{q}_1+\vec{q}_2)} ... G_{-(\vec{q}_1+...+\vec{q}_{N-1})} \varepsilon_{\vec{q}_N}, \tag{3}$$

where

$$G_{\vec{q}} = \frac{\omega}{c} \left[(\vec{k}+\vec{q})^2 \mathbb{1} - (\vec{k}+\vec{q})(\vec{k}+\vec{q}) - \bar{\varepsilon} \right]^{-1}, \tag{4}$$

$\mathbb{1}$ is the 3×3 identity matrix, $(\vec{k}+\vec{q})(\vec{k}+\vec{q})$ is a dyadic product, and the vectors $\vec{q}_n$ satisfy the following relations:

$$\vec{q}_1 + ... + \vec{q}_N = 0, \quad \sum_{n=1}^{N'} \vec{q}_n \neq 0 \quad \forall\, N' < N. \tag{5}$$

The terms given by Eq. (3) can be interpreted as the effect of the multiple scattering within the periodic structure, and the equation (5) states that only the *forward* scattering with multiplicity $N \geq 2$ gives a contribution to $\tilde{\varepsilon}$. The formal expression of $\tilde{\varepsilon}$ can be written as:

$$\tilde{\varepsilon} = \bar{\varepsilon} + \sum_{N=2}^{\infty} \sum_{\vec{q}_1} ... \sum_{\vec{q}_{N-1}} \varepsilon_{\vec{q}_1} G_{-\vec{q}_1} \varepsilon_{\vec{q}_2} G_{-(\vec{q}_1+\vec{q}_2)} ... G_{-(\vec{q}_1+...+\vec{q}_{N-1})} \varepsilon_{-(\vec{q}_1+...+\vec{q}_{N-1})}. \tag{6}$$

The dependence of $\tilde{\varepsilon}$ on $\vec{k}$ comes from the tensors $G_{\vec{q}}$, as shown by Eq. (4). The dominant contribution to $\tilde{\varepsilon}$ is given in general by the terms with $N = 2$, *i.e.* by the double scattering (two-photon scattering), a fact that allows to greatly simplify Eq. (6). A further simplification is obtained by expanding $G_{\vec{q}}$ in power series of p/λ, where p is an average period of the crystal lattice, since only the first terms of such expansion are expected to give a non-negligible contribution to $\tilde{\varepsilon}(\vec{k})$. This gives

$$\tilde{\varepsilon} = \bar{\varepsilon} + \tilde{\varepsilon}^{(0)} + \left(\frac{p}{\lambda}\right) \tilde{\varepsilon}^{(1)} + \left(\frac{p}{\lambda}\right)^2 \tilde{\varepsilon}^{(2)} + ..., \tag{7}$$

For the simple case of crystals periodic in only one direction, say x_3 (1D crystals), the vectors $\vec{q}$ depend on a single index r, and:

$$\vec{q}_r = rq\hat{x}_3, \quad \tilde{\varepsilon}_0^{(m)} = \sum_{r\neq 0} \varepsilon_r G_{-r}^{(m)} \varepsilon_r;$$

where:

$$G_r^{(0)} = -\frac{1}{\varepsilon_{33}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad G_r^{(1)} = -\frac{1}{r}\frac{1}{\varepsilon_{33}} \begin{pmatrix} 0 & 0 & n_1 \\ 0 & 0 & n_2 \\ n_1 & n_2 & g_{33} \end{pmatrix}, \tag{8}$$

($r = 0, \pm 1, \pm 2, ...$), $q = 2\pi/p$, $\vec{n} = \vec{k}/k_0$, $g_{33} = -\frac{2}{\varepsilon_{33}}(\varepsilon_{13}n_1 + \varepsilon_{23}n_2)$, and only the two-photon scattering has been considered. For chiral structures, the most important term is the one containing $\tilde{\varepsilon}^{(1)}$ and scaling as p/λ, since it is related to the optical activity of the structure. Interestingly, it is identically zero for light propagating along the periodicity axis of 1D crystals, *i.e.* for $\vec{n} = n_3\hat{x}_3$, as clearly shown by the expression of $G_r^{(1)}$, Eq. (8). The higher order terms give in general small corrections to the preceding ones, but in some particular case they can give qualitatively new effects. This actually occurs in cubic crystals (and in particular in the blue phases), because the term scaling as $(p/\lambda)^2$ gives a small anisotropy to the structure. This also occurs along the periodicity axis of 1D crystals, whose optical rotation is related to a term scaling as $(p/\lambda)^3$, since the term scaling as p/λ is zero. In fact, for real $\varepsilon(\vec{r})$ only the terms scaling as $(p/\lambda)^m$ with odd m-values give a contribution to the optical activity.

3. Discussion

(i) The method developed here allows to easily obtain $\tilde{\varepsilon}(\vec{k})$ in all the cases where the function $\varepsilon(\vec{r})$ has only few non-zero Fourier components, a fact that actually occurs in many liquid crystal phases. The simplest and most important one is the choelsteric phase, where only the Fourier components ε_r with $r = 0, \pm 1$ are different from zero. For such crystals:

$$\tilde{\varepsilon}^{(0)} = \begin{pmatrix} \tilde{\varepsilon}_o & 0 & 0 \\ 0 & \tilde{\varepsilon}_o & 0 \\ 0 & 0 & \tilde{\varepsilon}_e \end{pmatrix}, \quad \tilde{\varepsilon}^{(2)} = \varepsilon' \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \tilde{\varepsilon}^{(3)} = \varepsilon'' n_3 \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{9}$$

and $\tilde{\varepsilon}^{(1)} = 0$, where $\tilde{\varepsilon}_o = (\varepsilon_o + \varepsilon_e)/2$, $\tilde{\varepsilon}_e = \varepsilon_e$, $\varepsilon' = (\varepsilon_e - \varepsilon_o)^2(2\varepsilon_o - n_1^2 - n_2^2)/8$ and $\varepsilon'' = i(\varepsilon_e - \varepsilon_o)^2(2\varepsilon_o - n_1^2 - n_2^2)/(4\varepsilon_o)$. The pedices o and e refer to ordinary and extraordinary, respectively. We recall that cholesteric liquid crystals are locally uniaxial, with the optic axis orthogonal to x_3, and that the pitch of their helical structure is equal to $2p$, where p is the lattice period. The Eqs. (9) show that effective medium is still uniaxial, but with the optic axis parallel to x_3. The term scaling as (p/λ) is identically zero for any direction of the light beam: the optical activity of cholesteric is therefore given by the term scaling as $(p/\lambda)^3$. This unusual property constitutes a further demonstration of the unique optical properties of the cholesteric phase.

One may notice that the equations given here can straightforwardly give the expression of $\tilde{\varepsilon}$ up to terms scaling as $(p/\lambda)^3$, whereas the standard effective-medium theories generally only give terms scaling as $(p/\lambda)^0$ and $(p/\lambda)^1$.

(ii) The presence of $\vec{k}$-dependent terms give non-easy problems for the search of plane wave solutions and for the boundary conditions. In fact for any given direction of $\vec{k}$ the dispersion

relation becomes a polynomial that could have, in principle, any number of roots. However, only four solutions, $\pm k_1$ and $\pm k_2$, corresponding to plane waves with different polarization states, have physical meaning. The difficulty for the boundary conditions is due to the fact that the terms of $\widetilde{\varepsilon}(\vec{k})$ depending on the m-power of $\vec{k}$ come from the space derivatives of order m of the electric field. The constitutive equations, written in the Landau form, are in fact:

$$\widetilde{\varepsilon}_{ij} = \widetilde{\varepsilon}_{ij}^{(0)} + \gamma_{ijk}\partial/\partial x_k + \chi_{ijkl}\partial^2/\partial x_k \partial x_l + ...; \qquad \mu = \mu_0.$$

Interestingly, the Bloch wave method gives the actual expression of $\widetilde{\varepsilon}(\vec{k})$ without any use of higher order tensors. In the presence of derivative-depending terms, the usual conditions of continuity for the tangential components of $\vec{E}$ and $\vec{H}$ are no more valid. The problem has been at least partially solved only for the simple case of first-order derivatives [5], by writing the constitutive equations in the Post or Tellegen form [6].

(iii) The limits of validity of the macroscopic model have been carefully tested. For achiral crystals, the approximation $\widetilde{\varepsilon}(\vec{k}) \approx \widetilde{\varepsilon}(\vec{0})$, where all the $\vec{k}$-dependent terms are omitted, is in most cases good enough up to $p/\lambda \approx 0.1$. The full expression of $\widetilde{\varepsilon}(\vec{k})$ allows to extend these limits up to $p/\lambda \approx 0.5$, except for the optical properties directly related to the spatial dispersion (as, for instance, the rotatory power). Such properties require a more careful analysis, that has been partially given on the basis of a different approach in Ref. [7]. The limit $p/\lambda \approx 0.5$ is due to the fact that for higher p-values the periodic medium can give Bragg diffraction, where the dispersion curves show forbidden bands. Obviously, homogeneous models are not able to account for the Bragg diffraction bands, even though they could still give approximated expressions for some optical properties at higher p-values.

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