Bloch wave approach to the optics of crystals

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The Bloch wave method is used to find the effective permittivity tensor $\tilde{\varepsilon}$ of periodic liquid crystals and artificial structures whose period p is short with respect to the light wavelength λ and whose optical properties are defined by a permittivity field $\varepsilon(\mathbf{r})$. The main role of the multiple scattering within the periodic medium is evidenced, and very general expressions of $\tilde{\varepsilon}$, based on expansions in ascending powers of the ratio p/λ and of the light wave vector $\mathbf{k}$, are found. Such expansions allow to discuss the general properties of $\tilde{\varepsilon}$, to clarify the role of the spatial dispersions, i.e., to separate the part of $\tilde{\varepsilon}$ explicitly depending on $\mathbf{k}$ from its $\mathbf{k}$-independent part, and to find some interesting properties of crystals that are (i) periodic in only one direction, or (ii) locally isotropic. Finally, the limits of validity of the macroscopic model are discussed. Within these limits only a few terms of the power expansions are required, and their expressions are explicitly given. The obtained results are also useful to better understand the macroscopic optical properties of solid crystals.

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I. INTRODUCTION

The optical properties of crystals have been the object of intense research in the last century. The results obtained till 1954 are summarized in the classical treatise of Born and Huang [1] where the macroscopic properties are obtained in the framework of the adiabatic (or Placzek's) approximation, by the method of long waves. At low frequencies the electromagnetic interactions reduce to the unretarded Coulomb forces, but already in the infrared frequency region the retardation effects and the radiative energy are no more negligible. The new energy terms greatly complicates the theory, so that the effective permittivity tensor $\tilde{\varepsilon}$ is found in Ref. [1] under simplifying assumptions (*locality* in the macroscopic sense and *homogeneity*) such that all the optical properties related to the spatial dispersion, and in particular the optical activity of chiral media, are lost. To take into account the nonlocality of the electromagnetic interactions and the microscopic inhomogeneity of matter, two different problems must be solved. The first one, that is common to the liquid and crystal phases, has been the object of many excellent monographs [2–4] and papers [5,6]. The other one, that is specific of crystals, has been approached in its full generality recently [7–10].

The two types of difficulties can be well understood in the framework of the phenomenological approach of Agranovich and Ginsburg [11]. The liquids are treated as homogeneous in space and time even from a microscopic point of view, because of the averaging due to the thermal fluctuations. The tensor $\tilde{\varepsilon}$ is therefore defined on the statistical ensemble. It is obtained by decomposing the electromagnetic field in plane waves and it explicitly depends on the wave vector $\mathbf{k}$ and on the frequency ω of the plane-wave components, i.e., $\tilde{\varepsilon} = \tilde{\varepsilon}(\mathbf{k},\omega)$. The assumption of homogeneity is not allowed for crystals, whose permittivity tensor also depends on $\mathbf{r}$ and satisfies the relation $\varepsilon(\mathbf{r},\mathbf{k};\omega)=\varepsilon(\mathbf{r}+\ell,\mathbf{k};\omega)$, where ℓ is a lattice vector. This fact poses the additional problems to find the conditions under which it may be presumed that the tensor $\varepsilon(\mathbf{r},\mathbf{k};\omega)$ is well approximated by an *effective* tensor $\tilde{\varepsilon}(\mathbf{k},\omega)$. In any case, an *approximation* is required. The aim of this paper can now be stated as follows: we only consider the spatial dispersion (neglecting the dependence on ω of the permittivity tensors) and neglect the dependence on $\mathbf{k}$ of $\varepsilon(\mathbf{r};\mathbf{k})$. Our problem reduces therefore to find the macroscopic permittivity tensor $\tilde{\varepsilon}(\mathbf{k})$, by assuming that the optical properties of the crystal are fully defined by a continuous permittivity function $\varepsilon(\mathbf{r})$. For solid crystals, the assumption that $\varepsilon(\mathbf{r})$ is independent of $\mathbf{k}$ is certainly not valid. It is done for sake of simplicity, and because it is important to separate the contribution to $\tilde{\varepsilon}(\mathbf{k})$ of the periodic inhomogeneity of the medium from the other contributions. For liquid crystals the assumption is well motivated. It is in fact well known that the dominant contribution to the optical activity (and more generally to the spatial dispersion) of periodic liquid crystals is related to the helical arrangement of their molecules. In general, the intrinsic contribution defined by $\varepsilon(\mathbf{r};\mathbf{k})$, and due to the chirality of the constituent molecules, is negligible. Moreover, helical structures made of achiral molecules have been recently discovered [12–14]. Their chirality and optical activity are therefore entirely due to the periodic superstructure. In short period liquid crystals, the continuous function $\varepsilon(\mathbf{r})$ defines the optical properties on a *mesoscopic scale*, where the details of the structures on the molecular scale are disregarded. In solid crystals, $\varepsilon(\mathbf{r})$ defines the optical properties of the statistical ensemble.

The standard method to define $\tilde{\varepsilon}(\mathbf{k})$ as a function of $\varepsilon(\mathbf{r})$ is based on the following facts. A plane-wave decomposition of the optical field is suitable in homogeneous media, because the plane waves are eigenmodes of the electromagnetic field. In periodic media, the eigenmodes are Bloch waves and can be decomposed in interacting plane waves. The effective (homogeneous) medium is implicitly defined by only considering the long-wavelength component, that represents the macroscopic field [15]. The wavelengths of the neglected

components are in the order of magnitude of the lattice period p or smaller. We therefore expect that they have very small macroscopic effects in solid crystals, because $p \ll \lambda$. In liquid crystals the ratio p/λ can in principle have any value, so that the definition of the limits of validity of the homogeneous model becomes essential.

The Bloch wave method has been largely used for solid crystals [1,11,15]. It has been extended to liquid crystals by Zeldovich and Tabiryan [16] and by Galatola [8]. This paper is a continuation of the research done by Galatola. It gives a contribution to the solution of one of the problems posed but not solved by Born and Huang [1], and treats quantitatively some of the problems that have been qualitatively discussed by Agranovich and Ginsburg in their phenomenological approach to the spatial dispersion in crystals [11].

II. BASIC EQUATIONS

The effective dielectric tensor $\tilde{\varepsilon}$ of the macroscopic model is a function of the Fourier components of $\varepsilon(\mathbf{r})$, i.e., of the tensors $\varepsilon_{\mathbf{q}}$ defined through

$$\varepsilon(\mathbf{r}) = \sum_{\mathbf{q}} \varepsilon_{\mathbf{q}} \exp(i\mathbf{q}\cdot\mathbf{r}). \tag{1}$$

The component of order zero is the space average of $\varepsilon(\mathbf{r})$, i.e.,

$$\varepsilon_{\mathbf{0}} = \overline{\varepsilon}, \tag{2}$$

and constitutes a first rough approximation for $\tilde{\varepsilon}$. The other approximations are obtained by adding to $\overline{\varepsilon}$ terms having the general structure,

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

where

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

and where the vectors $\mathbf{q}_n$ satisfy the relations

$$\mathbf{q}_n \neq \mathbf{0}, \tag{5}$$

$$\mathbf{q}_1+\mathbf{q}_2 \neq 0, \ldots, \quad \mathbf{q}_1+\cdots+\mathbf{q}_{N-1} \neq \mathbf{0}, \quad \text{and}$$

$$\mathbf{q}_1+\cdots+\mathbf{q}_N = 0. \tag{6}$$

In Eq. (4), $\mathbf{k}$ is the wave vector of the plane-waves propagating in the effective homogeneous medium, $\mathbb{I}$ is the 3×3 identity matrix, and $(\mathbf{k}+\mathbf{q})(\mathbf{k}+\mathbf{q})$ is a dyadic product.

The formal expression of $\tilde{\varepsilon}$ can be written as

$$\tilde{\varepsilon} = \overline{\varepsilon} + \sum_{N=2}^{\infty} \sum_{\mathbf{q}_1} \cdots \sum_{\mathbf{q}_{N-1}} \varepsilon_{\mathbf{q}_1} G_{-\mathbf{q}_1} \varepsilon_{\mathbf{q}_2} \times G_{-(\mathbf{q}_1+\mathbf{q}_2)} \cdots G_{-(\mathbf{q}_1+\cdots+\mathbf{q}_{N-1})} \varepsilon_{-(\mathbf{q}_1+\cdots+\mathbf{q}_{N-1})}. \tag{7}$$

The summations in Eq. (7) take into account the multiple scattering within the medium. More precisely, they give the additional contribution to $\tilde{\varepsilon}$ arising from the inhomogeneity of the periodic medium on the mesoscopic scale. In fact, Eq. (5) states that only the components $\varepsilon_{\mathbf{q}}$ with $\mathbf{q} \neq \mathbf{0}$, which define the inhomogeneous part of the medium, are involved (the homogeneous contribution is already included into $\overline{\varepsilon}$). Equations (6) define the multiplicity N of the scattering and state that only the forward scattering contributes to $\tilde{\varepsilon}(\mathbf{k})$ [17].

The equations given here are a simple generalization of the equations derived by Galatola [8], who only considers the double scattering. Terms that contain the $\mathbf{q}$ components of the polarization current already appear in the microscopic approach of Born and Huang, in Sec. 44 of Ref. [1], but they are neglected in those following sections where the optical properties of solid crystals are considered.

III. THE DIFFERENT APPROXIMATIONS

As discussed in the introduction, the definition of the effective tensor $\tilde{\varepsilon}(\mathbf{k})$ in crystals requires some approximations. The Bloch wave method neglects the short-wavelength components of the normal modes. The very structure of the Eq. (7) requires a further approximation, since the summation over N has to be stopped at some finite value N_{max} in actual computations. If the modulation amplitude of $\varepsilon(\mathbf{r})$ and consequently the components $\varepsilon_{\mathbf{q}}$ are small with respect to $\overline{\varepsilon}$, the dominant terms in the summation are the ones corresponding to $N=2$ (double scattering). In general, the approximation already considered by Galatola is good enough. However, it will be shown in Sec. V that for locally isotropic crystals the terms corresponding to $N=3$ could be the dominant ones.

It is evident that the complexity of the expression of $\tilde{\varepsilon}(\mathbf{k})$ essentially depends on the number of the relevant Fourier components $\varepsilon_{\mathbf{q}}$ of $\varepsilon(\mathbf{r})$. The most important liquid crystals are periodic in only one direction (1D crystals) and have a very limited number of nonzero components: three in the cholesteric phase and five in the chiral smectic phases C^*. For such crystals, the expression of $\tilde{\varepsilon}(\mathbf{k})$ is simple. However, the structure of the matrices $G_{\mathbf{q}}$ is such that it is not easy to have an intuitive feeling of the dependence of $\tilde{\varepsilon}(\mathbf{k})$ on the relevant parameters, as for instance the period of the structure and the light wave vector. It is therefore convenient to find approximate expressions for $G_{\mathbf{q}}$.

In one-dimensional (1D) crystals having period p and periodicity direction x_3, Eq. (1) writes

$$\varepsilon(x_3) = \sum_{r} \varepsilon_r \exp(irqx_3), \tag{8}$$

where $q = 2\pi/p$. The matrices $G_{\mathbf{q}}$ only depend on the integer number r and the matrix $G \equiv G_1$, corresponding to $\mathbf{q} = q\hat{x}_3$ (i.e., to $r=1$), can be written as

$$G=\left(\frac{p}{\lambda}\right)^2\left[\left(\hat{x}_3+\frac{p}{\lambda}\mathbf{n}\right)^2\mathbb{I}-\left(\hat{x}_3+\frac{p}{\lambda}\mathbf{n}\right)\left(\hat{x}_3+\frac{p}{\lambda}\mathbf{n}\right)-\left(\frac{p}{\lambda}\right)^2\overline{\overline{\varepsilon}}\right]^{-1}, \tag{9}$$

where

$$\mathbf{n}=\frac{\mathbf{k}}{k_0}\equiv\frac{c}{\omega}\mathbf{k}. \tag{10}$$

The expression (9) of G suggests to expand this matrix in a Fourier series of the parameter p/λ as

$$G=G^{(0)}+\left(\frac{p}{\lambda}\right)G^{(1)}+\left(\frac{p}{\lambda}\right)^2G^{(2)}+\cdots. \tag{11}$$

The matrix G_r, corresponding to the reciprocal vector $\mathbf{q}=rq\hat{x}_3$, only differs from G for the fact that one must set rq instead of q. This is equivalent to substitute p with p/r in Eq. (9), and $G^{(m)}$ with

$$G_r^{(m)}=r^{-m}G^{(m)} \tag{12}$$

in Eq. (11). The tensor $\tilde{\varepsilon}$ depends on p/λ only through the matrices $G_r^{(m)}$, and it can be written as

$$\tilde{\varepsilon}=\tilde{\varepsilon}^{(0)}+\left(\frac{p}{\lambda}\right)\tilde{\varepsilon}^{(1)}+\left(\frac{p}{\lambda}\right)^2\tilde{\varepsilon}^{(2)}+\cdots, \tag{13}$$

where the coefficients $\tilde{\varepsilon}^{(m)}$ contain in principle an infinite number of summations, corresponding to N-photon scattering with $N=2,3\ldots$. Up to $N=3$, the matrices $\tilde{\varepsilon}^{(0)}$, $\tilde{\varepsilon}^{(1)}$, and $\tilde{\varepsilon}^{(2)}$ are

$$\tilde{\varepsilon}^{(0)}=\overline{\varepsilon}+\sum_{r\neq0}\varepsilon_rG^{(0)}_{-r}\varepsilon_{-r}+\sum_{r\neq0}\sum_{r'\neq0,-r}\varepsilon_rG^{(0)}_{-r}\varepsilon_{r'}\times G^{(0)}_{-(r'+r)}\varepsilon_{-(r'+r)},$$

$$\tilde{\varepsilon}^{(1)}=\sum_{r\neq0}\varepsilon_rG^{(1)}_{-r}\varepsilon_{-r}+\sum_{r\neq0}\sum_{r'\neq0,-r}(\varepsilon_rG^{(0)}_{-r}\varepsilon_{r'}G^{(1)}_{-(r'+r)}\times\varepsilon_{-(r'+r)}+\varepsilon_rG^{(1)}_{-r}\varepsilon_{r'}G^{(0)}_{-(r'+r)}\varepsilon_{-(r'+r)}), \tag{14}$$

$$\tilde{\varepsilon}^{(2)}=\sum_{r\neq0}\varepsilon_rG^{(2)}_{-r}\varepsilon_{-r}+\sum_{r\neq0}\sum_{r'\neq0,-r}(\varepsilon_rG^{(0)}_{-r}\varepsilon_{r'}G^{(2)}_{-(r'+r)}\times\varepsilon_{-(r'+r)}+\varepsilon_rG^{(1)}_{-r}\varepsilon_{r'}G^{(1)}_{-(r'+r)}\varepsilon_{-(r'+r)}+\varepsilon_rG^{(2)}_{-r}\varepsilon_{r'}G^{(0)}_{-(r'+r)}\varepsilon_{-(r'+r)}),$$

where

$$G^{(0)}=-\frac{1}{\overline{\overline{\varepsilon}}_{33}}\begin{pmatrix}0&0&0\\0&0&0\\0&0&1\end{pmatrix},$$

$$G^{(1)}=-\frac{1}{\overline{\overline{\varepsilon}}_{33}}\begin{pmatrix}0&0&n_1\\0&0&n_2\\n_1&n_2&g^{(1)}_{33}\end{pmatrix},$$

$$G^{(2)}=-\frac{1}{\overline{\overline{\varepsilon}}_{33}}\begin{pmatrix}\overline{\varepsilon}_{33}-n_1^2&-n_1n_2&g^{(2)}_{13}\\-n_1n_2&\overline{\varepsilon}_{33}-n_2^2&g^{(2)}_{23}\\g^{(2)}_{13}&g^{(2)}_{23}&g^{(2)}_{33}\end{pmatrix}, \tag{15}$$

and

$$g^{(1)}_{33}=-\frac{2}{\overline{\overline{\varepsilon}}_{33}}(\overline{\varepsilon}_{13}n_1+\overline{\varepsilon}_{23}n_2),$$

$$g^{(2)}_{13}=(n_3-g^{(1)}_{33})n_1-\overline{\varepsilon}_{13},$$

$$g^{(2)}_{23}=(n_3-g^{(1)}_{33})n_2-\overline{\varepsilon}_{23}, \tag{16}$$

$$g^{(2)}_{33}=-g^{(1)2}_{33}+\frac{1}{\overline{\overline{\varepsilon}}_{33}}[\overline{\varepsilon}^2_{13}+\overline{\varepsilon}^2_{23}+\overline{\varepsilon}_{11}n_1^2+\overline{\varepsilon}_{22}n_2^2-2(\overline{\varepsilon}_{13}n_1n_3+\overline{\varepsilon}_{23}n_2n_3-\overline{\varepsilon}_{12}n_1n_2)].$$

A new set of approximations is obtained by only considering in Eq. (13) the terms up to $(p/\lambda)^m$. The lower order approximations, corresponding to $m=0,1$ and $N=2$, are the most important ones and are indeed given by very simple expressions.

A different and even more important way to represent $\tilde{\varepsilon}(\mathbf{n})$ is an expansion in power series of the normalized wave vector $\mathbf{n}$ as

$$\tilde{\varepsilon}_{ij}(\mathbf{n})=\tilde{\varepsilon}_{ij}(\mathbf{0})+\alpha_{ijl}n_l+\beta_{ijlm}n_ln_m+\cdots, \tag{17}$$

that gives another set of possible approximations. It is important to observe that the different terms of this last expansion do not strictly correspond to the terms of the expansion in power series of p/λ, because the tensor $G^{(m)}$ contains terms scaling as $(p/\lambda)^{m'}$, with $m'=m,m-2,\ldots$. This fact appears evident if we consider the definition of $G^{(m)}$ given by Eqs. (9) and (11) [or the expression of $G^{(2)}$, Eq. (15)]. In particular, the second rank tensor $\tilde{\varepsilon}_{ij}(\mathbf{0})$ is obtained by adding to $\varepsilon^{(0)}$ terms scaling as $(p/\lambda)^2,(p/\lambda)^4,\ldots$, and the third rank tensor $\tilde{\varepsilon}_{ijl}$ contains terms scaling as $(p/\lambda),(p/\lambda)^3,\ldots$.

In conclusion, the Bloch wave method generates many approximations for $\tilde{\varepsilon}(\mathbf{n})$ that depend on the maximum values of three parameters: the multiplicity of the scattering, the power of (p/λ) and the rank of the tensors appearing in Eq. (17). The last two are obtained by expanding the matrix $G(p/\lambda,\mathbf{n})$ in power series of (p/λ) or of $\mathbf{n}$. They are not strictly necessary, because $G(p/\lambda,\mathbf{n})$ is already given by an analytic expression, but they are of the most importance for the study of the optical properties of crystals.

The above analysis is easily extended to 3D crystals. In a cartesian frame with $\hat{x}_3\equiv\hat{q}$, the tensor $G_{\mathbf{q}}$ is still given by

Eq. (9), where (p/λ) is substituted by $2\pi(|\mathbf{q}|\lambda)^{-1}$. Each tensor $G_{\mathbf{q}}$ can be expanded in power series of this last parameter, but an expansion of all the tensors $G_{\mathbf{q}}$ in power series of $(\overline{p}/\lambda)$, where $\overline{p}$ is an average dimension of the lattice cell, is more convenient and easily obtained. As it is evident, the expression of $\tilde{\varepsilon}$ becomes rather involved, except for some cubic crystals. However, the fact to be able to express $\tilde{\varepsilon}$ by means of series expansions of the type (13) and (14) allows to find some important properties of these tensors and of $\tilde{\varepsilon}(\mathbf{n})$.

IV. OPTICAL ACTIVITY

We discuss here the properties of $\tilde{\varepsilon}(\mathbf{n})$, by assuming that all the elements of $\varepsilon(\mathbf{r})$ *are real*. This means that we are considering *nondissipative* and *locally nonchiral crystals*. The reality of $\varepsilon(\mathbf{r})$ has the following important implication: in Eq. (13), the terms scaling as $(p/\lambda)^m$ are *real if m is even and purely imaginary if m is odd*.

For the two-photon scattering, this property is immediately found by adding the two terms that contain $\varepsilon_{\mathbf{q}}$ and $\varepsilon_{-\mathbf{q}}$ and taking into account the fact that real $\varepsilon(\mathbf{r})$ implies real $G_{\mathbf{q}}$ with

$$G^{(m)}_{-\mathbf{q}}=(-1)^m G^{(m)}_{\mathbf{q}}, \tag{18}$$

because in defining Eq. (9) a sign change of $\mathbf{q}=q\hat{x}_3$ is fully equivalent to a sign change of p/λ and

$$\varepsilon_{ij}(-\mathbf{q})=\varepsilon^*_{ij}(\mathbf{q}). \tag{19}$$

The extension to the *N*-photon scattering is straightforward.

The effective dielectric tensor of a crystal defined by a real tensor $\varepsilon(\mathbf{r})$ can therefore be written as $\tilde{\varepsilon}=\tilde{\varepsilon}'+i\tilde{\varepsilon}''$, where

$$\tilde{\varepsilon}'=\overline{\varepsilon}+\sum_{m\ even}\left(\frac{p}{\lambda}\right)^m\tilde{\varepsilon}^{(m)},\quad \tilde{\varepsilon}''=\sum_{m\ odd}\left(\frac{p}{\lambda}\right)^m\tilde{\varepsilon}^{(m)}. \tag{20}$$

We recall that in the Born-Landau formalism, used here, the imaginary part of $\tilde{\varepsilon}(\mathbf{n})$ defines the optical activity of the medium, that is the most important optical property related to the spatial dispersion. Since the tensor of rank *m*, in the expansion (17) of $\tilde{\varepsilon}(\mathbf{n})$, contains terms scaling as $(p/\lambda)^{m'}$, with $m'=m,m+2,\ldots$, Eq. (20) implies that such tensors are real if *m* is even, purely imaginary if *m* is odd. We recover here a well known property of $\tilde{\varepsilon}(\mathbf{n})$, which in the Landau approach [18] is a consequence of the fact that the quantity $n_l=(\omega/c)k_l$ appearing in the expression of $\tilde{\varepsilon}(\mathbf{n})$ comes from the space derivative $\partial E_j/\partial x_l$, that for plane waves reduces to ik_lE_j.

Our analysis allows us to find many interesting properties of crystals, and in particular the following ones, related to the optical activity. In the two-photon approximation, the contribution of $\varepsilon_{\mathbf{q}}$ to the optical activity of the terms linear in (p/λ) is zero for light parallel to $\mathbf{q}$. For the particular case of 1D crystals, all the *terms of* $\tilde{\varepsilon}$ *scaling as* p/λ *are identically zero for light propagating along the periodicity direction*. This property is a simple consequence of the fact that the terms appearing in the expression of $\tilde{\varepsilon}^{(1)}$, Eq. (14), contain the tensor $G^{(1)}$ that, according to Eqs. (15) and (16), becomes identically zero if $\mathbf{n}=\mathbf{k}/k_0$ is parallel to x_3. Let us now discuss some interesting consequences of this important property.

Cholesteric liquid crystals are known for their huge rotatory power for light parallel to their helix axis, in the wavelength range from $p|n_e-n_o|$ to p, where n_e and n_o are the local extraordinary and ordinary refractive indices [19]. For higher wavelengths the medium can still rotate the polarization plane of light, but the rotatory power abruptly drops, becoming negligible for $\lambda>5p$. This unusual property is known since 1951 [20], but it has never received a simple explanation. According to our analysis, it is a particular case of a very general property, valid for any 1D crystal. In the absence of the term scaling as (p/λ), the main term giving optical activity scales as $(p/\lambda)^3$. To evidence the practical consequences of this fact we recall that the most important effect of the optical activity is the uniform rotation of the polarization plane of light, a fact that in anisotropic crystals only occurs for light propagating along their optic axes, because in the other directions linearly polarized light becomes elliptically polarized and the major ellipse's axis oscillates around a given direction. The simplest and most known 1D crystals, as for instance cholesterics, chiral smectic C, most TGB crystals, and Reusch piles [21], are macroscopically uniaxial with the optic axis along the periodicity direction. In the limit of small p/λ values, the uniform optical rotation of such crystals is therefore in general of little interest for applications, because it comes from a term scaling as $(p/\lambda)^3$, which gives a rotatory power scaling as p^3/λ^4. The optical activity in, short period, 1D crystals can therefore be of interest for applications if and only if *an optic axis of the effective medium is not in their periodicity direction*.

V. LOCALLY ISOTROPIC CRYSTALS

In what follows, we will consider the simple case of locally isotropic crystals, whose dielectric tensor can be written as $\varepsilon(\mathbf{r})\mathbb{I}$, where $\varepsilon(\mathbf{r})$ is a scalar quantity and $\mathbb{I}$ is the 3×3 identity matrix, that in the following will be omitted. The homogeneous medium is in general anisotropic, owing to the presence of the tensors $G^{(m)}_{\mathbf{q}}$ in the expression of $\tilde{\varepsilon}$, Eq. (7). This equation greatly simplifies because the Fourier component $\varepsilon_{\mathbf{q}}$ commutes with $\varepsilon_{\mathbf{q}'}$ and with the matrices $G^{(m)}_{\mathbf{q}}$. As a consequence of this fact and of the property (18), the terms of $\tilde{\varepsilon}$ coming from two-photon scattering cancel each other and give no contribution to the optical activity. In fact the quantity

$$\varepsilon_{-\mathbf{q}}G^{(m)}_{\mathbf{q}}\varepsilon_{\mathbf{q}}+\varepsilon_{\mathbf{q}}G^{(m)}_{-\mathbf{q}}\varepsilon_{-\mathbf{q}}=G^{(m)}_{\mathbf{q}}[\varepsilon_{-\mathbf{q}}\varepsilon_{\mathbf{q}}+(-1)^m\varepsilon_{\mathbf{q}}\varepsilon_{-\mathbf{q}}] \tag{21}$$

is identically zero for odd *m*. *The contribution to the optical activity of locally isotropic crystals only comes from scattering involving at least three photons* or from the intrinsic chirality of the constituent molecules.

A further simplification, concerning the tensors $G^{(m)}$, comes from the fact that the tensor $\bar{\varepsilon}$ appearing in their expressions is now a scalar quantity. In a coordinate system where $\hat{x}_3 \equiv \hat{q}$, the first five tensors $G^{(m)}$ are represented by the matrices

$$G^{(0)} = -\frac{1}{\bar{\varepsilon}}\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad G^{(1)} = -\frac{1}{\bar{\varepsilon}}\begin{pmatrix} 0 & 0 & n_1 \\ 0 & 0 & n_2 \\ n_1 & n_2 & 0 \end{pmatrix}, \quad G^{(2)} = \frac{1}{\bar{\varepsilon}}\begin{pmatrix} \bar{\varepsilon}-n_1^2 & -n_1 n_2 & n_1 n_3 \\ -n_1 n_2 & \bar{\varepsilon}-n_2^2 & n_2 n_3 \\ n_1 n_3 & n_2 n_3 & n_1^2+n_2^2 \end{pmatrix},$$

$$G^{(3)} = \frac{1}{\bar{\varepsilon}}\begin{pmatrix} -2n_3(\bar{\varepsilon}-n_1^2) & 2n_1 n_2 n_3 & n_1 a \\ 2n_1 n_2 n_3 & -2n_3(\bar{\varepsilon}-n_2^2) & n_2 a \\ n_1 a & n_2 a & -2n_3(n_1^2+n_2^2) \end{pmatrix}, \tag{22}$$

$$G^{(4)} = \frac{1}{\bar{\varepsilon}}\begin{pmatrix} (\bar{\varepsilon}-n_1^2) & -n_1 n_2 & n_1 n_3 c \\ -n_1 n_2 & (\bar{\varepsilon}-n_2^2) & n_2 n_3 c \\ n_1 n_3 c & n_2 n_3 c & (n_1^2+n_2^2) \end{pmatrix}(2n_3^2-a),$$

where

$$a = n_1^2+n_2^2-n_3^2-\bar{\varepsilon},$$

$$c = -(2n_3^2+3a)/(2n_3^2-a). \tag{23}$$

If the spatial dispersion is neglected, the effective medium is uniaxial with the optic axis coincident with x_3, because of the full rotational symmetry (C_∞) of $\varepsilon(\mathbf{r})$ around this axis. The spatial dispersion breaks the uniaxial symmetry of $\tilde{\varepsilon}(\mathbf{n})$, except for the particular case of light propagating along x_3.

VI. LIMITS OF VALIDITY OF THE MACROSCOPIC MODEL: THE ROLE OF SPATIAL DISPERSION

As repeatedly stated in the preceding sections, many approximations are required to define the effective permittivity tensor $\tilde{\varepsilon}(\mathbf{n})$ in crystals. The definition of the limits of validity of the macroscopic models is therefore as important as the definition of $\tilde{\varepsilon}(\mathbf{n})$. It is particularly interesting for liquid crystals, since the ratio (p/λ) can have any value. In this section we consider the models that take into account the multiple scattering up to $N=4$ and the powers of (p/λ) up to $m=\infty$. The cases with $m=2$ and $m=4$ are considered in the next section.

To test the limits of validity of the macroscopic models we compare the optical field generated by an external plane wave within a semi-infinite crystal and the field generated in the corresponding homogeneous medium. This is a good test, but it poses problems that are not easy to be resolved.

For the homogeneous medium, the main difficulty comes from the boundary conditions required in the presence of spatial dispersion. As far as we know, such problem has never been approached in its full generality. The difficulty is due to the fact that the terms of $\tilde{\varepsilon}(\mathbf{k})$ depending on the m power of $\mathbf{k}$ come from the space derivatives of order m of the electric field. In the presence of these derivatives, the usual conditions of continuity for the tangential components of the field vectors $\mathbf{E}$ and $\mathbf{H}$ are no more valid. The problem has been at least partially solved only for the simple case of first order derivatives [22–25]. To avoid the use of the boundary conditions, we consider the wave vectors $\mathbf{k}_j (j=1,2)$ of the internal plane-waves generated by the external one. For a macroscopically uniaxial medium with the optic axis x_3 parallel to the incidence plane (Fig. 1), the vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ refer to the TE and TM polarized waves. The component k_x is equal to the tangential component of the external wave (phase matching condition), the z components are $k_0 n_j$, where n_j are the eigenvalues of the 4×4 matrix B appearing in the propagation equation:

$$\frac{d\beta}{dz} = ik_0 B(\mathbf{k})\beta, \tag{24}$$

where β is a four-dimensional column vector that, in the Berreman formalism [26], is defined as the transpose of the row vector $(e_x, h_y, e_y, -h_x)$, where $\mathbf{e}=(\mu_0/\epsilon_0)^{(-1/4)}\mathbf{E}$ and $\mathbf{h}=(\mu_0/\epsilon_0)^{(1/4)}\mathbf{H}$. In the optical geometry of Fig. 1, the equation system (24) splits in the two independent systems

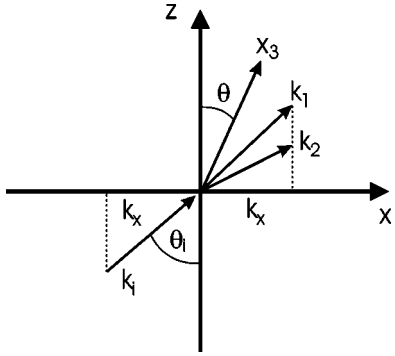

FIG. 1. Optical geometry: $\mathbf{k}_i$ is the wave vector of the input light, x_3 is the symmetry axis of the crystal.

$$\frac{d}{dz}\begin{pmatrix} e_x \\ h_y \end{pmatrix} = ik_0 \begin{pmatrix} -n_x\tilde{\varepsilon}_{xz}/\tilde{\varepsilon}_{zz} & 1-n_x^2/\tilde{\varepsilon}_{zz} \\ \tilde{\varepsilon}_{xx}-\tilde{\varepsilon}_{xz}\tilde{\varepsilon}_{zx}/\tilde{\varepsilon}_{zz} & -n_x\tilde{\varepsilon}_{xz}/\tilde{\varepsilon}_{zz} \end{pmatrix} \begin{pmatrix} e_x \\ h_y \end{pmatrix}, \tag{25}$$

$$\frac{d}{dz}\begin{pmatrix} e_y \\ -h_x \end{pmatrix} = ik_0 \begin{pmatrix} 0 & 1 \\ \tilde{\varepsilon}_{yy}-n_x^2 & 0 \end{pmatrix} \begin{pmatrix} e_y \\ -h_x \end{pmatrix}, \tag{26}$$

corresponding to TM and TE polarization states, respectively. The difficulty of our problem reduces now to the fact that the matrix B explicitly depends on its eigenvalues through the quantities $\tilde{\varepsilon}_{ij}(\mathbf{k})$, i.e., $B=B(\mathbf{k})$. We have computed k_z with the following iteration procedure. We first insert in Eqs. (25) and (26) the tensor $\tilde{\varepsilon}(0)$ obtained by neglecting the $\mathbf{k}$-dependent part of $\tilde{\varepsilon}(\mathbf{k})$, to obtain a first approximation for $k_z=k_0 n$. Each one of these k_z values is now inserted in $\tilde{\varepsilon}(\mathbf{k})$ to obtain a better approximation. The iteration of this procedure gives rapidly converging values for k_z.

For the periodic medium, general and reasonably simple methods giving the internal field are available for 1D crystals. We have used the method developed in [27], that makes use of a propagation equation formally identical to Eq. (24), where the vector β and the matrix B become infinite dimensional. The eigenvectors of B define the Bloch waves generated within the crystal by the external one, the corresponding eigenvalues n_j ($j=-\infty,\infty$) define $k_{j,z}=k_0 n_j$. Here $\mathbf{k}_j$ is the wave vector of the long-wavelength component of the j-Bloch wave (the wave vectors of the other components are $\mathbf{k}_j+\mathbf{q}$). In general, only a limited number of eigenvalues n_j are real. They correspond to propagating modes and define the bulk properties of the electromagnetic field within the medium. The other ones are only present in a boundary layer having finite thickness. They are responsible for some interesting boundary effects [9,25], that are not considered here.

To compare the wave vectors given by the mesoscopic and macroscopic models, we have considered the simple case of a locally isotropic medium whose dielectric tensor is given by $\varepsilon_{ij}=\varepsilon(x_3)\delta_{ij}$, with

$$\begin{aligned} \varepsilon(x_3) &= \overline{\varepsilon} + 2\varepsilon_1 \cos(qx_3) \\ &\equiv \overline{\varepsilon} + \varepsilon_1[\exp(-iqx_3)+\exp(iqx_3)]. \end{aligned} \tag{27}$$

The local isotropy and the fact that x_3 is an axis of full rotational symmetry ensure that the TM and TE polarizations are not coupled. Since $\varepsilon(\mathbf{r})=\varepsilon(-\mathbf{r})$, the structure is achiral and macroscopically uniaxial, with the optic axis parallel to x_3 (we have purposely considered an achiral system, because the validity of the macroscopic model for chiral systems has already been discussed in [9,10]).

The Figs. 2 and 3 give the plots vs p/λ of the quantity $n_z=k_z/k_0$, that represents an effective refractive index if $k_x=k_y=0$. In Fig. 2, the $\mathbf{k}$-dependent terms of $\tilde{\varepsilon}$ are neglected, i.e., $\tilde{\varepsilon}=\tilde{\varepsilon}(0)$, whereas they are taken into account in Fig. 3. The full curves give the n_j values of the propagating modes in the periodic medium, which are exactly the same in the two figures. The Fig. 2 shows that the approximation $\tilde{\varepsilon}=\varepsilon(0)$ is not valid for large p/λ values, thus evidencing the main role played by the spatial dispersion for $p/\lambda>0.1$. In Fig. 3, two full curves are rather well fitted by the model. Despite this fact, it is evident that the macroscopic model loses any meaning for $p\geqslant\lambda$, where more than two full curves are present. Here the periodic medium gives Bragg diffraction, and the lower n_j curves give the k_z values of the diffracted waves.

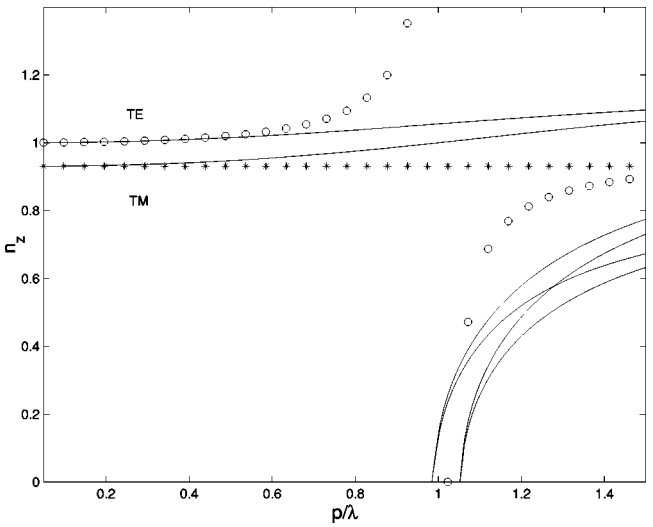

FIG. 2. Components n_z of the normalized wave vector $\mathbf{n}=\mathbf{k}/k_0$ for the running modes in the periodic medium (full lines) optically defined by Eq. (27) and for the plane waves in a homogeneous medium with dielectric tensor $\tilde{\varepsilon}(0)$, in the optical geometry of Fig. 1, and with the following parameters: $\vartheta_i=0$, $\vartheta=\pi/2$, $\overline{\varepsilon}=1$, $\varepsilon_1=0.25$. In the two upper curves the quantity n_z has the meaning of an effective refractive index. The figure also describes a medium with $\overline{\varepsilon}\neq 1$, if we substitute ε_1 with $\varepsilon_1/\overline{\varepsilon}$, n_z with $n_z\sqrt{\overline{\varepsilon}}$ and λ with $\lambda\sqrt{\overline{\varepsilon}}$.

At oblique incidence, a diffracted beam of nonzero order can appear at lower p values, thus restricting the range of

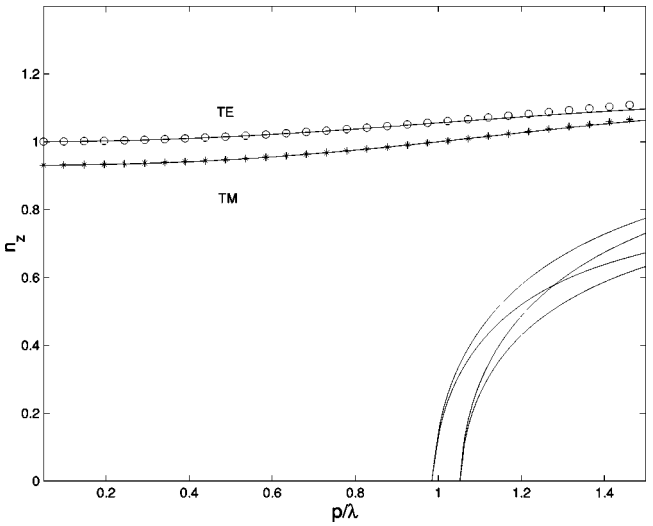

FIG. 3. Same as Fig. 2, with $\tilde{\varepsilon}(\mathbf{k})$ instead of $\tilde{\varepsilon}(0)$.

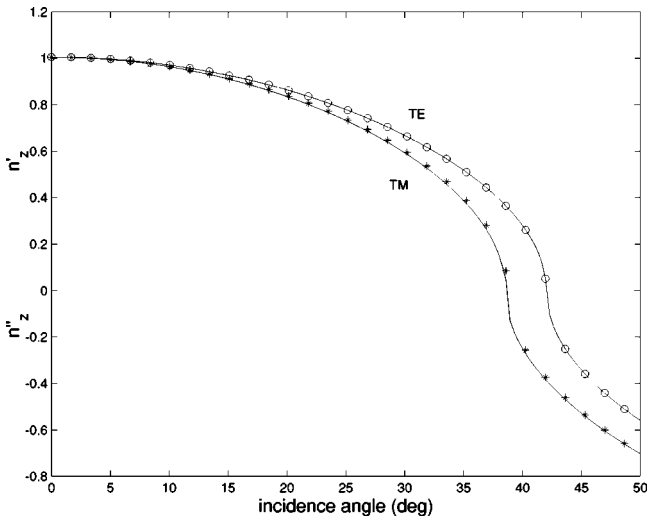

FIG. 4. Real part n_z' and imaginary part n_z'' of $n_z = k_z/k_0$ vs the incidence angle for the Bloch waves within the periodic medium (full lines) and for the plane waves within the macroscopic medium defined by $\tilde{\varepsilon}(\mathbf{k})$. The periodic medium is the same as in Fig. 2, with $p = 0.25\lambda$. The optical geometry is given in Fig. 1, with $\vartheta = 0$. The refractive index of the external medium is $n_i = 1.5$. The quantity n_z is real at the left side of the total reflection angle, purely imaginary at the right side.

validity of the model by a factor 2. In the considered geometry, with x_3 orthogonal to the layer normal ($\vartheta = \pi/2$), the model is valid up to p value nearly equal to one-half of the internal wavelength, *namely, in the whole p interval where the external wave can never give rise to Bragg diffraction* (see Fig. 4).

Let us now consider the case with $\vartheta \neq \pi/2$. If x_3 makes an angle ϑ with z, a sample of the crystal between the planes $z = z_0$ and $z = z_0 + d$ behaves as a grating with grating constant $a = p/\sin\vartheta$. For $a > \lambda$, i.e., for $p/\lambda > \sin\vartheta$, it gives diffracted beams of order $m \neq 0$. Despite this fact, our computations suggest that the macroscopic model can still be usefully applied in this interval, up to $p/\lambda \approx 0.5$, because in this range the grating works in the Raman-Nath regime and the intensity of diffracted beams is generally very small.

VII. LIMITS OF VALIDITY OF THE LOWER ORDER APPROXIMATIONS

The use of the macroscopic model can avoid the heavy calculations required by the actual periodic crystal. However the full expression of $\tilde{\varepsilon}(\mathbf{k})$ given by Eq. (7) is still rather involved and requires a numerical analysis. In this section, we compare the $\mathbf{k}$ values given by the full expression of $\tilde{\varepsilon}(\mathbf{k})$ with the approximations obtained by considering the first terms of its expansion in a power series of (p/λ). To this purpose, we consider the crystal defined by Eq. (27) and make use of a reference frame x_1, x_2, x_3 with x_3 along the periodicity axis. In this frame, the tensor $\tilde{\varepsilon}(0)$ is diagonal, with $\tilde{\varepsilon}_{11} = \tilde{\varepsilon}_{22} = \tilde{\varepsilon}_{\perp}$, $\tilde{\varepsilon}_{33} = \tilde{\varepsilon}_{||}$. In the two-photon approximation, such quantities are

$$\tilde{\varepsilon}_{\perp} = \overline{\varepsilon} + 2\varepsilon_1^2 \left(\frac{p}{\lambda}\right)^2 + 2\varepsilon_1^2 \overline{\varepsilon} \left(\frac{p}{\lambda}\right)^4,$$

$$\tilde{\varepsilon}_{||} = \overline{\varepsilon} - 2\varepsilon_1^2 \overline{\varepsilon}. \tag{28}$$

The tensor $\tilde{\varepsilon}(\mathbf{n})$ is given by $\tilde{\varepsilon} = \tilde{\varepsilon}(0) + \tilde{\varepsilon}^{(2)}(\mathbf{n})(p/\lambda)^2 + \tilde{\varepsilon}^{(4)}(\mathbf{n})(p/\lambda)^4$, where $\mathbf{n} = \mathbf{k}/k_0$ and the tensors $\varepsilon^{(2)}(\mathbf{n})$, $\varepsilon^{(4)}(\mathbf{n})$ are easily derived from the equations given in Secs. IV and V.

In the optical geometry defined by Fig. 1, with $x_3 \equiv x$ and normally incident light, the effective refractive index n satisfies the equations

$$\tilde{\varepsilon}_{||} + \frac{\varepsilon_1^2}{2\overline{\varepsilon}} n^2 \left(\frac{p}{\lambda}\right)^2 + \frac{\varepsilon_1^2}{2\overline{\varepsilon}} n^2 (\overline{\varepsilon} - n^2) \left(\frac{p}{\lambda}\right)^4 - n^2 = 0,$$

$$\tilde{\varepsilon}_{\perp} - \frac{\varepsilon_1^2}{2} n^2 \left(\frac{p}{\lambda}\right)^4 - n^2 = 0, \tag{29}$$

for the TM and TE polarizations, respectively. Interestingly, the first equation is biquadratic and gives therefore two couples of solutions. In general, the dispersion relation in presence of spatial dispersion can give many solutions, but only four are physically acceptable. Here, a couple of solutions of the first equation diverges for $p/\lambda \to 0$, giving an internal wavelength smaller than p, i.e., outside the limits of validity of the model (the conditions under which a third couple of solutions could be physically meaningful are discussed in Ref. [11]).

The solutions given by the above approximation have been compared with the ones obtained in Sec. VI in the approximation up to $(p/\lambda)^{\infty}$. Figure 5 shows that even the simple $(p/\lambda)^2$ approximation, where the terms scaling as $(p/\lambda)^4$ are neglected, is good enough to any practical purpose in the whole range of validity of the macroscopic model (at least for the periodic medium considered here).

VIII. SUMMARY AND CONCLUDING REMARKS

A very general and fully analytic expression is found for the tensor $\tilde{\varepsilon}$ of short periodic crystals, that is formally written as the sum of the space average $\overline{\varepsilon}$ of $\varepsilon(\mathbf{r})$ and of terms that take into account the multiple scattering due to the inhomogeneity of the structure on a mesoscopic scale. In general, all the terms corresponding to scattering with multiplicity $N \geqslant 2$ give a contribution to $\tilde{\varepsilon}$, whose imaginary part $\tilde{\varepsilon}''$ is related to the optical activity of the medium and is identically zero for achiral media. However for locally isotropic and chiral crystals, the contribution to $\tilde{\varepsilon}''$ of the terms with $N = 2$ is identically zero. As a consequence of this fact, the optical activity of such crystals scales as $(\delta\varepsilon)^3$, where $(\delta\varepsilon)$ is the difference between the maximum and the minimum value of $\varepsilon(\mathbf{r})$.

The macroscopic medium displays strong spatial dispersion, i.e., $\tilde{\varepsilon} = \tilde{\varepsilon}(\mathbf{k})$, where $\mathbf{k}$ is the light wave vector. Both $\tilde{\varepsilon}(\mathbf{k})$ and its nondispersive part $\tilde{\varepsilon}(0)$ are expanded in power series of the ratio p/λ, to better describe the optical proper-

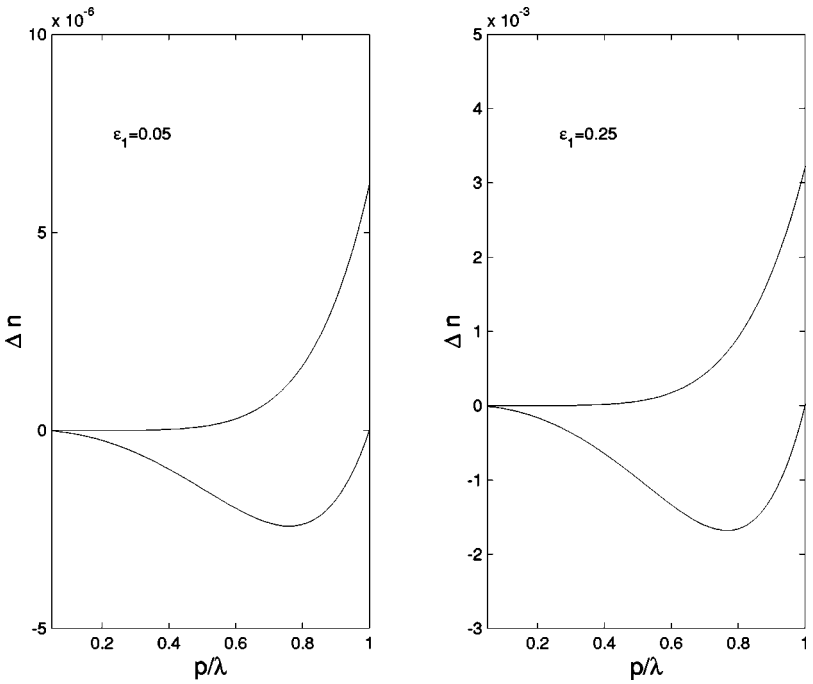

FIG. 5. Difference between the refractive indices of the homogeneous medium computed at any order of p/λ and the ones given by the approximation with terms up to $(p/\lambda)^2$ for two different values of ε_1. The periodic medium is the same as in Fig. 2 and the optical geometry is given in Fig. 1, with $\vartheta=\pi/2$.

ties of the medium and to find simple approximate expressions for these tensors. The obtained equations show that the optical activity of a crystal, that is periodic in only one direction x_3, is negligibly small for light propagating along x_3. More precisely, it scales as $(p/\lambda)^3$. This unusual scaling law, already known for cholesteric liquid crystals, receives here an explanation and is generalized to all 1D crystals.

The limits of validity of macroscopic models, already discussed in [9,10], are reconsidered on the basis of the equations given here. For achiral crystals, the approximation $\tilde{\varepsilon}=\tilde{\varepsilon}(0)$ is generally valid up to $p/\lambda\approx 0.1$. This approximation is particularly interesting because it allows to make use of the usual and well known boundary conditions for the field vectors **E** and **H**. The full expression $\tilde{\varepsilon}(\mathbf{k})$ extends the validity of the model up to $p/\lambda\approx 0.5$. Even the lowest order approximations, which consider only a few terms in the $(p/\lambda)^m$ expansion, are generally very good. However, the model could fail in some very particular optical geometries, as discussed at the end of Sec. VI.

Let us now recall some still open problems, and suggest some possible developments of the research. The most important unsolved problem concerns the definition of a self-consistent set of bulk and boundary conditions in the presence of spatial dispersion. This problem is under study, together with the important problems of the boundary effects in crystals and of the limits of validity of homogeneous models for 2D and 3D crystals.

As an obvious continuation of this research, we are considering the application of the found equations to crystals whose tensor field $\varepsilon(\mathbf{r})$ is known, and in particular to some interesting periodic liquid crystals phases and artificial structures. For such crystals, the equations given here are perhaps the most convenient published up to today, at least for the bulk properties. For this purpose, we observe that in the *LC* literature the trivial approximation $\tilde{\varepsilon}=\overline{\varepsilon}$ is generally used for periodic crystals with $p<\lambda$, because of the lacking of better and reasonably simple approximations.

We also presume that an extension of the theory to crystals with some degree of randomness and to the acoustic waves could be of interest.

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