Spatial dispersion and rotatory power of short-pitch periodic dielectric media

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By a Bloch-wave decomposition, we deduce a general expression for the effective optical dielectric tensor of a short-pitch periodic dielectric medium. The effective dielectric tensor describes an equivalent homogeneous medium presenting spatial dispersion. We consider in detail the case of a cholesteric liquid crystal, deformed by a flexoelectric torque induced by an electric field orthogonal to the helical axis. The distorted structure displays a true rotatory power that scales as the ratio between the pitch of the cholesteric and the vacuum light wavelength. The approximate analytical results are compared with exact numerical calculations, showing a good agreement even far from the limits of validity of the theory. Possible generalizations of these results are discussed. [S1063-651X(97)04104-4]

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I. INTRODUCTION

Natural optical activity is usually associated wth molecules, or arrangement of molecules, having a helical structure [1]. This fact was recognized by Pasteur [2] who argued that the optical activity of natural substances comes from atomic arrangements inside the molecules that differ from their mirror image: he dubbed this property *dissymétrie moléculaire.* One of the most striking examples in this sense is constituted by the DNA double helix. More generally, it can be shown that optical activity arises from spatial inhomogeneities with atomic distances small with respect to the light wavelength [3]. Such inhomogeneities give rise to an imaginary antisymmetric contribution to the dielectric tensor that displays spatial dispersion, i.e., depends on the light wavevector, or, equivalently, on the gradients of the electric field. This imaginary part is responsible for the rotatory power, and can be described in terms of a second-rank gyration pseudotensor [3]. By general symmetry considerations one can infer the permitted nonzero elements of the gyration tensor for the different crystalline classes. However, the actual determination of the optical properties of specific dielectric structures requires elaborate numerical computations [4].

Chiral liquid crystals [5], and, in particular, cholesteric and chiral smectic-*C* liquid crystals, offer a remarkable macroscopic model of the microscopic helical structure of chiral molecules. They also attract much interest for their potential applications, such as the realization of fast electro-optic devices [6]. The optics of chiral liquid crystals has been thoroughly studied from a long time ago [5]. In particular, exact analytical solutions exist for light propagation in cholesteric liquid crystals along the helical axis [7]. They show a large *pseudo*-rotatory-power close to the Bragg reflection band, which changes its sign at the center of the band. In the case of oblique propagation, instead, numerical calculations are required [8]. Recently, it has been numerically shown that interesting unexplored properties arise when the pitch is much shorter than the light wavelength [9]: certain structures acquire a *true* rotatory power for *off-axis* light propagation, which scales as the ratio between the pitch and the wavelength, thus rapidly overcoming the usual *pseudo*-rotatory-power for propagation *along* the helical axis, which scales instead as the cube of the ratio between the pitch and the wavelength. This rotatory power is not simply related to the helical arrangement of the molecules, but it depends on the spatial correlations of neighboring molecules. In fact, it is absent in unperturbed cholesteric liquid crystals.

In this paper, we derive a general formula that allows one to compute the optical properties of an arbitrary short-pitch dielectric periodic medium, by modeling it as an effective spatially-dispersive homogeneous medium that displays optical activity. We apply this model to the case of a cholesteric liquid crystal deformed by a flexoelectric coupling with a dc electric field orthogonal to the unperturbed helical axis [10]. We show analytically that in the distorted structure a *true* rotatory power arises, that scales as the ratio between the pitch of the cholesteric and the light wavelength, similarly to the case of a short-pitch chiral smectic-*C* liquid crystal [11]. The comparison of these results with exact numerical calculations reveals a good agreement even far from the limits of validity of the theory.

In Sec. II we derive a general expression for the effective dielectric tensor, in the limit of short-pitch and small dielectric tensor modulations; in Sec. III we apply this expression to the case of a cholesteric liquid crystal deformed by a flexoelectric coupling with an external electric field; in Sec. IV we obtain an exact numerical solution for the cholesteric and we compare it with the predictions of our analytical model. Finally, in Sec. V we summarize and discuss our results.

II. THEORY

Let us consider a periodic dielectric medium whose relative complex dielectric tensor $\boldsymbol{\epsilon}(\boldsymbol{r})$ reads

$$\boldsymbol{\epsilon}(\boldsymbol{r}) = \sum_{\boldsymbol{q}} \boldsymbol{\epsilon}_{\boldsymbol{q}} \exp(i\boldsymbol{q}\cdot\boldsymbol{r}), \tag{2.1}$$

where the summation runs over all the $\boldsymbol{q}$ vectors of the reciprocal lattice. Any electromagnetic field vector $\boldsymbol{F}$ can then be decomposed in Bloch waves, given by

$$\boldsymbol{F}=\sum_{\boldsymbol{q}} \boldsymbol{F}_{\boldsymbol{q}} \exp[i(\boldsymbol{k}+\boldsymbol{q})\cdot \boldsymbol{r}], \tag{2.2}$$

where $\boldsymbol{k}$ is the Bloch vector and a time dependence of the kind $\exp(-i\omega t)$ is assumed. By inserting (2.1) and (2.2) into Maxwell's equations, one obtains the following propagation equation for each $\boldsymbol{q}$ component of a Bloch wave of the electric field

$$(k_{\boldsymbol{q}}^2 \boldsymbol{I}-\boldsymbol{k}_{\boldsymbol{q}}\otimes \boldsymbol{k}_{\boldsymbol{q}}-k_0^2 \boldsymbol{\epsilon}_{\boldsymbol{0}})\cdot \boldsymbol{E}_{\boldsymbol{q}}=k_0^2 \boldsymbol{\epsilon}_{\boldsymbol{q}}\cdot \boldsymbol{E}_{\boldsymbol{0}}+k_0^2 \sum_{\boldsymbol{q}'\neq \boldsymbol{0},\boldsymbol{q}} \boldsymbol{\epsilon}_{\boldsymbol{q}-\boldsymbol{q}'}\cdot \boldsymbol{E}_{\boldsymbol{q}'}, \tag{2.3}$$

where $k_0=\omega/c$ is the modulus of the vacuum wavevector, $\boldsymbol{I}$ is the 3×3 identity tensor, and we put $\boldsymbol{k}_{\boldsymbol{q}}=\boldsymbol{k}+\boldsymbol{q}$. Equation (2.3) is similar to the wave equation used in the dynamical theory of X-ray scattering [12], except that in the latter, one usually writes the equation in terms of the dielectric displacement $\boldsymbol{D}$ and considers the case of isotropic media. Moreover, in the dynamical theory of x-ray scattering a two-wave approximation close to a Bragg reflection band is considered. Here, instead, we shall suppose that we are far from a Bragg band and that the modulation of the dielectric tensor is sufficiently small, $|\boldsymbol{\epsilon}_{\boldsymbol{q}}|\ll 1$ for $\boldsymbol{q}\neq \boldsymbol{0}$, such that we can solve perturbatively Eq. (2.3) by first neglecting the summation in the right-hand side, i.e., the effect of multiple scattering events: at this stage this amounts to a Born approximation. We can thus express each Fourier component $\boldsymbol{q}\neq \boldsymbol{0}$ of the $\boldsymbol{k}$-th Bloch wave of the electric field as a function of the zeroth-order component alone

$$\boldsymbol{E}_{\boldsymbol{q}}=k_0^2 \boldsymbol{G}(\boldsymbol{q})\cdot \boldsymbol{\epsilon}_{\boldsymbol{q}}\cdot \boldsymbol{E}_{\boldsymbol{0}}, \tag{2.4}$$

where we have defined the tensor

$$\boldsymbol{G}(\boldsymbol{q})=(k_{\boldsymbol{q}}^2 \boldsymbol{I}-\boldsymbol{k}_{\boldsymbol{q}}\otimes \boldsymbol{k}_{\boldsymbol{q}}-k_0^2 \boldsymbol{\epsilon}_{\boldsymbol{0}})^{-1}. \tag{2.5}$$

The electric field components (2.4) will in turn induce a polarization and thus give rise to the dielectric displacement $\boldsymbol{D}$, whose Fourier components are given by

$$\boldsymbol{D}_{\boldsymbol{q}}=\epsilon_0 \sum_{\boldsymbol{q}'} \boldsymbol{\epsilon}_{\boldsymbol{q}-\boldsymbol{q}'}\cdot \boldsymbol{E}_{\boldsymbol{q}'}, \tag{2.6}$$

where ϵ_0 is the vacuum permittivity. Let us now focus our attention on the case in which the pitch of the periodic medium is much shorter than the light vacuum wavelength, $q=|\boldsymbol{q}|\gg k_0$. In this situation, for almost all practical purposes, the meaningful quantity is not the exact value of the electromagnetic field at each point, but a suitable average over a volume sufficiently large with respect to the pitch of the periodic medium, and at the same time sufficiently small with respect to the light wavelength [3]. Such an effective field corresponds to the zeroth-order Fourier component of the Bloch wave. From Eq. (2.6) with $\boldsymbol{q}=\boldsymbol{0}$ and (2.4), one thus sees that in this limit the periodic medium behaves as an effective homogeneous medium whose relative dielectric tensor is given by

$$\boldsymbol{\epsilon}_{\mathrm{eff}}=\boldsymbol{\epsilon}_{\boldsymbol{0}}+k_0^2 \sum_{\boldsymbol{q}\neq \boldsymbol{0}} \boldsymbol{\epsilon}_{-\boldsymbol{q}}\cdot \boldsymbol{G}(\boldsymbol{q})\cdot \boldsymbol{\epsilon}_{\boldsymbol{q}}. \tag{2.7}$$

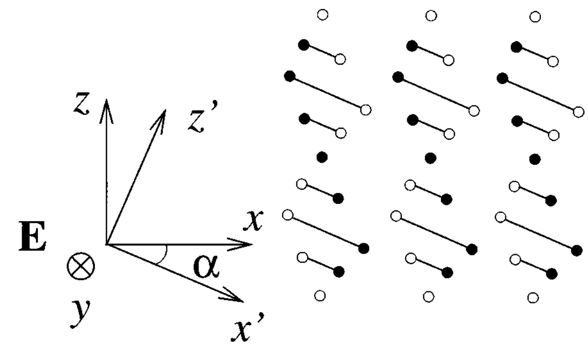

FIG. 1. Geometry of the flexoelectrically distorted cholesteric. The unperturbed helical axis is along z. The static electric field is applied along y. The local optical axis $\hat{\boldsymbol{n}}$ uniformly rotates about z in the plane (x',y), where x' is a direction in the (x,y) plane rotated by an angle α with respect to x.

This is the starting point of our analysis, that extends the quasi-static approach developed in [13]. The correction to the effective dielectric tensor from the average value $\boldsymbol{\epsilon}_{\boldsymbol{0}}$, expressed by the summation on the right-hand side of (2.7), describes the effect of two-photon scattering events. In the following we shall see on a specific example how, in general, the effective homogeneous dielectric medium described by Eq. (2.7) displays a spatial dispersion, accompanied by a rotatory power [3], that scales as the ratio between the vacuum wavelength and the pitch of the periodic medium.

III. CHOLESTERIC LIQUID CRYSTALS

Locally, a cholesteric liquid crystal has the same symmetry as the nematic phase. However, in the cholesteric phase the nematic director $\hat{\boldsymbol{n}}$, which gives the average orientation of the molecules and coincides with the local optical axis of the system, describes a helix around a fixed axis, that we take as the z axis, uniformly rotating perpendicularly to it [5].

Some time ago it has been shown [10] that a linear flexoelectric coupling with a dc electric field, orthogonal to the unperturbed helical axis, turns the plane of rotation of the molecules around the direction of the applied static electric field by the angle

$$\alpha=\tan^{-1}(\overline{e}E/q_0 K), \tag{3.1}$$

where $\overline{e}$ is an average flexoelectric coefficient, E is the amplitude of the static electric field, q_0 is the unperturbed wavevector of the cholesteric, and K is an elastic constant. Therefore, if we take the static electric field along the y axis, the director $\hat{\boldsymbol{n}}$ will uniformly rotate about the z direction in the plane (x',y), where x' is the direction counterclockwise rotated, around the y direction, by the angle α from the x axis in the (x,z) plane (see Fig. 1). In the laboratory frame (x,y,z), the director $\hat{\boldsymbol{n}}$ is given by [10]

$$\hat{\boldsymbol{n}}=\cos\alpha\cos(qz)\hat{\boldsymbol{x}}+\sin(qz)\hat{\boldsymbol{y}}-\sin\alpha\cos(qz)\hat{\boldsymbol{z}}, \tag{3.2}$$

where $q=q_0/\cos\alpha$. The relative dielectric tensor corresponding to the periodic structure is

$$\boldsymbol{\epsilon}=\epsilon_a \hat{\boldsymbol{n}}\otimes \hat{\boldsymbol{n}}+\epsilon_\perp \boldsymbol{I}, \tag{3.3}$$

where $\epsilon_a = n_e^2 - n_o^2$ is the optical anisotropy and $\epsilon_\perp = n_o^2$, where n_o (n_e) is the ordinary (extraordinary) index of refraction. In Eq. (3.3) we have disregarded any biaxiality and intrinsic molecular optical activity as they are largely negligible in practical cases [14].

The reciprocal lattice of the periodic medium has wave vectors $\boldsymbol{q} = mq\hat{\boldsymbol{z}}$, with $m = 0, \pm 1, \pm 2, \ldots$. The only nonzero Fourier components of the dielectric tensor are the components $m=0$ and $m=\pm 2$, with

$$\boldsymbol{\epsilon}_0 = \begin{pmatrix} \epsilon_\perp + \frac{\epsilon_a}{2}\cos^2\alpha & 0 & -\frac{\epsilon_a}{4}\sin 2\alpha \\ 0 & \epsilon_\perp + \frac{\epsilon_a}{2} & 0 \\ -\frac{\epsilon_a}{4}\sin 2\alpha & 0 & \epsilon_\perp + \frac{\epsilon_a}{2}\sin^2\alpha \end{pmatrix}, \tag{3.4}$$

$$\boldsymbol{\epsilon}_2 = \boldsymbol{\epsilon}_{-2}^* = \begin{pmatrix} \frac{\epsilon_a}{4}\cos^2\alpha & -\frac{i}{4}\epsilon_a\cos\alpha & -\frac{\epsilon_a}{8}\sin 2\alpha \\ -\frac{i}{4}\epsilon_a\cos\alpha & -\frac{\epsilon_a}{4} & \frac{i}{4}\epsilon_a\sin\alpha \\ -\frac{\epsilon_a}{8}\sin 2\alpha & \frac{i}{4}\epsilon_a\sin\alpha & \frac{\epsilon_a}{4}\sin^2\alpha \end{pmatrix}. \tag{3.5}$$

In the asymptotic limit $q \to \infty$, the tensor (2.5) becomes then

$$\boldsymbol{G}(m) = \frac{-2}{k_0^2(2\epsilon_\perp + \epsilon_a \sin^2\alpha)} \times \begin{pmatrix} 0 & 0 & \frac{k_x}{mq} \\ 0 & 0 & \frac{k_y}{mq} \\ \frac{k_x}{mq} & \frac{k_y}{mq} & 1 + \frac{\epsilon_a k_x \sin 2\alpha}{mq(2\epsilon_\perp + \epsilon_a\sin^2\alpha)} \end{pmatrix} + O(q^{-2}). \tag{3.6}$$

From Eq. (2.7) and Eqs. (3.4)–(3.6) one readily obtains that the real part $\boldsymbol{\epsilon}'_{\mathrm{eff}}$ of the effective dielectric tensor is uniaxial, with optical axis along the direction z' (cf. Fig. 1). In the rotated frame $(x', y' \equiv y, z')$ it reads explicitly

$$\boldsymbol{\epsilon}'_{\mathrm{eff}} = \begin{pmatrix} \overline{\epsilon_\perp} & 0 & 0 \\ 0 & \overline{\epsilon_\perp} & 0 \\ 0 & 0 & \overline{\epsilon_\parallel} \end{pmatrix}, \tag{3.7}$$

with

$$\overline{\epsilon_\perp} = \epsilon_\perp + \frac{\epsilon_a}{2} - \frac{\epsilon_a^2 \sin^2\alpha}{4(2\epsilon_\perp + \epsilon_a\sin^2\alpha)}, \tag{3.8}$$

$$\overline{\epsilon_\parallel} = \epsilon_\perp . \tag{3.9}$$

Therefore, if the cholesteric is a locally positive uniaxial crystal, as it is usually the case, the real part of the effective dielectric tensor turns out to correspond to a negative uniaxial crystal [9]. Moreover, differently from the case of a chiral smectic-C liquid crystal [11], it is not possible to reverse the sign of the optical anisotropy by changing the angle α.

The rotation of the macroscopic optical axis has been experimentally detected at normal incidence [10], and provides the means for detecting the deformation of the cholesteric structure under the influence of the applied electric field. However, in the distorted cholesteric ($\alpha \neq 0$) the effective dielectric tensor also displays a nonzero imaginary part $i\boldsymbol{\epsilon}''_{\mathrm{eff}}$. In the same rotated frame it is given by

$$\boldsymbol{\epsilon}''_{\mathrm{eff}} = \frac{k_x}{q} \frac{\epsilon_a^2 \epsilon_\perp \sin 2\alpha}{4(2\epsilon_\perp + \epsilon_a \sin^2\alpha)^2} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{3.10}$$

This imaginary part, which comes from the spatial variation of the dielectric tensor, displays a spatial dispersion, i.e., depends on the light wave vector $\boldsymbol{k}$, and gives rise to an optical activity [3]. According to Eq. (3.10), the optical activity is maximum for light propagating along the x direction, i.e., along the direction perpendicular to the unperturbed helical axis and to the applied static electric field. The imaginary part of the dielectric tensor is usually interpreted by supposing that, for a spatially nonuniform linear dielectric medium, the average optical dielectric displacement $\langle \boldsymbol{D} \rangle \equiv \boldsymbol{D_0}$ depends not only on the average electric field $\langle \boldsymbol{E} \rangle \equiv \boldsymbol{E_0}$, but also on its spatial derivatives [3]. In Cartesian coordinate components

$$\langle D_i \rangle = \epsilon_0 \left[\overline{\epsilon_{ij}} \langle E_j \rangle + \gamma_{ijm} \frac{\partial \langle E_j \rangle}{\partial x_m} \right] = \epsilon_0 (\overline{\epsilon_{ij}} + i\gamma_{ijm} k_m) \langle E_j \rangle, \tag{3.11}$$

where summation over repeated indices is implied and the last equality holds true for a plane wave whose wavevector is $\boldsymbol{k}$. It can be shown [3] that the third-rank tensor γ_{ijm} is antisymmetric with respect to the first two indices $\gamma_{ijm} = -\gamma_{jim}$ and can therefore be written as a function of the second-rank gyration pseudotensor $\boldsymbol{g}$, having Cartesian components g_{ij}, according to

$$\gamma_{ijm} = k_0^{-1} e_{ijn} g_{nm}, \tag{3.12}$$

where e_{ijn} is the Levi-Civita antisymmetric unit pseudotensor. In the present case, in the rotated frame in which the real part of the dielectric tensor is diagonal, the gyration tensor reads

$$\boldsymbol{g}=g_x\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ -\cos\alpha & 0 & -\sin\alpha \end{pmatrix}, \tag{3.13}$$

with

$$g_x=\frac{k_0}{q}\,\frac{\epsilon_a^2\epsilon_\perp\sin 2\alpha}{4(2\epsilon_\perp+\epsilon_a\sin^2\alpha)^2}. \tag{3.14}$$

Therefore the rotatory power depends on the handedness of the cholesteric helix and on the sign of the rotation angle α, i.e., on the sign of the applied field; it does not depend on the sign of the optical anisotropy ϵ_a. This rotatory power scales as the ratio between the pitch of the cholesteric and the light wavelength. For typical values of the indices of refraction of liquid crystals, g_x can reach values of the order of 10^{-2}, to which they correspond huge rotatory powers of the order of a degree per wavelength. We note that the gyration tensor (3.13) is not symmetric; however, in the determination of the optical activity, only its symmetric part enters.

IV. EXACT NUMERICAL SOLUTIONS

In order to check the limits of validity of the previously obtained expressions for the effective dielectric tensor of the distorted cholesteric, we performed a fully numerical simulation of the optical behavior of a finite cholesteric slab sandwiched between two equal isotropic dielectric media having index of refraction n_1.

In this section we take the z axis as the sample normal, with the cholesteric comprised between the planes $z=0$ and $z=d$, the light impinging from the $z<0$ half-space, the unperturbed helical axis along the x direction and finally the static electric field applied along z, such that the optical axis of the effective medium rotates in the (x,y) plane. In this geometry the sample behaves as a phase diffraction grating; to numerically analyze its optical properties we follow the procedure employed in [15], that we briefly recall for the case at hand. The electromagnetic field is expressed as the Bloch wave

$$\boldsymbol{E}=Z_0^{1/2}\sum_{m=-\infty}^{+\infty}\boldsymbol{e}_m\exp[i(\boldsymbol{k}_m\cdot\boldsymbol{r}-\omega t)]+\text{c.c.}, \tag{4.1a}$$

$$\boldsymbol{H}=Z_0^{-1/2}\sum_{m=-\infty}^{+\infty}\boldsymbol{h}_m\exp[i(\boldsymbol{k}_m\cdot\boldsymbol{r}-\omega t)]+\text{c.c.}, \tag{4.1b}$$

where $Z_0=\sqrt{\mu_0/\epsilon_0}$ is the vacuum characteristic impedance, c.c. indicates complex conjugate and the wave vectors $\boldsymbol{k}_m$ are given by

$$\boldsymbol{k}_m=k_0[(p_i+mp)\hat{\boldsymbol{x}}+q_i\hat{\boldsymbol{y}}], \tag{4.2}$$

where, as in Eq. (3.2), $k_0p=q$ is the wavevector of the nematic director—and thus of the dielectric tensor—and p_i and q_i are the normalized x and y components of the incident wavevector, defined in terms of the polar angles ϑ_i and φ_i of the incident beam by

$$p_i=n_1\sin\vartheta_i\cos\varphi_i, \tag{4.3a}$$

$$q_i=n_1\sin\vartheta_i\sin\varphi_i. \tag{4.3b}$$

With decomposition (4.1), Maxwell's equations can be cast in the matrix form

$$\frac{d\psi}{dz}=ik_0D\psi, \tag{4.4}$$

where ψ is the column matrix

$$\psi=\begin{pmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{pmatrix}, \tag{4.5}$$

in which each element is in turn an infinite column matrix containing the various Fourier components of the transverse part of the fields (4.1)

$$E_x=\begin{pmatrix} \vdots \\ e_{x-1} \\ e_{x0} \\ e_{x1} \\ \vdots \end{pmatrix},\quad E_y=\begin{pmatrix} \vdots \\ e_{y-1} \\ e_{y0} \\ e_{y1} \\ \vdots \end{pmatrix},\quad H_x=\begin{pmatrix} \vdots \\ h_{x-1} \\ h_{x0} \\ h_{x1} \\ \vdots \end{pmatrix},$$

$$H_y=\begin{pmatrix} \vdots \\ h_{y-1} \\ h_{y0} \\ h_{y1} \\ \vdots \end{pmatrix}, \tag{4.6}$$

and D is the propagation matrix

$$D=\begin{pmatrix} -PE_{zz}^{-1}E_{zx} & 1-PE_{zz}^{-1}P & -PE_{zz}^{-1}E_{zy} & 0 \\ E_{xx}-E_{xz}E_{zz}^{-1}E_{zx} & -E_{xz}E_{zz}^{-1}P & E_{xy}-E_{xz}E_{zz}^{-1}E_{zy} & 0 \\ 0 & 0 & 0 & 1 \\ E_{yx}-E_{yz}E_{zz}^{-1}E_{zx} & -E_{yz}E_{zz}^{-1}P & E_{yy}-E_{yz}E_{zz}^{-1}E_{zy}-P^2 & 0 \end{pmatrix}. \tag{4.7}$$

Here 1 and 0 are the identity and the null matrix, respectively. P is an infinite diagonal matrix having diagonal elements equal to p_i+mp ($m=-\infty,\ldots,+\infty$), and $E_{\alpha\beta}$ ($\alpha,\beta=x,y,z$) are infinite square matrices with elements

$$(E_{\alpha\beta})_{mn}=\epsilon_{\alpha\beta,(m-n)}, \tag{4.8}$$

where $\epsilon_{\alpha\beta,(m-n)}$ is the $(m-n)$ Fourier harmonic of the $\alpha\beta$ component of the dielectric tensor (2.1).

The matrix Eq. (4.4) is solved by considering only a limited set $-M,-M+1,\ldots,-1,0,1,\ldots,M-1,M$ of the Fourier components. The propagation matrix D is constant in the external homogeneous dielectric media and inside the periodic slab; in the external media its eigenvectors ψ_{em} represent plane waves whose nonzero components are given by

$$e_{xm}=\frac{1}{\sqrt{2}}\frac{i_m c_m|r_{em}|^{1/2}}{n_1}, \tag{4.9a}$$

$$h_{ym}=\frac{1}{\sqrt{2}}\frac{c_m n_1}{|r_{em}|^{1/2}}, \tag{4.9b}$$

$$e_{ym}=\frac{1}{\sqrt{2}}\frac{i_m s_m|r_{em}|^{1/2}}{n_1}, \tag{4.9c}$$

$$-h_{xm}=\frac{1}{\sqrt{2}}\frac{s_m n_1}{|r_{em}|^{1/2}}, \tag{4.9d}$$

for the transverse magnetic (TM) waves and by

$$e_{xm}=\frac{1}{\sqrt{2}}\frac{s_m}{|r_{em}|^{1/2}}, \tag{4.10a}$$

$$h_{ym}=\frac{1}{\sqrt{2}}i_m s_m|r_{em}|^{1/2}, \tag{4.10b}$$

$$e_{ym}=\frac{-1}{\sqrt{2}}\frac{c_m}{|r_{em}|^{1/2}}, \tag{4.10c}$$

$$-h_{xm}=\frac{-1}{\sqrt{2}}i_m c_m|r_{em}|^{1/2}, \tag{4.10d}$$

for the transverse electric (TE) waves. Here

$$c_m=\cos\varphi_m, \tag{4.11a}$$

$$s_m=\sin\varphi_m, \tag{4.11b}$$

$$\varphi_m=\tan^{-1}\left(\frac{q_i+mp}{p_i}\right), \tag{4.11c}$$

where φ_m represents the azimuthal propagation angle of the m-diffracted beam, r_{em} is the eigenvalue associated to the eigenvector ψ_{em}, that gives the z component of the wave vector normalized to k_0

$$r_{em}=\pm\sqrt{n_1^2-(p_i+mp)^2-q_i^2}, \tag{4.12}$$

and, finally, i_m is a coefficient equal to $+1$ for forward waves ($r_{em}>0$), -1 for backward ones ($r_{em}<0$), and $\pm i$ for inhomogeneous waves ($r_{em}^2<0$). The eigenvectors have been normalized in such a way that the forward (backward) solutions have energy flux in the z direction equal to $+1$ (-1).

Inside the periodic medium, the eigenvectors ψ_{pm} of the propagation matrix (4.7) can be found only numerically. Truncating the maximum Fourier components to $\pm M$, we obtain $4(2M+1)$ different eigenvectors. Each of them represents a proper wave propagating according to

$$\psi_{pm}(z)=\exp(ik_0\lambda_m z)\psi_{pm}(0), \tag{4.13}$$

where λ_m is the eigenvalue associated with ψ_{pm}. Real eigenvalues correspond to propagating solutions, complex ones to inhomogeneous waves. In the limit of short pitch with respect to the light wavelength ($p\to\infty$), both outside and inside the medium, only four eigenvalues remain real, corresponding to two forward and to two backward solutions. However, the other inhomogeneous waves cannot be neglected, as they are responsible for the rotatory power that we are looking for.

As $p\to\infty$, the imaginary parts $\mathrm{Im}(\lambda_m)$ of the complex eigenvalues λ_m grow very rapidly, giving diverging propagation factors $\exp(ik_0\lambda_m d)$ for $\mathrm{Im}(\lambda_m)<0$. In order to cope with this divergence, we divide the eigenvalues (and the corresponding eigenvectors) in two sets: λ^+, containing the eigenvalues with $\mathrm{Im}(\lambda_m)\geqslant 0$, and λ^-, containing the eigenvalues with $\mathrm{Im}(\lambda_m)<0$. The field $\psi(z)$ inside the periodic medium is then expressed as a function of the eigenvectors of the propagation matrix D as

$$\psi(z)=\sum_{\lambda_m^+} a_m^+\exp(ik_0\lambda_m^+ z)\psi_{pm}^+ + \sum_{\lambda_m^-} a_m^-\exp[ik_0\lambda_m^-(z-d)]\psi_{pm}^-. \tag{4.14}$$

In other words, we take as unknowns the amplitudes a_m^+ (a_m^-) of the exponentially decaying (growing) waves in $z=0$ ($z=d$); this ensures that finite values of the field inside the periodic medium correspond to finite values of the unknown amplitudes, thus removing any fictitious divergence in the solution of the equations.

The eigenvectors ψ_{pm} can be arranged in columns to form the $4(2M+1)\times 4(2M+1)$ square matrix T_p, where we choose to order them in such a way that the ψ_{pm}^+ come first. Then, from Eq. (4.14) we can write the values of the transverse fields on the two internal boundaries $z=0^+$ and $z=d^-$ as

$$\psi(0^+)=T_p\Lambda_0 a, \quad \psi(d^-)=T_p\Lambda_d a, \tag{4.15}$$

where a is the column matrix containing the amplitudes a_m^+ and a_m^- arranged in the same order as the corresponding eigenvectors in the matrix T_p, and the diagonal square matrices Λ_0 and Λ_d contain the propagation factors of the eigenvectors

$$\Lambda_0 = \mathrm{diag}(1, \ldots, 1, \exp(-ik_0\lambda_1^- d), \exp(-ik_0\lambda_2^- d), \ldots), \tag{4.16a}$$

$$\Lambda_d = \mathrm{diag}(\exp(ik_0\lambda_1^+ d), \exp(ik_0\lambda_2^+ d), \ldots, 1, \ldots, 1), \tag{4.16b}$$

where it is meant that in Λ_0, a 1 in the diagonal appears for each coefficient a_m^+, and a term $\exp(-ik_0\lambda_m^- d)$ appears for each corresponding a_m^-, and similarly for Λ_d.

The unknown amplitudes a can be found by imposing the continuity of the transverse field ψ across the boundaries. Precisely, the eigenvectors ψ_{em} of the external media can be arranged in columns in the square matrix T_e, that plays the same role as T_p. We order the eigenvectors in such a way that the forward solutions (i.e., those having $r_{em}>0$ for the propagating solutions and $\mathrm{Im}(r_{em})>0$ for the inhomogeneous waves) come first. Then, since for $z>d^+$ only forward solutions must be present, while for $z<0^-$ both forward (incident) and backward (reflected) waves are present, the values of the transverse fields on the external boundaries can be written as

$$\psi(0^-) = T_e(b_i + P_r b), \quad \psi(d^+) = T_e P_t b, \tag{4.17}$$

where b_i is the column vector containing the known amplitudes of the incident field, P_r (P_t) is the projection matrix on the subspace of the reflected (transmitted) waves; it is a diagonal matrix whose first (last) $2(2M+1)$ diagonal elements are equal to 0 (1), and the remaining are equal to 1 (0) and finally b is the column vector containing the amplitudes of the unknown transmitted [in the first $2(2M+1)$ rows] and reflected [in the last $2(2M+1)$ rows] plane waves. Imposing the continuity of Eqs. (4.15) and (4.17), with the help of the projection matrices, we get the system of $4(2M+1)$ linear equations

$$P_r T_e^{-1} T_p \Lambda_d a = 0, \quad P_t T_e^{-1} T_p \Lambda_0 a = b_i, \tag{4.18}$$

which determine the unknown amplitudes a of the internal proper waves as a function of the known amplitudes of the external incident plane waves b_i. We stress that in Eq. (4.18) only $4(2M+1)$ equations are present, in fact, in the first matrix equation, the first $2(2M+1)$ rows are identically zero, as are the last $2(2M+1)$ rows of the second matrix equation. We also note that the matrices T_e and T_p can be very easily inverted using the orthogonality relationships of the proper waves of the D matrix [15]. The transmitted and reflected amplitudes are then obtained from the remaining $4(2M+1)$ relationships

$$b_t \equiv P_t b = T_e^{-1} T_p \Lambda_d a, \quad b_r \equiv P_r b = P_r T_e^{-1} T_p \Lambda_0 a. \tag{4.19}$$

A comparison between the exact numerical calculations and the effective homogeneous model is shown in Fig. 2. There we plotted, as a function of the incidence polar angle ϑ_i, the rotation and the ellipticity of the transmitted beam for incident linear TE polarization. The curves corresponding to the homogeneous effective model were computed from the effective index of refraction obtained in the previous section using a standard Berreman matrix approach [16]. Since the transmitted beam is generally elliptically polarized, due to the birefringence of the medium, we define the polarization rotation as the angle of the major axis of the polarization ellipse of the transmitted beam with respect to the direction of the incident linear polarization; similarly, the ellipticity is defined as the ratio of the intensities of the transmitted beam along the major and minor axes, respectively. In order to minimize the effect of the birefringence, the incidence plane is taken parallel to the direction of the optical axis of the effective homogeneous medium; in Fig. 2 the distortion angle of the cholesteric is chosen in order to maximize the rotatory power, $\alpha=45°$ and the azimuthal incidence angle is then taken as $\varphi_i=\alpha=45°$. The other parameters are chosen to correspond to typical experimental values: Precisely, the index of refraction of the external media is $n_1=1.5$. The indices of refraction of the cholesteric are $n_0=1.5$ and $n_e=1.7$. The thickness of the cell is $d=10\lambda$, where $\lambda=2\pi/k_0$ is the vacuum wavelength. Finally, the pitch of the cholesteric is $2\pi/q=0.33\lambda$. From Fig. 2 it is apparent that the homogeneous effective model works quite well even far from its expected limits of validity: in fact with our parameters the optical anisotropy is quite large ($\epsilon_a=0.64$) and the pitch of the cholesteric is only one third of the light vacuum wavelength. We also note that, according to Fig.

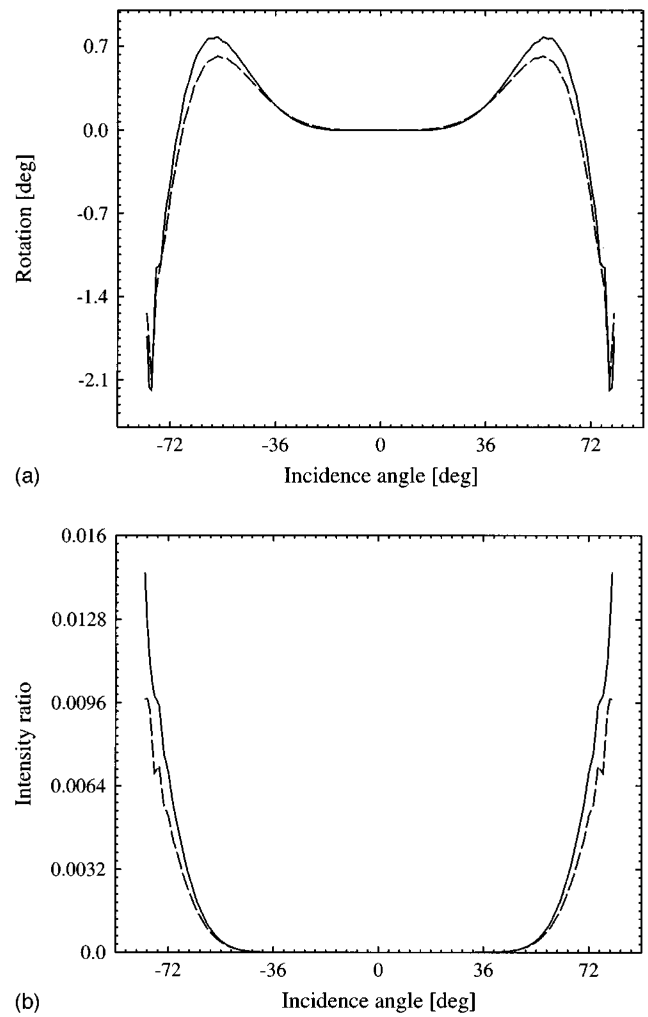

FIG. 2. (a) Rotatory power and (b) ellipticity of the transmitted light as a function of the incidence angle ϑ_i for TE incidence. Solid lines: effective homogeneous model; dashed lines: exact numerical calculation.

2(a), the rotatory power seems not to have a definite sign; actually, the apparent inversion of the rotatory power at large incidence angles is an effect of the birefringence that dephases the local ordinary and extraordinary waves as the light travels inside the sample. As it is apparent from Fig. 2(b), the ellipticity of the transmitted beam is always quite small: this is due to our choice of the incidence plane, which is parallel to the direction of the optical axis of the equivalent homogeneous medium. For light traveling along the optical axis of the equivalent homogeneous medium, the cholesteric would behave as an isotropic optically active material.

V. CONCLUSIONS

To conclude, we have obtained a general expression for the effective dielectric tensor of an arbitrary periodic dielectric medium having a short pitch with respect to the light wavelength. This expression is valid in the limit of small modulations of the dielectric tensor, as it accounts only for two-photon scattering events. By applying it to a particular case, we have shown that the effective medium displays a spatial dispersion related to an optical activity. This optical activity arises from the spatial correlations of neighboring points, i.e., from multiple scattering events, and scales as the ratio between the pitch of the periodic medium and the light wavelength. Higher-order corrections should be taken into account for large modulations of the dielectric tensor. However, the numerical simulations have shown that the effective dielectric tensor accounts for the main optical properties of the periodic medium also for relatively large dielectric anisotropies and for pitches up to one half of the light wavelength inside the medium. For pitches close to the light wavelength, new phenomena, such as selective Bragg reflections [7], occur, which cannot be accounted for within the model of a homogeneous optically active dielectric medium. Another situation in which the effective homogeneous model cannot properly describe the periodic structure is when the latter is sandwiched between boundary planes orthogonal to one of the axes of the reciprocal lattice. In the case of the cholesteric, this situation corresponds to having the helical axis perpendicular to the boundary planes. In this degenerate case, the transmission and reflection properties crucially depend on the position of the boundary planes, i.e. on the phase of the optical dielectric tensor. The homogeneous model only gives the optical properties of the finite sample averaged over all these possible phases.

The optical properties of an undeformed cholesteric liquid crystal can be computed analytically for light propagating along the helical axis [7]. A pseudo-rotatory-power is found that, for short helical pitches with respect to the light wavelength, scales as $(k_0/q)^3$, and therefore becomes rapidly negligible [9]. Here we have shown, instead, that in the presence of a flexoelectric distortion, the cholesteric acquires a true rotatory power, that scales as k_0/q, in the direction orthogonal to the direction of the unperturbed helical axis and of the applied electric field. Such a rotatory power, similar to what found in undeformed chiral smectic-*C* liquid crystals [9,17] has not yet been experimentally detected and could be important also for the realization of practical electro-optic devices [6]. Finally, we note that the expression (2.7) of the effective dielectric tensor can be easily generalized to aperiodic systems, by replacing the discrete sum over the reciprocal lattice points with a Fourier integral.

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