Modulations in the diffracted intensity in chiral smectic-*C* liquid crystals

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Recently [K. A. Suresh, Yuvaraj Sah, P. B. Sunil Kumar, and G. S. Ranganath, Phys. Rev. Lett. **72**, 2863 (1994)] the intensity and polarization features associated with the optical diffraction in chiral smectic-*C* liquid crystal were studied in the phase grating mode. In the computations it was found that the diffracted intensity, as a function of sample thickness, has modulations of different length scales. In this paper, using a perturbative approach, we show that these modulations are a consequence of a coupling between different orders of scattering. [S1063-651X(96)07608-8]

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The chiral smectic-*C* liquid crystal (Sm-*C**) has a helical stack of molecular layers. In each layer the molecules are uniformly tilted at a constant angle with respect to the layer normal. The optical periodicity of Sm-*C**, in general, corresponds to a 2π rotation of the index ellipsoid. For the light propagation perpendicular to the twist axis the medium acts as a one-dimensional phase grating resulting in the diffraction of light.

An earlier paper [1] presented some experimental and theoretical studies on the diffraction of light in Sm-*C** in this mode. There the experimentally observed intensity and polarization features were accounted for by applying the rigorous theory of Rokushima and Yamakita [2]. The computations indicated that in any diffraction order there are modulations in the diffracted intensity as a function of sample thickness. The period of these modulations depends on the order of diffraction, geometry of diffraction, and material parameters. In this paper, using a perturbation technique, we show that these modulations are the consequence of coupling between different orders of scattering.

The Rokushima-Yamakita (RY) theory has been discussed in detail elsewhere [2,3]. Here we present the theory briefly. We consider Sm-*C** with its twist axis along the y direction. When a linearly polarized plane wave front is incident in the z direction normal to the twist axis, the medium acts as a one-dimensional phase grating with the grating vector along the y direction. Here we assume the medium to be infinite in extent in the xy plane and to have a thickness d in the z direction. We can write [2,3] Maxwell's equations in the form

$$\frac{d\Psi(z)}{dz}=ik_0D\Psi(z), \tag{1}$$

where

$$\Psi(z)=\begin{bmatrix} e_x \\ h_y \\ e_y \\ h_x \end{bmatrix}.$$

e_x, e_y, h_x, and h_y are submatrices of the infinite column matrix $\Psi(z)$ at any point z and contain the various Fourier components of the transverse field. Also $k_0=2\pi/\lambda$, λ being the wavelength of the incident light. The propagation matrix D is an infinite square matrix [1,3].

According to the modal analysis of Galatola, Oldano, and Sunil Kumar [3], the solution of Eq. (1) is

$$\Psi(d)=\exp(ik_0dD)\Psi(0)=U\Psi(0). \tag{2}$$

Usually it is convenient to write $\Psi(z)$ in terms of the modes in the bounding isotropic media. We assume the bounding region to have the refractive index equal to $(n_1+n_2)/2$, i.e., the mean refractive index of the Sm-*C** medium. Here n_1 and n_2 are the principle values of the local index ellipsoid. Then $\Psi(z)$ in these regions can be written as

$$\Psi(z)=T\phi(z), \tag{3}$$

where $\phi(z)$ is the column vector containing the strength of different modes in the isotropic media arranged in a particular order and T is the matrix having the elements T_{lj}, which are the lth component of the jth eigenvector of the bounding isotropic media. The columns of the T matrix are arranged in the same order as in $\phi(z)$. Then, from Eq. (2), for a sample of thickness d we get

$$\phi(d)=T^{-1}UT\phi(0)=S\phi(0). \tag{4}$$

The matrix S is called the scattering matrix and it can yield the features of the diffraction pattern. The vector $\phi(0)$ is the sum of the reflected and incident components, while $\phi(d)$ is the transmitted component. Thus we write

$$\phi(0)=\phi_r+\phi_i, \tag{5}$$

$$\phi(d)=\phi_t. \tag{6}$$

According to the standard procedure [3], from Eqs. (4)–(6) we get

$$\phi_r=\mathcal{R}\phi_i,\quad \phi_t=\mathcal{T}\phi_i, \tag{7}$$

where $\mathcal{R}$ and $\mathcal{T}$ are the reflection and transmission matrices, respectively.

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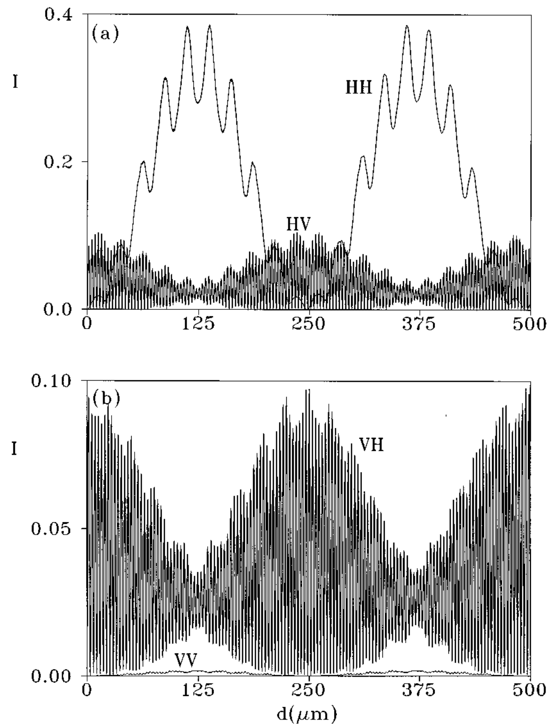

FIG. 1. Computed normalized intensity I for the first-order diffraction as a function of sample thickness for (a) the HH and HV geometries, and (b) the VH and VV geometries. Here, in the HV and VH geometries one can see modulations of periods 250, 25, and 4 μm. In the HH geometry modulations of periods 250 and 25 μm can be seen. The parameters used in the calculation are pitch $p=5$ μm, $n_1=1.535$, $n_2=1.715$, $\lambda=0.633$ μm, and tilt angle $\theta=18°$.

In Figs. 1(a) and 1(b) we have given the computed intensity of the first-order diffraction in different geometries using the transmission matrix in Eq. (7). One can notice that, in Fig. 1(a) for the HV geometry (i.e., the incident wave with the TM polarization and the diffracted wave analyzed for the TE polarization), the diffracted intensity has fine fringes of width 4 μm with intensity modulation of periods of about 25 and 250 μm. For the HH geometry (i.e., the incident wave with the TM polarization and the diffracted wave also analyzed for the TM polarization) the periods of the modulation are 250 and 25 μm. Also for the VH geometry [Fig. 1(b)], one may again notice such modulations in the intensity. Such effects are also present in second order. We use a perturbation technique [3] to account for these modulations. This technique is a simpler but approximate method of computing the diffracted intensity from the scattering matrix S. In this procedure we look at the propagation equation for the S matrix. To get this propagation equation, we start with Eqs. (3) and (1) to get

$$\frac{d\phi(z)}{dz}=ik_0G\phi(z), \tag{8}$$

where $\phi(z)=S\phi(0)$ at any point z and G is a new propagation matrix. Since T matrix is independent of z, G is equal to $T^{-1}DT$. From Eq. (8) we can get the propagation equation for the scattering matrix S as

$$\frac{dS}{dz}=ik_0GS. \tag{9}$$

The matrix G can be written as a sum of its diagonal matrix G_0 and an off-diagonal matrix g. G_0 contains the strength of the zeroth-order Fourier components of the dielectric tensor of an effective homogeneous anisotropic medium. The matrix g contains the strength of the higher-order Fourier components that arises due to the z dependence part of the dielectric tensor. It may be noticed that, mathematically, the above equation is rather analogous to the time-dependent Schrödinger equation [4] wherein $-i/\hbar$ is equivalent to ik_0 and the time evolution is equivalent to the thickness variation of the grating in the z direction. Based on this analogy, we use the time-dependent perturbation method of quantum mechanics [4] to solve this problem.

The first-, second-, and third-order scattering contributions to the amplitude of the diffracted light, $(A_{S_{\mathrm{I}}})_{lj}$, $(A_{S_{\mathrm{II}}})_{lj}$, and $(A_{S_{\mathrm{III}}})_{lj}$, from the lth and the jth scattered waves are given by

$$(A_{S_{\mathrm{I}}})_{lj}=ik_0\int_0^d dz\ \exp[ik_0E_j(d-z)]g_{lj}\exp(ik_0E_lz), \tag{10}$$

$$(A_{S_{\mathrm{II}}})_{lj}=(ik_0)^2\sum_k\int_0^d dz\int_0^z dz'\exp[ik_0E_j(d-z)]g_{jk}\times\exp[ik_0E_k(z-z')]g_{kl}\exp(ik_0E_lz'), \tag{11}$$

$$(A_{S_{\mathrm{III}}})_{lj}=(ik_0)^3\sum_k\sum_m\int_0^d dz\int_0^z dz'\int_0^{z'} dz''\times\exp[ik_0E_j(d-z)]g_{jk}\exp[ik_0E_k(z-z')]g_{km}\times\exp[ik_0(z'-z'')]g_{ml}\exp(ik_0E_lz''). \tag{12}$$

The summations are over all the scattered waves. The element g_{lj} of the g matrix represents the coupling between the lth and the jth-order diffracted waves. E_l and E_j are the eigenvalues of the corresponding waves of a particular polarization in the effective homogeneous anisotropic medium.

The matrix g can be treated as a perturbation over the matrix G_0. In our case the g matrix does not explicitly depend on z, so the integrals in Eqs. (10)–(12) can be analytically solved. They give, respectively, the first-, second-, and third-order scattering contributions to the amplitude of the diffracted wave for a particular polarization. Figure 2(a) shows the contributions of different orders $(A_{S_{\mathrm{I}}},A_{S_{\mathrm{II}}},A_{S_{\mathrm{III}}},\dots)$ of perturbations to the intensity of the first-order diffraction due to the coupling between zeroth-order diffracted TM and first-order diffracted TE polarizations, respectively (HV geometry). We see that the first-order perturbation $[|(A_{S_{\mathrm{I}}})_{0_H1_V}|^2]$ results only in the fine modulations of 4 μm. The third-order perturbation $[|(A_{S_{\mathrm{I}}})_{0_H1_V}+(A_{S_{\mathrm{II}}})_{0_H1_V}+(A_{S_{\mathrm{III}}})_{0_H1_V}|^2]$ leads to modulation with a larger period of 25 μm. Figure 2(b) shows the first- $[|(A_{S_{\mathrm{I}}})_{0_H1_H}|^2]$ and third- $[|(A_{S_{\mathrm{I}}})_{0_H1_H}+(A_{S_{\mathrm{II}}})_{0_H1_H}+(A_{S_{\mathrm{III}}})_{0_H1_H}|^2]$ order perturbation contributions to the intensity in the first-order diffraction due to the coupling between

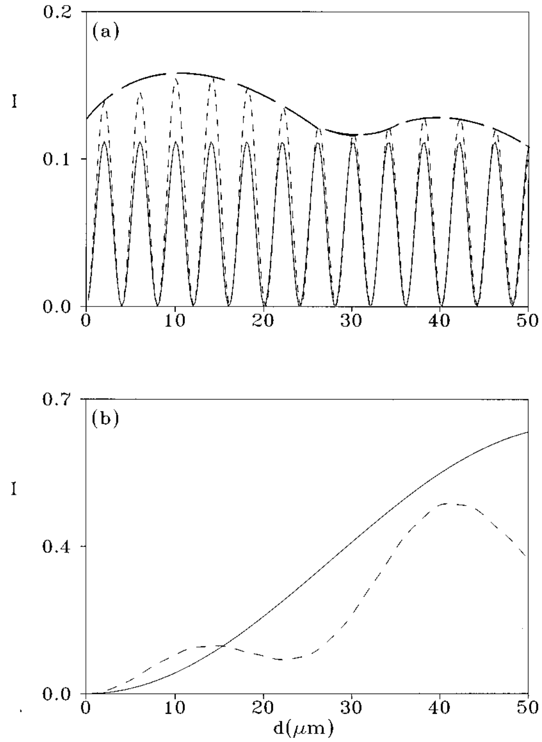

FIG. 2. Normalized intensity I of the first-order diffraction as a function of sample thickness computed using a perturbation theory for (a) the *HV* geometry and (b) the *HH* geometry. The full line represents the contribution from the first-order perturbation and the dashed line represents the third-order perturbation contribution to the diffracted intensity. In (a), a long-dashed line is drawn over the peaks of the dashed curve as a guide to the eye to show the 25-μm modulation.

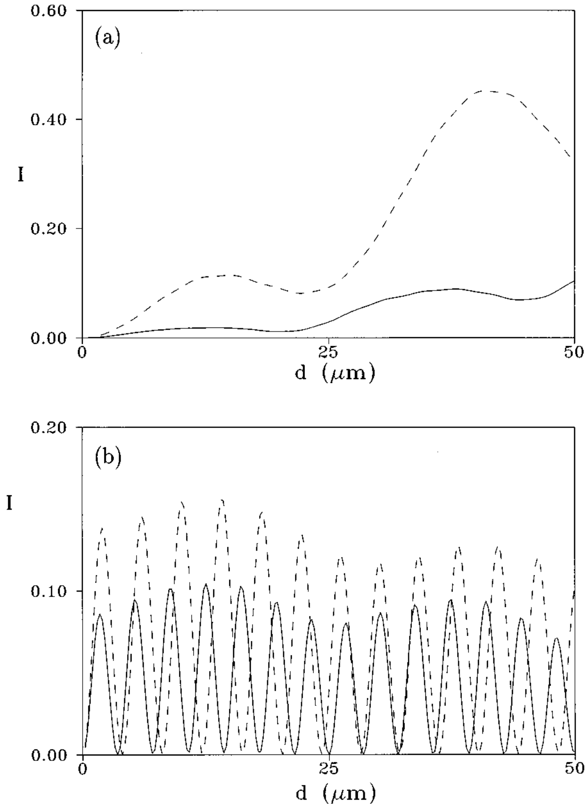

FIG. 3. Comparison of the intensities I obtained from the two theories as a function of sample thickness for (a) the *HH* geometry and (b) the *HV* geometry. The full line represents the calculation from the RY theory. The dashed line represents the calculation from the perturbation theory with contributions up to the third order to the diffracted intensity.

the zeroth-order TM polarization and the first-order TM polarization (*HH* geometry). Here we find that the first-order perturbation contribution corresponds to the modulation of the 250-μm period and the third-order perturbation has, in addition, the modulation of the 25-μm period seen in Fig. 1(a) for the *HH* geometry. For the parameters that we used, the contributions due to second-order perturbation are not very different from those due to the first-order perturbation.

We would like to mention that the perturbation technique does not incorporate the effect of reflectance. However, we find from the rigorous theory that the reflectance effects are negligible as can be calculated from the reflection matrix [Eq. (7)]. For the material parameters used, our calculations show that the contribution from diffraction to reflectance is 10^3 times weaker as compared to transmittance. We also find that the boundary effects do not alter the scale of the modulations present in the diffracted intensity.

In Fig. 3 we have given the intensities obtained from the RY theory and the perturbation theory for direct comparison. One can see that the perturbation calculations are valid only for thin samples. Also the discrepancy in intensity can be partly attributed to the fact that in the perturbation theory, contributions up to third order only have been considered. However, this technique gives a qualitative understanding of the modulations in the diffracted intensity obtained from the rigorous theory.

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