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The flexoelectro-optic effect in cholesterics

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The flexoelectro-optic effect in short-pitch cholesterics [1] is analysed in terms of applied electric field strengths and material parameters such as the two flexoelectric coefficients e_s and e_b and the three elastic constants. Starting from the free energy density of the uniformly lying-helix (ULH) configuration, including the flexoelectric polarization term, the equation describing the field-induced tilt angle of the bulk optic axis is derived. It is convenient to introduce the flexoelectric ‘anisotropy’ Δe as the difference between the splay and bend flexoelectric coefficients, hence defined by $\Delta e = e_s - e_b$. Our results then show that Δe is the essential material parameter controlling the sign and magnitude of the electrically induced tilt. In the region of linear approximation, the tilt is proportional to Δe and to the electric field E , and inversely proportional to the helical wave vector k , as well as the effective elastic constant ($K_{11} + K_{33}$). The individual values of the elastic constants K_{11} , K_{22} and K_{33} do have an influence on the magnitude of the effect, but not on its linearity. The Δe dependence conforms in the simplest way to the physical requirement that the flexoelectro-optic effect must be particularly pronounced in the case that e_s and e_b are of opposite sign.

1. Introduction

A short-pitch cholesteric with its helix axis unidirectionally aligned parallel to the cell glass plates (Uniformly Lying-Helix texture, ULH texture) acts macroscopically as a uniaxial birefringent plate with optic axis coinciding with the helix axis. An applied electric field, normal to the helix axis, induces a rotation of the optic axis in a plane parallel to the cell glass plates due to the flexoelectric effect, as demonstrated in 1987 by Patel and Meyer [1]. The angle of rotation of the optic axis is experimentally found to be linearly proportional to the applied field, giving a linear electro-optic effect between crossed polarizers when the ULH cell zero-field optic axis is set at an angle of 22.5° to the polarizer axis.

The most general form for the expression of flexoelectric polarization density in a nematic is given by [2]

$$\mathbf{P}_{\text{flex}} = e_s \hat{\mathbf{n}}(\nabla \cdot \hat{\mathbf{n}}) + e_b (\nabla \times \hat{\mathbf{n}}) \times \hat{\mathbf{n}} \quad (1)$$

where $\hat{\mathbf{n}}$ is the director and e_s and e_b are the flexoelectric coefficients for splay and bend deformations, respectively. In the case of e_s and e_b having opposite signs or very different magnitudes, a domain texture with alternating splay and bend deformations may occur in a nematic. In a cholesteric in the ULH texture, this periodic splay–bend deformation pattern is easily achieved just by rotating the director a certain angle around a direction parallel to the electric field and perpendicular to the helix axis; this field-induced director

rotation gives the effective tilt of the macroscopic optic axis. The build-up of splay–bend deformation at increasing electric field strength is illustrated in figure 1. The left hand side of the figure corresponds to cuts through the ULH cell showing the periodic splay–bend deformation plane for different applied field strengths, $0 < E_1 < E_2 < E_3$. The cuts are perpendicular to the plane of the cell and the periodicity of the field-induced deformation decreases with increasing field strength. The right hand side of the figure shows the plane of the ULH cell. The cuts (marked as lines) are then normal to the paper. The macroscopic optic axis is always perpendicular to the plane of deformation and the induced tilt of the optic axis increases with decreasing periodicity of the splay–bend pattern.

By introducing $e_b = -e_s = e_f$, in a way corresponding to the one-constant approximation for the elastic coefficients K_{ii} , and putting the elastic constants for splay and bend deformations as equal, $K_{11} = K_{33}$, the field-induced tilt, ϕ , of the optic axis can be expressed by [3–5]

$$\tan \phi = \frac{[e_f E - (K_{11} - K_{22})k \sin \phi]}{K_{22}k} \quad (2)$$

where k is the magnitude of the helical wave vector (including sign) of the cholesteric liquid crystal. In the one-constant approximation $K_{ii} = K$, one then gets the simple relation $\phi = \arctan(e_f p E / 2\pi K)$, where p is the cholesteric pitch, $2\pi/k$, or for $\phi \ll 1$

$$\phi = \frac{e_f p E}{2\pi K} \quad (3)$$

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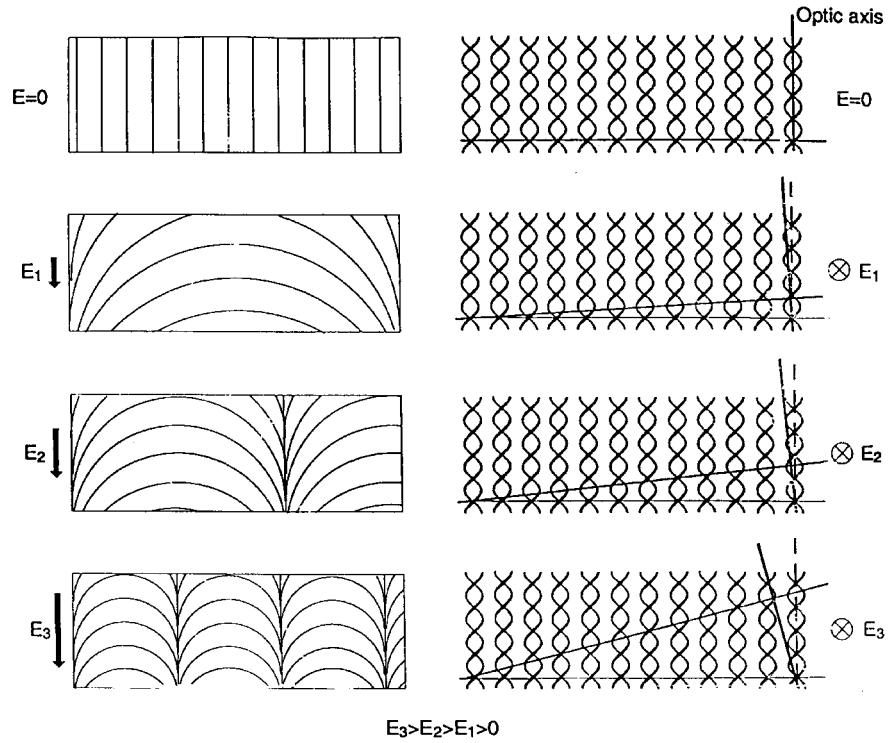


Figure 1. The field-induced periodic deformation pattern (left) and the corresponding induced tilt angle of the optic axis (right) in the ULH texture.

In earlier work [6] we have shown that this effect can give 100% modulation of the transmitted light intensity due to the field-induced tilt of the optic axis being capable of exceeding $\pm 22.5^\circ$. Such a large modulation, however, requires the dielectric anisotropy $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$ to be very close to zero in order to rule out the dielectric field-induced unwinding of the cholesteric helix for high electric fields [7]. The influence from the two individual flexoelectric coefficients has, so far, not been studied. Here we derive a more general expression for the field-induced tilt angle of the optic axis in the bulk, investigating the influence from the flexoelectric coefficients e_s and e_b separately, and we also discuss the influence from K_{11} , K_{22} and K_{33} on the effect.

2. Theory

In this section the expression for the tilt, ϕ , of the helical axis as a function of the applied electric field is calculated, without making any assumptions about the values of e_s and e_b . The sum of the average elastic and flexoelectric free energy density of the system is calculated and minimized with respect to ϕ . This sum, for a given spatial variation of the director, is given by

$$w = \frac{1}{2}K_{11}(\nabla \cdot \hat{\mathbf{n}})^2 + \frac{1}{2}K_{22}(\hat{\mathbf{n}} \cdot \nabla \times \hat{\mathbf{n}} + k)^2 + \frac{1}{2}K_{33}(\hat{\mathbf{n}} \times \nabla \times \hat{\mathbf{n}})^2 - \mathbf{E} \cdot \mathbf{P} \quad (4)$$

where \mathbf{P} is the induced flexoelectric polarization given by equation (1).

In order to keep the linearity of the electro-optic response, the dielectric coupling between the field and the molecules has to be ruled out by choosing a material with very low dielectric anisotropy. We therefore study the case of $\Delta\varepsilon = 0$. Tilting the helical axis by the angle ϕ , as depicted in figure 2, the director can be written as

$$\hat{\mathbf{n}} = (\cos kz \cos \phi, \sin kz, \cos kz \sin \phi) \quad (5)$$

where $k = 2\pi/p$ is the helical wavevector.

Substituting (5) into (1) and (4), the total free energy density of the system is given by

$$w^{\text{tot}} = \frac{1}{2}K_{11}k^2 \sin^2 \phi \sin^2 kz + \frac{1}{2}K_{22}k^2(1 - \cos \phi)^2 + \frac{1}{2}K_{33}k^2 \sin^2 \phi \cos^2 kz + (e_s \sin^2 kz - e_b \cos^2 kz)Ek \sin \phi \quad (6)$$

Averaging w^{tot} over the whole system, the average free energy per unit volume of the sample is

$$\langle w^{\text{tot}} \rangle = \frac{1}{2}(e_s - e_b)Ek \sin \phi + \frac{1}{4}(K_{11} + K_{33})k^2 \sin^2 \phi + \frac{1}{2}K_{22}k^2(1 - \cos \phi)^2 \quad (7)$$

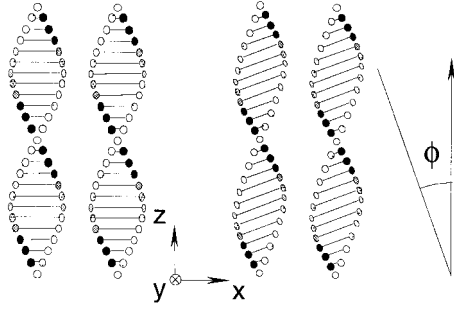


Figure 2. Coordinate system used in the calculations; $p > 0$ corresponds to a right-handed helix.

Before proceeding, we rewrite equation (7) into a dimensionless form. By doing so, the number of free parameters governing the behaviour of the solution of the equation is reduced to two. The dimension of the quantity $K_{22}k^2$ is energy per unit volume, and thus we introduce the dimensionless quantity \tilde{w} according to

$$\tilde{w} = \frac{\langle w^{\text{tot}} \rangle}{K_{22}k^2} \quad (8)$$

Further, two parameters α and β are introduced as

$$\alpha = \frac{\Delta e E}{2K_{22}k} \quad (9)$$

$$\beta = \frac{K_{11} - 2K_{22} + K_{33}}{2K_{22}} \quad (10)$$

We may formally look upon the difference, $e_s - e_b$, between the splay and bend flexoelectric coefficients as a flexoelectric ‘anisotropy’ Δe , being a characteristic parameter for each material. Note that this is in close analogy to the adopted definition of dielectric anisotropy $\Delta \epsilon = \epsilon_{\parallel} - \epsilon_{\perp}$, because e_s expresses the induced flexoelectric polarization along the molecular axis, whereas e_b expresses the corresponding polarization perpendicular to the long direction of the molecule. The flexoelectric anisotropy may be positive or negative and may in certain materials even change sign with temperature [8]. With this convention, the parameter α , which can adopt any value, reflects the strength of the applied electric field for a given set of material parameters. One can thus regard α as the ratio of the applied field E and a characteristic ‘field’ $E^* = 2K_{22}k/\Delta e$ of the system. Now, \tilde{w} can be expressed as

$$\begin{aligned} \tilde{w} &= \alpha \sin \phi + \frac{1}{2}(\beta + 1)\sin^2 \phi + \frac{1}{2}(1 - \cos \phi)^2 \\ &= \alpha \sin \phi + \frac{1}{2}\beta \sin^2 \phi + 1 - \cos \phi \end{aligned} \quad (11)$$

From the definition of β , it can be seen that $\beta > -1$, K_{ii} being positive definite. Furthermore, in the one-constant approximation $K_{11} = K_{22} = K_{33}$, the parameter β equals zero. The parameter ranges of α and β are thus given by

$$-\infty < \alpha < \infty, \beta > -1, \quad (12a, b)$$

In addition we conclude that

$$\beta < 0 \text{ if } K_{22} > \frac{1}{2}(K_{11} + K_{33}), \quad (13a)$$

$$\beta = 0 \text{ if } K_{11} = K_{22} = K_{33}, \quad (13b)$$

$$\beta > 0 \text{ if } K_{22} < \frac{1}{2}(K_{11} + K_{33}). \quad (13c)$$

Experimental data [9] seem to indicate that for most materials $K_{22} < K_{11}$, K_{33} , and hence we expect $\beta > 0$ to be the typical case. This will be adopted in the following.

Minimizing \tilde{w} given by equations (7) and (8) with respect to ϕ gives the equation

$$\tan \phi = -\alpha - \beta \sin \phi. \quad (14)$$

In the specific case that $e_s = -e_b$ and $K_{11} = K_{33}$, this conforms with the result derived by Patel and Meyer. In general, this equation has to be solved numerically and the solution for various values of the parameters α and β is discussed in the next section. Before proceeding to the numeric solution we note that, assuming the applied electric field to be sufficiently small for the assumption $\phi \ll 1$ to be valid, the solution is given by

$$\phi = -\frac{\alpha}{1 + \beta}. \quad (15)$$

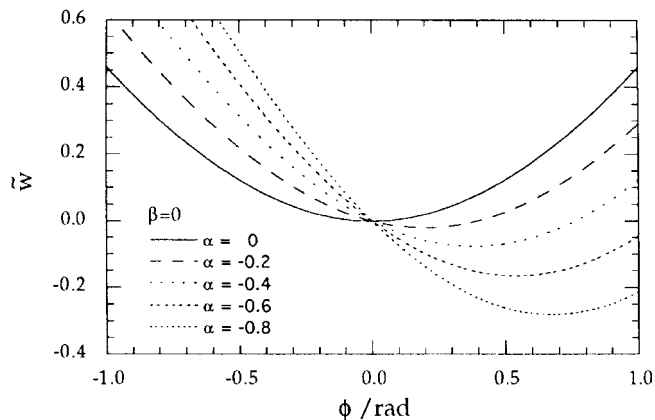
Using the definitions (9) and (10) of the parameters α and β , equation (15) reads

$$\phi = -\frac{\Delta e}{(K_{11} + K_{33})k} E, \quad (16)$$

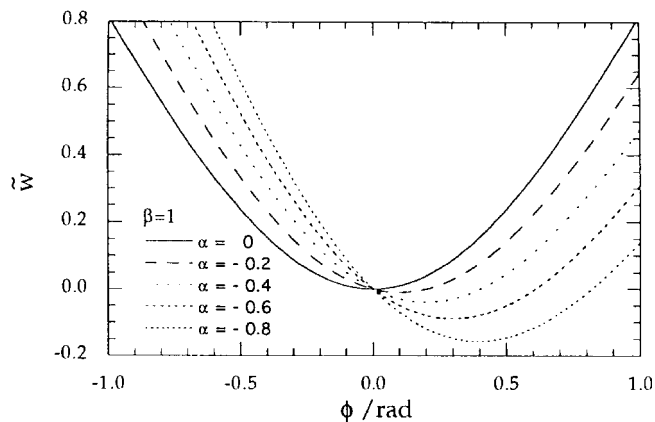
giving the slope of the tilt versus field curve in the linear regime. The twist elastic constant K_{22} has in general an influence on $\phi(E)$, equation (14), but for $\phi \ll 1$ only the splay and bend elastic constants (K_{11} and K_{33}) play significant roles in influencing the induced tilt angle, as seen from equation (16).

3. Results and discussion

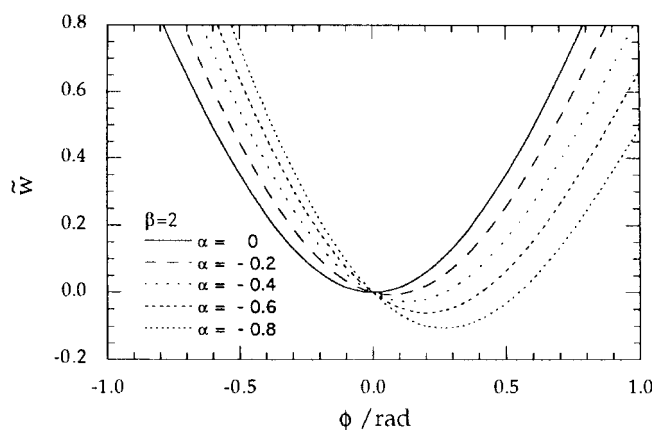
Figures 3(a), 3(b) and 3(c) show the dimensionless free energy \tilde{w} as a function of the tilt angle ϕ for different values of the parameters α and β , calculated from equation 11. Minimizing \tilde{w} by taking $\partial \tilde{w} / \partial \phi = 0$ gives ϕ as function of α , shown in figure 4. Since $\alpha = \Delta e E / K_{22}k$, this can be regarded as the field dependence of the optic axial tilt, because the parameter combination $\Delta e / K_{22}k$



(a)



(b)



(c)

Figure 3. Dimensionless free energy \bar{w} as function of α for (a) $\beta=0$, (b) $\beta=1$ and (c) $\beta=2$.

is a characteristic constant of the material. As also known from experiment, the tilt is a linear function of the applied field around $E=0$ and the linear range in tilt angle versus field is dependent on β .

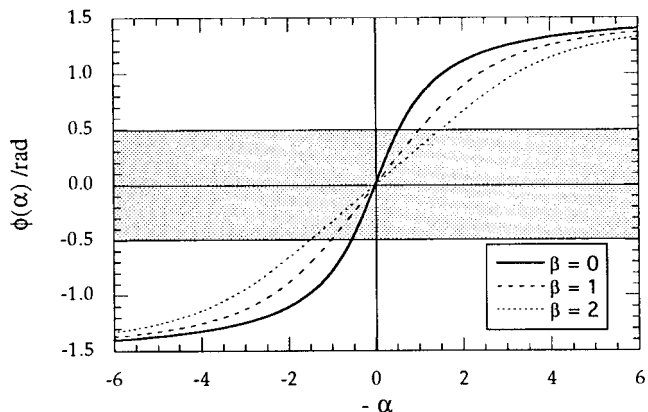


Figure 4. Field-induced tilt angle as a function of $\alpha = \Delta e E / 2K_2 k$. The relevant electro-optic interval $|\phi| < 0.5$ radians (roughly corresponding to 30 degrees) has been shaded in the figure.

From an application point of view, large tilt angles for low fields are desired. As we see from equation (16), the proportionality factor giving the induced tilt per unit of field is the parameter combination $\Delta e / Kk$, where $K \equiv K_1 + K_3$. We may thus increase this ratio by decreasing K and k , both parameters having a possible variation of about a factor of two, the lower limit for k being given by $k^{-1} < \lambda$, the wavelength of light (in practice, visible or infrared). This increase occurs at the cost of lower response speed, with the characteristic electro-optic response time being given by [4]

$$\tau = \frac{y_1}{Kk^2} \quad (17)$$

Considering that the flexoelectro-optic effect basically has a much higher response speed than that for the twisted nematic effect, that it is capable of giving a 100% modulation linear in E and that it has the added advantage (like the SSFLC effect) of being colour neutral, it would for many applications certainly be advantageous to optimize both parameters K and k (and in particular to lower the value of $K = K_1 + K_3$ as much as possible, while increasing τ in the same proportion) to achieve the full modulation amplitude for applied voltages as small as possible.

However, the more interesting option is to increase Δe . Very little is known today about this parameter and the relationship between molecular structure and flexoelectricity, and it is most important to develop international interest in this subject and in flexoelectricity in general. The measurement of Δe and of the separate flexoelectric coefficients stands out as a most important and timely research challenge.

The flexoelectric effect has nothing to do with chirality. The interest in cholesteric materials for the flexoelectro-

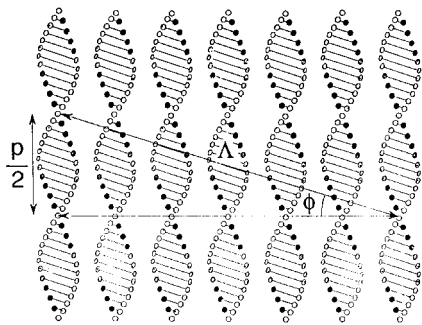


Figure 5. Periodicity Λ of the splay–bend deformation in the ULH texture under a uniform rotation ϕ of the molecules in the plane of the cell.

optic effect does not stem from the chirality of the medium, but from the fact that the twisted medium allows for a homogeneously space filling structure of splay–bend. The first hint of this fact came from Bouligand's remarkable analysis of biological tissue [10] in 1969 in which he showed that if a cut is made in a cholesteric structure, not perpendicular, but at an oblique angle to the helical axis, an 'arc pattern' (which is a periodic splay–bend) will be observed for the projection of the director field in the cut plane. The difference in the present case relative to that of Bouligand is that now the electric field turns the director into the oblique plane so that the director field itself, and not the projection, forms the stripe-like domain pattern (the 'arcs' of figure 1). In his first paper on flexoelectricity [2], Meyer observed that if e_s and e_b are of opposite signs, this could produce a strip-like domain pattern with alternating regions of splay and bend. Putting $e_s = -e_b = e_f$, $K_1 = K_3 = K$ and $\Delta\epsilon = 0$, he calculated the period Λ of the splay–bend deformation, i.e. the distance between consecutive splay deformations separated by a bend deformation, as a function of the applied electric field. He found it to be

$$\Lambda = \frac{\pi K}{e_f E} \quad (18)$$

It is interesting to compare this with the geometrical features of the present ULH texture under applied field, which is illustrated for $\phi \ll 1$ in figure 5. If we tilt the optic axis by an angle ϕ in the ULH texture (corresponding to $E \neq 0$), we see that the periodicity or the domain width Λ of the field-induced splay–bend flexoelectric deformation is given by

$$\sin \phi = \frac{p/2}{\Lambda} \quad (19)$$

Combining equations (18) and (19) gives

$$\sin \phi = \frac{e_f p E}{2\pi K} \quad (20)$$

which for $\phi \ll 1$ leads to the same expression for the tilt as stated before in equation (3).

4. Conclusions

We have studied the field-induced rotation of the optic axis in the bulk of a ULH cholesteric layer making no special assumptions about the values of the splay and bend flexoelectric coefficients e_s and e_b . We have included the influence from neither the surface alignment nor the surface regions. The magnitude of the tilt under a certain field E is shown to be linear in what we call the flexoelectric anisotropy $\Delta e = e_s - e_b$. This can be expected from the geometry of the problem (changing sign of Δe corresponds to a mirror splay–bend in a plane containing the electric field). The elastic coefficient $K = K_1 + K_3$ has the same influence on the induced tilt of the optic axis and on the response time, in the first approximation not perturbing the linearity of the electro-optic effect. Materials development would be desirable, with emphasis on the synthesis of highly flexoelectrically anisotropic materials for optimizing the effect and on measurement methods for separate determination of e_s and e_b or, eventually, e_s/K and e_b/K .

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