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On the flexoelectric effect in nematics

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Flexoelectricity is a general and fundamental phenomenon in liquid crystals. It describes the linear coupling between an applied electric field and gradients in the director field. Whereas flexoelectricity has for decades been regarded as only of academic interest, we think it is time to point out its considerable application potential, for instance in the case of the flexoelectro-optic effect, and to urge a revival of interest in the subject. As a result of long-time neglect, published data on flexoelectric coefficients are scarce and inconsistent, even with regard to the sign of the reported effect. In this paper we critically review the possible definitions of flexocoefficients in order to propose an international standard. We point out that the absence of such a standard obstructs the understanding of the physical basis, microscopically as well as macroscopically, of the effect, and leads to the introduction of nonsensical concepts like 'flexoelectric anisotropy'. Based on the only natural convention, we finally propose a simple method for measuring sign and magnitude of the effective flexoelectric coefficient which is the control parameter in electro-optic effects.

1. Introduction

In 1969 Robert Meyer first described the coupling between an electric field and gradients in the director field in nematic liquid crystals [1]. The phenomenon was later named flexoelectricity (rather than piezoelectricity as originally proposed) by P. G. de Gennes [2]. Basically, it consists of the following: if the molecules in addition to permanent dipoles also possess shape polarity, i.e. if they are drop-like or banana-like, there will be an induced polarization in a nematic subjected to splay or bend deformations, respectively. The effect is analogous to the piezoelectric effect in solids, but of a different origin. Whereas piezoelectric polarization is induced by strain (first order spatial derivatives), the flexoelectric polarization is due to curvature deformations (curvature strain, second order spatial derivatives). We may also illustrate another fundamental difference in the following way.

Let us begin with the piezoelectric effect and ask under what conditions a polarization may be induced if we apply a deformation to a medium. According to Neumann's principle, the deformed medium must be non-centrosymmetric because a property represented by an arrow, in this case the polarization \mathbf{P} which is a polar vector, does not have inversion symmetry. In order to see how the medium transforms under the deformation, we apply the Curie principle which may be stated as follows: a medium subjected to an external action changes its point symmetry so as to preserve only the symmetry elements common with those of the influencing action. In the case of the piezoelectric effect the external action is a stress. The symmetry of stress is mmm with three mirror planes, three two-fold axes and a centre of symmetry as symmetry elements. Therefore, according to the Curie principle, the centre of symmetry (centre of inversion) will always be retained in the deformed medium if the unstrained medium has a centre of symmetry. It follows that the piezoelectric effect can appear only in non-centrosymmetric media. It may be further pointed out that lack of inversion symmetry is only a necessary condition, not a sufficient condition, for piezoelectricity, due to other existing symmetries of the medium which together may have an equivalent effect as a centre of inversion [3].

The situation is very different for the flexoelectric effect in nematics. The unstrained nematic is centrosymmetric which is expressed by the full rotational symmetry around the director **n**, together with the statement that $\mathbf{n} \rightarrow -\mathbf{n}$ is a symmetry operation of the medium. But now the symmetry of the deformation is instead very different and, in particular, none of the three basic curvature strains of splay, twist and bend, in which we can compose any bulk deformation, has a centre of symmetry, cf. figure 1. In principle, therefore, each of them could be associated with a local polarization. Looking more closely we see that twist and bend preserve the local director inversion invariance. Hence, any polarization connected with these deformations has to be

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Figure 1. Symmetry of the three basic bulk deformations of splay, twist and bend. The drawings express only the symmetry features and not the fact that the local material density is preserved.

perpendicular to the director, $\mathbf{P}^{\perp}\mathbf{n}$. In the splay deformation, this local inversion symmetry $(\mathbf{n} \rightarrow -\mathbf{n})$ is broken; thus in this case a polarization along the director is admitted, $\mathbf{P} \| \mathbf{n}$, whereas the deformation has a rotational symmetry around \mathbf{n} excluding any perpendicular component. Finally, we see that the twist is a particular case: because there is always an axis perpendicular to the helix axis (for a fully developed helix, as in a cholesteric, there are an infinite number of such axes) which is a two-fold rotation axis, any polarization along the twist axis has to be zero. Hence, the twist is not, in contrast to splay and bend, connected with a local polarization of the medium.

The flexoelectric effect is thus compatible with the centrosymmetric character of the nematic and a characteristic of liquid crystals in general. It is connected to splay and bend deformation and has nothing to do with chirality. In contrast, local polarization in a non-deformed liquid crystal which obeys the $n \rightarrow -n$ invariance requires chirality, as exemplified by tilted chiral smectics, for instance smectic C*.

2. Sign conventions

According to the above, total flexoelectric polarization **P** may in its simplest form be written as

$$\mathbf{P} = e_{\rm s} \mathbf{S} + e_{\rm b} \mathbf{B} \tag{1}$$

where **S** and **B** are vectors describing the splay and bend deformations. **S** and **B** not only describe the magnitude of splay and bend but, since they are vectors, relate the deformations to certain directions. The flexoelectric coefficients for splay and bend, e_s and e_b , are proportionality constants describing the relation with a certain deformation and the resulting polarization. With S = $\mathbf{n}(\nabla \mathbf{n})$ and $\mathbf{B} = (\nabla \times \mathbf{n}) \times \mathbf{n}$ we obtain the expression

$$\mathbf{P} = {}_{e_{s}}\mathbf{n}(\nabla \mathbf{n}) + {}_{e_{b}}(\nabla \times \mathbf{n}) \times \mathbf{n}, \qquad (2)$$

as originally given by Meyer. The coefficients e_s and e_b may both be positive or negative and the meaning of either sign is implicit in equation (2). If e_s and e_b are

taken as positive, the splay contribution to **P** is along and in the sense of the director, whereas the bend contribution is perpendicular to the director and directed towards the centre of curvature. However, the almost universally adopted convention for writing a bend deformation, e.g. in the Oseen–Frank expression for the elastic energy, is $\mathbf{n} \times (\nabla \times \mathbf{n})$. With this convention we instead arrive at the alternate expression

$$\mathbf{P} = e_{s}\mathbf{n}(\nabla \mathbf{n}) + e_{b}\mathbf{n} \times (\nabla \times \mathbf{n})$$
(3)

corresponding to a different meaning for the interpretation of a positive e_b . The sign convention is important, because in most experimental cases it is the difference $e_{\rm s} - e_{\rm b}$, in the sign convention based on equation (2), that is responsible for the observed phenomenon. This has led many authors, including ourselves [4, 5], to introduce the concept of 'flexoelectric anisotropy' $\Delta e \equiv e_{\rm s} - e_{\rm b}$, in analogy with dielectric anisotropy $\Delta \varepsilon =$ $\varepsilon_{\parallel} - \varepsilon_{\perp}$. As we will show below, however, this concept is inconsistent or contradictory or, at least misleading, and therefore should be abandoned. Moreover, the fact that it is experimentally very hard to measure the coefficients $e_{\rm s}$ and $e_{\rm b}$, together with the ambiguity in their sign, has given rise to a considerable confusion in the values of e_s and $e_{\rm b}$ (or $e_{\rm s}/K$ and $e_{\rm b}/K$ which are the quantities normally measurable; K is here an elastic constant). The lack of uniqueness and the fact that the definition used is seldom clarified in published articles make us suggest that one convention should be adopted, i.e. that by convention it should be determined whether P should be defined according to equations (2) or (3). This would allow a meaningful discussion of e_s and e_b values related to molecular properties and also to a simple method for determining their sign, as well as their values, as we will propose in the following.

As an illustration of the present state, the sign of the flexoelectric bend coefficient for MBBA is reported as both positive and negative in the literature. P. G. de Gennes [2] uses the definition given by expression (2) when he describes the flexoelectric effect, but then refers to the measurement of e_b for MBBA by Schmidt *et al.* [6], not being aware of the fact that they use (3) in the definition of the sign of e_b . As for Meyer, he used expression (2) in the original paper, but in later work [7] has adopted a sign convention corresponding to (3).

Despite the difficulties of measuring flexoelectric coefficients, the reason for ambiguous signs appearing in the literature is often just the different ways that the authors implicitly define the bend coefficient e_b . In this work we want to set a standard in the convention for the sign of the flexoelectric coefficients. We begin by studying two examples in §3 and §4—in fact the two most important experimental geometries—in order to

show that when the contributions of splay and bend are combined, the effect would be counter-intuitive in the case of definition (2), whereas it comes out very naturally if (3) is adopted. The first example is the hybrid-aligned nematic cell and the second is the flexoelectro-optic effect in cholesterics and, as a matter of fact, we let these two examples be the basis for the convention to be adopted.

Based on this convention we also propose a simple way to measure the sign of the flexoelectric polarization in nematic liquid crystals.

3. The flexoelectric polarization density

As already mentioned, the nature of flexoelectricity makes \mathbf{P}_{splay} parallel to the local director \mathbf{n} and \mathbf{P}_{bend} perpendicular to it, i.e. \mathbf{P}_{splay} is at right angles to \mathbf{P}_{bend} in every point of a deformed nematic. The flexocoefficients e_s and e_b are the only material parameters involved in equation (1) and therefore they themselves or a combination of them often describe the direction and magnitude of not only the flexoelectric polarization density, but also the total flexoelectric polarization of a nematic sample under deformation. If we take the cases of pure splay and bend separately and write

$$\mathbf{P}_{\text{splay}} = e_{\text{s}} \mathbf{S}, \\ \mathbf{P}_{\text{bend}} = e_{\text{b}} \mathbf{B}$$

we see that if the local polarization density is parallel/ antiparallel to the deformation vector, then the corresponding coefficient should be positive/negative. Thus, the sign of the flexoelectric coefficients is directly coupled to the definition we choose for the directions of the deformation vectors S and B. This leaves us with four possible ways to combine the directions and signs of the deformation vectors and the flexoelectric coefficients (figure 2). For a pure splay (bend) deformation described by the vector S(B), the induced polarization P is given for a certain type of molecular shape polarity. The sign of the corresponding flexoelectric coefficient is then chosen from the direction of the deformation vector. We may now choose any one of the four combinations $(S_I, B_I), (S_I, B_{II}), (S_{II}, B_I)$, and (S_{II}, B_{II}) for describing the total flexoelectric polarization density. The combination normally used in literature is (S_I, B_{II}) , but as we shall see it would be advantageous to change to (S_I, B_I) .

4. The in-plane polarization in a hybrid-aligned nematic cell

Let us first consider the hybrid-aligned nematic (HAN) cell shown in figure 3. Assuming strong boundary conditions, the director will be subjected to splay-bend deformation going from pure bend at the hometropic side to pure splay at the planar side. The deformation will give a flexoelectric polarization in the volume of the







Figure 2. The four possible definitions of sign for \mathbf{S} , \mathbf{B} , e_s and e_b .

sample. Let us, in particular, look at the net flexoelectric polarization in the plane along the cell glass plates, i.e. horizontally in the figure. In figures 3(a) to 3(d) are shown four cells in which the possible flexoelectric polarizations in the pure splay and bend regions are inserted. In figures 3(a) and 3(d) the polarization contributions from the splay and bend regions are pointing in the same direction and hence the total flexoelectric polarization is maximized, whereas in figures 3(b) and 3(c) they point in different directions, so that the total polarization is lowered or even cancelled. Dozov et al. [8] have used this geometry and, on applying an electric field perpendicular to the director and parallel to the cell plane, they deflect the director field by acting on the flexoelectrically induced polarization. Their theoretical analysis shows that a twist $\Delta \phi$ is induced around an axis normal to the cell plates with $\Delta \phi$ proportional to $e_{\rm s} - e_{\rm b}$, and this is also confirmed by the experiment. This means that the splay and bend contributions to the observed effect are added constructively if e_s and e_b are of opposite sign, and destructively if they have the same sign. In other words, in accordance with the expression (2), which is the basis of the work of Dozov et al. as well as of later work [5], the observed effect due to the flexoelectric polarization in (2) is maximized, not minimized when e_s and e_b have the opposite sign. Such a convention, in our opinion, is not convenient and rather obstructs the physical understanding of the effect, microscopically as well as macroscopically. Instead we suggest that when the splay and bend contributions work in the same sense, i.e. reinforcing the effect, the flexoelectric coefficients for splay and bend should have the same sign, and when the contributions are working against each other, the flexo-coefficients should have opposite sign. Comparing with figure 2 we conclude that this leaves us only with the two possibilities (S_I, B_I) , and (S_{II}, B_{II}) to be used for describing the flexoelectric polarization density in a nematic. However, we still have no reason for choosing one above the other, and we must therefore define what should be the positive direction of the splay-bend deformation. One way to do this is to study the mechanism of the field-induced rotation of the optic axis in the flexoelectro-optic effect in cholesteric liquid crystals. This is an example of the inverse flexoelectric effect, where an applied electric field polarizes the medium, leading to a flexoelectrically induced deformation.

5. The flexoelectro-optic effect in cholesterics

For certain materials, especially in the case where the molecular shape is a combination of drop and banana shape and the net dipole is directed as in for instance figure 4, the application of an electric field may induce a periodic splay-bend deformation pattern schematically



Figure 4. Molecules promoting periodic splay-bend deformation when subjected to an electric field.

described in figure 5. (The material can of course also be a mixture of drop- and banana-like molecules.) In reality, the formation of such a periodic pattern requires rotation of the director and the creation and motion of defects, and therefore has not been observed starting from a uniformly aligned nematic.

Starting from a cholesteric liquid crystal, however, the intrinsically helicoidal medium allows for a space filling structure of splay-bend if the molecules are rotated around an axis perpendicular to the helix axis. In 1969, Bouligand, showed [9] that if a cut is made in a cholesteric structure, at an oblique angle to the helical axis, an 'arc pattern' (which is a periodic splay-bend) will be observed as the projection of the director field in the cut plane. Therefore, when the molecules become tilted relative to the helix axis, the director field changes from uniform to arc-shaped, connected with a polarization density $\mathbf{P}\neq 0$, with $\hat{\mathbf{P}}$ along the tilt axis. Thus, in a cholesteric, a periodic flexoelectric splay-bend deformation is formed with no threshold under application of an electric field E perpendicular to the helix axis, causing the director to rotate through a certain angle ϕ around the field axis and making the director lie in the plane of the 'Bouligand cut', cf. figure 6. This effect was first established by Patel and Meyer [7].

Consider a short-pitch cholesteric with its helix unidirectionally aligned in the plane of a sandwich cell (uniformly lying helix, ULH, structure). For E=0, the optic axis is collinear with the helix axis. For $\mathbf{E}\neq 0$, the macroscopic optic axis, being everywhere normal to the director, swings out or tilts at the same angle ϕ as the director, in the plane of the cell and the tilt is driven by the flexoelectric coupling. The tilt of the optic axis is experimentally and theoretically found to be linear in the applied field, and the sign of ϕ for a fixed direction of \mathbf{E} is determined by the handedness of the pitch and the flexoelectric coefficients of the medium. Theoretical analysis shows that the tilt is proportional to the sum of the flexoelectric coefficients if the conventions (S_I, B_I), or (S_{II}, B_{II}) are used, and proportional to the difference between the flexoelectric coefficients if (S_I, B_{II}), or (S_{II}, B_I) are used. The total induced polarization (splay and bend contributions added vectorially) is parallel to the applied field and the tilt of the optic axis is maximized when the splay and bend deformations 'work together'. Therefore, as in the HAN cell, again it seems more appropriate



Figure 6. Field-induced rotation of the macroscopic optic axis, which is everywhere normal to the director, in a uniformly lying helix (ULH) structure. The figure is drawn for $\tilde{e} > 0$. The grey plane in the figure is the Bouligand cut which is the plane of the director for $E \neq 0$. The director field has a characteristic periodic splay-bend deformation.

to use the convention that the flexoelectric coefficients have the same sign when splay and bend reinforce the effect, and different signs when they act against each other. Hence, again (S_I, B_I), or (S_{II}, B_{II}) is the convention to be used. Let us now define the total flexoelectric coefficient \tilde{e} in the following way:

For a positive rotation ϕ of the optic axis about the direction of a positive field **E** for a positive wavevector **k** we have a positive total flexoelectric coefficient $\tilde{e} = (e_{\rm s} + e_{\rm b})$.

Using the right-hand rule we may say that for a *right-handed helix with* $\tilde{e} > 0$, *the optic axis will rotate in the direction of the index finger if we apply the electric field in the direction of our right thumb.* Alternatively, we may say that for $\tilde{e} > 0$, the wave vector **k** rotates into the new

direction \mathbf{k}' in the same sense as a right-handed screw rotates when it propagates in the direction of **E**. This particular situation is shown in figure 6 where the corresponding Bouligand cut is shown (see also figure 7).

We may now conclude from figures 2 and 6 that the convention to be used is (S_I, B_I) . The splay and bend vectors that give the appropriate directions of the deformations are then

$$\mathbf{S} = \mathbf{n}(\nabla \mathbf{n}), \tag{4}$$
$$\mathbf{B} = \mathbf{n} \times (\nabla \times \mathbf{n})$$

and hence, the flexoelectric polarization density should be written as

$$\mathbf{P} = e_{s}\mathbf{n}(\nabla \mathbf{n}) + e_{b}(\mathbf{n} \times \nabla \times \mathbf{n}).$$
(5)



Figure 7. Relation between E and ϕ for k > 0 and $\tilde{e} =$ $e_{\rm s} + e_{\rm b} > 0$. Positive field is into the paper.

Compared with the more common sign convention described by expression (2), we have in (5) merely changed the sign of the flexoelectric bend contribution. The proposed convention for e_b and **B** is, as already mentioned, the one used by Schmidt et al. where they also use the helpful picture that for $e_b > 0$, \mathbf{P}_{bend} is in the direction of the arrow if the bend deformation is considered the bow. Their conclusion that $e_b > 0$ for MBBA then of course means that if they had used the definition (2) they would have obtained the result that $e_b \leq 0$. The convention (5) is also used by Meyer and Patel in their first paper on the flexoelectro-optic effect.

6. The normal case of only one flexoelectric parameter

Except for rare cases, es and eb cannot be separated in the observed physical effects. As mentioned already, these typically depend only on one single flexoelectric parameter. Such is the case both in the HAN structure and in the cholesteric structure. With the convention codified in expression (2), this parameter comes out as the flexoelectric 'anisotropy' Δe . For instance, the tilt of the optic axis in the flexoelectro-optic effect is for $\phi \ll 1$ given by [4]

$$\phi = \frac{\Delta e}{(K_{11} + K_{33})k} E. \tag{6}$$

As we have shown, however, the interpretation of the flexoelectric interaction on a molecular level cannot give any other result than that this convention is completely unnatural. The interpretation of Δe as anisotropy is made on the very superficial basis that e_s is 'along' the director, like ε_{\parallel} , and that $e_{\rm b}$ is perpendicular, like ε_{\perp} . In fact, e_s and e_b , as they relate to different deformations, are not really compatible, and besides, each of them already represents a very strong kind of anistropy, but of completely different character and origin from the different anisotropies which are characteristics of the director symmetry of a nematic. With the convention (3), the question of 'anisotropy' never arises, because the observed effects depend on $e_s + e_b$, the sum of the coefficients. In fact, the most natural interpretation of all is found if we then introduce the *average value* e of e_s and e_b . With

$$e \equiv \frac{1}{2} \left(e_{\rm s} + e_{\rm b} \right) \tag{7}$$

the expression (6) for the tilt of the optic axis then takes the form

$$\phi = \frac{eE}{Kk} \tag{8}$$

where K is the average value of the splay and bend elastic constants

$$K \equiv \frac{1}{2} (K_{11} + K_{33}). \tag{9}$$

Also with these parameters, the dynamics of the flexo-



Figure 8. The linear electro-optic response in the mixture CF1 at three temperatures. The sign reversal of the response at $T \approx 26^{\circ}$ C reveals a sign change of e.

electro-optic effect obtain their most simple expressions. As an example, the characteristic response time of the axis deflection turns out to be

$$\tau = \frac{\gamma}{Kk^2} \tag{10}$$

where γ is a characteristic viscosity.

7. A simple method for measuring the sign of *e* for nematic liquid crystals

In order to get a linear electro-optic response, for the flexoelectro-optic effect described in §4, the ULH cell must be placed between crossed polarizers at an angle of 22.5° between the zero-field optic axis (the uniformly lying helix axis) and the polarizer axis. The sign of ϕ is easily observed with a two-channel oscilloscope displaying the values of the applied field and the transmitted light intensity, and hence the sign of the total flexoelectric coefficient e is directly determined if we know the handedness (the sign of the helical wavevector k) of the material. This is even possible, for instance, in the cholesteric mixture Roche CF1 showing a very small flexoelectro-optic response indicating that e is close to zero. The material is right-handed and has a constant pitch $p \approx 0.3 \,\mu\text{m}$ over the whole cholesteric range $\approx 0^{\circ}\text{C}$ to $\approx 50^{\circ}$ C. Yet, studying the weak response as a function of temperature we observed a sign reversal in the electrooptic response in the middle region. In figure 8 is shown the oscilloscope screen displaying the applied voltage (upper curves) and the transmitted light intensity (lower curves) for three different temperatures, 10°C, 26°C and 50° C; in (a) the electro-optic signal is out of phase, in (b) it is zero and in (c) it is in phase with the applied field. Since the handedness of the helix does not change between the three temperatures, the phase shift of the electro-optic response at $T \approx 26^{\circ}$ C is due to a sign reversal of the flexocoefficient e. The noise is due to the high amplification of the very low signal amplitude. In figure 8, the set-up is arranged in such a way that e > 0corresponds to an electro-optic signal out of phase with the applied voltage and e < 0 gives a signal in phase. Hence we may conclude that for

$$\begin{array}{ll} T < 26^{\circ} \text{C}, & e > 0 \\ T = 26^{\circ} \text{C}, & e \approx 0 \\ T > 26^{\circ} \text{C}, & e < 0. \end{array}$$

The cholesteric mixture CF1 is a multicomponent mixture designed to give the same wavelength of the selectively reflected light over the whole N* range, and we do not know the structures of the constituent molecules. Hence, we cannot draw any conclusions about the relation between molecular structure and flexoelectric properties from this experiment. However, we have shown that certain liquid crystals may exhibit a sign reversal of the flexoelectric coefficient e with temperature.

One even more interesting option is to use a mixture like CF1 with $e\approx 0$ as a cholesteric matrix to which we may add nematics of known molecular structure and study the sign of the flexoelectro-optic response originating now from the flexoelectric properties of the added nematic. In a preliminary study we made two mixtures containing 90 wt % of CF1 and 10 wt % of MBBA and 8 OCB, respectively, and the electro-optic response in the ULH texture reveals that e is positive for MBBA and negative for 8 OCB. The electro-optic response for the two mixtures is shown in figure 9. The set-up is here arranged in such that a way e > 0/e < 0 gives in phase/out





Figure 9. Electro-optic response for mixtures of CF1/MBBA (*a*), and CF1/8 OCB (*b*). The response indicates that *e* has a different sign for MBBA and 8 OCB.

of phase response. The results are consistent with known results from the literature. The contribution from the very low flexocoefficient for CF1 (also far from the sign reversal point) is negligible compared with the flexoelectric strength of the nematic additives, and therefore the sign of *e* for the mixtures should be given merely by the sign of *e* for the nematic under study.

A slight modification of the method would be to create the necessary helical structure by means of adding a chiral dopant directly to the nematic instead of inserting the nematic into a cholesteric matrix as we have done. It should not matter whether the pitch is not very short, as long as we can determine the sign of the rotation of the director in the ULH structure. With knowledge of the elastic constants and the pitch of the chiralized system probably also the magnitude of eshould be determinable from the slope of the field versus field curve. Since the flexoelectro-optic effect is fast, $\tau_{\rm r} \approx 10-100 \,\mu$ s, we can work with a.c. fields at frequencies high enough such that there is no ionic screening of the flexoelectric polarization, a well-known problem when measuring flexoelectric coefficients.

8. Conclusions

In the above, we have argued the case for the adoption of a standard convention of sign for the flexoelectric coefficients. The suggested way of writing the flexoelectric polarization density

$$\mathbf{P} = e_{s}\mathbf{n}(\nabla \mathbf{n}) + e_{b}(\mathbf{n} \times \nabla \times \mathbf{n})$$

gives as a result that when e_s and e_b have the same sign

the studied effect is particularly large and is a function of the total flexoelectric coefficient $e_{\rm s} + e_{\rm b}$, which can equally well be described as the mean value $e \equiv \frac{1}{2} (e_s + e_b)$. We propose a simple method for determining the sign and possibly also the magnitude of e for different materials. Determination of the sign and magnitude of e for a number of nematics with different molecular structures is being carried out and results will be published elsewhere.

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