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Comment on “The limits of flexoelectricity in liquid crystals” [AIP Advances 1, 032120 (2011)]

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In their paper, using the principle of energy conservation, F. Castles, S. M. Morris, and H. J. Coles [AIP Advances 1, 032120 (2011)] establish inequalities involving the elastic and dielectric constants and flexoelectric coefficients of liquid crystals. They then argue that recently measured values of flexoelectric coefficients by Harden *et al.* do not obey these inequalities, hence they violate the principle of energy conservation. In this comment, we point out that in their calculation, Castles *et al.* use an inappropriate value for an elastic constant, hence their conclusions, predicated on the outcome of this calculation, are not justified. *Copyright 2013 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License.* [<http://dx.doi.org/10.1063/1.4774285>]

In their paper, the authors of Ref. 1 set out to investigate the “flexoelectric conversion of mechanical to electrical energy in nematic liquid crystals. . . using continuum theory”. They argue that “since the electrical energy produced cannot exceed the mechanical energy supplied, and vice-versa, upper bounds are imposed on the magnitudes of the flexoelectric coefficients in terms of the elastic and dielectric coefficients.” The authors then go on to derive the inequalities which impose these upper bounds. Finally, they conclude that recently reported measurements² violate these inequalities, and hence they are irreconcilable with the continuum theory of nematics and with the principle of energy conservation.

The purpose of this comment is not to address the bounds on the magnitudes of flexoelectric coefficients of nematics nor the validity of the experimental results reported in Ref. 2. It is simply to point out that the logic of the authors argument is flawed, and hence their conclusions do not hold.

The authors’ argument consists of three assertions:

(For simplicity, I focus here only on the splay deformation.)

1. The mechanical energy required, per volume, to distort the nematic, is

$$F_d = \frac{1}{2} \frac{K_1^D}{\rho^2} \quad (1.1)$$

where ρ is the radius of curvature and K_1^D is the “elastic constant K_1 at constant electric displacement”.

2. The flexoelectric energy density is

$$F_E = \frac{e_1^2}{2\varepsilon_{\parallel}^a \rho^2} \quad (1.2)$$

where e_1 is the splay flexoelectric coefficient and $\varepsilon_{\parallel}^a$ is the dielectric permittivity “parallel to $\pm \mathbf{n}$ ” where \mathbf{n} is the nematic director.

3. Energy conservation requires that $F_E \leq F_d$, hence if

$$e_1^2 > \varepsilon_{\parallel}^a K_1^D \quad (1.3)$$

the principle of energy conservation is violated.



This argument is sound, provided that Eq. (1.1) indeed describes the work per volume required to produce the deformation; that is, that K_1^D is the elastic constant which incorporates the energy cost of the flexoelectric polarization.

Meyer, in his original paper,³ gives the “electric enthalpy”

$$H_E = \frac{1}{2}k_{11}S^2 - e_{1z}\mathbf{E} \cdot \mathbf{S} - \frac{1}{2}\varepsilon_{zz}(\mathbf{E} \cdot \mathbf{L})^2 \quad (1.4)$$

and the electric displacement

$$\mathbf{D} = e_{1z}\mathbf{S} + \varepsilon_{zz}\mathbf{L}(\mathbf{E} \cdot \mathbf{L}) \quad (1.5)$$

where $\mathbf{S} = \frac{\hat{\mathbf{r}}}{r}$ is the splay, \mathbf{L} is the director, and ε_{zz} the component of the dielectric tensor along the director. (I have omitted terms involving twist and bend, and changed the units to SI). Since $\mathbf{D} = 0$, $e_{1z}\mathbf{S} = -\varepsilon_{zz}\mathbf{L}(\mathbf{E} \cdot \mathbf{L})$, and the enthalpy becomes

$$H_E = \frac{1}{2}k_{11}S^2 + \frac{1}{2}\varepsilon_{zz}(\mathbf{E} \cdot \mathbf{L})^2 \quad (1.6)$$

The mechanical work to distort a nematic is thus clearly the sum of the bare elastic energy, without the flexoelectric contribution, and the energy required to produce the flexoelectric polarization, as indicated in Meyer’s original paper³ and adhered to in subsequent work by others.⁴⁻⁶ If the director field is radial, $\mathbf{E} \cdot \mathbf{L} = \frac{-e_1}{\varepsilon_{zz}r}$, and one can write

$$H_E = \frac{1}{2}k_{11}S^2 + \frac{1}{2}\frac{e_{1z}^2}{\varepsilon_{zz}}S^2 = \frac{1}{2}k_{11}\left(1 + \frac{e_{1z}^2}{k_{11}\varepsilon_{zz}}\right)S^2 \quad (1.7)$$

In the notation of Ref. 1, this becomes

$$F_d = \frac{1}{2}\frac{K_1^D}{\rho^2} = \frac{1}{2}\frac{K_1}{\rho^2} + \frac{1}{2}\frac{e_1^2}{\varepsilon_{zz}^a\rho^2} = \frac{1}{2}\frac{K_1}{\rho^2} + F_E \quad (1.8)$$

where $K_1 = k_{11}$ and $K_1^D = k_{11}\left(1 + \frac{e_{1z}^2}{k_{11}\varepsilon_{zz}}\right)$. K_1^D is therefore the effective splay constant, renormalized to include the flexoelectric contribution.

The problem with the argument in Ref. 1 is that the authors do not clearly distinguish between K_1^D and K_1 . They write, correctly, K_1^D in their inequality in Eq. (1.3), but in evaluating the terms, instead of using the value of K_1^D , they use, as an estimate, a value of the bare elastic constant K_1 taken from the literature. It is not surprising, therefore, that the inequality is violated; the bare elastic energy density need not be greater than the energy density of the flexoelectric polarization. Energy conservation only requires that $F_d > F_E$. Since Castles *et al.* use the wrong elastic constant in their analysis, the conclusion they draw is invalid.

It may be useful to make some observations about the experimental determination of elastic constants.

Consider Scenario 1, where a nematic sample with homeotropic orientation is between flat plates, as shown in of Fig. 1a (top) of Ref. 1. If the plates are then deformed so that they form sections of concentric cylinders, as shown in of Fig. 1a (bottom) of Ref. 1, flexoelectric polarization $P_f = \frac{e_1}{\rho}$ will arise in the radial direction due to the splay induced by the deformation. In addition, there will be dielectric polarization $P_d = (\varepsilon_{||}^a - \varepsilon_o)E$, and an electric field $E = -\frac{e_1}{\varepsilon_{||}^a\rho}$ produced by surface charges due to polarization at the plates. Although the polarization and the electric field depend on ρ , the electric displacement $D = 0$ for all ρ . The work to deform the nematic was therefore done a constant \mathbf{D} , and the work per volume is given by Eq. (1.8),

$$F_d = \frac{1}{2}\frac{K_1^D}{\rho^2} = \frac{1}{2}\frac{K_1}{\rho^2} + \frac{1}{2}\frac{e_1^2}{\varepsilon_{||}^a\rho^2} \quad (1.9)$$

So, in principle, by measuring the work done in Scenario 1, one can determine K_1^D , the elastic constant at constant D .

Consider now Scenario 2, which differs from configuration above only in that the plates are conducting and they are electrically connected, so that the potential difference V between the plates

is zero. If the plates are then deformed as before, flexoelectric polarization $P_f = \frac{e_1}{\rho}$ will arise again, but now $E = 0$ since the surface charges at the plates due to polarization are cancelled by those now on the plate (after having been transferred from the other plate) and $D = \frac{e_1}{\rho}$. Since $E = 0$, there is now no stored electrical energy, and the work per volume to deform the nematic at constant V is just

$$F_d = \frac{1}{2} \frac{K_1^V}{\rho^2} = \frac{1}{2} \frac{K_1}{\rho^2} \quad (1.10)$$

where I have introduced K_1^V , the elastic constant at constant voltage. So, in principle, by measuring the work done in Scenario 2, one can determine the bare K_1 , which is shown here to be the elastic constant at constant V .

In practical terms, however, although the experimental realization of Scenarios 1 and 2 is straightforward, the measurement of the work of deforming the sample is not practical due to the contributions from the plates. Practical measurements of elastic constants are typically done neither at constant D nor at constant V ; this allows for the possibility of different experiments yielding different results. Since flexoelectric polarization is mandated by symmetry, it is always present in deformed nematics, but its magnitude varies. When flexoelectric effects are small, $K_1^D \simeq K_1^V \simeq K_1$, and the distinction between them is unimportant. When flexoelectric effects are large, however, specifying the electrical constraints during elastic constant measurements becomes essential. It would be of interest to compare K_1^D and K_1^V for various liquid crystals, but we are unaware of any such measurements or comparisons to date.

It is worth noting that the determination of the flexoelectric coefficient e_1 is much more straightforward. It can be simply obtained via Scenario 1 from the potential difference V between the plates;

$$e_1 = \frac{\varepsilon_{\parallel}^a V}{\ln(\rho_2/\rho_1)} \simeq \frac{\varepsilon_{\parallel}^a V \Delta\rho}{\rho} \quad (1.11)$$

where $\Delta\rho$ is the plate separation; or via Scenario 2, from the charge q transferred from one plate to the other;

$$e_1 = \frac{2\pi\rho q}{A} \quad (1.12)$$

where A is the area of the plates.

In summary, when flexoelectric effects are large, it is essential to distinguish between elastic constants measured under different electrical constraints. Since the authors of Ref. 1 failed to do this, their conclusion, that the results of Ref. 2 are “irreconcilable with the theoretical limitations established using Oseen-Frank continuum theory of LCs and the conservation of energy.”, does not hold.

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⁴W. Helfrich, “Inherent Bounds to Elasticity and Flexoelectricity of Liquid-Crystals,” *Mol Cryst Liq Cryst*, **26**, 1–5 (1974).

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⁶S. A. Pikin, *Structural transformations in liquid crystals* (Gordon and Breach Science Publishers. xii, New York, 1991), p. 423.