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Soft materials for linear electromechanical energy conversion

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We briefly review the literature of linear electromechanical effects of soft materials, especially in synthetic and biological polymers and liquid crystals (LCs). First we describe results on direct and converse piezoelectricity, and then we discuss a linear coupling between bending and electric polarization, which maybe called bending piezoelectricity, or flexoelectricity.

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Piezoelectricity

Piezoelectricity is a linear coupling between electric and mechanical properties in special non-centrosymmetric materials. It has been known in crystals since the work of Pierre and Jacques Curie in 1880 [1]. Piezoelectric properties were also found in amorphous and polycrystalline materials, such as ferroelectric ceramics [2–4]. The existence of piezoelectricity in certain synthetic and biological polymers [5], such as bone and tendon has been extensively studied [6,7]. Piezoelectric effects in polymers are generally small, but can be increased when they are subjected to a strong DC electric field at elevated temperatures [8]. Owing to their flexibility and the possibility to prepare films of large area, these materials (especially polyvynilidene fluoride, PVDF) have been utilized as the active element in many applications ranging from infrared detectors to loudspeakers. Certain cellular polymers internally charged by corona discharge (ferroelectrets) have been found to behave like soft sensitive piezoelectrics [9–11]. Similarly large and soft piezoelectricity was found in fiber mats composed of ferroelectric barium titanate (BT) nanoparticles dispersed in polylactic acid (PLA) [12•].

As the type of materials with experimentally observed piezoelectric coupling, so the definition of piezoelectricity extended considerably in time. Originally it was used only for crystals in connection with compressions, but later it was generalized to polymers and other materials for any strains and stresses, including shear. The *direct piezoelectric effect* (when electric polarization is produced by mechanical stress) mathematically can be expressed by the equation:

$$P_i = \sum_{jk} d_{i,jk} T_{jk}, \tag{1}$$

where P_i is the ith component of the polarization induced by the stress tensor T_{jk}. The third rank tensor coefficients $d_{i,jk}$ are called piezoelectric coefficients.

In the *converse* (or 'inverse') *effect* the material becomes strained when electric field is applied. Mathematically this is expressed as

$$S_{jk} = \sum_{i} d_{i,jk} E_i. \tag{2}$$

Here $S_{jk} = \partial s_j / \partial x_k$ is the strain tensor (s_j is the displacement of a volume element from its equilibrium position), and E_i is the external electric field.

By symmetry not only solids, but also liquid crystals (LC) can lack inversion symmetry owing to the chirality of rod-shape molecules containing carbon stereo-centers, or owing to asymmetric packing of molecules with special (bent, banana) shapes [13]. The chiral liquid crystal phases are the cholesteric (N*), chiral smectic (e.g. SmC* of rod-shape molecules [14], or SmCP of bent-core molecules [15]) and the chiral columnar phases.

Table 1 lists the LC phases that have no inversion symmetry, that is, which can have linear couplings between electric field and mechanical strain. We see that several of them (SmC* of chiral rod-shape, the tilted columnar phase of chiral disc shape and the SmCP of achiral bent-core or tilted bowl-shape molecules) have C_2 symmetry, which allows 8 independent piezoelectric coefficients.

In the SmA* and N* phases a shear induces a tilt, which, combined with the lack of mirror plane owing to the molecular chirality, allows the existence of a polar vector normal to the shear plane. This is illustrated in the upper part of Figure 1.

The inverse effect, when an electric field is applied perpendicular to the director, results in a shear strain, which would also lead to a tilt of the director. The optical consequence of this effect, rotation of the optic axis (the

Table 1

List of the most important liquid crystalline phases with their symmetry and non-vanishing piezoelectric coupling constants

Phase	Molecular shape	Symmetry	Non-zero Piezo constants
N*, SmA*	Chiral rod	D_∞	$d_{1,23} = -d_{2,13}$
SmAP	Bent shape	$C_{2\nu}$	$d_{3,11}$; $d_{3,22}$; $d_{3,33}$, $d_{1,13}$; $d_{2,23}$
SmC*	Chiral rod	C_2	$d_{3,11}$; $d_{3,22}$; $d_{3,33}$, $d_{1,13}$; $d_{2,23}$; $d_{1,23}$; $d_{2,13}$; $d_{3,12}$
SmCP	Bent shape	C_2	$d_{3,11}$; $d_{3,22}$; $d_{3,33}$, $d_{1,13}$; $d_{2,23}$; $d_{1,23}$; $d_{2,13}$; $d_{3,12}$
SmCG	Bent shape	C_1	All 18 $d_{i,jk}$
Col^*_{tilt}	Chiral disc	C_2	$d_{3,11}$; $d_{3,22}$; $d_{3,33}$, $d_{1,13}$; $d_{2,23}$; $d_{1,23}$; $d_{2,13}$; $d_{3,12}$
P_h	Bowl shape	$C_{\infty\nu}$	$d_{3,33}$; $d_{3,11} = d_{3,22}$
P^*_h	Chiral bowl	C_∞	$d_{3,33}$; $d_{3,11}$; $d_{3,22}$
P_{tilt}	Tilted bowl	C_2	$d_{3,11}$; $d_{3,22}$; $d_{3,33}$, $d_{1,13}$; $d_{2,23}$; $d_{1,23}$; $d_{2,13}$; $d_{3,12}$

director) proportional to the electric field, is known as electro-clinic effect [16–18].

The ***direct piezoelectric*** (or linear mechano-electrical) effect in SmC* materials is owing to the shear-induced polarization [19,20], and is arising from the distortion of the helix. In the ground state (no-shear) the polarization averages out in bulk, but shear unwinds the helix and leads to a polarization normal to the shear plane. Direct piezoelectric effects were also observed in various biological membranes [21] and in aqueous lyotropics, where the highly conductive water is often replaced by ethylene glycol [22–24]. Linear mechano-electrical effects were also found on non-aqueous lyotropic phases [21,25] Recently neat phospholipids were also shown to be piezoelectric [26,27].

Figure 1

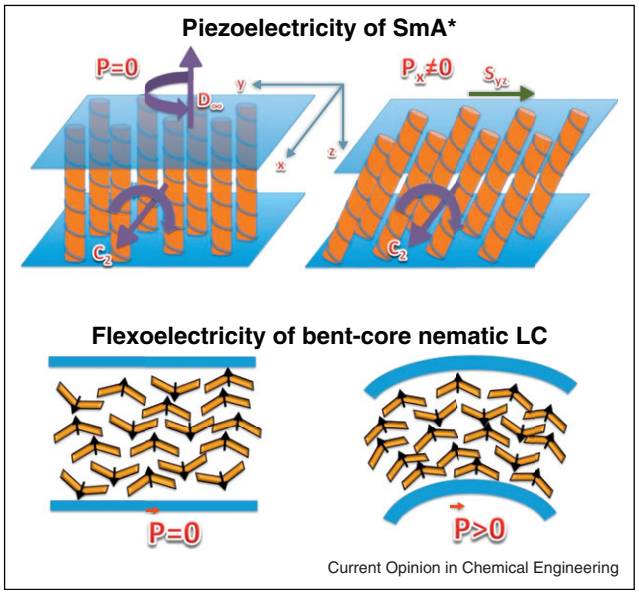


Illustration of the main physical mechanism of piezoelectricity and flexoelectricity in LCs.

The ***converse piezoelectric*** (linear electro mechanical) effect in SmC* materials [28–30] may result in audible acoustic effects implying their possible use in electro-mechanical transducers [31,32]. A systematic study [33] with accurate control of the alignment revealed that the vibrations parallel to the smectic layers and the film surface are generally the strongest, especially if the polarization is also parallel to the plates.

Linear electro-mechanical effects of free-standing ferroelectric SmC* films were studied by light scattering while applying AC electric fields along the film surface [34–37]. In addition, they were also investigated in ferroelectric columnar LCs [38], SmC* elastomers [39,40] and glasses [41], in various biological systems [42], lyotropic LCs and in membranes [21]. Converse piezoelectricity was also invoked [43] to explain the swelling of membranes in response to voltage changes [44]. Converse piezoelectric measurements in ferroelectric SmCP bent-core LCs [45] and a summary of experimental studies in SmC* elastomers [46] have been published only recently. An electroclinic effect related field-induced twist, whose direction changes sign with that of the electric field, was also reported on SmA* elastomers [47,48].

Flexoelectricity

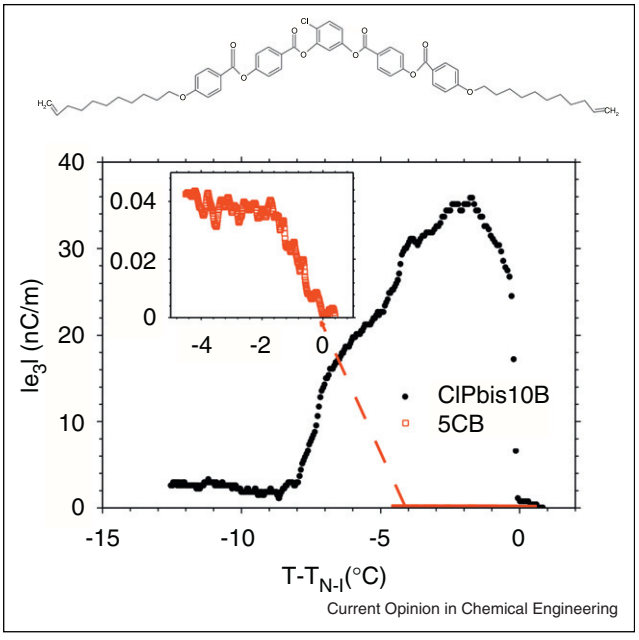
Another linear electromechanical phenomenon, flexoelectricity, connects bending and electricity. Initially it was regarded as bending piezoelectricity [49,50], however, in contrast to piezoelectricity, flexoelectricity does not require the lack of inversion symmetry and the coupling constants e_{ijkl} are given by a fourth rank tensor:

$$P_i^{flexo} = e_{ijkl}\frac{\partial S_{jk}}{\partial x_l}; \quad A\frac{\partial T_{ij}}{\partial x_k} = e_{ijkl}E_l, \tag{3}$$

corresponding to direct and converse flexoelectricity, respectively. S_{jk} (T_{ij}) are the elements of the strain (stress) tensor, and A is the area of the sample. In crystals the first experiments [51] in 1988 gave $e \sim 10^{-11}$ to 10^{-10} C/m. In perovskite-type [52••] and relaxor ferroelectric ceramics [53–55] and in polyvinylidene fluoride (PVDF) film [56•,57•] the flexoelectric coefficients are 4 orders of magnitude larger than in dielectric crystals, and the large polarization induced by flexing is evidently of ferroelectric origin.

In fluid LCs stresses and strains are related mostly to orientational elasticity. R.B. Meyer predicted flexoelectricity [50] for nematic LCs composed of dipolar molecules with asymmetric shapes, such as pear-shape or bent-core. In the absence of electric fields the molecular dipoles average out and the net polarization is zero.

Figure 2



Variation of the flexoelectric coefficient on a relative temperature scale $T–T_{N-I}$ for the bent-core liquid crystal ClPbis10BB (molecular structure seen above the graph), and for the calamitic liquid crystal 5CB measured in cells of $A = 1$ cm^2 active areas. The inset shows part of the figure ($|e_3|$ of 5CB) at a magnified scale.

However, when the director is subjected to splay or bend deformations, respectively, the material become macroscopically polar:

$$P^{flexo} = e_1 \mathbf{n}(\mathrm{div}\,\mathbf{n}) - e_2 \mathbf{n} \times (\mathrm{curl}\,\mathbf{n}) \quad (4)$$

This effect is illustrated in the lower row of Figure 1.

Molecular statistical theories based on molecular packing considerations [58–60] predicted for the splay and bend flexo-coefficients e_1 and e_3 to be about 1 pC/m for nearly rod-shape molecules, and about an order of magnitude larger e_3 values for banana shape molecules. Actual measurements on rod-shape LCs [61–69] typically give about $e_1 \sim e_3 \sim 10$ pC/m, owing to a contribution of a quadrupolar mechanism [70]. These values are similar to those measured on elastomers subjected to a deformation gradient [51].

A 'peculiar kind of piezoelectric effect' was measured on black lipid membranes (BLM) [71–73] and found that the molecular basis of the flexoelectricity of lipid bilayers is an asymmetric redistribution of charges, dipoles and the splayed uniaxial orientation of their quadrupolar moments [74]. Converse flexoelectric measurements on BLM [75] and vesicles [76] found $e_1 \sim 100$ pC/m, which is an order of magnitude larger than typical for calamitic thermotropic LCs. The sensory mechanism of outer hair cell composite membranes [77,78] can be understood by the flexoelectric properties of the lipid bilayer [79]. The converse of this effect, that is, voltage-generated curvature has also been observed [80].

The bend flexoelectric coefficient determined from mechanical bending induced electric current measurements in bent-core LCs [81,82] show 3 orders of magnitudes larger values than in normal dielectric LCs. An example on 4-chloro-1,3-phenylene bis4-[4′-(9-decenyloxy) benzoyloxy] benzoate (ClPbis10BB) in comparison with a typical rod-shape liquid crystal pentyl cyano biphenyl (5CB) is shown in Figure 2.

This increase is similar to that of ferroelectric ceramics [55] and indicates the presence of ferroelectric smectic clusters [83–85,86•] in bent-core nematics.

Conclusion

The physical effects reviewed above represent couplings between electrical and mechanical phenomena, which might allow energy conversion.

These concepts turned out to be useful in interpreting some functions of living organisms, successfully developed during evolution. These effects are already utilized in state-of-the-art sensor and actuator technologies, and might have potential for further practical applications. The latter hopefully prompt designing and synthesizing more appropriate functional materials (presumably polymers or elastomers) with larger coupling coefficients and easier processability.

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