Flexoelectricity: A Perspective on an Unusual Electromechanical Coupling

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The ability of certain materials to convert electrical stimuli into mechanical deformation, and vice-versa, is a prized property. Not surprisingly, applications of such so-called piezoelectric materials are broad—ranging from energy harvesting to self-powered sensors. In this perspective, written in the form of question-answers, we highlight a relatively under-studied electromechanical coupling called flexoelectricity that appears to have tantalizing implications in topics ranging from biophysics to the design of next-generation multifunctional nano materials.

What is flexoelectricity and how does it differ from piezoelectricity?

A *uniform* mechanical strain electrically polarizes a piezoelectric material. There is extensive literature on the formal development of the phenomenological theory of piezoelectricity however, simply put, this phenomenon may be described by the following linearized relation between the components of the polarization vector ($\mathbf{P}$) and the strain tensor (ε): $P_i \sim d_{ijk}\varepsilon_{jk}$. The piezoelectric property tensor ($\mathbf{d}$) is third order. Group theory tells us that only non-centrosymmetric crystals may exhibit properties dictated by third order tensors [1] and accordingly, common insulators which possess a centrosymmetric crystal structure, such as Si and NaCl, are non-piezoelectric.

However, as schematically alluded to in Fig.1, a non-uniform strain may break the mirror symmetry even in otherwise centro-symmetric crystals. The relation of the polarization to the extent of the non-uniformity of the strain field, or strain gradient, is known as *flexoelectricity*: $P_i \sim d_{ijk}\varepsilon_{jk} + f_{ijkl}\frac{\partial \varepsilon_{jk}}{\partial x_l}$; where f_{ijkl} are the components of the so-called flexoelectric tensor. While the piezoelectric property is non-zero only for selected materials, the strain gradient-polarization coupling (i.e. flexoelectricity tensor) is in principle non-zero for all (insulating) materials. This implies that under a non-uniform strain, *all* dielectric materials are capable of producing a polarization. The reader is referred to the following articles for further information: Refs.[2, 3, 4, 5, 6, 7, 8, 9].

What are some examples of real materials in which flexoelectricity has been observed?

The flexoelectric mechanism is well-illustrated by the non-uniform straining of a graphene nanoribbon—a manifestly non-piezoelectric material (Fig.2(a))[10, 11]. Several works have now appeared [12, 13, 14, 15] that have addressed 2D materials e.g. boron nitride, carbon nitride among others. As another widely studied two dimensional soft material, biological membranes also show flexoelectricity (Fig.2(b))[16]. A review on flexoelectricity in 2D materials has recently been provided by [9]. Flexoelectricity has been experimentally confirmed in several crystalline materials such as NaCl, strontium titanate, and ferroelectrics like barium titanate among others [17, 18, 19, 20, 21]. Some recent experiments have also measured a flexoelectric response in several polymers[22, 23, 24].

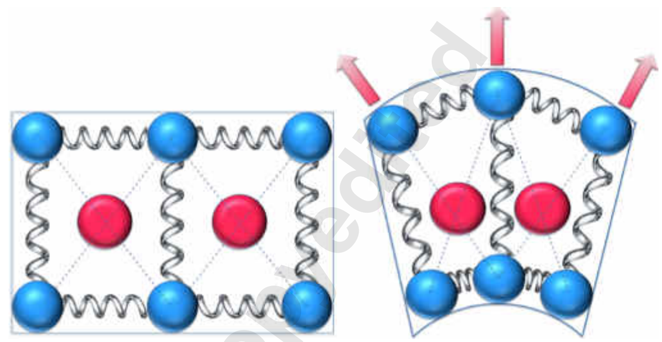

Fig. 1. Illustration of induced polarization due to non-uniform deformation of a centro-symmetric (non-piezoelectric) material.

How "significant" is the flexoelectric effect?—is it just an "exotic" phenomenon or something that may have some compelling implications?

The jury is still out on how important flexoelectricity may turn out to be. Even though *all* dielectrics are flexoelectric, the effect *may* be negligibly small and is dictated by the strength of the flexoelectricity tensor ($\mathbf{f}$). In other words, symmetry considerations *guarantee* that flexoelectricity *exists* but not how important it will be in a given material. Flexoelectricity becomes important when one or more of the following (sometimes overlapping) situations occur:

1. The material's flexoelectricity coefficients are unusually large: This is usually the case for high dielectric-constant materials like ferroelectrics and complex oxide ceramics cf. [4, 17, 18, 19, 20, 21].

2. The more traditional form of electromechanical coupling e.g. piezoelectricity, is absent: In such a case, flexoelectricity then provides perhaps the only significant route to couple mechanical deformation and electrical stimuli. For example, biological membranes have no crystalline symmetry that would permit piezoelectricity. Accordingly, flexoelectricity—which relates changes in curvature to the

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development of polarization—becomes quite important.

3. The feature size in the structure of interest is ”small”: Large strain gradients can induce a strong flexoelectric response even if the magnitude of flexoelectric coefficients is not large. For a given strain field, large strain *gradients* are generated easily only at the nanoscale. Here we mention that the precise scale at which this effect becomes prominent depends (largely) on the relative strength of the elastic properties, dielectric coefficients and flexoelectric coefficients. Usually sub-10 nm characteristic length scales are required c.f. [8, 9, 25], albeit in some contexts (e.g. soft materials or ferroelectrics), this effect can also manifest with feature sizes of several hundreds of nanometers [18, 26].

4. Soft materials: Strain gradients scale inversely with the elastic stiffness. Experiments appear to indicate that the flexoelectric coefficients of soft materials (such as polymers) are at least the same order of magnitude as hard crystalline materials, if not stronger [22, 23, 24]. However, the elastic stiffness of soft materials can be several orders of magnitude smaller than hard ceramics. Accordingly, there is an expectation that flexoelectricity will be important for soft materials. Preliminary analysis of Deng et. al. [26, 27] appear to confirm this where flexoelectricity was shown to create artificial piezoelectric materials whose apparent piezoelectric strength is nearly twenty times larger than the hard ferroelectrics like barium titanate.

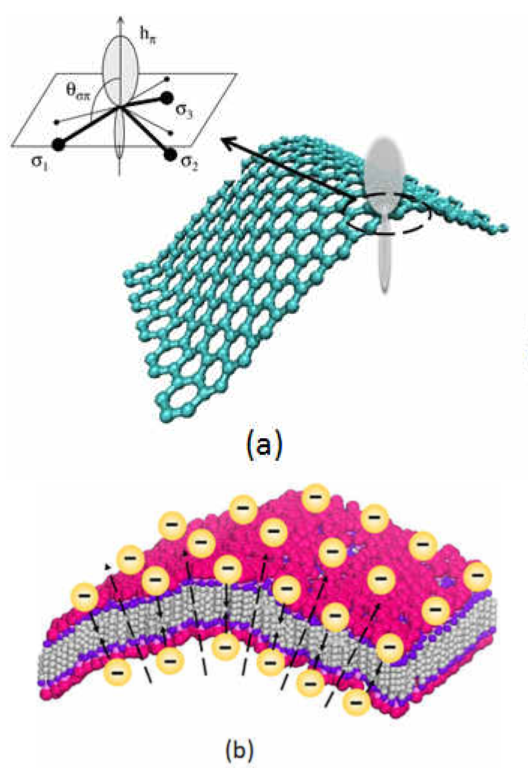

Fig. 2. [Reprinted with permission from Ref. [26]]: Flexoelectricity in membranes. (a) Bending of a (dielectric) graphene nano-ribbon: bending deformation leads to symmetry-breaking of the electron distribution at each atomic site leading to the development of a dipole moment normal to the ribbon plane. (b) Bending of a lipid bilayer membrane: Due to bending, the asymmetry in both the charge and dipole densities in the upper and lower layers causes the normal polarization in the bilayer membrane. .

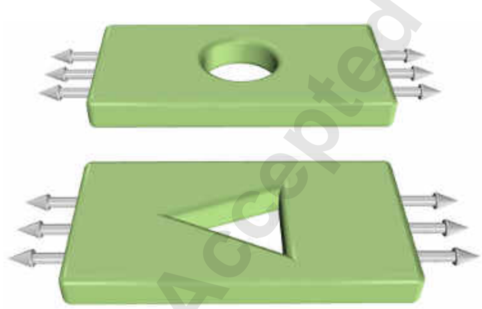

Fig. 3. [Adapted from [13] and reprinted with permission from [9]]. The first figure depicts a non-piezoelectric 2D sheet with circular pores. Under uniform stretching, strain gradients develop in the vicinity of the holes and therefore the local polarization due to flexoelectricity is non-zero, however the net or average polarization remains zero, and thus overall there is no emergent piezoelectric response. The second figure shows the same sheet with triangular pores. In this case, again, locally, in the vicinity of the triangular holes, polarization develops. Unlike the previous case, however, there also exists now a net non-zero polarization and thus this hypothetical material with triangular holes exhibits an apparent piezoelectricity even though the native material itself is non-piezoelectric.

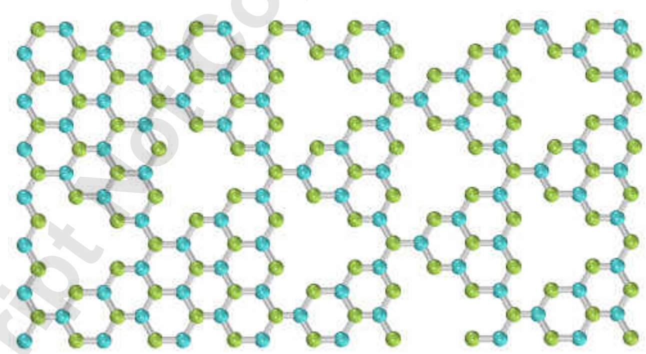

Fig. 4. [Reprinted with permission from [9]] Graphene nitride nano sheet, riddled by triangular holes, was experimentally and computationally shown to exhibit an apparent piezoelectric response.

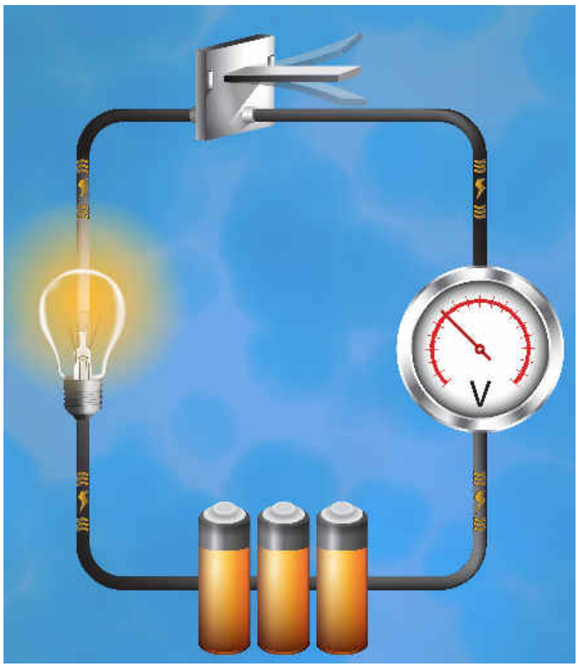

Fig. 5. A centrosymmetric flexoelectric energy harvester under base excitation can function in a manner similar to a piezoelectric harvester.

What is the connection between nanotechnology and flexoelectricity?

As alluded to in the response to the previous question, strain gradients are most easily achievable at the nanoscale and accordingly for appreciable flexoelectricity, nano structures and nano materials are highly relevant. For example, Ref. [28] provides a study of how the dynamic and energy harvesting response of a flexoelectric beam changes with size—non-trivial results are usually obtained only at nanoscale dimensions.

How can be flexoelectricity be exploited to "design" multifunctional materials?—how to create "apparently piezoelectric materials without using piezoelectric materials"

Arguably, one of the most interesting applications of flexoelectricity is to *create* apparently piezoelectric materials without using piezoelectric materials [29]. The central idea underpinning this is quite simple: Consider a material consisting of two or more different non-piezoelectric dielectrics—as a concrete example that has been studied in the past we may think of a (dielectric) graphene nano ribbon impregnated with holes (Fig.3)[11]. Upon the application of uniform stress, differences in material properties at the interfaces of the materials will result in the presence of strain gradients. Those gradients will induce polarization due to the flexoelectric effect. As long as certain symmetry rules are followed, the net average polarization will be nonzero. Thus, the artificially structured material will exhibit an electrical response under uniform stress behaving therefore like a piezoelectric material. Regarding symmetry: Topologies of only certain symmetries can realize the aforementioned concept. For example, circular holes distributed in a material will not yield apparently piezoelectric behavior even though the flexoelectric effect will cause local polarization fields. Due to circular symmetry, the overall average polarization is zero (Fig.3(a)). A similar material but containing triangular shaped holes (or inclusions) for example, and aligned in the same direction, will exhibit the required apparent piezoelectricity. In a similar vein, a finite bilayer or multilayer configuration may also be used (Fig.3(b))–see discussions in [25]. Zelisko et. al. [13] characterized graphene nitride nano-sheets (g-C_3N_4) both experimentally and via *ab initio* simulations. Intrinsically, pristine graphene nitride nano sheets are non-piezoelectric however, in one of its stable form, the sheets are riddled with triangular holes [?], as shown in (Fig.4). In their work, it was confirmed that indeed flexoelectricity, together with triangular defects cause graphene nitride to exhibit an apparent piezoelectricity.

What are some possible implications and applications of flexoelectricity?

The idea of designing artificial piezoelectric materials has already been explained in the response to the preceding questions. Here are some other implications and applications of flexoelectricity:

1. Energy Harvesting: To date, research on energy harvesting is centered around piezoelectric materials e.g. [30]. Several recent works have appeared that have illustrated the potential for the use of flexoelectricity in energy harvesting [28, 31, 32, 33]. More recently Ref. [28] have developed a complete theoretical continuum model for flexoelectric nanoscale energy harvesting (Fig.5). When a cantilever beam, covered by conductive electrodes on its top and bottom surfaces, undergoes bending vibrations, an alternating potential differtence is generated across the electrodes. Many conventional piezoelectrics (which are often ferroelectrics) loose their piezoelectricity above the so-called Curie temperature. Flexoelectricity, in contrast can persist to fairly high temperatures and can be fruitfully exploited to circumvent this limitation of conventional piezoelectrics [34]. In that context, flexoelectricity is a possible solution in situations where piezoelectric materials cannot be used.
2. Material Behavior: Flexoelectricity has been found to play a prominent role in a range of phenomena exhibited by ferroelectric nano structures or bulk specimens with nanoscale features. Some examples are: polarization rotation in thin-films [35], indentation size-effect [36], fracture toughness [37], defects[38] among others.
3. Sensors and Actuators: Very few works have actually exploited the concept of flexoelectricity to create sensors and actuators—-a natural application of any multifunctional coupling. Some examples are, Bhaskar et. al. [39] who have created the so-called electromechanical strain diode as well as a MEMs device on Silicon[40] and Wang et. al. 2013 who fabricated a ferroelectric micro machined diaphragm.

What is the role of flexoelectricity in biology?

Two representative examples of non-uniform strain modes are bending and torsion. Relatively little energy is required to

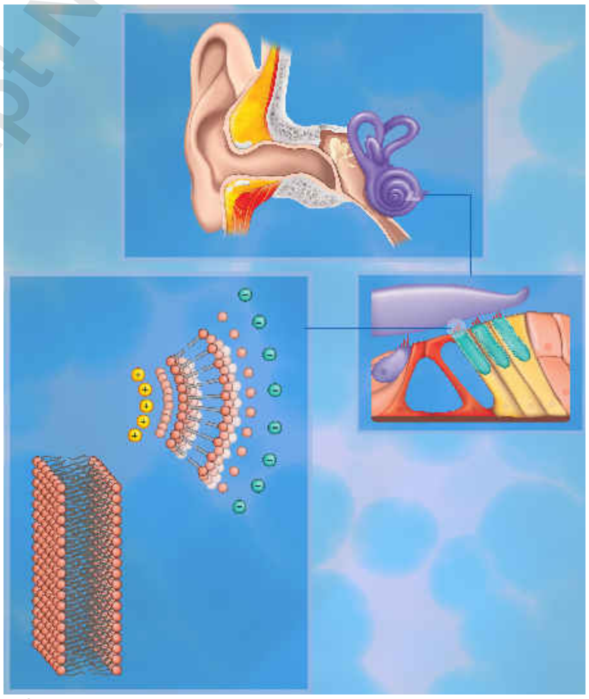

Fig. 6. [Caption quoted from text in Ahmadpoor and Sharma[9]] Hair bundles consist of several stereocilia that are connected by thin fibers called tip links and organized in rows of decreasing height. The axes of hair bundles point away from the center of the cochlea. Mechanosensitive ion channels are located within the wall of the stereocilia near the top and tethered to adjacent stereocilia by tip link tension. Bending of the hair bundle toward the tallest row imposes tip link tension on channels in the shorter neighbor causing them to open and make the cellular inner environment more electrically positive. Similarly, bending the bundle in the opposite direction, closes the channel, causing the cell become more negative. During these processes, a voltage difference emerges across the thickness of the stereocilia membrane and due to the flexoelectric response of the cellular membrane, the radius of the stereocilia changes. Accordingly, the height of the stereocilia increases (or decreases) to maintain the fixed volume.

induce curvature in soft biomembranes whose bending modulus is only slightly higher ($10 - 20k_BT$) than the thermal energy scale. In the context of biomembranes, flexoelectricity takes the following form: $\mathbf{P} = f\kappa\mathbf{n}$, where $\mathbf{n}$ is the normal vector to the membrane mid-plane. Given the absence of any plausible micromechanism for piezoelectricity, flexoelectricity is most likely the key mechanism underpinning the electromechanical coupling in biomembranes. It has been found to be relevant for studying ion channels, thermal fluctuations, the equilibrium shape of the vesicle, and electromotility [26, 42, 43, 44, 45, 46, 47]. Based on several hypotheses and experiments [48, 49, 50, 51, 52, 53, 54, 55], the mammalian hearing mechanism appears to be one of the most exciting implications of flexoelectricity in biology. Hair cells are the primary sensory receptors in the auditory system that transform the mechanical vibrations of sound into sensible electrical action potential. Though, the corresponding mechanism is still not *fully* understood, one possible explanation is that flexoelectricity is the electromechanical coupling mechanism in the outer hair cells of the mammalian ears (Fig.6).

What are some open areas of research in flexoelectricity?

Despite the emergence of active interest in this area in both the mechanics and physics community, the topic of flexoelectricity is wide-open. We mention here just a few topics that may be of interest to the mechanics community.

The connection of flexoelectricity to traditional mechanics topics such as defects and fracture is only just being touched upon. Likewise, while a few works (cited elsewhere in this article) have exploited flexoelectricity to design novel forms of multifunctional materials, this path has hardly been exhausted.

A tantalizing future direction in the case of flexoelectricity is in the realm of soft materials. As indicated previously, larger strain gradients are easily possible in soft materials and thus the prospects of a stronger flexoelectric response along with the possibility of large deformations is attractive. While flexoeletricity, both from a quantum view point (c.f. [56]) as well as classical (c.f. [7]), appears to now be well-understood, a clear microscopic picture underpinning flexoelectricity in soft materials is still lacking. Perplexingly, experiments appear to indicate flexoelectricity to be both large and small in a variety of polymers[22, 23, 24]. Currently, the Maxwell stress effect is exploited for electromechanical actuation in soft materials. However, the latter suffers from some disadvantages: the Maxwell stress effect is a one-way coupling i.e. mechanical deformation does not produce an electric field, large electric fields are required for actuation and finally, reversal of electric field does not reverse the direction of the deformation. In contrast, appropriately used flexoelectric-response will not suffer from any of these disadvantages.

Perhaps the biggest need currently is in the development of flexoelectricity based applications and careful materials characterization experiments. In general, theoretical and computational work has far outpaced experimental efforts in this direction. Having said that, even from a computational viewpoint, atomistic calculations of flexoelectric constants is nontrivial due to the fact that periodic boundary conditions make the imposition of strain gradients rather difficult.

With the exception of graphene, boron nitride and (to some degree) graphene nitride[14, 12, 13], a characterization of the flexoelectricity in other 2D materials is still missing. In particular, we note that to date, flexoelectricity has not been *experimentally* evaluated for any of the 2D inorganic materials —however, as described in the main text, considerably more progress has been made in the case of lipid bilayers[57, 58, 59, 60, 61].

Finally, the connection of flexoelectricity to other multifunctional materials such as magnetoelectrics, liquid crystal elastomers is wide open.

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