Flexoelectricity in dielectrics: Materials, structures and characterizations*

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Flexoelectricity in dielectrics suggests promising smart structures for sensors, actuators and transducers. In this review, dielectric materials, structures and the associated flexoelectric characterization methods are presented. First of all, we review structures and methods to measure different flexoelectric coefficients, including μ_{1122} , μ_{1111} , μ_{1211} , μ_{3121} , μ_{2312} , μ_{1123} , etc., via direct or converse flexoelectric effect. The flexoelectric materials in the form of bulk, thin films and 2D materials and the reported flexoelectric properties of these dielectrics will then be discussed. Semiconductor materials and the associated flexoelectric studies will also be reviewed. The progress of flexoelectric device study will next be presented, followed by the flexoelectricity research challenges and future trend.

Keywords: Flexoelectricity; dielectrics; semiconductor; thermal dependence; health monitoring.

1. Principles of Flexoelectricity

The rapid development in nanotechnology has satisfied human beings' fantasy of daily life. At the same time, it demands high-performance electronics, integrated circuits and micro/nanoelectromechanical systems.¹ Electromechanical coupling effect, referring to the transformation from mechanical variables to electric variables or vice versa, underpins the functionality of an extremely broad range of materials and related devices. Currently, piezoelectricity is the main conversion mechanism between mechanical energy and electrical energy in those electromechanical devices, such as accelerometer,² field effect transistors,^{3,4} self-powered nanogenerators,^{5–7} sensors and actuators.⁸ However, the materials selection is very limited due to the crystalline asymmetry constraint of piezoelectricity.

The flexoelectric effect describes the generation of an electric polarization response under a mechanical strain gradient (direct flexoelectric effect) or the mechanical response under an electric field gradient (converse flexoelectric effect). The existence of flexoelectricity in solid symmetric crystalline materials was originally discussed by Kogan in 1964.⁹ As strain gradient can break the inversion symmetry, flexoelectricity allows the generation of electric polarization from lattice deformations in all dielectric

materials. In mathematics, the flexoelectric effect is usually written in the form of

$$\begin{aligned} P_i &= \mu_{ijkl}^{\text{dir}} \frac{\partial S_{jk}}{\partial x_l}, & P_i &= f_{ijkl}^{\text{dir}} \frac{\partial T_{jk}}{\partial x_l}, \\ T_{ij} &= \mu_{ijkl}^{\text{con}} \frac{\partial E_k}{\partial x_l}, & S_{ij} &= f_{ijkl}^{\text{con}} \frac{\partial E_l}{\partial x_k}, \end{aligned} \quad (1)$$

where P_i is the induced polarization, T_{jk} is the stress, S_{ij} is the induced strain and E_k , E_l are the electric fields, x is the axis of coordinate, μ_{ijkl} and f_{ijkl} are the flexoelectric coefficients with respect to strain gradient and stress gradient, respectively. The superscripts “dir” and “con” indicate the direct and converse flexoelectric effects, respectively. The relationship between coefficients μ_{ijkl} and f_{ijkl} can be deduced via the elastic constant:

$$\mu_{ijkl} = c_{ijmn} \cdot f_{mnkl}. \quad (2)$$

The concept of flexoelectricity was first originated from liquid crystals.^{10,11} The irregularly-shaped polarized molecules existing in liquid crystals will be reorientated by the mechanical strain gradient. Later, similar strain gradient-induced polarization phenomenon was also found in crystalline dielectrics, especially in the perovskites, as shown in Fig. 1. The B -site of the lattice will be deviated from the

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Fig. 1. Origin of flexoelectric effect in solids. (a) Two-dimensional (2D) structure of elementary charges without dipole moment. (b) Under uniformly tensile strain for each unit cell, the tension gradually varies from one cell to another. (c) Under inhomogeneous deformation, a dipole moment via the flexoelectric effect was induced within the unit cell.¹³

central position when the material is under an inhomogeneous strain.¹²

Theoretically, the flexoelectric effect in polymers such as polyvinylidene fluoride films (PVDFs) is similar to that in liquid crystals. However, the flexoelectric effect in polymer films is more complicated than that in solid crystals because both residual piezoelectricity effect and the flexoelectric effect should be considered. Thus the mechanism of flexoelectric effect in polymers has not been adequately understood so far.

In the nanoelectronic devices, strong strain gradients are often present, which imply the potential promising applications for flexoelectricity.^{14–16} Huang *et al.* showed that the flexoelectricity is a size-dependent effect due to the increasing of strain gradient when the size goes down.¹⁷

In this review, both experimental and theoretical studies will be included on the flexoelectricity in dielectrics, especially thermal effects on flexoelectricity. Furthermore, the flexoelectric responses of semiconductors are summarized and discussed. Furthermore, we summarize the current works on flexoelectric applications.

2. Reported Experimental Studies and Theoretical Models (μ_{1122} , μ_{1111} , μ_{1211} , μ_{3121} , μ_{2312} and μ_{1123})

To discover the materials with advanced flexoelectricity, it is essential to measure the flexoelectric coefficient of dielectrics, thin films, semiconductor materials and structures. In the last decades, experiments on a series of ferroelectrics have been performed due to their high dielectric constants as suggested by the lattice dynamic theory.¹² For cubic crystals,

mostly the ferroelectrics, only three independent components of the flexoelectric coefficient matrix are nonzero, which are μ_{1122} , μ_{1111} and μ_{1211} (transverse, longitudinal and shear). For thin-film polymer, whose behavior is similar to liquid crystal, there are more nonzero components. In this section, we will summarize the experimental studies in two categories: direct flexoelectric measurement, through applying mechanical input and measuring the electrical polarization output; and converse flexoelectric measurement, where an external electrical voltage is applied to the materials and the mechanical output is monitored.

2.1. Direct flexoelectric measurement

Transverse and longitudinal flexoelectric coefficients (μ_{1122} and μ_{1111}) of dielectric perovskite ceramics were the first two components of the flexoelectric coefficient matrix being characterized. Cross and his co-worker first investigated μ_{1122} experimentally by using a cantilever system and a four-point bending system, shown in Fig. 2(a).¹⁸ Since then, various ferroelectric and paraelectric perovskite materials have been studied, such as barium titanate (BT),¹⁹ barium strontium titanate (BST),²⁰ lead magnesium niobate (PMN)^{18,21} and lead zirconate titanate (PZT).²² Similar method has been adopted to study STO by Zubko *et al.*²³ To eliminate the contributions of longitudinal and shear components, the sample was constrained to be a slender bar (length/width > 5). In addition, due to the single-layer plate shape, the remnant piezoelectric contribution is symmetrically canceled.

μ_{1111} was studied by compressing a truncated pyramid specimen, shown in Fig. 2(b).²⁴ This truncated pyramid shape was designed to generate longitudinal strain gradient when stress is applied. The average strain gradient in this truncated pyramid can be calculated from Eq. (3):

$$\frac{\partial \varepsilon_{11}}{\partial x_1} = \frac{\Delta \varepsilon_{11}}{\Delta x_1} = \frac{\varepsilon_{11}^u - \varepsilon_{11}^l}{h}. \quad (3)$$

Other than using continuous stress and strain, many other attempts on measuring the flexoelectric coefficient of ferroelectrics were made. The shock wave method succeeded in measuring the flexoelectric coefficient of bulk barium titanate.²⁵

The shear flexoelectric coefficient (μ_{1211}) for bulk ceramic material has not been measured until very recently. It has been a technical barrier to generate a shear strain gradient on bulk ceramics. However it was easy to generate the shear strain gradient on polymers like PVDF. This material has been the one that was measured for the components of its flexoelectric coefficient matrix, including μ_{1211} , μ_{3121} , μ_{2312} and μ_{1123} .^{26–29}

In addition to this shear component, the giant transverse and longitudinal flexoelectric coefficients in PVDF were observed.^{15,30–32} The difference apart from the flexoelectric characterization for solid material is that the residual piezoelectricity cannot be ignored for PVDF. The residual piezoelectricity is due to the residual electric polarization,

Fig. 2. The experimental setups for (a) μ_{1122} and (b) μ_{1111} .^{18,24}

microstructural effects such as defects, cracks and the interaction between the α -phase and the amorphous phase in the film.

2.2. Converse flexoelectric measurement

Comparing with the direct flexoelectric measurement, the converse flexoelectric measurement is more difficult to achieve mainly due to two critical issues. One is that the flexoelectric deformations have always been in the picometer scale for bulk ceramics, which is too small to detect. The other one is the simultaneous electrostriction effect. Unlike the piezoelectricity, the electrostriction effect has no symmetry constraint. The centrosymmetric materials are selected to exclude piezoelectricity but not the electrostriction effect. The electrostriction effect is a quadratic effect while the flexoelectric effect is a first-order effect. It is suggested that the electrostriction effect can be filtered out with a lock-in filter. However, Fu *et al.*'s initial experiments did not suggest so.³³

To verify the converse flexoelectricity in BST ceramics, both longitudinal and lateral models were adopted. Fu *et al.*³³ reported a longitudinal model by using an optical system to measure the displacement with the resolution as small as $10\text{E}-5\text{ nm}$. The optical system was based on the scanning Michelson laser beam interferometer, shown in Fig. 3.

There were two groups composed for the test: one was the flexoelectric group, where the electric field gradient existed; while the other was the control group where the electric field gradient was absent. As mentioned above, the flexoelectric group was expected to have first harmonic response, while only the second harmonic response should have appeared for the control group. But significant harmonic response appeared in the results for both the groups. The work was concluded as that the first harmonic response in samples of the control group was due to the first coupling of electrostriction effect with the DC electrical noise signal. The other work on converse flexoelectric effect was conducted on the

Fig. 3. An optical measurement system based on the scanning Michelson laser beam interferometer; PD: photodiode and BS: beam splitter.

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SrTiO_3 single crystal,³⁴ which was structured as a cantilever beam. By applying a high-voltage (0–3 kV) pulse for 2–3 s, the bending curvature was measured by the deflection angle of the laser beam reflected from the surface of the unfixed edge. Instead of discussing the electrostriction effect, the temperature dependence of the effect within 77–450 K was obtained.

The lateral model was designed and reported by Shu *et al.* in order to isolate the converse flexoelectric effect from the electrostriction effect.³⁵ A trapezoid BST block was used and the electrodes were attached to the lateral faces of this structure, where a nonparallel plate capacitor was created, as

Fig. 4. Schematic for converse flexoelectric measurement of the shear strain along x_1 -direction generated by the electric field gradient along the x_3 -direction.

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shown in Fig. 4. The expected result was that the electrostriction contribution should be canceled out because the product of the longitudinal electric field (E_1) and the transverse electric field (E_3) would be zero. However, the first harmonic response showed up in the results of both trapezoid and rectangle samples. By assuming all the coefficients for different shapes are the same, a calculated shear flexoelectric coefficient of $124 \pm 14 \mu\text{C/m}$ was reported.

Besides the experimental studies for converse flexoelectric measurement, Shu *et al.* intended to make the converse flexoelectric coefficient inclusive by studying the relation between the converse coefficient and the direct coefficient.³⁶ Their theoretical derivation with Gibbs free energy density function suggested that, in crystalline mediums and systems, the direct and converse flexoelectric coefficients are equivalent. Due to the absence of practical experiments, this conclusion has assisted researchers to explore the applications of converse flexoelectric effects.

2.3. Theoretical methods

For flexoelectric (nonpiezoelectric and centrosymmetric) dielectrics, the most general bulk thermodynamic potential density^{37–40} considering both polarization gradient and strain gradient is written as

$$U = \frac{1}{2\chi} P_i P_i + \frac{1}{2} b_{ijkl} P_{i,j} P_{k,l} + \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} + \frac{1}{2} g_{ijklmn} \varepsilon_{ij,k} \varepsilon_{lm,n} + h_{ijkl} \varepsilon_{ij,k} P_l + k_{ijkl} \varepsilon_{ij} P_{l,k} - E_i P_i - \sigma_{ij} \varepsilon_{kl}, \quad (4)$$

where χ is the dielectric susceptibility. Also c_{ijkl} , h_{ijkl} , k_{ijkl} , b_{ijkl} and g_{ijklmn} are the material parameter tensors. P_i , $P_{i,j}$ denote the polarization and polarization gradient tensors, respectively. E_i denotes the electric field and σ_{ij} is the stress tensor. ε_{ij} , $\varepsilon_{ij,k}$ are the strain and strain gradient tensors, respectively, defined as

$$\varepsilon_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i}). \quad (5)$$

Here, u_i represents the displacement vector and the comma in the subscript indicates differentiation with respect to the spatial variables, i.e., $(\dots)_{,i} = \partial(\dots)/\partial x_i$.

In the bulk thermodynamic potential density, the terms $\frac{1}{2} b_{ijkl} P_{i,j} P_{k,l}$ and $\frac{1}{2} g_{ijklmn} \varepsilon_{ij,k} \varepsilon_{lm,n}$, respectively, represent the pure polarization and strain gradients effects, which are not included in the classical piezoelectric theory. The term $h_{ijkl} \varepsilon_{ij,k} P_l + k_{ijkl} \varepsilon_{ij} P_{l,k}$ describes the flexoelectric coupling effect, which can be expressed in other forms using the integration by parts. Here, the most used three other forms^{41–43} are

$$\begin{aligned} & h_{ijkl} \varepsilon_{ij,k} P_l + k_{ijkl} \varepsilon_{ij} P_{l,k} \\ &= \frac{h_{ijkl} - k_{ijkl}}{2} (\eta_{ijk} P_l - \varepsilon_{ij} P_{l,k}) + \frac{h_{ijkl} + k_{ijkl}}{2} (\varepsilon_{ij} P_l)_{,k} \\ &= (h_{ijkl} - k_{ijkl}) \eta_{ijk} P_l + k_{ijkl} (\varepsilon_{ij} P_l)_{,k} \\ &= (k_{ijkl} - h_{ijkl}) \varepsilon_{ij} P_{l,k} + h_{ijkl} (\varepsilon_{ij} P_l)_{,k}. \end{aligned} \quad (6)$$

In each form, the last term will be treated as a surface term because the whole derivative cannot be included in the bulk energy density. These expressions lead to the same governing equations but with different natural boundary conditions.

Applying the Euler equations $\partial H / \partial A - (\partial H / \partial A_{,i})_{,i} = 0$, where A stands for P_i or ε_{ij} , the bulk constitutive electro-mechanical equations will be obtained:

$$P_i - \chi b_{ijkl} P_{k,lj} = \chi E_i + \chi (k_{kjli} - h_{kjli}) \varepsilon_{jk,l}, \quad (7)$$

$$\sigma_{ij} = c_{ijkl} \varepsilon_{kl} + (k_{ijkl} - h_{ijkl}) P_{l,k} - g_{ijklmn} \varepsilon_{lm,nk}. \quad (8)$$

In Eq. (7), the first term in the right-hand side describes the classical dielectric response to an electric field. The second term describes the direct flexoelectric response. In Eq. (8), the first term in the right-hand side describes the classical Hook's law and the second term describes the converse flexoelectric response. In addition, it should be noted that $\chi b_{ijkl} P_{k,lj}$ and $g_{ijklmn} \varepsilon_{lm,nk}$, which are derived from the pure polarization and strain gradients terms, respectively, represent the higher-order effects. These higher-order effects could influence the polarization field and the stress field in the nanoscale or even in the microscale provided the characterized size of the structure (for example, the thickness of a

dielectric cantilever beam) is comparable to the effective range of these effects.^{44–46} Unfortunately, the coefficients of these higher-order terms are scarcely quantified in most dielectrics, which makes it difficult to verify the effective range of these higher-order effects. Generally, these higher-order terms are neglected especially in the experiment (the characteristic size is tens or hundreds of microns). Substituting Eq. (7) into Eq. (5) and neglecting the higher-order effects, we find the most general constitutive equations characterizing the flexoelectric effects:

$$\begin{aligned} P_i &= \chi E_i + \mu_{ijkl} \varepsilon_{jk,l}, \\ \sigma_{ij} &= c_{ijkl} \varepsilon_{kl} + \mu_{jik} E_{l,k}. \end{aligned} \quad (9)$$

Here, $\mu_{ijkl} = \chi(k_{kjli} - h_{kjli})$ is the flexoelectric coefficient. The tensor $(k_{kjli} - h_{kjli})$ is called the flexoelectric coupling coefficient that can be represented by an independent tensor f_{ijkl} . It is obvious that the flexoelectric coefficient is proportional to the relative dielectric constants and the flexoelectric coupling coefficients, the values of which were theoretically estimated by Kogan,⁹ Tagantsev¹² and Hong and Vanderbilt^{47,48} to be around several tens of volts for simple ideal ionic solids. These may be called the intrinsic (or lattice-based) flexoelectric coupling coefficients. However, more and more experimental results showed that the apparent (or effective) flexoelectric coupling coefficients in many dielectrics can reach up to several hundred or even several thousand volts.^{17,19,49} Therefore, it appears there are other dominant origins contributing to the apparent flexoelectric responses.

Many researchers have done a lot of work to investigate the anomaly-enhanced flexoelectric responses and some possible origins were proposed.^{23,50–56} First, the initial strain gradients formed around the interface, grain boundary or defects have been reported to possibly enhance the flexoelectric responses. These local strain gradients can reach up to as large as 10^6 – 10^7 m^{−1}, which are 7–8 orders of magnitude larger than those reported for bulk solids. For example, using grazing-incidence in-plane X-ray diffraction, Lee *et al.* measured the strain gradient, induced by a strain relaxation with tens of nanometers of the film–substrate interface due to the lattice mismatch, to be 10^5 – 10^6 m^{−1}.⁵¹ Zubko *et al.* believed the strain gradient near the defect can be as large as 10^7 m^{−1}.²³ The extremely large strain gradients will induce large local polarization and should contribute to the effective flexoelectric responses. Another main origin of the anomalously large effective flexoelectric responses may come from the residual ferroelectricity, of which the polar nanoregion is one important source. Narvaez and Catalan found that the ferroelastic switching induced by the stress combined with the intrinsic flexoelectric field may pole the relaxor ferroelectrics such as PMN–PT even at the room temperature.⁵⁰ Similar enhancement of apparent flexoelectric responses due to the residual ferroelectricity is also found in the BST ceramics by Garten *et al.*⁵² and much recently in

0.3Pb(In_{1/2}Nb_{1/2})O₃–0.35Pb(Mg_{1/3}Nb_{2/3})O₃–0.35PbTiO₃ (PIN–PMT–PT) by Shu *et al.*⁵⁴

All in all, due to the complex microscale or nanoscale structures existing in the dielectrics, the effective flexoelectric responses may be mixed effects of many factors such as the surface piezoelectricity, intrinsic piezoelectricity, residual ferroelectricity and defects, which still need to be verified in terms of both theory and experiment.

3. Flexoelectricity of Dielectrics

3.1. Bulk material

Both experimental studies and theoretical derivation have suggested that the enhanced flexoelectricity may possibly exist when the materials have a high dielectric permittivity. All measured flexoelectric coefficients up to date are summarized in Table 1, along with their dielectric permittivity values. It is worth mentioning that more different topographic structures of dielectric materials were not studied to measure the longitudinal or shear flexoelectric component.

Instead, the temperature dependence has caught the attention. It has been reported that flexoelectricity is positively correlated with the dielectric constant in solid crystalline materials.²⁴ The temperature dependence of flexoelectric coefficients has been investigated since the dielectric permittivity has a peak value at the Curie temperature. Furthermore, comparing to the piezoelectric materials, it is expected that flexoelectric materials can work in a broader temperature. It was found in BST that the flexoelectric coefficient approaches its peak at the phase transition point.²⁴ Later on, the temperature dependence of flexoelectricity in BaTiO₃–0.08Bi(Zn_{1/2}Ti_{1/2})O₃ (BT–8BZT), BaTiO₃ and SrTiO₃ perovskite nanostructures were also measured.^{58,59} For BST, BaTiO₃ and SrTiO₃, the rapid drop in the dielectric permittivity in the paraelectric phase leads to an inconsistent flexoelectricity as temperature increases. The narrow and sharp dielectric peak occurring at the ferroelectric-to-paraelectric phase transition limits the enhanced flexoelectric effect within a narrow temperature range. All these experiments characterized the flexoelectricity under 70°C, which are not qualified for the high-temperature application.

Thus, to find a material whose dielectric permittivity would smoothly decline would be the direction of material

Table 1. Measured flexoelectric coefficient values and the dielectric permittivities of dielectric materials (at room temperature).

Material	ε_r	μ_{1122} (μC/m)	μ_{1111} (μC/m)	μ_{1212} (μC/m)
PMN	11,720	3.4 ¹⁸		
PZT	2130	1.4 ²²		
BST	13,200	20–100 ^{17,20}	120 ³³	110(f_{1212}) ³⁵
BT	2300	9 ¹⁹		
STO	300	4E–3 ²³	9E–3 ²³	3E–3 ²³
PMN–PT	~5000	70.8–100.8 ⁵⁷		

hunting. The gradual decline of flexoelectric coefficient μ_{12} with increasing temperature in BT–8BZT was reported by Huang *et al.*⁵⁹ The temperature range goes beyond 200°C. The measured flexoelectric coefficient μ_{12} of BT–8BZT is 25 $\mu\text{C}/\text{m}$ at 25°C and can remain above 12 $\mu\text{C}/\text{m}$ at temperatures up to 200°C. This suggests a new group of compositions in the BiMeO₃–BaTiO₃ system to be studied for its flexoelectricity, since these compositions exhibit temperature-stable permittivity over a much wider range.⁶⁰

On the other hand, a high Curie temperature material can also be considered for flexoelectricity. Very recently, Shu *et al.* have reported the flexoelectric behavior in PIN–PMT–PT single crystal.⁵⁴ The Curie temperature of PIN–PMT–PT is at 229°C but the crystal undergoes a macrodomain-to-microdomain transformation at 209°C. The maximum effective flexoelectric coefficient is 125 $\mu\text{C}/\text{m}$ measured at 209°C. But above Curie temperature, it decrease to 27 $\mu\text{C}/\text{m}$.

Although significant advances of flexoelectricity have been made in the past decades, the origin of the enhanced flexoelectricity has not been fully understood. In ceramics, interference factors, such as the grain boundary, were discussed because of their influence on the flexoelectricity.⁶¹ Single crystals, on the other hand, could minimize such effects. But fewer studies on single crystals were reported. Whether flexoelectricity is a bulk effect or a surface effect has been in debate. Catalan and co-workers' recent work on BaTiO₃ single crystal has emphasized the flexoelectricity with enlarged residual surface piezoelectricity.⁶² Shu *et al.* discovered the frequency dispersion phenomenon in PMN–PT at a relatively low frequency.⁵⁷ This dispersion occurred with a time delay but had a positive relationship with frequency. Thus it is proved that the flexoelectric effect acts more like a bulk effect rather than a surface effect. Meanwhile, it was revealed that for the single crystal, the frequency is highly related to the crystal structure, and it is easier to enhance by the ferroelectricity.

3.2. Thin films

Because the strain gradient is always inversely proportional to the material size, the induced flexoelectricity by strain gradient is expected to be larger in thin films compared with bulk materials. Due to large flexoelectric coefficient of BST bulk ceramics, the flexoelectricity in BST thin film was studied.⁶³ Also, from the principle, if the deformation applied to one structure is larger, the strain gradient would be larger.⁶⁴ Then the flexoelectric response is stronger. Polymer thin films, especially PVDF, have their advantages in this aspect. All the reported components of the flexoelectric coefficient matrix are listed in Table 2.

3.3. Two-dimensional crystalline membrane

As mentioned, flexoelectricity is strongly scale-dependent. Compared to thin film, 2D crystalline membranes are nearly

Table 2. Flexoelectric coefficient matrix components of PVDF.

μ_{1122} ($\mu\text{C}/\text{m}$)	μ_{1111} ($\mu\text{C}/\text{m}$)	μ_{1211} ($\mu\text{C}/\text{m}$)	μ_{3121} ($\mu\text{C}/\text{m}$)	μ_{1123} ($\mu\text{C}/\text{m}$)
34.5, 37.3 ³¹	−78.19, 1.6E−2 ^{65,66}	7.318E−4 ²⁶	1.037E−2 ²⁷	(9.2–9.6)E−3 ²⁹

atomistically thin. Though the flexoelectricity is negligibly small for many dielectrics on the conventional scale, it becomes dominant at the nanoscale. The scaling prediction was first proposed by Majdoub *et al.* and recently confirmed by various experimental evidences.¹⁴ Those experimental evidences indicated that the flexoelectricity has a promising application potential in charge transportation, domain engineering, random access memory and nanoscale actuating device.⁶⁷ For membranes, flexoelectric polarization can be simplified down to a simple proportional relation with the mean curvature of the membrane in the direction normal to the membrane. Similar to a thin paper-like structure, it is easy to induce a strain gradient by bending, so the researchers have focused on its potential applications. There have been rapid computational developments but rarely reported experimental results. Piezoelectricity of such materials was first explored for an enhanced electromechanical coupling performance. Graphene nitride nanosheet, riddled by triangular holes, was experimentally and computationally shown to exhibit an apparent piezoelectric response.⁶⁸ Piezoelectricity in materials such as BN, MoS₂ and WS₂ was calculated as well.^{67,69} The characterization data of flexoelectricity in these two-dimensional materials are yet missing.

Sharma *et al.* have published a thorough review on this topic including the biological membrane recently and their focus was primarily on mathematical and computational developments. They pointed out that the flexoelectricity has a number of implications in biology including ion transport, electromotility and mammalian hearing mechanism.^{67,73} They also summarized experimental findings in organic materials such as lipid bilayers.⁷³

3.4. Semiconductors and their flexo-like response

Although the flexoelectricity has always been studied in dielectric insulators, the insulation is not a formal requirement for the property. Theoretically, the polarization induced by strain gradients would be responsible for the redistribution of the free charge in semiconductors.

Catalan and co-workers reported the significantly enhanced flexoelectricity in vacuum-annealed BaTiO₃ semiconductor, which was attributed to a barrier layer mechanism.⁶² The mechanism of enhanced flexoelectricity in semiconductive, treated BTO single crystals is shown in Fig. 5. As reported, the effective flexoelectric coefficient reached 1 mC/m, increased by orders of magnitude since all conventional flexoelectric coefficients were reported in the order of $\mu\text{C}/\text{m}$.

Fig. 5. Effective flexoelectric behavior of semiconductive BaTiO₃.

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Serry and Sakr have reported a semiconductor structure with graphene–metal–silicone for multimodal energy conversion.⁷⁴ Flexoelectric behavior can be observed and further modulated in the fabrication of composite structures. During the fabrication, a mismatch occurred in the mechanical and thermal properties between different layers. At the same time, residual stresses exist at the interface of the structural layers induced from the thin-film deposition processes, which were tested through pure bending and thermomechanical tests. Therefore mechanical and thermal energy conversions are achieved through both flexoelectric and thermionic effects.

4. Applications

Although the studies of flexoelectricity in materials have been proceeding, the flexoelectric effect is still not as large as the piezoelectric effect. Designing new flexoelectric materials has become a fundamental issue in this newly arising field. Engineering the current materials to obtain great flexoelectric coefficient has become one trend to satisfy the standard of real applications, especially in passive sensing devices. Meanwhile, people started to introduce flexoelectricity into energy harvesting devices and structural health monitoring (SHM) sensors. In this section, the works in these two branches will be summarized.

4.1. Materials design

The generation of strain gradients should come from the external deformation of the material or the internal stress of the materials. Characterization techniques always introduce external force into the materials in order to observe large deformation. The internal stress of the material has rarely been engineered. Chu and co-workers have demonstrated a material type named as flexoelectric/piezoelectric metamaterials by using NBBT, PZT-51, PZT-81 and BST

ceramics.^{75–78} The fabrication method was asymmetric reduction, which was adopted from the fabrication of RAINBOW and THUNDER piezoelectric materials. Because of the reduction-induced chemical inhomogeneity, a strain gradient was generated so that the apparent electromechanical coupling effect was enhanced. As a result, its effective piezoelectric coefficients approached 57 pC/N at the temperature near Curie temperature, where the nominal piezoelectric device would become nonexistent.

On the other hand, design of composites using the currently known flexoelectric material has been undergoing. Chu *et al.* initially proposed a two-dimensional flexure composite model with BST sheets and tungsten balls.⁷⁹ Due to the existence of strain gradient, a giant electric polarization was exhibited in this composite and the effective piezoelectric coefficients were calculated to exceed 4300 pC/N. Shu and co-workers recently followed this model and systematically designed a flexure composite consisting of BST ceramic and the supporting material based on a parametric sweep finite element method (FEM) simulation. The design parameters are shown in Fig. 6.⁸⁰ The optimized composite structure can enhance the performance of the initial model up to 50 times by FEM simulation, with the thickness/length ratio of BST at 1/150, and with silicon carbide as the supporting material at 2.5 mm distance from the top and 2.1 mm distance from the bottom.

4.2. Devices: Sensors, energy harvester and microphone

Previously, in 2013, the potentials of nanogenerators were summarized and discussed.⁸¹ Since then, there have been some investigations on materials which exhibit high flexoelectric coefficients as we mentioned previously. However, the proper degradation of nanoscale structures is still needed for inventing flexoelectric devices. Up to now, flexoelectric devices have been designed mainly in two application

Fig. 6. Schematic of the flexure composite with geometry varied supporting material.

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directions, actuators and sensors, like nominal electromechanical converters by piezoelectricity.

Structural health monitoring in mechanical, architectural and transport vehicles has been the key area for flexoelectric detections. In such systems, defects like cracks would cause

catastrophic accidents. Because of the scaling effect of gradient, the sensitivity and accuracy of flexoelectric sensors could stand out among existing detection technologies. Huang and co-workers published the flexoelectric strain gradient sensor for structural health monitoring and crack

(a)

(b)

(c)

Fig. 7. (a) Optical image of an array of SrTiO_3 nanocantilevers. (b) Three-dimensional image of one SrTiO_3 nanocantilever with color scale corresponding to the out-of-plane displacement. (c) The digital holographic microscope experiment setup.

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detection.^{82–85} Due to the stress concentration at the surrounding of a crack, the strain gradient varies abruptly in the vicinity of the crack. The sensor was attached to the neighborhood of a cracking site. With varied tension stress, the induced charge due to flexoelectricity was measured to predict the cracking site. Tzou and co-workers have reported a distributed system for structural sensing by flexoelectricity.⁸⁶ A group of BST ceramic sensors were distributed on a cantilever flexoelectric beam and their flexoelectric constants were found consistent by the open-circuit model. An alternative for dynamic measurements of mechanical structures was provided.

The performance of micro/nanofabricated sensors can also be improved by adopting the flexoelectric effect. Kwon *et al.* first designed and fabricated a highly sensitive flexoelectric microphone, which can work at an extremely wide working frequency range based on barium strontium titanate ceramic.⁸⁷

As for energy harvesting, Zhang *et al.* reported a curved resonant flexoelectric actuator fabricated with PVDF.⁸⁸ Tzou and co-workers claimed the static nanocontrol beams.⁸⁹ Catalan and co-workers fabricated a pioneer cantilever actuator with strontium titanate integrated onto silicon, shown in Fig. 7.⁹⁰ A figure of merit (curvature divided by electric field) of 3.33 MV^{-1} was reported to be comparable to that of state-of-the-art piezoelectric bimorph cantilever. Similar structure was also made with PZT which showed the flexoelectricity can either enhance or suppress the piezoelectric response of the cantilever depending on the ferroelectric polarity and lead to a diode-asymmetric electromechanical response.

Besides all above known areas where flexoelectricity can show its advantages, thermal gradient-induced flexoelectric effects were first studied by Jiang and co-workers in bulk BST ceramics.⁹¹ Wave propagation within the microstructured solid was observed by Shen's group and Wei's group.^{92,93} These new applications have truly broadened the research on flexoelectricity.

5. Future Work and Challenges

So far, in the research field of flexoelectricity, researchers had still mainly focused on bulk dielectrics and some thin films. Only few findings have been reported in other material categories. How to further optimize the flexoelectric performance and how to design flexoelectric devices have become critical in this newly arising field. Nanoscale materials like 2D materials and some semiconductors may be advanced in terms of flexoelectricity. It may lead to an edge that flexoelectric effect would go beyond the conventional piezoelectric effect due to its scale and its thermal advantages. On one hand, the studies of its origin and the characterization of flexoelectric coefficient matrix in bulk dielectric are not convincing and completed. On the other hand, the potential applications in sensing and actuating devices urge developing

a classical flexoelectric material that is excluded from the piezoelectric materials. The composite design and the material search should keep going. Furthermore, more energy harvesting devices made with current known materials with high flexoelectricity should be designed and tested. The scaling down trend of miniature devices or the lab-on-chip industry needs new type of electromechanical devices that can be easily integrated with other elements on the chip at the nanoscale.

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