ABSTRACT

KWON, SEOL RYUNG. Flexoelectricity of Barium Strontium Titanate and Its Applications. (Under the direction of Xiaoning Jiang).

Electromechanical effects including piezoelectric, piezoresistive, capacitive and flexoelectric effects are ubiquitous in biological and material systems. Among them, the flexoelectric effect, the fundamental electromechanical coupling effect between electric polarization and mechanical strain/stress gradient, has drawn researchers’ attention because of its uniqueness. In crystallography, flexoelectricity exhibits in all dielectric solids while piezoelectricity can only be found in non-centrosymmetric crystal system. Moreover, flexoelectric effect is size-dependent effect that becomes more significant in micro/nano scale. Thus, it is important to explore flexoelectric effect of materials and to explore the associated sensor applications. In this dissertation research, the characterizations of barium strontium titanate, bulk ceramic and thin films, were carried out. Furthermore, several types of flexoelectric sensors for vibration and sound pressure monitoring were designed, fabricated and tested.

Firstly, BST unimorph type micro-cantilevers with the thickness ranging from 25 μm to 100 μm were prepared for measurement of the transverse flexoelectric coefficient $\mu_{12}$. The measured $\mu_{12}$ of bulk BST was about 10 μC/m, suggesting that BST possesses excellent flexoelectric properties.

In order to exploit the flexoelectric effect in BST thin films, BST thin film with thickness of 130 nm was deposited on silicon wafers using the RF magnetron sputtering
process. The flexoelectric coefficients of the thin films were then determined experimentally. It was revealed that the BST thin films possessed a transverse flexoelectric coefficient of 24.5 μC/m at Curie temperature (~ 28 °C) and 17.44 μC/m at 41 °C, respectively. The measured flexoelectric coefficients are comparable to that of bulk BST ceramics, which were reported to be 10 - 100 μC/m.

The relatively low flexoelectric coefficients of ferroelectrics inhibit the potential in developing flexoelectric sensing devices. Therefore, multilayered structure using BST ceramic was attempted to enhance effective flexoelectric coefficients. The performance of piezoelectric and flexoelectric cantilevers under the same dimensions and conditions was first compared analytically. Owing to the flexoelectric scaling effect, under the same force input, BST flexoelectric structure would generate higher charge output than that of its piezoelectric counterparts. Also, amplification of charge output using multilayered structure was experimentally verified. The prototyped structure consisted of three layers of 350 μm-thick BST plates with a parallel electric connection. The charge output of the multilayered structure was approximately 287 % of that obtained using a single-layer structure with the same total thickness and under the same end deflection input.

In order to demonstrate the sensing applications with the characterized flexoelectric BST ceramics, flexoelectric accelerometer was designed, fabricated and tested for vibration monitoring. The flexoelectric sensor is configured as a trapezoidal shape micro-unimorph with a BST layer bonded onto a steel substrate. A seismic mass was attached to the unimorph tip to amplify the transverse flexoelectric response of the BST layer. The theoretical model
was developed and validated by vibration tests using the prototyped flexoelectric micro-unimorph. The prototyped accelerometer showed a stable sensitivity of 0.84 pC/g in the frequency range of 100 Hz - 1.6 kHz.

A flexoelectric bridge-structured microphone using BST ceramic was designed and fabricated. The prototyped flexoelectric microphone was tested in an anechoic box using a loud speaker as the pressure source. Charge sensitivity of the flexoelectric microphone was measured and calibrated using a reference microphone. The 1.5 mm × 768 μm × 50 μm flexoelectric microphone showed a sensitivity of 0.92 pC/Pa, and the resonance frequency of 98.67 kHz. The signal to noise ratio was measured to be 74 dB. The analytical and experimental results suggest that the flexoelectric microphone is featured with both high sensitivity and broad bandwidth.

The findings from this dissertation research strongly suggest that micro/nano flexoelectric sensing is promising for a broad range of applications.
Flexoelectricity of Barium Strontium Titanate and Its Applications

by
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DEDICATION

To my father and mother who always love and support me.

I always thank God for being your daughter.

I always love you.
BIOGRAPHY

Seol ryung Kwon was born on June 24, 1983 in Daegu, South Korea. She received her Bachelor of Science in Mechanical Engineering from Kyungpook National University, Korea, in 2006. She received her Master of Science in Mechanical Engineering from the same university under the direction of Dr. Seunghan Yang in 2008. The title of her MS thesis was “Development of 6 D.O.F geometric error measurement system using PSDs for long travel range in mMT”.

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The following journal and conference proceeding publications were authored or co-authored by Seol ryung Kwon during her study at North Carolina State University.
Journal Articles


Pending Journal Articles

1. S. Kwon, W. Huang, S. Zhang, F. Yuan, and X. Jiang, "Study on a flexoelectric microphone using barium strontium titanate".
Conference Proceedings


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I would like to appreciate my advisory committee members, Dr. Fuh-Gwo Yuan, Dr. Paul Ro and Dr. Jon-Paul Maria for their valuable insight and suggestions. I always thank Dr. Fuh-Gwo Yuan whom I spent two and half years for the project with for his comments and advices during project meetings. Dr. Paul Ro has been my spiritual support since he is a religiously and professionally excellent example. Without the support from Dr. Jon-Paul Maria, I could not have accomplished the flexoelectricity study on BST thin films. Also, I’d like to acknowledge Dr. Shujun Zhang at Penn State for his helpful advices and support.

I am sincerely appreciative to my M.S. course advisor, Dr. Seung Han Yang. He led me to pursue higher academic career and enlightened me academically humanely. He also pushed me when I had a moment that I almost gave up pursuing Ph.D degree. Dr. Yongrae Ro is the person who guided me to this moment. I always thank him for taking care of me all the time.

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to my brother and sister-in-law for cheering me up and being proud of me all the time. Also, Yezi is my sister like friend. She is always there for me when I’m happy or feel alone. I’m a really lucky person to have their unconditional love and support. I really thank them for being proud of me. Without them, I could not have accomplished this Ph. D. course.
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1. INTRODUCTION

1.1. Flexoelectricity

1.1.1. Flexoelectric effect

Flexoelectric effect, defined as the linear coupling effect between the strain gradient and the induced electric polarization [1], has drawn scientific interest since it was first introduced 50 years ago. The direct flexoelectric effect describes the generation of an electric polarization response under a mechanical strain gradient and the converse flexoelectric effect describes the mechanical response under an electric field gradient. It can be simply explained in an ionic crystal structure as shown in Figure 1.1 [2-4]. If the ionic crystal is in the free status as shown in Figure 1.1 (a), it shows zero polarization. When the ionic crystal is under the homogeneous strain (Figure 1.1 (b)), the centers of the negative and positive charges coincide again resulting in a zero net polarization. However, if the cell is under inhomogeneous strain as shown in Figure 1.1 (c), the displacement of the centers of the negative charge and positive charge differs from each other, creating a dipole moment in the opposite direction to the strain gradient.

As an example, flexoelectric effect can be further explained as shown in Figure 1.2. When a plate is bent, the upper part of the plate undergoes a tensile stress/strain and the lower part experiences a compressive stress/strain. Then, the strain gradient is formed in the material along the thickness direction and the flexoelectric polarization $P$ associated charges is induced on the top and bottom surfaces.
1.1.2. Flexoelectric constitutive equation and constants

The concept of flexoelectric effect was first introduced in the 1960s by Kogan [3]. Originally, flexoelectric effect was considered as one type of piezoelectric effect, which refers to the electric polarization induced by the stress. However, the cause of electric polarization by the flexoelectric effect was found to be different from piezoelectric effect. Unlike piezoelectric effect, flexoelectric effect is associated with the spatial derivative of strain, that is, the strain gradient.

In general, a polarization from dielectric materials can be induced from both flexoelectric and piezoelectric effects. The total electric polarization $P$ induced by the two effects is expressed as

$$ P_i = d_{ijk} \sigma_k + \mu_{ijkl} \frac{\partial \varepsilon_{jk}}{\partial x_i} $$  \hspace{1cm} (1.1)

where $d_{ijk}$, $\sigma_k$, $\mu_{ijkl}$, $\varepsilon_{jk}$ and $x_i$ are the piezoelectric constant, stress, flexoelectric constant, mechanical strain and position coordinate, respectively [3, 5, 6]. The first term is the polarization induced by the piezoelectric effect and the second is the one by the flexoelectric effect. As indicated, the piezoelectric polarization is induced by the mechanical stress, while the flexoelectric polarization is associated with the strain gradient. Piezoelectric effect exists in only 20 non-centrosymmetric crystal point groups out of 32, while the flexoelectric effect exists in all 32 point groups. Thus, piezoelectric effect is absent in centro-symmetric materials leading to the following equation.

$$ P_i = \mu_{ijkl} \frac{\partial \varepsilon_{jk}}{\partial x_i} $$  \hspace{1cm} (1.2)
As the strain gradient can break the inversion symmetry, flexoelectric polarization can be induced from the lattice deformation in all dielectric materials. Through the harmonic decomposition and Cartan decomposition of the group theory, the number and types of the rotational symmetries for flexoelectric tensor are given as

\[ \text{[7]. Among these symmetry groups, 432, } 4m3, m\bar{3}m \text{ cubic materials are generally chosen for the study of flexoelectricity because of the high permittivity, broad material choice and wide temperature range. As shown in Table 1.1, there are three independent components } \mu_{1111}, \mu_{1122}, \mu_{1212} \text{ (} \mu_{11}, \mu_{12}, \mu_{44} \text{ - matrix notion) in these cubic materials.}

The tensor matrix of flexoelectric constants of the cubic materials can be written as

\[
\begin{pmatrix}
\mu_{1111} & \mu_{1122} & \mu_{1212} & 0 & 0 & 0 \\
\mu_{1122} & \mu_{1111} & \mu_{1122} & 0 & 0 & 0 \\
\mu_{1212} & \mu_{1122} & \mu_{1111} & 0 & 0 & 0 \\
0 & 0 & 0 & \mu_{1212} & 0 & 0 \\
0 & 0 & 0 & 0 & \mu_{1212} & 0 \\
0 & 0 & 0 & 0 & 0 & \mu_{1212}
\end{pmatrix}
\]

(1.3)

\( \mu_{1111} \text{ or } \mu_{11} \) corresponds to longitudinal mode, \( \mu_{1122} \text{ or } \mu_{12} \) to transverse and \( \mu_{1212} \text{ or } \mu_{44} \) to shear mode as shown Figure 1.3.

The flexoelectric coefficients were further studied by Tagantsev [1]. In his studies, it was found that the flexoelectric coefficient scales with the dielectric susceptibility of the material as follows

\[
\mu \sim \tilde{\chi} \frac{e}{a}
\]

(1.4)
where \( \chi \) is the susceptibility of the dielectric, \( e \) is the electron charge and \( a \) is the lattice constant. According to Tagantsev, there are several factors that can contribute to this phenomenon, such as the static bulk flexoelectricity, dynamic bulk flexoelectricity and surface flexoelectricity. However, Resta [8] discovered that the dynamic contribution due to the long wavelength phonons found to be the same as the bulk contribution. And in his study of centrosymmetric materials, there does not exist surface contribution from the non-zero quadrupole moment associated with ions which reside in the thin surface layer as Tagantsev suggested. But the existence of surface contribution is still unclear. Nonetheless, the effect of dielectric susceptibility in flexoelectric effect were experimentally verified [9-12]. Ma and Cross’ group revised the Eq. (1.4) for the coefficient and included the material dependent scaling factor \( \gamma \) as follows

\[
\mu_{ijkl} = \gamma \chi_{ijkl} \frac{e}{a}
\]  

(1.5)

Originally, Tagantsev proposed that \( \gamma = 1 \), however, none of ceramics showed this behavior. The studies by Ma and Cross’ group reported the scaling factors of several ceramics. The scaling factors of BST ceramic, barium titanate, lead magnesium niobate and lead zirconate titanate were found to be \( \gamma_{\text{BST}} = 9.3 \), \( \gamma_{\text{BT}} = 12 \), \( \gamma_{\text{PMN}} = 0.65 \) and \( \gamma_{\text{PZT}} = 0.57 \) [5]. Up to now, reasons of this factor remain unclear. However, these observations suggests that the chemical makeup of the perovskite structure could cause the difference in flexoelectricity [13].
1.2. Merits of flexoelectric effect

The flexoelectric effect has received comparatively less attention than piezoelectric effect. As mentioned before, in large scale, such as centimeter or millimeter scales, the flexoelectric effect does not play a big role because of the weak flexoelectric coupling in the macro domain. However, the flexoelectric effect has attracted increasing attention for several decades because very strong flexoelectric effect in several ferroelectrics and in micro/nano scale was found [14-17].

1.2.1. Aging rate

It has been known that piezoelectric ceramics have a long-term polarization related aging problem after polarization [18]. Hence, when these materials are used in sensors and actuators, the decrease of efficiency or sensitivity should be considered and compensated properly to maintain the accuracy of devices. Aging of piezoelectric ceramics occurs rapidly in the first few hours after removal of the poling field. After few days, the changes of piezoelectric constants become much more slowly. Generally, the aging rate \( A \) can be defined by the equation, \( \frac{y(t)}{y(t_0)} = 1 + A \cdot \log\left(\frac{t}{t_0}\right) \), where \( y \) is the concerned materials property, \( t_0 \) is an arbitrary time when the measurement started, and \( t \) is the elapsed time [19].

Unlike piezoelectric ceramics, flexoelectric materials do not have the polarization related aging problem due to the absence of poling process during utilization of the flexoelectric materials. To demonstrate the zero aging rate of flexoelectric materials, \( \mu_{12} \) coefficient was measured with time and compared with the reported aging properties of the piezoelectric coefficients in piezoelectric materials [20, 21]. The piezoelectric coefficients \( (d_{33} \text{ and } d_{31}) \)
and flexoelectric coefficient ($\mu_{12}$) were normalized by deriving the real time values to that measured just after poling. As shown in Figure 1.4, the fit of BST has 0.0004 % per decade aging rate that is in the error range and can be considered as zero. Compared to other piezoelectric materials, flexoelectric properties are invariant over time.

1.2.2. Scaling effect

The flexoelectricity has a unique intrinsic effect called the scaling effect. Due to the fact that it is based on strain gradient, not strain, the larger polarization can be generated if the structures are scaled down. This fact makes the flexoelectric sensors more attractive in micro/nano scales [22].

The flexoelectric coefficients can be converted to effective piezoelectric coefficients for comparison. Huang et al. [14] calculated the effective piezoelectric coefficient $d_{33}^{\text{eff}}$ of a flexoelectric BST microcantilever. It is expressed as

$$d_{33}^{\text{eff}} = \frac{6\mu_{12}l^3}{Eh^3}$$

(1.6)

Here $h$ is the thickness of the beam, $E$ is the Young’s modulus of BST ($E=153$ GPa), $l$ is the length of the beam and $\mu_{12}$ is flexoelectric coefficient. In the equation, $d_{33}^{\text{eff}}$ is inversely proportional to the cubic of the thickness of the beam. Thus, it can be significantly enhanced in micro/nano scale. In addition, the BST cantilevers were compared to other piezoelectric bimorphs of ZnO, PZT and PMNT single crystal. $d_{33}^{\text{eff}}$ of these materials are shown in Figure 1.5 as a function of thickness $h$. The effective piezoelectric coefficient of flexoelectric structures can be larger than those of piezoelectric materials with high piezoelectric
coefficients when the thickness of cantilevers is scaled down. Therefore, flexoelectric micro/nano structures can be promising because of their significantly enhanced effective piezoelectric coefficients due to the intrinsic scaling effect of the flexoelectric effect.

1.3. Dissertation outline

The main goal of this research is to investigate the characterization of flexoelectric materials and the design, fabrication and testing methods of flexoelectric sensors, including the acceleration and sound pressure sensing. The dissertation consists of 7 chapters and references. Each chapter is briefly described as follows.

Chapter 1 reviews the fundamentals of the flexoelectric effect, derivation of flexoelectric constitutive equations and advantages as sensing materials. Chapter 2 presents the measurement of BST transverse flexoelectric coefficient \( \mu_{12} \). It includes the characterization method, fabrication, test method and results. In Chapter 3, the characterization of BST thin film that was fabricated by RF magnetron sputtering is presented. The characterization of dielectric behavior, piezoelectricity, topography and flexoelectric coefficient of the fabricated BST thin film was carried out. Chapter 4 investigates the multi-layered BST structure. The comparisons of a flexoelectric structure vs. a piezoelectric counterpart and a single layered structure vs. a multilayered structure are presented in the chapter showing the enhancement in flexoelectricity. Chapters 5 and 6 show applications of flexoelectric effect. In Chapter 5, a flexoelectric accelerometer was designed, fabricated and tested. A unimorph type of a flexoelectric accelerometer with a proof mass at the tip was investigated. Chapter 6 presents a flexoelectric microphone. In order to satisfy high resonance and sensitivity simultaneously,
a BST ceramic bridge structure was designed to have a thickness of 50 μm. The optimization for electrode shape is also discussed. Finally, Chapter 7 summarizes the major conclusions of this dissertation, and recommendations for future research.
Figure 1.1. Schematic illustration of flexoelectric effect of an ionic crystal. (a) Free status without strain, (b) Under homogeneous strain and (c) Under inhomogeneous strain.

Figure 1.2. Schematic illustration of a flexoelectric structure. When the plate is bent, polarization is induced by strain gradient between the compressive (red arrows) strain under the neutral surface and the tensile (blue arrows) strain above the neutral surface.
Figure 1.3. Three modes of flexoelectric effect of the cubic materials. The yellow areas are electrodes.

Figure 1.4. Aging of the piezoelectric and flexoelectric coefficients [23].
Figure 1.5. Effective piezoelectric coefficient ($d_{33}^{\text{eff}}$) of piezoelectric bimorphs and flexoelectric microcantilevers [14].
Table 1.1. Numbers of the non-zero independent components of flexoelectric coefficient of solid materials in point and Curie group.

<table>
<thead>
<tr>
<th>Point and Curie groups</th>
<th>Numbers of independent components of $\mu_{ijkl}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1, $\bar{1}$</td>
<td>54</td>
</tr>
<tr>
<td>2, $m$, $2/m$</td>
<td>28</td>
</tr>
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<td>3, $\bar{3}$</td>
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<td>4, $4\bar{m}$, $4/m$</td>
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<td>4$mm$, $42m$, 422, $4/mmm$</td>
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<td>6, $\bar{6}$, $6/m$, $\infty$, $\infty m$</td>
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<td>23, $m3$</td>
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2. CHARACTERIZATION OF FLEXOELECTRIC BULK CERAMICS

2.1. Background

Flexoelectricity exists in all dielectric materials. A variety of dielectric materials have been studied for their flexoelectricity. Cross’ group has studied lead magnesium niobate (PMN, \( \mu_{12} \sim 4 \ \mu\text{C/m} \) at room temperature) [10], barium strontium titanate (BST, \( \mu_{12} \sim 100 \ \mu\text{C/m} \) at Curie temperature) [11], lead zirconate titanate (PZT, \( \mu_{12} \sim 1.4 \ \mu\text{C/m} \) at room temperature) [24] and barium titanate (BT, \( \mu_{12} \sim 50 \ \mu\text{C/m} \) at Curie temperature) [13]. Zubko [9] reported that SrTiO\(_3\) single crystal exhibited a large dielectric polarization induced by bending, with flexoelectric coefficients on the order of \( 10^{-9}-10^{-8} \ \text{C/m} \). Flexoelectricity in polymers, such as polyvinylidene fluoride (PVDF, \( \mu_{12} \sim 13 \ \text{nC/m} \) at room temperature), polyethylene terephthalate (PET, \( \mu_{12} \sim 9.9 \ \text{nC/m} \) at room temperature), polyethylene (\( \mu_{12} \sim 5.8 \ \text{nC/m} \) at room temperature) and epoxy (\( \mu_{12} \sim 2.9 \ \text{nC/m} \) at room temperature) were experimentally studied. Results suggest that polymers with low flexoelectricity can still be used to obtain high effective \( d_{33} \) by taking advantage of their low elastic modulus [16]. Recently, complete flexoelectric tensor of SrTiO\(_3\) was reported [9]. Of all the aforementioned materials, BST has garnered the most attention because of its largest reported flexoelectric properties at its Curie temperature of 21 °C among all studied dielectric materials as shown in Table 2.1.
2.1.1. Characterization method for $\mu_{11}$

There are several reported methods to characterize axial flexoelectric coefficient $\mu_{11}$. A truncated pyramid structure and trapezoid shaped cantilever were adopted for characterization of $\mu_{11}$ in order to generate strain gradient in the thickness direction.

As shown in Figure 2.1, Cross [25, 26] introduced a truncated pyramid of Ba$_{0.67}$Sr$_{0.33}$TiO$_3$ ceramic to characterize $\mu_{11}$. The truncated pyramid has an upper square face of $a_1$ and lower face of $a_2$ and the sidewall is configured so that $a_2$ is a linearly increasing function of $d$ the depth from $a_1$ to $a_2$. Force $F$ is applied normal to upper and lower surfaces. Stress in the upper surface is $T_{3(1)} = F / a_1^2$ and strain is $S_{3(1)} = F / a_1^2 c_{11}$ where $c_{11}$ is the elastic constant of the truncated pyramid. Similarly for the lower surface, $T_{3(2)} = F / a_2^2$ and $S_{3(2)} = F / a_2^2 c_{11}$.

Strain gradient along thickness direction becomes

$$\frac{\partial S_3}{\partial d} = \frac{S_{3(1)} - S_{3(2)}}{d} = \frac{F}{d} \left( \frac{1}{a_1^2} - \frac{1}{a_2^2} \right) \frac{1}{c_{11}}$$

(2.1)

Then polarization due to flexoelectricity is

$$P_3 = \mu_{11} \frac{\partial S_3}{\partial d} = \mu_{11} \frac{F}{d} \left( \frac{1}{a_1^2} - \frac{1}{a_2^2} \right) \frac{1}{c_{11}}$$

(2.2)

With Eq. (2.2), axial flexoelectric coefficient can be calculated from measured polarization and force and given dimensions. Cross measured $\mu_{11}$ of BST to be 100 $\mu$C/m. This value corresponds to piezoelectric coefficient $d_{33}$ by 60 pC/N for $a_1=50$ $\mu$m, $a_2=250$ $\mu$m and $d=250$ $\mu$m case and by 600 pC/N for $a_1=5$ $\mu$m, $a_2=25$ $\mu$m and $d=25$ $\mu$m case. It shows much higher value due to the scaling effect.
Baskaran et al. [27] characterized $\mu_{11}$ of $\alpha$-phase polyvinylidene fluoride (PVDF) films. In order to generate strain gradient, a trapezoid shaped cantilever was fabricated. $\alpha$-phase PVDF was used to exclude piezoelectricity because it is non-piezoelectric material unlike $\beta$-phase PVDF. 13.5 $\mu$m thick PVDF thin film was diced into two shapes of pieces with the rectangle shape and the trapezoid shape as shown Figure 2.2(a) and Figure 2.2(c), respectively. The setups are shown in Figure 2.2(b) and Figure 2.2(d). The strain of the rectangle shape film can be estimated as

$$S_{\text{ww}} = \Delta W / W_r$$  \hspace{1cm} (2.3)

here, $W_r$ is defined in Figure 2.2 and $\Delta W$ is the measured deformation by a tensile drawing tester (TDT). The strains of the trapezoid shape film at both ends are

$$S^B_{\text{ww}} = \Delta W / W_r \text{ (at the B end)}$$  \hspace{1cm} (2.4)

$$S^A_{\text{ww}} = (\Delta W \times h_B) / (W_r \times H_r) \text{ (at the A end)}$$  \hspace{1cm} (2.5)

Then, the average strain and strain gradient are

$$\bar{S}_{\text{ww}} = \frac{S^B_{\text{ww}} + S^A_{\text{ww}}}{2}$$  \hspace{1cm} (2.6)

$$\nabla S_{\text{ww}} = \frac{S^B_{\text{ww}} - S^A_{\text{ww}}}{W_r}$$  \hspace{1cm} (2.7)

The induced polarization of the trapezoid shape film comes from the residual piezoelectricity and flexoelectricity as follows

$$P_1 = d_{33} E \bar{S}_{\text{ww}} + \mu_{14} \nabla S_{\text{ww}}$$  \hspace{1cm} (2.8)
where, $E$ is Young’s modulus of PVDF. From a rectangular PVDF film, $d_{33}$ of PVDF was found to be approximately 1.58 pC/N. With measured polarization from a lock-in-amplifier and deformation information from TDT, the $\mu_{11}$ of PVDF was measured to be 81.5 $\mu$C/m.

2.1.2. Characterization methods for $\mu_{12}$

Figure 2.3(a) shows a free-standing cantilever type test setup [10] for characterization of transverse flexoelectric coefficient $\mu_{12}$, where one end of a cantilever was clamped and the other end was actuated by a loudspeaker with a sinusoidal signal. When the cantilever is under excitation, the strain gradient was generated along the thickness direction of the cantilever. The deflection $W(x_1)$ of the cantilever was detected by a displacement sensor. As shown in Figure 2.3 (b), the strain gradient along the thickness direction can be calculated as

$$\frac{\partial \varepsilon_{11}}{\partial x_3} = \frac{\partial^2 W(x_1)}{\partial x_1^2}$$

(2.9)

Due to the strain gradient, the polarization induced by flexoelectric effect can be inverted to the form of an electric current. The relationship between the induced polarization and the current is described as

$$P_3 = \frac{i}{2\pi fA}$$

(2.10)

where $P_3$, $i$, $f$ and $A$ are the induced polarization, current, driving frequency and electrode area. Then, the flexoelectric coefficient $\mu_{12}$ can be calculated from the strain gradient and observed current.
Finally, the flexoelectric coefficient \( \mu_{12} \) has a linear relationship between the induced polarization and strain gradient.

There is another method to characterize flexoelectric coefficient \( \mu_{12} \). Zubko et al. [9] used three point bending method to characterize single crystal paraelectric strontium titanate (SrTiO\(_3\) - STO). As shown in Figure 2.4, a tensile stress was generated on the bottom side and compressive stress on the top so that the strain gradient induced along the thickness direction, resulting in a flexoelectric polarization. Thermally controlled chamber was adopted for ruling out piezoelectric and ferroelectric contribution by increasing temperature above Curie temperature.

As a result, three flexoelectric coefficient \( \mu_{1111}, \mu_{1122}, \) and \( \mu_{1212} \) of STO were experimentally measured and found to be \( \mu_{1111} = 0.2 \ \text{nC/m}, \mu_{1122} = 7 \ \text{nC/m}, \) and \( \mu_{1212} = 5.8 \ \text{nC/m}. \)

2.2. Unimorph cantilever method

The unimorph type cantilever was adopted for characterization of \( \mu_{12} \) because of the fact that free-standing BST cantilevers with thickness of tens of microns cannot be easily realized without any support layer due to the material fragility. As shown in Figure 2.5, the unimorph cantilever structure is composed of one flexoelectric layer and one supporting layer. The tip force at one end creates a normal strain gradient along the thickness direction, inducing the electric polarization.
Based on Euler-Bernoulli beam theory, the radius of curvature $R$ of a unimorph under a bending moment induced by force $F$ at the tip can be given as

$$\frac{1}{R(x_i)} = \frac{\partial^2 w(x_i)}{\partial x_i^2} = \frac{12Fs_{11}'s_{11}'(s_{11}'t_s + s_{11}'t_f)}{Kb}(L-x_i) \quad (2.12)$$

where

$$K = 4s_{11}'s_{11}'(t_f)^3 + 4s_{11}'s_{11}'(t_s)^3 + (s_{11}'(t_s)^2(t_f)^4 + (s_{11}'(t_f)^2(t_s)^4 + 6s_{11}'s_{11}'(t_s)^2(t_f)^2) \quad (2.13)$$

The superscript or subscript $s$ denotes the supporting layer and $f$ denotes the flexoelectric material. $x$, $s_{11}$, $t$, $b$ and $R$ are the axial distance from the clamped end, compliance, and thickness, width of the unimorph and radius of curvature, respectively.

And the tip deflection can be derived as

$$w_L = \frac{4s_{11}'s_{11}'(s_{11}'t_s + s_{11}'t_f)L^2}{Kb}F \quad (2.14)$$

Since the gradient of axial normal strain along the thickness direction of the unimorph can be given as

$$\frac{\partial \varepsilon_{11}}{\partial x_3} = \frac{\partial}{\partial x_3} \left( \frac{x_3}{R(x_i)} \right) = \frac{1}{R(x_i)} \quad (2.15)$$

The electric polarization $P$ from the unimorph can be written as follows [28, 29]

$$P_f = \mu_{11} \frac{\partial \varepsilon_{33}}{\partial z} + \mu_{12} \left( \frac{\partial \varepsilon_{11}}{\partial z} + \frac{\partial \varepsilon_{22}}{\partial z} \right) = [\nu\mu_{11} + (1+\nu)\mu_{12}] \frac{\partial \varepsilon_{11}}{\partial z} \quad (2.16)$$

Here, $\nu$ is the Poisson ratio of a flexoelectric ceramic. For pure bending of beam with $L \gg b \gg t$, the flexoelectric polarization due to the gradient of axial normal strain $\varepsilon_{11}$ in the thickness direction can be simplified as
\[ P_f = \mu_{12}^{\text{eff}} \frac{\partial e_{11}}{\partial z} \]  

(2.17)

\( \mu_{12}^{\text{eff}} \) denotes the effective transverse flexoelectric coefficient. For convenience, \( \mu_{12}^{\text{eff}} \) will be abbreviated to \( \mu_{12} \). Then, the total charge \( Q \) can be obtained by combining Eqs. (2.15), (2.16) and (2.17).

\[ Q = \mu_{12} \int_{-L}^{L} \frac{\partial e_{11}}{\partial x_3} b dx_1 = \frac{6\mu_{12} s_{11}^{s} s_{11}^{f} (s_{11}^{s} t_x + s_{11}^{f} t_f) L^2}{K} F \]  

(2.18)

Finally, the flexoelectric coefficient \( \mu_{12} \) can be calculated from the measured charge output and tip deflection as follows.

\[ \mu_{12} = \frac{2LQ}{3bwL} \]  

(2.19)

2.3. Fabrication of unimorphs

\( \text{Ba}_{0.65}\text{Sr}_{0.35}\text{TiO}_3 \) was prepared by conventional solid state processing using raw materials including \( \text{BaCO}_3 \) (99.9\%, Alfa Aesar), \( \text{SrCO}_3 \) (99.9\%, Alfa Aesar) and \( \text{TiO}_2 \) (99.99\%, Alfa Aesar). All the materials were weighted for the composition of BST to be \( \text{Ba}:\text{Sr}=65\%:35\% \). The powders were milled in anhydrous ethanol for one day, and then calcined at 1200 °C for 2 hours to decompose the carbonate and synthesize the BST powder. The obtained powder was milled and calcined again at the same temperature and then granulated through an 80-mesh sieve. The powder was pressed into pellets with a minimum of 1 inch diameter using cold isotropic pressing at 30 ksi. The pellets were sintered at 1300-1350 °C and cooled down to room temperature with a cooling rate of 3 °C/min in an oxygen atmosphere. The ceramic
had the relative dielectric constant of 8000 at room temperature, and its maximum dielectric constant is about 14000 at the Curie temperature of 18 °C.

In order to fabricate unimorphs, a BST plate was bonded on a silicon substrate and lapped down to various thicknesses as shown in Figure 2.6. Firstly, the silicon substrates were lapped down to 200 μm thick. Then, 100 Å of Ti and 1000 Å of Au were sputtered as electrodes on the topside of silicon substrates and bottom side of the BST plates. The sputtered sides of the BST and the silicon substrate were bonded with epoxy (EP301, Epoxy Technology, Billerica, MA). Normal pressure of 1 MPa was applied to produce the bonding layer thickness of about 4 ~ 5 μm. This also ensured a good adhesion, and hence good electric conductivity, between the BST and the silicon substrate. Then, the BST layers were lapped into 25, 50, 100 μm, respectively. And the top surface of the BST layer was sputtered for an electrode as mentioned above. Finally, the bonded Si-BST stacks were diced (DAD321, Disco Corp., Tokyo, Japan) into designed cantilever dimensions.

2.4. Experimental methods

The experimental setup is shown schematically in Figure 2.7. Firstly, the unimorphs were clamped rigidly at one end. Then, the piezoelectric actuator was placed at the tip to generate deflection of the cantilever. The actuator was driven by a power amplifier (Type 2706, Brüel & Kjær, Nærum, Denmark) and a function generator (AFG3101, Tectronix Inc., Beaverton, OR). A sinusoidal 2~5 μm peak-to-peak tip displacement was applied at the frequency of 2 Hz. Low frequency was chosen to ensure that the real deflection of the cantilever follows the input of the piezoelectric actuator. The deflection of the cantilever at the tip was measured by
a laser vibrometer (OFV-5000, Polytec, Dexter, MI). The generated polarization output signal was amplified by a charge amplifier (Type 2635, Brüel & Kjær, Nærum, Denmark) and the amplified signal was measured by a lock-in amplifier (SR830, Standard research system, Sunnyvale, CA). A lock-in amplifier was used to rule out other signals with the frequency not of interest.

2.5. Results and discussion

2.5.1. Dielectric behavior

The capacitance of the BST was measured at 1 kHz by an impedance analyzer (4294A, Agilent, Santa Clara, CA). Dielectric constants were calculated from the capacitance and are shown as a function of temperature, as shown in Figure 2.8. It was observed that the dielectric constant of the BST at room temperature is about 8000 and reaches a peak at 29 ºC. Above 29 ºC, the dielectric constant decreases again. This means that at temperature below 29 ºC, which is the Curie temperature, BST is in a ferroelectric phase and tetragonal structure. At temperature above 29 ºC, it is in a paraelectric phase and cubic structure, and thus, there is no piezoelectricity.

2.5.2. Flexoelectric coefficient

The flexoelectric coefficient $\mu_{12}$ of three unimorphs with different thicknesses was measured at 2 Hz at room temperature. Each experiment was performed 5 times. In Figure 2.9, all the points represent the averages and the bars represent standard deviation of each point.
25, 50 and 100 μm thick unimorphs showed linear relationship between strain gradient and polarization. The $\mu_{12}$ calculated from the slopes was found to be about 10 μC/m.

2.6. Conclusion
In summary, BST bulk ceramic - silicon unimorphs were fabricated and tested for dielectric and flexoelectric characterizations. The measured Curie temperature is 29 °C and peak dielectric constant is 8000. The measured transverse flexoelectric coefficient $\mu_{12}$ of 10 μC/m remains constant for unimorphs with various thicknesses. This large flexoelectricity in BST bulk ceramic is promising for flexoelectric sensing applications.
Figure 2.1. Schematic drawing of the flexoelectric composite for $\mu_{11}$ characterization [25].
Figure 2.2. Measurement setup for characterization of $\mu_{11}$. C: current measurement, FG: function generator, LIA: lock-in amplifier, MS: Microstrain DVRT probing sensor and TDT: tensile drawing tester. (a) The rectangle shape of PVDF film (b) The lock-in detection setup for piezoelectric measurement (c) The trapezoid shape PVDF film and (d) The lock-in detection setup for piezoelectric and flexoelectric measurement [30].
Figure 2.3. (a) Experimental setup for measurement of the flexoelectric effect and (b) Schematic diagram of a cantilevered beam and the axial arrangement [10].
Figure 2.4. Experimental setup for flexoelectric measurements [9].

Figure 2.5. Schematic of a flexoelectric BST unimorph.
Figure 2.6. Fabrication of BST unimorphs (a) Schematic cross-section of the unimorph and (b) photograph of a BST unimorph.
Figure 2.7. (a) Schematic view of the experimental setup for the transverse flexoelectric coefficient measurement and (b) Photograph picture of the experimental setup.
Figure 2.8. Dielectric behavior of the BST ceramic
Figure 2.9. Measured relationship between polarization and strain gradient of three unimorphs.
Table 2.1. Flexoelectric constant of reported ceramics and its characterization method.

<table>
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<tr>
<th>Author (Year)</th>
<th>Material</th>
<th>Characterization method</th>
<th>Type</th>
<th>Flexoelectric coefficient</th>
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<td>Ma (2001)</td>
<td>PbMg$<em>{1/3}$Nb$</em>{2/3}$O$_3$</td>
<td>Cantilever bending</td>
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<td>4.0 $\mu$C/m @ RT</td>
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<td>Ma (2002)</td>
<td>Ba$<em>{0.67}$Sr$</em>{0.33}$TiO$_3$</td>
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<td>PZT</td>
<td>Cantilever bending</td>
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<tr>
<td>Ma (2006)</td>
<td>BT</td>
<td>Cantilever bending</td>
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<td>50.0 $\mu$C/m @ Tc (120 °C)</td>
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<td>Zhu (2006)</td>
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<td>Zubko (2007)</td>
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<td>$\mu_{44}$</td>
<td>5.8 nC/m @ RT</td>
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3. CHARACTERIZATION OF BST THIN FILM

3.1. Background

As mentioned in section 1.2.2, flexoelectric effect inherently possesses the scaling effect [14, 15, 31]. To take advantage of this, the structure should be minimized as small as possible.

In order to obtain flexoelectric BST micro/nano-sensors, top-down or bottom-up micro/nanofabrications can be adopted. However, the top-down process using BST bulk materials remains to be challenge due to its material’s fragility and the low etch rate [32]. In addition, it is difficult to grow single crystals of ferroelectric materials with the high permittivity that leads to a high flexoelectric coefficient [11]. On the other hand, the bottom-up fabrication technique is preferred because that deposition of thin film dielectric materials has been reported with successes. Among various thin film fabrication methods [33-36], RF magnetron sputtering is favorable due to its ease of implementation, superior compositional reproducibility and controllability, good crystallinity, and desirable dielectric properties.

In this chapter, a flexoelectric thin film structure was designed and fabricated using the RF magnetron sputtering process. The dielectric, piezoelectric and flexoelectric properties of the fabricated BST thin film were then characterized to determine the flexoelectric transverse coefficient of BST thin films using a bending method.
3.2. Characterization of $\mu_{12}$

A beam bending method was developed to characterize the flexoelectric properties of BST thin films. For pure bending of a beam with $L \gg b \gg t$ (Figure 3.1(a)), the flexoelectric polarization due to the gradient of axial normal strain $\varepsilon_{11}$ in the thickness direction can be simplified and written as Equation (3.1) [28, 37]. Here, $\mu_{12}^{\text{eff}}$, $\nu$, $z$, and $\varepsilon$ denote the effective transverse flexoelectric coefficient, the Poisson ratio of BST thin film, position coordinate in $Z$ direction and elastic strain, respectively.

$$P_z = \mu_{11} \frac{\partial \varepsilon_{35}}{\partial z} + \mu_{12} \left( \frac{\partial \varepsilon_{11}}{\partial z} + \frac{\partial \varepsilon_{22}}{\partial z} \right) = [\nu \mu_{11} + (1 + \nu) \mu_{12}] \frac{\partial \varepsilon_{11}}{\partial z} = \mu_{12}^{\text{eff}} \frac{\partial \varepsilon_{11}}{\partial z} \quad (3.1)$$

From the Euler-Bernoulli beam theory, the strain gradient can be calculated as

$$\frac{\partial \varepsilon_{11}}{\partial z} = \frac{\partial^3 w(x)}{\partial x^3} = \frac{F(L-x)}{EI} \quad (3.2)$$

where, $x$, $w(x)$, $F$, $L$, $E$ and $I$ are the axial distance from the clamped end, deflection at $x$, applied force at the tip, beam length, Young’s modulus of substrate, and moment of inertia, respectively. Then, the total induced charge $Q$ can be calculated by integrating of the polarization. Here, $\delta$, $r$ and $l$ are the tip deflection, radius of top electrode and distance from the base of cantilever to the center of the top electrode as shown in Figure 3.1(a).

$$Q = \int P_z dA = \mu_{12}^{\text{eff}} \frac{\pi r^2 F(L-l)}{EI} = \mu_{12}^{\text{eff}} \frac{3\pi r^2 \delta(L-l)}{L} \quad (3.3)$$

Finally, the transverse flexoelectric coefficient, $\mu_{12}^{\text{eff}}$, can be calculated from the output charge and the displacement at the tip. The dimensions and material properties of the structure are shown in Table 3.1.
3.3. Fabrication of BST thin film

Firstly, a 22.5 nm of SiO$_2$ layer was grown on a silicon wafer to prevent charge leakage. A layer of TiO$_2$/Pt (72.4nm/130 nm) was then deposited as the bottom electrode. Due to its similar atomic lattice structure with BST, the TiO$_2$ layer acts as a template for the growth of the BST film and also aids in minimizing the residual strain induced by lattice mismatch. BST thin film was then deposited by RF magnetron sputtering using a Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ target. BST was deposited on the bottom electrode at 300 ºC with a shadow mask to expose some area of the bottom electrode for electrical connection. It was post annealed in air at 700 ºC for 20 hours. Finally, a top electrode of Ti/Au (1 nm/10 nm) was deposited in a circular shape with the diameter of 832 μm using a shadow mask. Detailed conditions of BST the thin film deposition and a schematic view of the structure are shown in Table 3.2 and Figure 3.1(b), respectively. The microstructure and crystallization of the BST thin film were examined by a scanning electron microscopy (JEOL, JSM-6400F) and an X-ray diffractometer (Rigaku, SmartLab) with Cu Kα radiation ($\lambda$=0.15406 nm) operating at 40 kV and 100 mA, respectively. The SEM picture of the cross section is illustrated in Figure 3.2. The thickness of the BST thin film was measured to be 130.5 nm. The XRD pattern of the BST thin film deposited on Pt(111)/TiO$_2$/SiO$_2$/Si(100) is shown in Figure 3.3. The pattern matches very well with previously reported works [38, 39].

\[
\mu_{12}^{\text{eff}} = \frac{QL^3}{3\pi r^2 \delta (L-l)}
\]
3.4. Results and discussion

3.4.1. Dielectric behavior

The capacitance of the BST thin film was measured at 1 kHz by an impedance analyzer (Agilent, 4294A). Figure 3.4 shows the temperature dependence of the dielectric constant and the dielectric loss tangent. It was observed that the dielectric constant of the BST thin film (327) is much lower than bulk BST (e.g. 8000 at room temperature). In principle, thicker film has a larger dielectric constant due to larger size of grains. Similarly, small grain size increases grain boundaries, resulting in higher leakage current and lower dielectric constant [34]. Around 28 °C (Curie temperature) a transition from the ferroelectric phase to the paraelectric phase is noted. The broad peak in the response of ferroelectric films can be attributed to the mechanical and electrical strains in the films. These strains cause different grains to have different transition temperatures and a non-ferroelectric surface layer at the grain boundaries and on the surface of the film [40]. The dielectric loss is less than 5 % throughout the entire measurement temperature range.

3.4.2. Piezoelectricity

Piezoelectricity shows the topography and piezoelectricity measured by a piezoresponse force microscopy (PFM) (MFP-3D, Asylum Research, Santa Barbara, CA) at room temperature. As seen in Figure 3.5, BST thin film is in its ferroelectric state at room temperature. Therefore, piezoelectricity might be present in this state even though the film had not been poled.
First, to verify the piezoelectricity of the film in $d_{33}$ mode, the magnitude of deformation and phase of the film under a 4V AC excitation was measured. This voltage was applied through a PFM cantilever tip. As shown in Figure 3.5(a), results indicate that the fabricated BST thin film exhibits a small-localized piezoelectricity with a maximum piezoelectric constant of about 3 pm/V. Figure 3.5(b) shows the phase between the piezoelectric response with the applied voltage across the scanned area, ranging from $-10^\circ$ to $50^\circ$. This suggests a weak positive domain distribution. The global piezoelectric coefficient of the thin film can be attained by averaging the measured responses over the whole area, which should be even smaller than the maximum measured value ($d_{33} \sim 3$ pm/V) [41].

In addition, the contribution of piezoelectricity in ($d_{31}$ mode) mode was measured. For this test, a 10V AC excitation was applied across the top and bottom electrodes at room temperature. The generated displacement at the tip of the cantilever was undetectable by a laser vibrometer (OFV-5000, Polytec, Dexter, MI) with resolution of 10 pm. A calculation for corresponding $d_{31}$ for this case was performed and it indicated a maximum $d_{31}$ value of 17.8 fm/V. The piezoelectric contribution was of the total charge output under bending test was calculated to be 0.01%. This indicates that the electric output of the bending test is dominantly attributed to the flexoelectricity of the BST thin film.

3.4.3. Flexoelectric constant

The measurement of the effective $\mu_{12}$ coefficient was carried out in a thermal chamber as shown in Figure 3.6. The flexoelectric beam structure was clamped rigidly at one end and deflected by a piezoelectric actuator at the tip. The actuator was driven by a power amplifier
(Brüel & Kjær, type 2706) under a 2 Hz-excitation from a function generator (Tectronix, Model AFG3101). The 2 Hz frequency was chosen because it is much lower than the resonance frequency of the cantilever beam and therefore the displacement does not significantly differ from the static case for beam deflection model derivation. The deflection of the cantilever was measured by a laser vibrometer (Polytec, OFV-5000). The generated output signal was amplified by a charge amplifier (Brüel & Kjær, type 2635) and the charge was measured by a lock-in amplifier (Stanford Research system, Model SR830). The transverse flexoelectric coefficient $\mu_{12}^{\text{eff}}$ was calculated from the charge output and tip deflection using Eq. (3.4).

Additionally, a contrast test with a silicon cantilever without BST thin film was performed. It was fabricated into the identical size to verify if the measured charge output was from the noise current or purely came from flexoelectricity. Under the 1 μm tip excitation at 2 Hz, the measured charge output was almost 0 C and the phase of the charge output was unlocked to the input frequency of the tip excitation. This means the charge output from the cantilever was not from piezoelectricity or electrostaticity. Thus, the measured charge output from the silicon cantilever was a noise current and it was not significant when $\mu_{12}$ of BST thin film is measured.

As seen in Figure 3.7, $\mu_{12}$ of BST thin film was measured at the temperature ranging from 24 to 41 ºC. $\mu_{12}$ in a function of dielectric constant is shown in the inset of Figure 3.7. At all measured temperatures, the strain gradient and electric polarization has linear relationship. Figure 3.8 shows the average $\mu_{12}$ (slope) from three measurement trials. At paraelectric phase
(41 °C), which is above the Curie temperature (28 °C), the BST thin film still possesses high 
$\mu_{12}$ of 17.44 $\mu$C/m.

3.5. Discussion

Although the dielectric constant of BST thin film was much smaller than that of BST 
ceramics, the magnitude of the flexoelectric constant of the BST thin film is similar to that of 
BST bulk material as seen in Figure 3.7. We suggest several reasons for this phenomenon. 
One reason might be that the low dielectric constant of the BST thin film is largely attributed 
to its small grain size configuration, which generates vast numbers of grain boundaries and 
incurs a tremendous external pinning effect. However, the inner atomic structure was not 
affected by the small grain size. This suggests that the strain gradient induced mismatch of 
positive and negative charge centroids remained the same, thus producing the same level of 
flexoelectric polarization as bulk materials. Another possibility lies in the lattice mismatch 
and annealing [42], which can results in residual stress in thin films. In the presence of 
residual stress, the relaxation along the thickness direction would cause an extremely high 
strain gradient compared with the bulk mechanical test [43]. The mechanism how it affects 
on flexoelectricity has not been clearly explained yet. However, the high intrinsic strain 
gradient in the BST thin film may cause the flexoelectricity more sensitive to the external 
strain gradient.

The measured flexoelectric coefficient of the BST thin film is compared with notable 
flexoelectric coefficients from the literature with similar characterization methods (Table 2.1). 
Among studied materials, BST exhibits relatively high flexoelectricity and the BST thin film
fabricated for this study shows a level of flexoelectricity similar to that of bulk BST. This fact makes the flexoelectricity in micro/nano domains greatly enhanced without sacrificing large flexoelectric coefficients and taking advantages of scaling effect simultaneously.

3.6. Conclusion

In summary, enhanced flexoelectric effect was observed in Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film. The measured effective transverse flexoelectric coefficient $\mu_{eff}$ was found to be 24.5 $\mu$C/m at Curie temperature (28 °C) and 17.44 $\mu$C/m at 41 °C, which is comparable to that of bulk BST ceramics. This result ensures the feasibility of enhanced flexoelectricity in micro/nano domains without sacrificing large flexoelectric coefficients. With proper design, the BST thin film is possible to be used in flexoelectric micro/nano scale sensing devices, which are preferable to macroscale devices due to the flexoelectric scaling effect.
Figure 3.1. (a) The configuration of a BST thin film cantilever. (b) Cross-section view across
A and A’.
Figure 3.2. Cross-section SEM picture of the BST thin film.

Figure 3.3. XRD pattern of RF sputtered Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film deposited on Pt.
Figure 3.4. Dielectric constant and dielectric loss of BST thin film at different temperatures.
Figure 3.5. The topography and piezoelectricity of BST thin film. (a) Magnitude. (b) Phase.
Figure 3.6. Experimental set-up for the measurement of the flexoelectric coefficient.

Figure 3.7. The transverse flexoelectric coefficients as a function of temperature and dielectric constant (Inset).
Figure 3.8. The linear relationship between the strain gradient and polarization at 41 °C.
Table 3.1. The dimensions and material properties of the BST thin film cantilever.

<table>
<thead>
<tr>
<th>$L$ (mm)</th>
<th>$l$ (mm)</th>
<th>$r$ (μm)</th>
<th>$t_{Si}$ (μm)</th>
<th>$t_{BST}$ (nm)</th>
<th>$E_{Si}$ (GPa)</th>
<th>$E_{BST}$ (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.43</td>
<td>4.45</td>
<td>320</td>
<td>525</td>
<td>130.5</td>
<td>125.5</td>
<td>185.0</td>
</tr>
</tbody>
</table>

Table 3.2. The BST thin film fabrication parameters.

<table>
<thead>
<tr>
<th>Item</th>
<th>Setting values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Substrate temperature</td>
<td>300 °C</td>
</tr>
<tr>
<td>Deposition rate</td>
<td>0.0181 Å/s</td>
</tr>
<tr>
<td>Annealing temperature</td>
<td>700 °C</td>
</tr>
<tr>
<td>Annealing time</td>
<td>20 hrs</td>
</tr>
<tr>
<td>Thickness</td>
<td>130.5 nm</td>
</tr>
</tbody>
</table>
4. ENHANCEMENT OF FLEXOELECTRICITY IN STRUCTURES

4.1. Background

Electromechanical sensors have been developed for automobile, consumer electronics, biomedical instruments and military systems in order to enhance overall system performance. These sensors need to be small (micro or nano scale) and lightweight to reduce the self-loading effect and the cost of manufacturing. Among these different types of sensors, piezoelectric sensors are widely used due to their fast response time, wide frequency bandwidth, easy to use, and low cost [18, 44-46]. However, piezoelectric sensors are being increasingly challenged: as the piezoelectric dimensions are scaled down to micro/nano domain, the piezoelectricity in thin films and piezo nanostructures are significantly degraded [18]. Furthermore, de-poling is another general concern over piezoelectric sensors [20]. Therefore, it is important to point out that recent research on the flexoelectric effect enlightened flexoelectric sensing as an alternative technique to piezoelectric sensing [23, 47].

From previous studies, flexoelectric sensing structures in millimeter and centimeter scale showed relatively low sensitivity compared to that of piezoelectric counterparts [23]. Therefore, output amplification mechanisms are of interest in enhancing overall performance of flexoelectric devices. Similar amplification mechanisms have been studied in piezoelectric sensing. For example, piezoelectric multilayered structure possesses advantages over single layer piezoelectric structures in many aspects, including high voltage/charge sensitivity and high resonant frequency [48-50].

In this chapter, a flexoelectric multilayered cantilever structure using BST ceramic was designed, fabricated and tested. Charge output analysis from piezoelectric and flexoelectric
multilayered structures was presented, followed by comparison between charge outputs from a flexoelectric single layered and a double layered cantilevers using an analytical model based on the Euler-Bernoulli beam theory. Finally, experiments on a flexoelectric multilayered cantilever were performed and the results were then compared to those from single-layer cantilever tests, followed by discussion and conclusions.

4.2. Comparison of piezoelectric and flexoelectric effects

The cantilever bimorph structures are usually composed of two active layers (piezoelectric or flexoelectric) and one supporting layer in between, as shown in Figure 4.1(a). In the piezoelectric case, when an external force is exerted at the end of a cantilever, axial normal stress in the piezoelectric layers induces electric polarization. Similarly, in the flexoelectric case, an end force creates a normal strain gradient along the flexoelectric layers’ thickness direction, inducing the electric polarization.

Based on Euler-Bernoulli beam theory, the curvature $\kappa$ of a cantilever under bending moment induced by force $F$ at the tip can be given as [51]

$$\kappa = \frac{1}{R} = \frac{12 F s_{111}^m s_{111}^s}{k b s_{11}^s} (L - x)$$

(4.1)

where,

$$k = 2s_{111}^s (3 t_s^2 t_m + 6 t_s t_m^2 + 4 t_m^3) + s_{11}^m t_s^3$$

(4.2)
The superscript or subscript $s$ denotes the supporting layer, $m$ denotes the top and bottom piezo or flexo material. $x$, $s_{11}$, $t$, $b$ and $R$ are the axial distance from the clamped end, compliance, and thickness, width of the cantilever and radius of curvature, respectively.

To amplify the charge output, two layer structures were designed with parallel electrical connection, as shown in Figure 4.1(b). The arrows in piezoelectric layers denote the poling direction for the piezoelectric cantilever.

The axial normal stress $\sigma_{11}$ in piezoelectric cantilever can be expressed as

$$\sigma_{11} = \frac{1}{s_{11}^m R} y_n = \frac{1}{s_{11}^m R} (t_s + t_m) / 2$$  \hspace{1cm} (4.3)

where, $y_n$ is the distance between the neutral axis and the top surface of the piezoelectric layer, which is half of the total thickness of the cantilever.

The electrical polarization $P_p$ from each piezoelectric layer is

$$P_p = d_{31} \sigma_{11}$$  \hspace{1cm} (4.4)

where, $d_{31}$ is the transverse piezoelectric coefficient. The total charge $Q_p$ from two piezoelectric layers can be calculated by combining Eqs. (4.1), (4.3) and (4.4).

$$Q_p = 2 \int_0^L P_p \, b \, dx = 6 \, d_{31} \, F \, L^2 (t_s + t_m) \, s_{11}^s / k$$  \hspace{1cm} (4.5)

For the flexoelectric cantilever, the gradient of axial normal strain along the thickness direction of the cantilever can be given as

$$\frac{\partial \varepsilon_{11}}{\partial z} = \frac{\partial}{\partial z} \left( \frac{z}{R} \right) = \frac{1}{R}$$  \hspace{1cm} (4.6)

where, $\varepsilon_{11}$ is axial normal strain.
The electric polarization $P_f$ from each flexoelectric layer can be written as follows [28]:

$$
P_f = \mu_{11} \frac{\partial \varepsilon_{33}}{\partial z} + \mu_{12} \left( \frac{\partial \varepsilon_{11}}{\partial z} + \frac{\partial \varepsilon_{22}}{\partial z} \right) = \left[ \nu \mu_{11} + (1 + \nu) \mu_{12} \right] \frac{\partial \varepsilon_{11}}{\partial z} \tag{4.7}
$$

Here, $\nu$ is the Poisson ratio of a flexoelectric ceramic. For pure bending of beam with $L \gg b \gg t$, the flexoelectric polarization due to the gradient of axial normal strain $\varepsilon_{11}$ in the thickness direction can be simplified as

$$
P_f = \mu_{12}^{\text{eff}} \frac{\partial \varepsilon_{11}}{\partial z} \tag{4.8}
$$

$\mu_{12}^{\text{eff}}$ denotes the effective transverse flexoelectric coefficient. For convenience, $\mu_{12}^{\text{eff}}$ will be abbreviated to $\mu_{12}$. Then, the total charge $Q_f$ from two flexoelectric layers can be obtained by combining Eqs. (4.1), (4.6) and (4.8).

$$
Q_f = 2 \int_0^L P_f b \, dx = 12 \, \mu_{12} \, F \, L^2 \, s_{11}^{s_m} / k \tag{4.9}
$$

The ratio between the charge outputs of the piezoelectric and flexoelectric cantilevers can be derived as

$$
\frac{Q_f}{Q_p} = \left| \frac{2 \, \mu_{12} \, s_{11}^{s_m}}{d_{31} \, (t_s + t_m)} \right| \tag{4.10}
$$

which is inversely proportional to the sum of the thickness of the supporting and piezo/flexoelectric layers when the elastic modulus of piezo/flexoelectric material, size and the supporting layer of cantilever are identical, meaning that flexoelectric cantilevers are favorable as micro/nano-sensing structures.
It is anticipated from Eq. (4.10) that the charge output of the flexoelectric cantilever will exceed that of the piezoelectric one in micro/nano scales. The ratio of charge output from a BST flexoelectric cantilever to that of a piezoelectric cantilever is shown in Figure 4.2 by using the reported material properties given in Table 4.1. When the performance of the BST cantilever is compared with that of a cantilever fabricated from a material with very high piezoelectric coefficients such as PMN-30PT, the thickness of the cantilever must be less than 1.43 μm to yield better performance. When piezoelectric materials with relatively low piezoelectric coefficients are used, the performance of the flexoelectric cantilever exceeds that of the piezoelectric case on larger scales. For example, the BST flexoelectric device shows much better performance than the piezoelectric device fabricated from P(VDF-TrFE) when the thickness decreases below 73.1 μm.

4.3. Comparison of single and multilayered structures

In the previous section, analysis showed that the scaling effect can make the flexoelectric devices favorable in micro/nano scale. Another approach to maximizing charge output from flexoelectric sensors involves adopting multilayer structures. Figure 4.3 shows a single layer cantilever and a 2-layered flexoelectric cantilever structure, where an external force applied at the cantilever tip generates a strain gradient in each layer.

From Bernoulli-Euler beam theory, the strain gradient of the single layer cantilever is

$$\frac{\hat{\varepsilon}_{11}}{\hat{z}} = \frac{1}{R} \frac{F_1}{R} \frac{s_{11}^m}{I} (L-x) \quad (4.11)$$
where $L$, $s_{11}^m I$ and $F_1$ are the length, compliance and moment of inertia of a single layered cantilever and applied force at the tip, respectively.

The tip deflection of the single layer cantilever $\delta_1$ can be written as

$$\delta_1 = \frac{F_1 s_{11}^m L^3}{3 I} \quad (4.12)$$

Then, the total electric charge $Q_1$ from the single layer cantilever is given as

$$Q_1 = \int_0^L \mu_{12} \frac{\partial \varepsilon_{11}}{\partial z} b dx = \frac{F_1 s_{11}^m b L^2}{2 I} = \mu_{12} \frac{3 b \delta_1}{2 L} \quad (4.13)$$

The charge output of the double layer cantilever can be obtained by Eq. (4.9) and the tip deflection $\delta_2$ when force $F_2$ is applied at the tip and it has the expression of

$$\delta_2 = \frac{4 F_2 L^3 s_{11}^m s_{11}^s}{k b} \quad (4.14)$$

where, $s_{11}^s$ is the compliance of the supporting layer.

By substituting Eq. (4.14) into Eq. (4.9), the charge output from the double layer cantilever $Q_2$ can be obtained to be

$$Q_2 = \mu_{12} \frac{3 b \delta_2}{L} \quad (4.15)$$

As shown in Eqs. (4.13) and (4.15), if the input deflection is identical for both cases, the charge output from the double layer cantilever can be amplified by 200 % compared to that from the single layer cantilever. Similarly, the charge output from a structure with more than two layers will be amplified and is proportional to the number of flexoelectric layers $N$ as follows.
\[ Q_n = \mu_{12} \frac{3N b \delta_s}{2L} \]  

(4.16)

4.4. Fabrication of multilayered BST structure

In order to fabricate multilayered structure, each single layer that consists of BST and silicon layers was fabricated first and three of single layers were stacked and bonded consecutively with conductive epoxy. Firstly, a layer of 1 μm of SiO₂ was sputtered on a 350 μm thick silicon wafer in order to prevent shorting of the electrical connection through the BST layer. Here, a 350 μm of silicon layer was chosen because it is easy to handle without breaking. Subsequently, 100 Å of Ti and 1000 Å of Au were sputtered as electrodes on the top and bottom side of the silicon substrate and one side of the BST. The sputtered sides of the BST and the silicon substrate were bonded with silver epoxy (EO-23M, Epoxy set, Lincoln, RI). Normal pressure of 1 MPa was applied to produce the bonding layer thickness of about 7~8 μm. This also ensured good adhesion and conductivity between the BST and the silicon substrate. Then, the BST layer was lapped to 25 μm and the top surface of the BST layer was electrode sputtered as mentioned above. Figure 4.4(a) shows the cross-section of the multilayered structure and detailed sequence of each layer, while Table 4.2 shows detailed dimensions of the multilayered flexoelectric structure.
4.5. Experimental setup

To experimentally verify the charge amplification of the flexoelectric multilayered cantilever, the charge output of the fabricated flexoelectric multilayered prototype was measured at room temperature. The experimental setup is shown schematically in Figure 4.5. Firstly, the multilayered flexoelectric structure was clamped rigidly at one end. Then, the piezoelectric actuator was placed at the tip to generate deflection of the cantilever. The actuator was driven by a power amplifier (Type 2706, Brüel & Kjær, Nærum, Denmark) and a function generator (AFG3101, Tectronix Inc., Beaverton, OR). The deflection of the cantilever at the tip was measured by a laser vibrometer (OFV-5000, Polytec, Dexter, MI). The generated output signal was amplified by a charge amplifier (Type 2635, Brüel & Kjær, Nærum, Denmark) and the amplified signal was measured by a lock-in amplifier (SR830, Standard research system, Sunnyvale, CA). The generated charge from the single layer and 3-layer cantilevers were recorded as a function of excitation frequency from 1 to 150 Hz. Low frequency was chosen to ensure that the real deflection of the cantilever follows the input of the piezoelectric actuator.

4.6. Results and discussion

The experiment to measure the charge output from the multilayered cantilever was carried out for sensitivity comparison with single layer cantilever. Charge outputs were measured under different deflections (1-6 μm) and frequencies (1-150 Hz). Experiments were repeated three times under each condition, and data point on the graph represents the average of the three trials. The linear relationship between the deflection at the tip and the charge output can
be seen in Figure 4.6. The equivalent transverse flexoelectric coefficient $\mu_{12}^e$ of the multilayered flexoelectric structure can be derived from Eq. (4.13) and calculated as

$$\mu_{12}^e = \frac{2QL}{3b\delta}$$

(4.17)

The average equivalent transverse flexoelectric coefficient $\mu_{12}^e$ can be calculated from the slope of Figure 4.6 and yields a value of approximately $59.5 \, \mu C/m$, which is about three times the magnitude of that of a single layered flexoelectric cantilever ($20 \, \mu C/m$ [23]). Therefore, when the total dimensions of the flexoelectric structure is the same with the single layered structure, the sensitivity that is directly related to the equivalent flexoelectric coefficient will be multiplied proportionally to the number of layers. Furthermore, the sensitivity can be more enhanced with a choice of intermediate layers with lower stiffness.

Figure 4.7 shows the measured equivalent $\mu_{12}$ (squares) of the multilayered flexoelectric structure as a function of excitation frequency and the percentage ratio (triangles) of equivalent $\mu_{12}$ of a multilayered structure to $\mu_{12}$ of a single layer structure. The dashed lines in the graph denote the average values of measurements. As seen in Figure 4.7, the transverse flexoelectric coefficient was amplified proportionally to the number of structure layers. The averaged multilayer $\mu_{12}$ is slightly lower than $300 \%$ of the $\mu_{12}$ of a single layered flexoelectric cantilever. This could be resulted from the non-ideal bonding between the BST layers and the supporting silicon layer, which would hinder full strain transfer between each layer. Also, non-perfect clamping could decrease charge output. The relatively low equivalent $\mu_{12}$ ($55.1 \, \mu C/m$) and the percentage ratio ($262.6 \%$) values at 1 Hz was likely caused by imperfect electric insulation of silicon substrates, which were chosen for
fabrication convenience. Charge leakage through the silicon layers could occur because silicon-BST acts as an RC circuit and signal degradation cannot be neglected at low frequency. This can be improved by choosing substrate materials with better insulating performance.

4.7. Conclusion

Flexoelectric sensors are becoming increasingly popular because of their small size, absence of de-poling and aging problem and lead-free composition. In this study, a flexoelectric multilayered cantilever was compared analytically to piezoelectric equivalent and displayed amplified charge output. As the thickness of the cantilever is reduced to the range of several microns, the flexoelectric cantilever is expected to perform with higher sensitivity compared to the piezoelectric cantilever. Also, theoretical analysis suggests that charge output of multilayer structure is proportional to the number of layers. Finally, a three-layered BST flexoelectric structure for the purpose of amplifying the charge output was successfully fabricated and tested using \( \text{Ba}_{0.65}\text{Sr}_{0.35}\text{TiO}_3 \) materials. The charge output was measured to be three times of that generated by the single layer structure, confirming the signal amplification mechanism of multilayered structure.
Figure 4.1. (a) A schematic of a cantilever composed of one supporting layer and two piezoelectric or two flexoelectric layers and (b) Side views of electric connections in piezoelectric (left) and flexoelectric (right) cantilevers.
Figure 4.2. The ratio of charge output from BST flexoelectric cantilevers to that of piezoelectric cantilevers as a function of the total beam thickness.

Figure 4.3. Schematics of a single (left) and multi (right)-layer cantilevers.
Figure 4.4. Fabrication of the multilayered structure. (a) A schematic cross-section of the three-layered flexoelectric structure, (b) Top view photograph and (c) Photograph of cross-section of a 3-layered flexoelectric structure.
Figure 4.5. A schematic of the experimental setup for the transverse flexoelectric coefficient measurement.

Figure 4.6. Deflection at the cantilever tip vs. charge output from the multilayered flexoelectric cantilever under different force excitation.
Figure 4.7. The measured equivalent $\mu_{12}$ of the multilayered flexoelectric structure and the ratio of multi to single layer.
Table 4.1. Piezoelectric and flexoelectric coefficients of various materials (see Ref [11, 52-56]).

<table>
<thead>
<tr>
<th>Material</th>
<th>$d_{31}$ (pC/N)</th>
<th>Compliance - $s_{11}$ ($10^{-12}$ m$^2$/N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lead-based piezoelectric materials</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PbTiO$_3$</td>
<td>-22.8</td>
<td>6.5</td>
</tr>
<tr>
<td>PZT (sol-gel)</td>
<td>-82</td>
<td>13.8</td>
</tr>
<tr>
<td>PZT (sputtering)</td>
<td>-100</td>
<td>13.3</td>
</tr>
<tr>
<td>PMN-30PT</td>
<td>-921</td>
<td>52.0</td>
</tr>
<tr>
<td>Lead-free piezoelectric materials</td>
<td></td>
<td></td>
</tr>
<tr>
<td>P(VDF-TrFE)</td>
<td>-18</td>
<td>740.1</td>
</tr>
<tr>
<td>PVDF</td>
<td>-28</td>
<td>370.4</td>
</tr>
<tr>
<td>BaTiO$_3$ (single crystal)</td>
<td>-33.4</td>
<td>7.4</td>
</tr>
<tr>
<td>$K_{0.48}Na_{0.52}NbO_3$ (sputtering)</td>
<td>-45.1</td>
<td>8.2</td>
</tr>
<tr>
<td>Flexoelectric material</td>
<td>$\mu_{12}$ ($\mu$C/m)</td>
<td>Compliance - $s_{11}$ ($10^{-12}$ m$^2$/N)</td>
</tr>
<tr>
<td>BST</td>
<td>100</td>
<td>6.54</td>
</tr>
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</table>

Table 4.2. Dimensions of the three-layered flexoelectric structure.

<table>
<thead>
<tr>
<th>Thickness of silicon ($t_s$ - μm)</th>
<th>Thickness of BST ($t_f$ - μm)</th>
<th>Total thickness ($t_{total}$ - mm)</th>
<th>Length ($L$ - mm)</th>
<th>Width ($b$ - mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>350</td>
<td>25</td>
<td>1.26</td>
<td>7.5</td>
<td>2</td>
</tr>
</tbody>
</table>
5. FLEXOELECTRIC ACCELEROMETER

5.1. Background

An accelerometer is a sensor that converts acceleration from motion or gravity to an electrical signal. There have been high demands for high performance and low cost micro-accelerometer in automotives, consumer electronics, biomedical devices, industrial monitoring and military applications [57], where accelerometers are used to monitor and control the force/acceleration of cars, aircrafts spaceships missiles and drones. Also, they are used to detect vibration in rotationary machinery to reduce vibration and noise. With incensement of markets of the mobile phone, portable electronics and video game controller, needs for micro-machined accelerometers have been significantly increased [58].

5.1.1. Current accelerometer technologies

Among several acceleration sensor structures including membrane (diaphragm), bridge and cantilever structures, cantilever structures are most compliant to applied force, vibration and pressure etc., thus, they are known with the highest sensitivity. Typical accelerometer cantilever structures as depicted in Figure 5.1 [58, 59].

The capacitive accelerometers measure capacitance change due to change of distance of two electrodes caused by vibration/acceleration [60-63] as shown in Figure 5.1(a). One of the two electrodes is fixed and one is on a flexible structure forming small gap between two electrodes. A proof mass is to amplify the vibration so that it can have larger signal. The performance of capacitive accelerometers is superior in low frequency range and they can be
operated in servo mode to achieve high stability and linearity. Capacitive accelerometers are also less prone to noise and variations with temperature, typically dissipate less power. However, it suffers from fragility and low bandwidth and requires on-chip electronics thus, increased cost.

As shown in Figure 5.1(b), the piezoresistive and piezoelectric accelerometers have cantilever beams with proof masses at the beam tips and piezoresistive or piezoelectric materials near the beam support where the strain is the maximum. The electric signal generated from the piezoresistive material due to change in resistance is proportional to the acceleration of the vibration. Piezoresistive accelerometers can be used to measure static accelerations and hence, are preferred in high shock applications without extra electronic circuitry. However, they suffer from limited temperature range since piezoresistive materials exhibit relatively large temperature coefficients, which require additional temperature compensation circuitry [18].

On the other hand, with piezoelectric accelerometers, stress change induces electric polarization shift that is proportional to the stress. Among the various types of accelerometers, piezoelectric accelerometers have been offering attractive features such as high output impedance, simple structures, rapid response, and wide dynamic range that is suitable for shock measurement as well as for almost imperceptible vibrations. However, piezoelectric accelerometers are incapable of measuring static accelerations, have limited temperature range, poor low-frequency response, aging, depolarization and decreased performance at micro-scale [64].
5.1.2. Motivation of microcantilever accelerometers

The microcantilever structure was firstly designed for the atomic force microscopy (AFM). And now, it has been adopted in physical, chemical and biological sensing applications [65-67]. In an AFM, a microcantilever senses the change in resonance response or deflection by detecting mass loading, surface stress variation or changes in damping conditions [14]. Various types of microcantilevers were adopted in AFMs for their simple structure and high precision such as capacitive [68], piezoresistive [69] and piezoelectric [70] sensors. Among these, piezoelectric readout technique has been widely used due to its simple fabrication process and low power consumption, thanks to the high impedance and low driving voltage for piezoelectrics. However, small-scale piezoelectric structures, such as micro or nano scales, show lower piezoelectric performance in comparison with their bulk counterparts. Therefore, it is difficult to obtain piezoelectric microcantilevers with high sensing performance. In order to improve the microcantilever sensitivity, flexoelectric microcantilever sensing structure has been investigated. Flexoelectric microcantilever structure is believed to possess the advantages of piezoelectric readout as well as other benefits mentioned in Chapter 1.

5.2. Flexoelectric accelerometers

5.2.1. Objectives

Similar to piezoelectric sensing, flexoelectric sensing is expected to have characteristics including fast response time, wide frequency bandwidth, easy to use and low cost [18, 44-46]. Thus, an accelerometer using flexoelectric effect was designed in order to take merits of
flexoelectric effect and at the same time, to overcome several problems of piezoelectric effect such as poling related aging and temperature limitations.

In this chapter, a flexural mode accelerometer based on the BST-steel unimorph with a seismic mass was designed, fabricated and tested. Theoretical analysis was conducted for flexoelectric vibration sensing. Frequency responses of the accelerometer were obtained using finite element analysis and compared with experimental measurements.

5.2.2. Configuration of the proposed flexoelectric accelerometer

Similar to piezoelectric accelerometers [64], there are typically three basic modes of flexoelectric transducers: compression mode, shear mode, and bending mode (e.g. cantilever mode). Figure 5.2 shows the corresponding structures of piezoelectric and flexoelectric accelerometers. In order to generate strain gradient instead of homogenous strain in the structures of compression or shear mode, the shape of the flexoelectric member needs to be tapered. In contrast to its piezoelectric counterpart, the electrode of the shear mode flexoelectric accelerometer should be patterned on the side walls of the sensor. These specific requirements of compression and shear mode flexoelectric transducers suggest fabrication challenges. Bending mode transducers, however, have better sensitivity and manufacturability for micro-accelerometer and can be fabricated using flexoelectric thin or thick film. For all these reasons, the bending mode design was adopted in this study. The following section will focus on the modeling of the bending type flexoelectric accelerometer. The microaccelerometer studied here is configured to be a flexoelectric unimorph with seismic mass attached at the tip, as shown in Figure 5.3. The unimorph consists of a
flexoelectric top layer and a suspension layer underneath. Rectangular shape cantilever usually have a large stress concentration in the base. As shown in Figure 5.4, a trapezoid shape cantilever could generate more uniform strain and strain gradient distribution along the length direction by eliminating the stress concentration, thus to improve the sensing range of the accelerometer \[71\]. Trapezoid shape cantilever also raises the resonance frequency of the sensor.

5.3. Analytical modeling
In case that an acceleration is exerted on the structure, the inertial force of the seismic masses could bend the unimorph, which induces a non-uniform transverse strain in the flexoelectric layer with a gradient along its thickness direction. Based on Bernoulli-Euler beam theory, the curvature \( \kappa \) of a flexoelectric unimorph cantilever under bending moment can be given as \[72\]

\[
\kappa(x_i) = \frac{12M(x_i)s_{t1}^fs_{t1}^s(s_{t1}^st_s + s_{t1}^ft_f)}{Kb(x_i)} \quad (5.1)
\]

where,

\[
K = 4s_{t1}^fs_{t1}^st_s(t_f)^2 + 4s_{t1}^fs_{t1}^ft_f(t_s)^3 + (s_{t1}^s)^2(t_s)^4 + (s_{t1}^s)^2(t_f)^4 + 6s_{t1}^fs_{t1}^st_s(t_s)^2(t_f)^2 \quad (5.2)
\]

The super or subscript \( f \) denotes the upper flexoelectric element and \( s \) denotes the lower steel element, while \( s_{t1}, t, b(x_1) \) are the elastic modulus, thickness, and width of the element, respectively. \( b(x_1) \) can be written as

\[
b(x_1) = \frac{a_1 - a_2}{L}(L - x_1) + a_2 \quad (5.3)
\]
where \(a_1\) and \(a_2\) are the width at the tip and clamping end of the unimorph, respectively. \(L\) is the length of the unimorph.

\(M(x_1)\) is the bending moment applied along the unimorph which can be expressed as

\[
M(x_1) = m_a(L - x_1)
\]  

(5.4)

where \(m_a\) is the mass of the seismic mass and \(a\) is the acceleration. The strain gradient of the unimorph can be determined as

\[
\frac{\partial \varepsilon_{11}}{\partial x_3} = \frac{\partial^2 w(x_1)}{\partial x_1^2} = \kappa(x_1)
\]  

(5.5)

where \(w(x_1)\) is the vertical deflection of the unimorph along the \(x_1\) direction. So the total charge induced can be obtained by integrating along the length as

\[
Q = \int_0^L \mu_{12} \frac{\partial \varepsilon_{11}}{\partial x_3} b(x_1) dx_1 = \frac{6 \mu_{12} s_{11}^s s_{11}^f (s_{11}^t s_{11}^t + s_{11}^f s_{11}^f) L}{K} m_a
\]  

(5.6)

The sensitivity \(S_Q\) of the flexoelectric accelerometer is as follows

\[
S_Q = \frac{Q}{a} = \frac{6 \mu_{12} s_{11}^s s_{11}^f (s_{11}^t s_{11}^t + s_{11}^f s_{11}^f) L}{K} m_a
\]  

(5.7)

5.4. Fabrication of flexoelectric accelerometer

BST plates with dimension of 10×10×1 mm\(^3\) was prepared for fabrication of flexoelectric accelerometers. Firstly, the bottom side of a 700 \(\mu\)m thick BST plate were sputtered with 50 \(\AA\) of Ti and 1000 \(\AA\) of gold as an electrode. The BST plates were bonded onto the 50 \(\mu\)m steel plate with high strength silver epoxy (EO-23M, Epoxy Set). Normal pressure of about 1 MPa was applied to make the bonding layer uniform and thin, thus to ensure good adhesion.
and electrical conductivity between BST and steel layers. BST layer was lapped down to 50 μm thick and the top surface of BST layer was then electroded. This composite was diced into the trapezoid shape with the dimensions shown in Table 5.1 using a Disco 321 dicing saw (DAD321, Disco Corp., Tokyo, Japan). Finally, 15 mg of a lead block was attached onto the tip of the accelerometer as a seismic mass using the superglue bonding method. The accelerometer was bonded to the plastic base with superglue to make it convenient to clamp in the vise. The prototyped accelerometer is shown in Figure 5.5.

5.5. Experimental setup

The experimental setup for sensor test is shown in Figure 5.6. The sensor was attached to a vibration exciter (Model ES020, KCF Tech, State College, PA). A function generator (AFG3101, Tectronix Inc., Beaverton, OR) was used to generate a sinusoidal signal that was amplified by a power amplifier (Type 2706, Brüel & Kjær, Nærum, Denmark), and then applied to the vibration exciter to generate vibration with designed frequency and amplitude. The output charge signal from the sensor was converted and amplified to voltage signal through a charge amplifier (Type 2635, Brüel & Kjær, Nærum, Denmark), which was recorded through a lock-in amplifier (SR830, Standard research system, Sunnyvale,CA). A commercial accelerometer (Model 352C22, PCB Piezotronics, Depew, NY) was used as a reference to measure the acceleration from the vibration exciter. This acceleration was also recorded on the oscilloscope through a signal conditioner (Model 482A16, PCB Piezotronics,
Depew, NY). The generated charge from the flexoelectric accelerometer was recorded as a function of vibration frequency (10-4000 Hz) and base acceleration (0.2 - 3g).

5.6. Results and discussion

5.6.1. Modeling results

The sensor was meshed automatically with free tetrahedral elements in normal-level size. The total number of elements is 2785. The first order resonance frequency of the accelerometer was found to be 3312 Hz. Excitation frequency was swept from 10 Hz to 4000 Hz to obtain the frequency response of the sensor. The finite element modeling results are shown in Figure 5.7. In the low frequency range that is far away from the resonance frequency of the system, the finite element analysis presents a flat frequency response with the sensitivity of 1.02 pC/g. To obtain the analytical results, the parameters in Table 5.1 were substituted into the Eq. (5.7). Charge sensitivity of the accelerometer at flat frequency response was calculated to be 0.89 pC/g. As can be seen from Eq. (5.7), the sensitivity of the accelerometer is determined by the seismic mass and the flexoelectric constant.

5.6.2. Experimental results

As shown in Figure 5.8, the experimental results reveal a stable sensitivity of 0.84 pC/g in the low frequency range. The working frequency with the flat frequency response reaches 1600 Hz. The resonance frequency of the sensor system is 2800 Hz. It is essential to stress
that Eq. (5.5) is only applicable to the cantilever with infinitesimal deflection, otherwise a large error would exist [27, 73]. The maximum deflection in the test was about 6.5 mm with the angle of 0.2°. Such a small deformation could ensure the precision of theoretical estimation. Figure 5.9 shows the sensor output as a function of acceleration at different frequencies. The lower charge sensitivity compared with the FEA and analytical results may be resulted from the absence of the rigid and precise clamping of the accelerometer. Non-ideal bonding between the BST layer and the suspension layer could inhibit the full strain transmission, thus diminishing the sensor sensitivity. Imprecise clamping could also decrease the natural frequency by increasing the effective sensor length. The additional mass of bonded electric wires could reduce the resonance frequency as well.

5.7. Conclusion
A trapezoidal shape flexoelectric unimorph accelerometer based on Ba$_{0.65}$Sr$_{0.35}$TiO$_3$ was designed, prototyped and tested. The analytic analysis and frequency response based on finite element analysis were conducted, and the results agreed well with the experimental measurements. The accelerometer provides a sensitivity of 0.84 pC/g in the working frequency up to 1.6 kHz. This flexoelectric micro-accelerometer showed the potential for future flexoelectric sensing applications.
Figure 5.1. Typical accelerometers with cantilever construction. (a) Capacitive accelerometer. (b) Piezoresistive and piezoelectric accelerometer.

Figure 5.2. Basic structures of flexoelectric accelerometers.
Figure 5.3. Schematic view of a flexoelectric unimorph accelerometer.

Figure 5.4. Distribution of strain gradient of different shapes in transverse direction.
Figure 5.5. Photographic and schematic pictures of the accelerometer. (a) Top view and (b) Side view.
Figure 5.6. Experimental setup for the flexoelectric acceleration test.

Figure 5.7. Finite element analysis of the sensor system using Comsol code indicating the frequency range and the charge sensitivity.
Figure 5.8. Comparison of measured frequency response with the analytical solution.
Figure 5.9. Sensor charge output under different frequencies and accelerations.
Table 5.1. The dimensions and material properties of the flexoelectric accelerometer.

<table>
<thead>
<tr>
<th>$a_1$ (mm)</th>
<th>$a_2$ (mm)</th>
<th>$L$ (mm)</th>
<th>$t_f$ (μm)</th>
<th>$t_s$ (nm)</th>
<th>$E_{BST}$ (GPa)</th>
<th>$E_{St}$ (GPa)</th>
<th>$m_s$ (mg)</th>
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<tr>
<td>7</td>
<td>2</td>
<td>3</td>
<td>50</td>
<td>50</td>
<td>185.0</td>
<td>210</td>
<td>15</td>
</tr>
</tbody>
</table>
6. FLEXOELECTRIC MICROPHONE

6.1. Background

Microphones are transducers that convert acoustic energy into electrical energy. Microphone applications include telephones, hearing aids, computers, live and recorded engineering, two-way radios, television broadcasting, speech recognition, voice over IP (VoIP) and non-acoustic purposes such as ultrasonic checking or knock sensors. During the last few decades, various types of micromachined microphones have been investigated. The advantages of micromachined microphones are accurate dimension control, a high degree of miniaturization and low cost due to batch processing [74].

6.1.1. Review of current technologies for microphones

Currently, there are many types of acoustic sensors being used as microphones, such as capacitive (condenser), fiber optic, piezoresistive and piezoelectric sensors. Each of these acoustic sensor types has advantages and drawbacks. Table 6.1 summarizes some of performance specifications for the three main types of microphones.

Capacitive microphones have two metal plates with an air gap between them as shown in Figure 6.1(a). One of these plates is made of very light material and act as the diaphragm. A polarizing voltage is applied to the diaphragm by an external power supply (battery or phantom power) or by the charge on an electret material in the diaphragm or on the back plate charging it with a fixed static voltage. The diaphragm and back plate, separated by a
small volume of air, form an electrical component called a capacitor (or condenser). The capacitance between these two plates changes as the freely suspended diaphragm is displaced by the sound wave. When the diaphragm vibrates in response to a sound, it moves closer to and farther away from the back plate. As it does so, the electrical charge that it induces in the back plate changes proportionally. The fluctuating voltage on the back plate is therefore an electrical representation of the diaphragm motion. Because the diaphragm of the condenser is not loaded down with the mass of a coil, it can respond very quickly to transients. Also, the condenser capsule can be made very small. Condensers generally have excellent sonic characteristics, and are widely used in high-quality professional microphones in sound reinforcement, measurement and recording. They are also known to have high sensitivity, high signal to noise ratio and high stability [62, 75, 76]. However, the complex structure and fabrication process can be challenging. In addition, they necessitate an external DC voltage supply and require high standby power consumption, leading to the inconvenience of frequent battery replacements [67].

Fiber optic microphones are relatively new compared to the capacitive, piezoresistive and piezoelectric microphones. They measure the changes in light intensity reflected from an ultra-sensitive silicon membrane [77]. As shown in Figure 6.1(c), light emitted by an LED is beamed via an optical path onto a special reflective membrane. Sound waves hitting the membrane cause it to vibrate, thereby changing the characteristics of the light reflection. The reflected light is transmitted through an optical path back onto a photo detector. With simple electronic processing, the light is then transformed into an audio signal representing clear, true-to-source sound. Fiber optic microphones [78, 79] have been used in harsh environments
because they are not associated with electrical, magnetic or radioactive fields and are tolerant to moisture and temperature. Also, the fiber optic microphones cannot be detected by electromagnetic means. Therefore, they are ideal for special purposes such as antidrug activity, military activity, and clear communications between the patients and the staffs during MRI scan. In addition, thanks to the nature of optic fiber light propagation, the distance between the light source of the microphone and its photo detector can be extended up to several kilometers without any preamplifier and other electrical devices. However, they have relatively low sensitivity and durability. In addition, fiber optic microphones are more costly and difficult to repair because the optical fiber are more difficult and expensive to be spliced than wires [80, 81]. For these reasons, the current trend is that fiber optic microphones are used just for the environments where other convectional microphones cannot be operated.

A piezoresistive microphone is shown in Figure 6.1(c) [82-86]. They are composed of a piezoresistive material with top and bottom electrodes and a membrane layer. The membranes are extremely compliant diaphragm, which is the part that moves in response to an applied sound pressure. When the sound waves incidents the membrane and the membrane vibrates, the mechanical strains in the membrane and piezoresistive materials change. The strain induces the change in resistance of piezoresistive material and the piezoresistive microphones can detect the sound waves.

A piezoelectric microphone has similar sensing structure with piezoresistive microphones [67, 87] with different sensing mechanism. The piezoelectric material generates the polarization according to the strain of the membrane, thus, the piezoelectric microphone detects the sound
waves. The piezoresistive and piezoelectric microphones have been desirable due to their simple and robust mechanical structures and easy to be integrated into semiconductor devices [67, 87]. However, they suffer from low sensitivity and narrow bandwidth (though, relatively wider than that of capacitive microphones) because the resonance of microphones is solely dependent on the dimensions and material properties, there is the trade-off relationship between the sensitivity and resonance. They also have a limited operating temperature range. Moreover, the most sensitive piezoelectric microphones are usually composed of lead based materials such as lead zirconate titanate (PZT), lead magnesium niobate – lead titanate (PMN-PT) and lead zinc niobate – lead titanate (PZN-PT). They are banned to use some countries in the world because of concerns of human health and environmental reasons [88].

6.2. Sensing structure

In order to utilize the flexoelectricity for a sensing mechanism, three different modes can be adopted. They are compression, shear and bending modes. The corresponded flexoelectric coefficients are $\mu_{11}, \mu_{44}$ and $\mu_{12}$, respectively. To generate a large strain gradient, bending or flexural mode was considered in this design. Figure 6.2 shows the schematic views of three main flexural mode sensing structures, namely membrane structure, bridge structure and cantilever structure, which are also main structures for piezoelectric microphones. Due to the symmetry of the structure, the strain gradient distribution has different signs at different locations, which could lead to cancellation of the electric charge output if a fully covered electrode pattern is used. To avoid this problem and to maximize the sensitivity, the electrode
layout should be optimized to collect the single signed flexoelectric polarization, either the positive or negative.

From Euler-Bernoulli beam theory, the deflection of the three flexural structures due to an external pressure can be estimated as

\[
\begin{align*}
\left\{ \begin{array}{l}
w_M(r) = \frac{Pa^4}{64D} \left( 1 - \frac{r^2}{a^2} \right)^2 \quad \text{where } D = \frac{Et^4}{12(1 - \nu^2)} \\
w_B(x) = \frac{PLx^3}{24EI} \left( 1 - \frac{x}{L} \right)^2 \\
w_C(x) = \frac{Px^2}{24EI} (6L^2 - 4Lx + x^2)
\end{array} \right.
\]

(5.8)

where \( w, P, t, a, L, \nu \) and \( EI \) represent the deflection, external pressure, thickness, radius of membrane, length of bridge and cantilever Poisson’s ratio and bending rigidity of inertia of bridge and cantilever. Here, super/subscript \( M, B \) and \( C \) indicate the membrane, bridge and cantilever structures, respectively. \( r \) and \( x \) are the arbitrary coordinates from the center of the circular membrane and clamped side of the bridge and cantilever structures, respectively.

The flexoelectric polarization of the bridge and cantilever structures due to the gradient of strain can be simplified as [28]

\[
P_{3B or C}^B = \mu_{11} \frac{\partial \varepsilon_{33}}{\partial z} + \mu_{12} \left( \frac{\partial \varepsilon_{11}}{\partial z} + \frac{\partial \varepsilon_{12}}{\partial z} \right) = [\nu \mu_{11} + (1 + \nu) \mu_{12}] \frac{\partial \varepsilon_{33}}{\partial z} = \mu_{12}^{\text{eff}} \frac{\partial \varepsilon_{11}}{\partial z}
\]

(5.9)

\[
= \mu_{12}^{\text{eff}} \frac{\partial}{\partial z} \left( \frac{z}{R} \right) = \mu_{12}^{\text{eff}} \frac{1}{R} = \mu_{12}^{\text{eff}} \frac{\partial^2 w}{\partial x^2}
\]

where \( R \) is the radius of the curvature. Similarly, the flexoelectric polarization of the circular membrane can be simplified as
\[ P_s^M = \mu _{12}^\text{eff} \frac{\partial ^2 w}{\partial r^2} \] (5.10)

The charge output induced by external pressure \( P \) of each structure can be calculated and simplified by the integration of the polarization.

\[
\begin{align*}
Q_a &= \int_0^{2\pi} \int_b^a P_s^M \, dr \, d\theta = \mu _{12}^\text{eff} \frac{P \pi b}{8D} (a^2 - b^2) \\
Q_b &= \int_0^w \int_0^{l_B} P_s^B \, dx \, dy = \mu _{12}^\text{eff} \frac{P w l_B (2l_B^2 - 3l_B L + L^2)}{12EI} \\
Q_c &= \int_0^w \int_0^{l_C} P_s^C \, dx \, dy = \mu _{12}^\text{eff} \frac{P w l_C (l_C^2 - 3l_C L + 3L^2)}{6EI}
\end{align*}
\] (5.11)

where \( b, l_B \) and \( l_C \) are the inner radius of electrode of the circular membrane, electrode lengths of the bridge and cantilever from the clamped sides, respectively.

The electrode sizes that maximize the charge output of each structure can be easily determined as follows.

\[
\begin{align*}
b &= a / \sqrt{3} \\
l_B &= (3L - \sqrt{3}L) / 6 \quad \text{two electrodes at both ends} \\
l_C &= L
\end{align*}
\] (5.12)

The sensitivity can be simply calculated by dividing the charge output by an external pressure. The optimized electrode size, sensitivity and resonance for three different structures are summarized and listed in Table 6.2. In order to compare the sensitivity and resonance of these three structures, the values of the bridge shape structure were used as a reference. It can be observed that the cantilever-structured microphone can generate the highest sensitivity, which is more than 10 times larger than the bridge structure. High flexibility of the cantilever structure lowers the resonance frequency by a factor of 0.16 than the bridge microphones.
The bridge structure was then chosen due to its wide bandwidth and moderate sensitivity. The following section will focus on the modeling of the bending type bridge-structured flexoelectric microphone.

6.3. Design of flexoelectric microphone

The configuration and side view of the flexoelectric microphone are shown in Figure 6.3. The resonance frequency of the flexoelectric microphone structure was first calculated since the resonance is usually associated with the bandwidth of a sensor. Considering a beam with two fixed ends, the first natural frequency can be calculated as

\[ f_0 = \frac{4.73^2}{2\pi} \sqrt{\frac{EI}{\rho A L^2}} \]  

(5.13)

where \( EI, \rho, A \) and \( L \) are the bending rigidity, density, cross section area and length of the BST bridge, respectively.

As mentioned earlier, the charge output reaches the maximum when \( l_b = (3 - \sqrt{3})L / 6 \) and the two separate electrodes are connected in parallel. Then the final charge output becomes

\[ Q = \mu_{12}^{\text{eff}} \frac{L^3w}{36\sqrt{3EI}}P \]  

(5.14)

The sensitivity of the flexoelectric microphone is calculated by dividing the charge output by the external pressure:

\[ S = \mu_{12}^{\text{eff}} \frac{L^3w}{36\sqrt{3EI}} \]  

(5.15)
With the dimensions given in Table 6.3, the resonance and sensitivity of the device become 93.73 kHz and 0.92 pC/Pa, respectively.

A prediction of the frequency response can be modeled as a second-order system with a transfer function given by

$$H(s) = \frac{A}{s^2 + Bs + C}$$  \hspace{1cm} (5.16)

Constants $A$, $B$ and $C$ are related to the microphone’s frequency response through $A=C S_{\text{low}}$, $B=2\pi f_0/Q_m$ and $C=(2\pi f_0)^2$, where $S_{\text{low}}$ is the low-frequency sensitivity and $Q_m$ is the quality factor. $Q_m$ will be calculated from experimental measurements.

$$Q_m = \frac{f_0}{f_2 - f_1}$$  \hspace{1cm} (5.17)

where $f_1$ and $f_2$ are two measured frequencies where the signal becomes 3 dB lower than that at resonance $f_0$.

6.4. Fabrication of flexoelectric microphone

To fabricate a flexoelectric microphone, Ti/Au electrodes on the top side of silicon wafer and bottom side of BST ceramic were deposited by e-beam sputtering with the thickness of 10 nm and 100 nm, respectively. A 1.5 mm wide trench was made by dicing (DAD321, Disco Corp., Tokyo, Japan), as shown in Figure 6.4(a), and then the BST ceramic was bonded onto the Si substrate with epoxy (EP301, Epoxy Technology, Billerica, MA) under the normal pressure of 1MPa (Figure 6.4(b)). The bottom of the BST ceramic was filled with wax (0CON-193, Logitech Limited, Glasgow, UK) to protect the BST ceramic during the
followed lapping and dicing steps (Figure 6.4(c)), then lapped down to 50 μm thick and the Ti/Au top electrode was deposited by the e-beam evaporation and patterned with a lift-off process (Figure 6.4(d)). The BST layer was next diced into the final dimensions (Figure 6.4(e)) shown in Table 6.3. Finally the wax was removed using the Ecoclear solution (0CON-178, Logitech Limited, Glasgow, UK) (Figure 6.4(f)). Figure 6.5 shows the photograph of the fabricated flexoelectric microphone.

6.5. Experimental setup

The test of the flexoelectric microphone was carried out at room temperature in an anechoic box. The experimental setup is shown schematically in Figure 6.6. The flexoelectric microphone was placed facing a loud speaker (KPSG-100, Kingstate Electronics Corp., New Taipei City, Taiwan), which was driven by a power amplifier (Type 2706, Brüel & Kjær, Nærum, Denmark), a function generator (AFG3101, Tectronix Inc., Beaverton, OR) generated sound pressure onto both the flexoelectric microphone and a reference microphone (46BG, G.R.A.S. Sound & Vibration, Holte, Denmark). The reference pressure was measured by processing the signal from the reference microphone with an oscilloscope (DSO7104B, Agilent Technologies Inc., Santa Clara, CA). The generated output signal from the flexoelectric microphone was monitored using a charge amplifier (Type 2635, Brüel & Kjær, Nærum, Denmark) connected with a lock-in amplifier (SR830, Stanford Research system, Sunnyvale, CA).
6.6. Results

The output charge from the flexoelectric microphone was measured by a lock-in-amplifier. The resulting sensitivity was then calculated by dividing the output charge by the pressure measured by the reference microphone. The test frequency was swept from 100 Hz to 130 kHz as shown in Figure 6.7. In addition to the experimental results, the analytical frequency response is plotted for comparison. It can be predicted from Eq. (5.16) by making analytical Bode plot of the expected response of the microphone with the measured value of \( Q_m \) and the expected resonance frequency and low frequency sensitivity. At low frequency range, e.g. between 1 ~ 20 kHz, the sensitivity of the flexoelectric microphone was measured to be 0.84 pC/Pa. Above 20 kHz, the sensitivity increases with frequency and approaches a peak at 105 kHz. It can be observed that the experimental results matched well with the analytical solution. Table 6.4 summarizes the measured parameters.

Figure 6.8 shows the sensor output as a function of pressure under different frequencies. Linearity between the pressure and charge output at 0.5, 1, 5, 10, 15 and 20 kHz can be observed, indicating a broad bandwidth of the sensor. Each measurement set at different frequencies is linear \((R^2=0.9997)\) and has sensitivity of 0.77 ~ 0.85 pC/Pa. The average is 0.81 pC/Pa and standard deviation is approximately 0.03 pC/Pa.

In addition to the sensitivity measurement, the signal-to-noise ratio at frequencies ranged from 1 to 20 kHz was measured. The lock-in amplifier measured the output charges while the function generator and power amplifier are on and the loud speaker is off. Figure 6.9 shows the signal-to-noise ratio of the flexoelectric microphone at 1 Pa. The charge output at 1 Pa is
5000 times larger than that of surrounding noise sources induced by the measurement equipment, corresponding to 74 dB.

6.7. Discussion

Discrepancies in the frequency response and sensitivity between experimental and analytical results were mostly caused by fabrication-related imperfections. For example, there might be a thin layer of residual wax that was used to protect the BST bridge from fracture during dicing. This residual wax on the bottom side of BST acts as an extra layer and hence, the resonance frequency increases and the sensitivity of the flexoelectric microphone decreases. Also, the location of the top electrode has an effect on the total charge. The charge output and thus the sensitivity can be reduced if the top electrode is not placed exactly at the designed location. A much lower sensitivity was observed below 1 kHz. This roll-off sensitivity toward low frequency is likely caused by the equalization of the pressure difference associated with acoustic wave [67].

Due to the trade-off between sensitivity and bandwidth in such microphones, it is hard to attain both the high sensitivity and a wide frequency range simultaneously. However, despite this trade-off, the flexoelectric microphone reported here is promising compared to reported piezoelectric microphones.[67, 87, 89-93] The microphone developed in this study is compared to the piezoelectric microphones and listed in Table 6.5 and Figure 6.10. The charge sensitivity was converted to voltage sensitivity for comparison.

\[ S_v = \frac{S_c}{C} \]  

(5.18)
6.8. Conclusion

In summary, a flexoelectric microphone with a barium strontium titanate (Ba$_{0.67}$Sr$_{0.33}$TiO$_3$) bridge structure was designed, fabricated and tested successfully. With the BST ceramic possessing $\mu_{12}$ of 30 $\mu$C/m, the microphone was designed to have a sensitivity of 0.92 pC/Pa at the audible frequency range. The experimental measurements of the fabricated microphone showed a sensitivity of 0.85 pC/Pa at the frequency range of 1 – 20 kHz, with the signal-to-noise ratio being at 74 dB. The prototyped BST microphone presents both high sensitivity and wide bandwidth, in comparison with other previously reported piezoelectric microphones. Thus, a flexoelectricity could provide an effective and promising alternative for better performance of various applications.
Figure 6.1. Typical microphones. (a) Capacitive (condenser) microphone. (b) Piezoresistive and piezoelectric microphone. (c) Optic fiber microphone.
Figure 6.2. Basic simplified structures for microphones using bending mode (a) Circular membrane, (b) Bridge, and (c) Cantilever (purple area: flexoelectric material, yellow area: electrode, gray area: silicon substrate, and blue line: electric connection).
Figure 6.3. Schematic view of a flexoelectric microphone. (a) Configuration of the microphone and (b) Side-view of the microphone across A and A’.
Figure 6.4. Fabrication process of the flexoelectric microphone.

Figure 6.5. Photograph of the flexoelectric microphone.
Figure 6.6. Experimental set-up for flexoelectric microphone tests.

Figure 6.7. The analytical and experimental sensitivity of the flexoelectric microphone at different frequencies.
Figure 6.8. Sensor charge output under different frequencies and pressure values.
Figure 6.9. The signal to noise ratio of the flexoelectric microphone at 1 Pa.
Figure 6.10. Comparison of sensitivity and resonance of piezoelectric and flexoelectric microphones from the academic literature.
Table 6.1. Comparison of basic performance characteristics of the three conventional microphones [94].

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<thead>
<tr>
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<th>Capacitive</th>
<th>Piezoresistive</th>
<th>Piezoelectric</th>
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<tr>
<td>Sensitivity (μV/Pa)</td>
<td>400 ~ 1000</td>
<td>0.1 ~ 100</td>
<td>10 ~ 500</td>
</tr>
<tr>
<td></td>
<td>Good</td>
<td>Low</td>
<td>Medium</td>
</tr>
<tr>
<td>Input power</td>
<td>Required</td>
<td>Required</td>
<td>None</td>
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<tr>
<td>Dynamic range</td>
<td>Narrow</td>
<td>Relatively wide</td>
<td>Wide</td>
</tr>
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</table>

Table 6.2. Electrode area, sensitivity and resonance for various structures

<table>
<thead>
<tr>
<th></th>
<th>Membrane</th>
<th>Bridge</th>
<th>Cantilever</th>
</tr>
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<tr>
<td>Electrode area</td>
<td>$\frac{2}{3} \pi r^2$</td>
<td>$2 \times \frac{3 - \sqrt{3}}{6} L \times w$</td>
<td>$L \times w$</td>
</tr>
<tr>
<td>Sensitivity</td>
<td>$a \frac{\pi (1 - \nu^2)}{\sqrt{3EI}} \approx 0.56 S_b$</td>
<td>$\mu_{12} \frac{wL^3}{36 \sqrt{3EI}} = S_b$</td>
<td>$\mu_{12} \frac{wL^3}{6EI} \approx 10.39 S_b$</td>
</tr>
<tr>
<td>Resonance</td>
<td>$2.94 \sqrt{\frac{E \nu^2}{2 \pi \rho a^4 (1 - \nu^2)}} \approx 2.93 f_b \frac{4.73^2}{2 \pi} \sqrt{\frac{EI}{\rho wtL^2}} = f_b \frac{1.875^2}{2 \pi} \sqrt{\frac{EI}{\rho wtL^2}} = 0.16 f_b$</td>
<td>$\mu_{12} \frac{wL^3}{6EI} \approx 0.16 f_b$</td>
<td></td>
</tr>
</tbody>
</table>
Table 6.3. Dimensions of the microphone structure and material properties of BST.

<table>
<thead>
<tr>
<th>$L$ (mm)</th>
<th>$l$ (μm)</th>
<th>$t$ (μm)</th>
<th>$w$ (μm)</th>
<th>$\mu_{12}$ (μC/m)</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$E$ (GPa)</th>
<th>$\varepsilon_r$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5</td>
<td>317</td>
<td>50</td>
<td>768</td>
<td>30</td>
<td>8200</td>
<td>153</td>
<td>8000</td>
</tr>
</tbody>
</table>

Table 6.4. Comparison of designed and measured resonance frequency ($f_0$), sensitivities and quality factor ($Q_m$)

<table>
<thead>
<tr>
<th></th>
<th>Designed</th>
<th>Measured</th>
<th>Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_0$ (kHz)</td>
<td>93.73</td>
<td>105</td>
<td>12.0 %</td>
</tr>
<tr>
<td>Sensitivity at 10 kHz (pC/Pa)</td>
<td>0.93</td>
<td>0.85</td>
<td>-8.6 %</td>
</tr>
<tr>
<td>Sensitivity at $f_0$ (pC/Pa)</td>
<td>2.32</td>
<td>2.10</td>
<td>-9.5 %</td>
</tr>
</tbody>
</table>
Table 6.5. Performance of the flexoelectric microphone compared to notable microphones from the literatures.

<table>
<thead>
<tr>
<th>Author</th>
<th>Material</th>
<th>Structure</th>
<th>Area×thickness (mm²×μm)</th>
<th>Sensitivity (mV/Pa)</th>
<th>Resonance (kHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Piezoelectric Material</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 Ried [89]</td>
<td>ZnO</td>
<td>Membrane</td>
<td>6.25×3.5</td>
<td>0.92</td>
<td>18.3</td>
</tr>
<tr>
<td>2 Ko [87]</td>
<td>ZnO</td>
<td>Membrane</td>
<td>9.0×3.0</td>
<td>0.51</td>
<td>7.3</td>
</tr>
<tr>
<td>3 Horowitz [90]</td>
<td>PZT</td>
<td>Membrane</td>
<td>2.54×3</td>
<td>0.002</td>
<td>59.0</td>
</tr>
<tr>
<td>4 Littrell [91]</td>
<td>AlN</td>
<td>Two cantilevers</td>
<td>0.62×2.3</td>
<td>1.82</td>
<td>40.0</td>
</tr>
<tr>
<td>5 Williams [92]</td>
<td>AlN</td>
<td>Membrane</td>
<td>0.54×3.84</td>
<td>0.030</td>
<td>129.5</td>
</tr>
<tr>
<td>6 Prasad [93]</td>
<td>ZnO</td>
<td>Membrane</td>
<td>9.61×30</td>
<td>0.335</td>
<td>4.0</td>
</tr>
<tr>
<td>7 Baumgartel [67]</td>
<td>ZnO</td>
<td>Cantilever</td>
<td>0.83×4.5</td>
<td>0.17</td>
<td>5.997</td>
</tr>
<tr>
<td>Flexoelectric material</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8 Current study</td>
<td>BST</td>
<td>Bridge</td>
<td>1.152×50</td>
<td>0.55</td>
<td>93.73</td>
</tr>
</tbody>
</table>
7. CONCLUSION AND RECOMMENDATIONS FOR FUTURE WORK

7.1. Conclusions

The primary goal of this dissertation was to investigate the flexoelectric effect and design, fabrication and testing of flexoelectric sensors.

The first part of this dissertation was focused on characterization of flexoelectric materials and structures. Specifically, the characterization of barium strontium titanate for the flexoelectricity study was carried out using both bulk ceramics and thin films. To overcome low charge/voltage output of flexoelectric sensing structures in millimeter scale, flexoelectric multilayered sensing structure was investigated for charge output amplifications. Based on the presented results, the following conclusions can be drawn.

**Bulk BST ceramics**

- BST bulk ceramic - silicon unimorphs were fabricated and tested for dielectric and flexoelectric characterizations. The measured Curie temperature is 29 ºC and peak dielectric constant is 8000. The measured transverse flexoelectric coefficient $\mu_{12}$ of 10 $\mu$C/m remains constant for unimorphs with various thicknesses. This large flexoelectricity in BST bulk ceramic is promising for flexoelectric sensing applications.
**BST thin film**

- Enhanced flexoelectric effect was observed in Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film. The measured effective transverse flexoelectric coefficient $\mu_{12}^{\text{eff}}$ was found to be 24.5 $\mu$C/m at Curie temperature (28 °C) and 17.44 $\mu$C/m at 41 °C, which is comparable to that of bulk BST ceramics. This result ensures the feasibility of enhanced flexoelectricity of BST thin films for promising flexoelectric micro/nano sensing devices.

**BST multilayer structure**

- A flexoelectric multilayered cantilever was compared analytically to piezoelectric equivalent and displayed amplified charge output. As the thickness of the cantilever is reduced to the range of several microns, the flexoelectric cantilever is expected to perform with higher sensitivity compared to the piezoelectric cantilever.

- Theoretical analysis suggests that charge output of a multilayer structure is proportional to the number of layers. Therefore, three-layered BST flexoelectric structure for the purpose of amplifying the charge output was successfully fabricated and tested using Ba$_{0.65}$Sr$_{0.35}$TiO$_3$ materials. The charge output was measured to be three times of that generated by the single layer structure, confirming the signal amplification mechanism of multilayered structure.
The second part of this dissertation presents the first study on utilizing the flexoelectricity for vibration and sound pressure sensing using BST ceramics, and the associated conclusions can be drawn as follows.

**Flexoelectric accelerometer**

- A trapezoidal shape flexoelectric unimorph accelerometer based on $\text{Ba}_{0.65}\text{Sr}_{0.35}\text{TiO}_3$ was designed, prototyped and tested. A trapezoid shape of an accelerometer was chosen since the shape can exploit higher strain at the root of the accelerometer.

- The analytic analysis and frequency response based on finite element analysis were conducted, and the results agreed well with the experimental measurements. The accelerometer provides a sensitivity of 0.84 pC/g in the working frequency up to 1.6 kHz.

**Flexoelectric microphone**

- A flexoelectric microphone with a barium strontium titanate ($\text{Ba}_{0.67}\text{Sr}_{0.33}\text{TiO}_3$) bridge structure was designed, fabricated and tested successfully. With the BST ceramic with $\mu_{12}$ of 30 μC/m, the microphone was designed to have a sensitivity of 0.92 pC/Pa at the audible frequency range. The experimental measurements of the fabricated microphone showed a sensitivity of 0.85 pC/Pa at the frequency range of 1 – 20 kHz, with the signal-to-noise ratio being at 74 dB. The analytical results and the experimental findings agreed reasonably well.
The prototyped BST microphone presents both high sensitivity and wide bandwidth, in comparison with other previously reported piezoelectric microphones. Thus, flexoelectric sensing could provide an effective and promising alternative for high performance piezoelectric sensing for a broad range of applications.

7.2. Recommendations for future work

It was verified that thin film has large flexoelectric constant $\mu_{12}$ of 24.5 $\mu$C/m at Curie temperature (28 °C) and 17.44 $\mu$C/m at 41 °C, therefore, flexoelectric thin film micro/nano sensors are expected with high performance due to the scaling effect. With the successful demonstration of bulk BST based flexoelectric accelerometer and microphone, BST thin film micro-accelerometer and microphone can be investigated as the next step. Generally, in micro/nano sensors, the disadvantage is known to be poor signal to noise ratio and weak responses at low frequency (<1 Hz). It is largely due to small mass and thermal noise. If a sensor’s sensitivity is enhanced via flexoelectricity, the signal output becomes significantly larger than the environmental and thermal noises, which will lead to advanced sensors for challenging applications.

Future work could also incorporate the sensor packaging and on-chip electronics to reduce size and environmental noise such as thermal effect and electromagnetic interference noise.
REFERENCES


