

This is the accepted version of the article:

Bhaskar U.K., Banerjee N., Abdollahi A., Wang Z., Schlom D.G., Rijnders G., Catalan G.. A flexoelectric microelectromechanical system on silicon. *Nature Nanotechnology*, (2016). 11. : 263 - . 10.1038/nnano.2015.260.

Available at: <https://dx.doi.org/10.1038/nnano.2015.260>

A flexoelectric microelectromechanical system on silicon

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KEYWORDS: flexoelectricity, MEMS, nanoscale, piezoelectrics

Flexoelectricity¹ is an electromechanical non-local piezoelectric effect² in which the magnitude of dielectric polarization is generated under an external force is insensitive to the absolute magnitude of deformation (ϵ), but is instead determined by the gradient of the an applied deformation ($\frac{\partial \epsilon}{\partial x}$)¹ or, conversely, a gradient of deformation (bending) is generated by a voltage².

Compared to piezoelectricity, flexoelectricity is a weak effect of little practical significance in

bulk materials. However, the roles can be reversed at the nanoscale³. Here, we demonstrate that flexoelectricity is a viable route to lead-free microelectromechanical and nanoelectromechanical systems (MEMS and NEMS). Specifically, we have fabricated a silicon compatible thin film cantilever actuator with a single flexoelectrically active layer of **strontium titanate** with a figure of merit (curvature divided by electric field) of 3.33 (MV)^{-1} , comparable to that of the state of the art piezoelectric bimorph cantilevers.

Main

Certain attributes of flexoelectricity point towards a favourable role in micro- and nano-electromechanical systems (MEMS and NEMS), for example: (i) Flexoelectricity is a universal phenomenon exhibited by materials of all symmetry groups, and thus flexoelectric devices can in principle be fabricated from silicon or any of its gate dielectrics in a completely **complementary metal oxide semiconductor (CMOS)** compatible environment; (ii) Any (strain) gradient scales inversely with the material dimension³, thus allowing flexoelectricity to match or even dominate over piezoelectricity at the nanoscale⁴, particularly in materials with high dielectric permittivity ϵ , such as ferroelectric thin film⁵ and composites⁶; (iii) High frequency bending resonators capable of functioning at extreme temperatures can be implemented; (iv) Flexoelectric devices can be made from simple dielectrics, with a performance that is therefore linear and non-hysteretic, and (v) A flexoelectric, unlike a piezoelectric bimorph actuator, does not need to be clamped to an elastic passive layer in order to bend: a single dielectric layer is sufficient to achieve field-induced bending, and this simplifies device design and removes the risk of delamination that can exist in standard piezoelectric bimorph actuators (**Figure 1a**).

In contrast, since the materials with the largest piezoelectric coefficients are ferroelectric, piezoelectric devices can suffer from their intrinsically hysteric nature and nonlinear behaviour at fields close to the coercive voltage, and in addition their properties are strongly temperature-dependent: they only work below their Curie temperature. Moreover, the ferroelectrics with largest piezoelectric coefficients are lead-based⁷, and lead toxicity poses serious problems for integration of such devices in biomedical applications, where MEMS-based energy harvesting devices would otherwise find a natural niche of applications⁸. In addition, bimorphs can also be restricted by the mechanical and thermal expansion mismatch between the piezoelectric and elastic layer, which can lead to progressive deterioration of the bonding between layers.

Despite the advantages offered by nanoscale flexoelectricity, research in this field is still in its infancy^{9,10}, and considerable effort is required before it can be established as a viable technology. On the fundamental front, we need a reliable catalogue of flexoelectric coefficients for all materials of technological interest, and proof that the magnitude of these coefficients remains constant at the nanoscale. On the practical front, we need to develop both nanofabrication and nano-characterization tools suitable for making and measuring flexoelectric nanodevices. This article addresses both of these issues.

We fabricated all-oxide nanocantilevers (**Figure 1b**) as capacitor structures consisting of a strontium titanate (SrTiO_3) active layer sandwiched between two layer of strontium ruthenate (SrRuO_3) for the top and bottom electrode; the complete capacitor stack (see Figure S1 and Figure S2) is epitaxially grown on a buffer of SrTiO_3 deposited by molecular beam epitaxy (MBE) on Si, which is currently an established template system for incorporating other epitaxial

oxide films on Si¹¹. Fabrication details are provided in the methods section. The centrosymmetric lattice of room temperature SrTiO_3 ensures that any measured bending moment arises purely from flexoelectricity; room temperature paraelectricity in SrTiO_3 is also confirmed by its linear and non-hysteretic mechanical response as a function of electric field; for comparison, in the supplementary materials (See Figure S3) we show the characteristic butterfly-shape hysteresis loop response of a ferroelectric lead zirconium titanate (PZT) cantilever grown by similar methods on silicon. SrTiO_3 is also currently the only (bulk) material for which the theoretical and experimental values, measured using the direct method are of the same order of magnitude¹², providing a good reference for testing two important questions: (i) whether the bulk flexoelectric coefficients retain their bulk value in thin films and (ii) whether the coefficients measured by us via the inverse method (actuator mode) are the same as those measured in bulk by the direct method (sensor mode) –something that is definitely true for piezoelectrics but is not obvious in flexoelectricity, where this question has been controversial¹³.

The most popular method currently used to characterize flexoelectric coefficients involves dynamically bending a cantilever and using lock-in techniques to instantaneously measure the charge generated by the bending. We refer to this as the direct method, and it has been employed on a variety of materials, including perovskite ceramics¹⁴, single crystals¹², and even polymers¹⁵. Its drawback is the difficulty of miniaturizing mechanical bending appliances down to the nanoscale. But, while direct flexoelectricity measures the polarization induced by bending, a converse or inverse effect also exists whereby polarizing a sample causes it to bend^{2,13,16,17,18}. The “inverse method” thus involves the application of an electric field to a cantilever or plate-shaped material, and measuring the induced bending^{16,18}. The curvature (k) induced via

flexoelectricity (μ) is related to the flexural rigidity (D) of the plate and the applied voltage (V) by ⁹ :

$$k = \frac{\mu V}{D} . \quad (1)$$

The flexural rigidity (D) of a cantilever is $\frac{Et^3}{12(1-\nu^2)}$, where E is the young's modulus, ν is the Poisson ratio, and t is the thickness. Hence, the flexoelectrically induced curvature k scales as the *cube* of the cantilever thickness, i.e., the voltage-induced bending multiplies by a factor of 8 –almost an order of magnitude- every time the thickness is halved. The inverse scaling of k with the Young's modulus also makes it pertinent to characterizing soft materials, which are expected to display giant electromechanical couplings¹⁹. On the practical side, achieving converse flexoelectricity only requires the fabrication of planar capacitive cantilevers, and we demonstrate that this requirement can be readily realized using existing MEMS techniques. Thus, inverse flexoelectricity is an optimum route to exploring and exploiting the flexoelectricity of nanodevices.

The observation of cantilever oscillations induced by an applied alternate voltage (V_{ac}) was made using a commercial digital holographic microscope^{20,21} (DHM) (schematically illustrated in **Figure 1c and 1d**) working in stroboscopic mode. The Fourier-filtered first harmonic displacement induced in the 16 x 40 μm **SrTiO₃** cantilever plate is plotted as a function of the AC excitation at 100 KHz and just above resonance (320 KHz) in **Figure 2(a)** and **2(b)** respectively (the unfiltered response at 100 KHz is shown in Figure S4). The curvature was calculated from the Fourier-filtered displacement²². In order to probe the dynamics further, the cantilever was excited with the same bias of 1 V but over a range of different sinusoidal frequencies (**Figure**

2(c)). The observed resonance frequency ~310 kHz corresponds quite well with the analytical estimation based on the geometry of the cantilever (see Figure S5), while the phase corresponds to the lag between the waveform of the excitation signal (voltage) and that of the flexoelectric response (deflection).

The first harmonic curvature measured as a function of applied AC field at ~100 KHz is plotted in **Figure 2(d)** and shows the expected linear behavior for a flexoelectric actuator. In order to demonstrate the stability of the measurements as a function of the frequency, we also include a complete curvature vs field measurement made at 10 KHz in figure S6. The value of the flexoelectric coefficient μ_{eff} calculated from the slope of curvature vs voltage using Equation 1 yields $\mu_{eff} \sim 4.6$ nC/m. This is an effective flexoelectric coefficient involving a geometry-dependent combination of the flexoelectric tensor components. Calculations using a self-consistent continuum model of flexoelectricity¹⁶ under the assumption that the ratio between μ_{11} and μ_{12} remains the same as in bulk¹² yield $\mu_{12} \sim 4.1$ nC/m. This is comparable to the μ_{12} for bulk **SrTiO₃** (100) crystals measured by the direct method ($\mu_{12} \sim 7$ nC/m)¹², particularly when factoring in the smaller relative permittivity of our SrTiO₃ thin film, which is ~four times smaller compared to that of bulk single crystals. Indeed, the quantity of physical significance⁹ is the flexocoupling ratio $f = \frac{\mu}{\epsilon}$, which we found to be 6 V for **SrTiO₃** nanocantilevers, in good agreement with the estimate proposed by Kogan of 1-10 V for ionic solids¹, and comparable to the value found for other perovskites such as lead magnesium niobate-lead titanate (PMN-PT)²³. The similarity of the coefficients measured by inverse and direct methods also provides experimental validation that flexoelectric devices will display the same coupling constant for operation as sensor and actuator¹³.

We now turn to the comparison between the actuation performance of the flexoelectric cantilevers and that of state of the art piezoelectric bimorph cantilevers fabricated using Zinc oxide (ZnO)²⁴, Aluminium nitride (AlN)²⁵, PZT²², and PMN-PT²⁶. This is shown in **Figure 3**. The electromechanical performance of our SrTiO_3 devices is comparable to or larger than that of devices fabricated using ZnO²⁴, and AlN²⁵ and PZT²². The performance of our flexoelectric devices is, however, lower than that of hyper-active PMN-PT²⁶ and an optimal ultra-thin device made with a 10 nm thick AlN²⁷ active layer. However, the *flexoelectric curvature/voltage* scales as the inverse of the cube of the thickness (eq. 1), so SrTiO_3 devices with the same thickness as the state of the art AlN²⁷ could exceed the performance of even the best piezoelectric and ferroelectric devices so far reported in literature. We have also programmed an open access App (https://umeshkbhaskar.shinyapps.io/FlexovsPiezo_app) to facilitate direct comparison between the expected performance of piezoelectric and flexoelectric actuators for different cantilever geometries and material specifications.

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Flexoelectricity represents a new technological paradigm for lead-free electromechanical actuators which can be integrated on silicon for MEMS and NEMS applications. Looking beyond SrTiO_3 , all high-k dielectric materials used in CMOS circuitry must in principle also be flexoelectric, because this is a property that is not restricted by material symmetry⁹. Flexoelectricity therefore opens up a new extensive catalogue of materials suitable for nanoscale electromechanical device applications, and provides a route to integrating “more than Moore” electromechanical functionalities within transistor technology.

FIGURES

Figure 1 Schematic illustration of flexoelectric actuation and the measurement setup. (a) Comparison between flexoelectric and piezoelectric actuation in nanoscale actuators; (b) Optical image of an array of SrTiO_3 nanocantilevers; (c) 3D image of one SrTiO_3 nanocantilever with colour scale corresponding to the out of plane displacement; (d) The digital holographic microscope splits a coherent laser beam in to an objective beam and a reference beam. The objective beam is focused on the sample, and the light reflected back is collected to form an interference pattern with the reference beam. Any difference in height along the sample surface results in a corresponding difference in phase of the light reflected back from it.

Figure 2 Experimental characterisation of flexoelectricity as a function of frequency and electric field. The AC voltage, and first harmonic displacement, for an applied voltage of 1 V plotted for the cantilever (a) below and (b) above the resonance frequency. (c) The Curvature/Voltage ratio as a function of the frequency for the SrTiO_3 nanocantilever at 1V excitation, showing the resonant peak at ~ 310 KHz. The quality factor Q is ~ 25 . The resonance is confirmed by the 180° phase change. (d) The first harmonic flexoelectric curvature shows a linear variance when plotted as a function of the applied AC field. The frequency of the measurement was 100 KHz, well below the resonant frequency amplification and close to the static performance calculated from the fit in (c).

Figure 3 Comparison of flexoelectric actuator with the state of the art piezoelectric bimorphs. The ratio of Curvature/Electric field is compared for a flexoelectric SrTiO_3 , and piezoelectric devices fabricated from ZnO^{24} , $\text{AlN}^{25,27}$, PZT^{22} , and PMN-PT^{26} . For all materials, we quote the intrinsic response measured out of resonance.

ACKNOWLEDGMENT

This work was funded by an ERC Starting Grant from the EU (Project No. 308023), a National Plan grant from Spain (FIS2013-48668-C2-1-P) and the Severo Ochoa Excellence programme. The authors are grateful to discussions with Etienne Cuche, Jerome Parent, Eduardo Solanas and Yves Emery from Lynceotec, Switzerland.

Author Contributions

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G.C and U.B conceived and designed the experiments; N.B designed and made the cantilevers under the supervision of G.R; U.B performed and analysed the inverse flexoelectric characterizations under the supervision of G.C; A.A performed the self-consistent finite field calculations; Z.W. performed the Molecular beam epitaxy growth of the template layer under the supervision of D.S; U.B. and G. C. wrote the paper with the help of all other authors. All authors discussed the results, commented on the manuscript and have given their approval to the final version of the manuscript.

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