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Author(s)	Shibamoto, Takeshi; Li, Yan; Kasai, Kohta et al.
Citation	Materials Today. 2026, 98, p. 103433
Version Type	VoR
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Cascading nanodomainization of bulk tetragonal BaTiO₃ triggered by mechanical stress of a few MPa

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ARTICLE INFO

Keywords:

Ferroelectrics
Polarization switching
Domain walls
Stress

ABSTRACT

Ferroelectrics enable the interconversion of mechanical, electrical, and thermal energies via control of spontaneous polarization. Here, we report nanodomainization of millimeter-scale bulk ferroelectric crystals induced by a few MPa of mechanical stress, where a tetragonal BaTiO₃ single domain evolves into an aggregate of nanodomains several hundred nanometers in size. The process features striking morphological evolution, including the emergence of domain-wall-related contrasts cascading across the entire surface, while the final visual appearance closely resembles the original pristine state. Owing to this internal complexity, the dielectric responses are significantly altered upon mechanical loading. Notably, small variations in loading conditions give rise to different domain configurations that lead to distinct dielectric responses. This mechanical approach opens a deterministic pathway for engineering nanoscale domain structures and their associated emergent properties in bulk ferroelectrics.

Introduction

Ferroelectric materials are widely utilized for energy conversion and functional applications, owing to their ability to interconvert electrical, mechanical, and thermal energies. They play essential roles in memory, logic, switching, and sensing devices that are integral to modern technologies [1,2]. A defining feature of ferroelectrics is the presence of spontaneous electric polarization, which can be reversed by the application of an external electric field [3]. In ferroelectrics, a region with uniform polarization direction is referred to as a domain, while the interface between adjacent domains is known as a domain wall (DW) [4]. Domains and domain walls serve as the fundamental units underlying the diverse functional properties of ferroelectrics [5–9], with domain walls often contributing significantly to the overall dielectric,

piezoelectric, and switching responses [6,9,10]. Although electric fields are most commonly used to control polarization in practical applications [4,5,11], magnetic fields [12] and polarized light [13] have also been shown to induce domain switching.

External mechanical forces are also known to drive DW motion and alter the domain structure in ferroelectrics, thereby modifying their material properties [14–17]. For example, Gao et al. achieved precise local directional regulation of vortex domains in hexagonal LuMnO₃ single crystals via nanoindentation and nanoscratch [16]. Lu et al. demonstrated mechanical writing of ferroelectric polarization in BaTiO₃ thin films using the stress gradient generated by an atomic force microscope tip, where the corresponding compressive uniaxial stress was estimated to be 3.2 GPa [17]. Deng et al. reported complete polarization reversal in single-crystalline BaTiO₃ nanopillars under a uniaxial

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<https://doi.org/10.1016/j.mattod.2026.103433>

Received 23 July 2025; Received in revised form 12 June 2026; Accepted 15 June 2026

Available online 20 June 2026

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compressive stress of 280 MPa [18]. The application of pre-stress has been shown to facilitate the electric-field-induced domain switching in ferroelectric single crystals, enabling giant electrostrictive strains [19,20] and elevated Curie temperatures [21]. In parallel, DW motion under various stress conditions has been investigated using numerical and theoretical approaches, including phase-field simulations [22], first-principles calculations [23], and molecular dynamics simulations [24].

Despite these advances, the mechanical threshold required for polarization switching, together with the polarization evolution from the onset of domain switching to complete reversal, remains insufficiently understood, even though these processes govern both the energetic cost and practical viability of ferroelectric functionalities [11,25]. In electric-field-driven switching, the coercive electric field, E_C , has long been established as a fundamental parameter [25]. In BaTiO₃, investigations of E_C across single crystals and thin films spanning length scales from millimeters to nanometers have provided a clear physical picture of its scaling behavior [26–30]. In comparison, mechanically driven polarization reversal has been studied predominantly in micrometer- and nanometer-sized samples [15,17,18,31]. The critical stress required for domain switching in millimeter-sized bulk crystals, as well as the corresponding evolution of polarization, remains largely unexplored. This limited understanding arises not only from the challenge of fabricating large single-domain crystals but also from the stereotype that brittle ferroelectric bulks cannot withstand external forces without fracturing.

In this study, a mechanical stress of 2–4 MPa was applied to induce extensive 90° domain switching in millimeter-sized BaTiO₃ crystals, leading to the transformation of a single-domain state into an aggregate of domains with characteristic sizes of several hundred nanometers. The observed nanodomain configuration deviates from the conventional equilibrium domain size predicted by Kittel's law, which we attribute to a stress-induced modification of the energetic balance governing domain periodicity. Remarkably, despite the presence of numerous

internal DWs, the dielectric permittivity along the polarization direction remained comparable to that of a large single domain. Moreover, the resulting domain configurations and their associated properties exhibited significant dependence on the applied loading conditions. These findings demonstrate that, in millimeter-scale ferroelectric materials, external mechanical force serves as a powerful means to control domain structures and their associated emergent properties, enabling low-cost and energy-efficient modulation of ferroelectric performance.

Results

Polarization switching by mechanical stress of a few MPa

Fig. 1A shows an optical image of the BaTiO₃ single-domain crystal used in this study. Prior to the application of mechanical force, the sample was colorless and optically transparent. Here, the sample surfaces perpendicular to the equivalent x - and y - axes in the lateral direction of the sample are referred to as F-surface and R-surface, respectively, and the surface perpendicular to the z -axis is designated as T-surface. Fig. 1B presents a typical stress–strain curve obtained from the uniaxial compression of the single-domain crystal, where an external force was applied at room temperature at a constant strain rate of 1×10^{-5} /s along the [001] polarization axis. In general, in the initial stage of deformation, there is an elastic deformation region in which the stress increases linearly with strain, followed by a plastic deformation region. However, in BaTiO₃ crystals loaded along the [001] direction, an unusual "stress plateau" appeared during the initial stage, where linear elastic deformation would typically be expected. In this stage, the stress remains nearly constant at a low level, showing minimal increase despite continued strain. Fig. 1C provides a magnified view of the stress plateau in Fig. 1B. This plateau stage begins when the stress reaches approximately 3 MPa and persists with a low stress of 2 to 4 MPa until

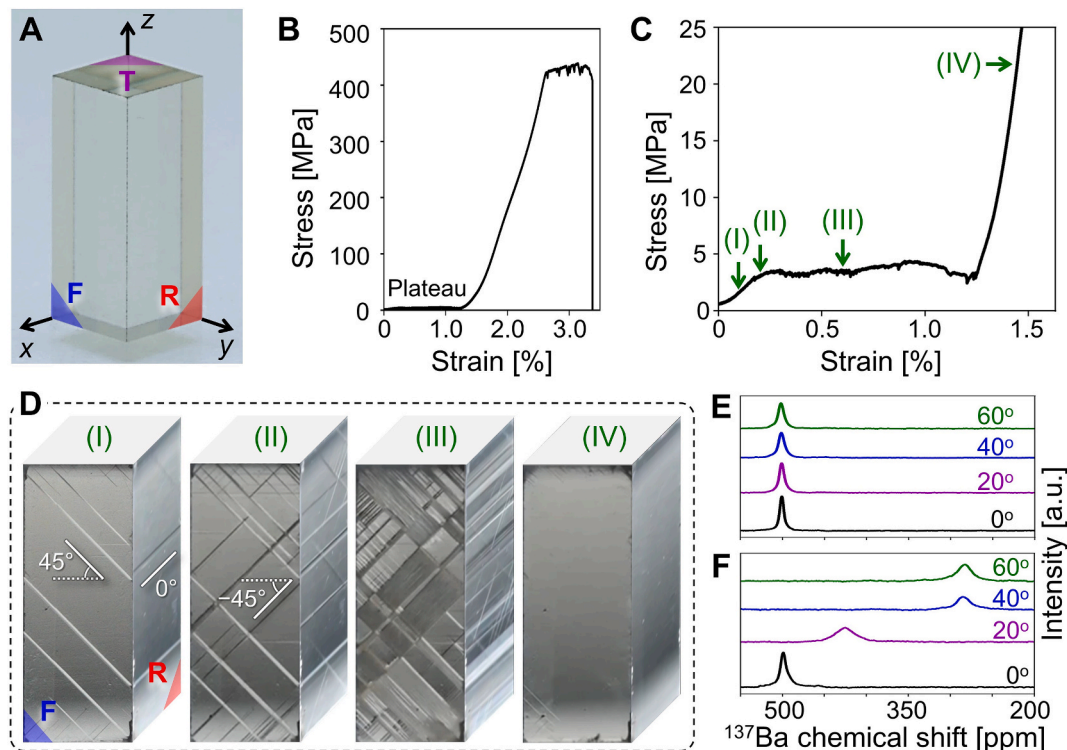


Fig. 1. (A) An optical image of the bulk sample before deformation. The sample surfaces along the x -, y -, and z -axes are named F-, R-, and T-surfaces, respectively. (B) The typical stress–strain curve obtained from a BaTiO₃ single-domain crystal loaded along the [001] direction at room temperature. (C) Magnified view of the initial plateau stage of the stress–strain curve. (D) In-situ optical images taken from the sample surfaces where (I), (II), (III), and (IV) correspond to the respective points indicated in (C) by the green letters. Note that states (I), (II), and (III) fall within the 2–4 MPa plateau regime, whereas state (IV) corresponds to the post-plateau stage reached upon further displacement-controlled compression. (E, F) NMR spectra for: (E) unloaded BaTiO₃, and (F) 1.5% strained BaTiO₃.

the strain reaches about 1%.

Fig. 1D presents the optical images of the sample surfaces acquired during and immediately after the stress plateau. The images (I)–(IV) in Fig. 1D correspond to the in-situ observations of the F- and R-surface at the respective points (I)–(IV) marked in Fig. 1C. At the initial loading point (I), several 45° line-like contrasts appear on the F-surface, while 0° line-like contrasts are observed on the R-surface. At point (II), marking the onset of the plateau, new 45° or 0° line-like contrasts appear on the F- and R- surfaces, respectively. Additionally, -45° line-like contrasts originate from the sample edges, connecting with the pre-existing 45° line-like contrasts at those edges. At point (III), the development of additional line-like contrasts on both the F- and R-surfaces becomes evident. State (IV) corresponds to the post-plateau stage reached upon further displacement-controlled compression. At point (IV), all line-like contrasts eventually disappear. Strikingly, the final visual appearance of the sample at point (IV) closely resembles its initial state, exhibiting the similar optical transparency and colorlessness as before mechanical loading. For videos showing the dynamic evolution of surface contrasts on the F- and R-surface, see the [Supplementary Materials \(movies S1 and S2\)](#).

The stress plateau extends over a strain range of approximately 1%. Notably, the lattice constants of the ferroelectric phase of tetragonal BaTiO_3 are $3.9958 \text{ \AA}$ along the a axis and $4.0363 \text{ \AA}$ along the c axis [32], yielding a c/a ratio of 1.011, corresponding to approximately 1% difference. This value nearly matches the 1% strain range observed in the plateau, suggesting that 90° polarization switching, triggered by a few MPa of mechanical loading, may occur during the plateau stage. To investigate the rotation of the polarization direction during deformation, solid-state nuclear magnetic resonance (NMR) measurements were performed on both unloaded and 1.5% strained samples. Solid-state NMR spectroscopy, a non-destructive technique, analyzes domain distributions in millimeter-scale BaTiO_3 bulks by examining the electric field gradient (EFG) tensor at barium sites (Supplementary Fig. S1), which determines NMR frequencies and reflects the spontaneous polarization orientations of a_1 , a_2 , and c domains [33,34]. The spectrum of the unloaded sample, shown in Fig. 1E, features a single peak across all rotation angles, with no measurable shift in the peak position. This aligns with the fact that the unloaded sample has a single c -domain structure with uniform polarization direction along the z -axis. Fig. 1F displays the NMR spectrum of the 1.5% strained sample. Similar to the unloaded sample, the spectrum contains only a single prominent peak. However, the position of the peak shifts with the rotation angle. The NMR data reveal that the strained sample has only in-plane a domains, indicating 90° polarization switching from c domain to a domains. For detailed NMR data together with X-Ray diffraction analyses, see [Supplementary Text A and B](#).

Thus, the original lateral y -axis became the new polarization direction as a result of applying only a few MPa of stress. This means that the polarization, initially aligned along the z -axis (Fig. 1A), underwent a 90° switching. The line-like contrasts observed in Fig. 1D can therefore be attributed to DWs associated with 90° polarization switching in individual domains. The stress required for such 90° domain switching is far below the mechanical threshold for plastic deformation; as shown in Fig. 1B, the yield stress for the onset of plasticity is 420 MPa, consistent with the high mechanical strength commonly observed in ferroelectric ceramics. While 90° polarization switching in micrometer- and nanometer-sized BaTiO_3 typically requires several hundred MPa of stress [18,31], it is noteworthy and surprising that in the millimeter-sized bulk samples studied here, switching occurred under stresses of only a few MPa, two orders of magnitude lower. This remarkably low stress is believed to be a unique feature of large domains on the millimeter scale. Polarization switching is typically driven by an electric field, and the work done in this process can be estimated as $2P^S E_C$, where P^S is the saturation polarization and E_C is the coercive electric field. For mechanically induced domain switching, the criterion is that the mechanical work, expressed as $\sigma \epsilon$, must exceed a critical threshold, taken as

$2P^S E_C$ [35]. Using representative values of P^S and E_C in reference [34], the stress required to induce 1% strain is calculated to be 2.65 MPa, in good agreement with the experimental observations. This low stress suggests that, in principle, 90° domain switching in BaTiO_3 can be accomplished with minimal mechanical input.

Morphological evolution during polarization switching

We then examined the in-situ evolution of the line-like contrasts and the associated microstructures with increasing strain. Figs. 2A to 2F show optical images taken from the center of the F-surface, where Figs. 2A, 2B, 2D, and 2F correspond to points (I), (II), (III), and (IV) in Fig. 1D, respectively. Fig. 2C was captured at a strain between points (II) and (III), while Fig. 2E was taken at a strain between points (III) and (IV). To the right of each optical image, a schematic depicts the corresponding domain configuration. Each schematic was constructed so as to minimize head-to-head or tail-to-tail polarization arrangements, thereby ensuring electrical stability. Here, the blue and green lines denote the initially formed 45° and subsequently developed -45° line-like contrasts, respectively. Red areas correspond to the crossing areas of the two sets of contrasts. As shown in Fig. 2A, 45° line-like contrasts first emerged on the surface. With increasing strain, additional 45° line-like contrasts appeared and progressively thickened. Meanwhile, -45° line-like contrasts developed, accompanied by the emergence and expansion of the red crossing areas (see Figs. 2B and 2C). As the strain further increased, neighboring crossing areas approached one another and merged into larger connected areas (Figs. 2D and 2E). Ultimately, the integration of the crossing areas cascaded across the entire surface, resulting in a colorless and transparent state of the sample, as shown in Fig. 2F, corresponding to point (IV) in Fig. 1D.

Cascading nanodomainization

Surprisingly, the final colorless and transparent state shown in Fig. 2F appears optically similar to the initial single-domain state, even though the domain structure has undergone substantial restructuring. Fig. 3 presents observations of the final polarization distribution and schematics of microscopic domain evolution. Figs. 3A and 3C correspond to piezoresponse force microscopy (PFM) phase images of the F-surface (in-plane mode) and R-surface (out-of-plane mode), respectively. The PFM characterization was conducted after stress relief. Fig. 3B shows the sampling locations where the PFM images were acquired. In PFM, the vibration response of a cantilever tip in contact with the sample surface is detected under an applied AC electric bias, enabling the amplitude and phase of the piezoresponse to be mapped [36,37]. The in-plane mode probes in-plane polarization components through torsional vibration of the cantilever tip, whereas the out-of-plane mode detects deflection associated with surface-normal displacement to map the out-of-plane polarization distribution [36,37]. In Fig. 3A, red or blue regions represent domains with rightward or leftward polarization, while in Fig. 3C, the red or blue regions indicate domains with polarization outward or inward relative to the screen. The images from both the F- and R-surfaces reveal a layered domain structure in the final state. Approximately 8–16 alternating layers are observed along the vertical direction in Figs. 3A and 3C, corresponding to an average layer thickness of 63–125 nm. Along the lateral direction, approximately 3–7 domains are observed, corresponding to an average domain length of 143–333 nm. The observed nanodomain configuration does not originate from topographic artifacts; a detailed discussion is provided in [Supplementary Text A](#). In the unloaded state, the BaTiO_3 single crystal exhibits a macroscopic single-domain configuration with dimensions of $2.5 \times 2.5 \times 6.0 \text{ mm}^3$. Upon applying a few MPa of mechanical stress, the domain structure undergoes a dramatic refinement. Although this nanodomain structure is entirely different from the initial single-domain configuration before mechanical loading, the optical transparency remains unchanged. Moreover, the nanodomain

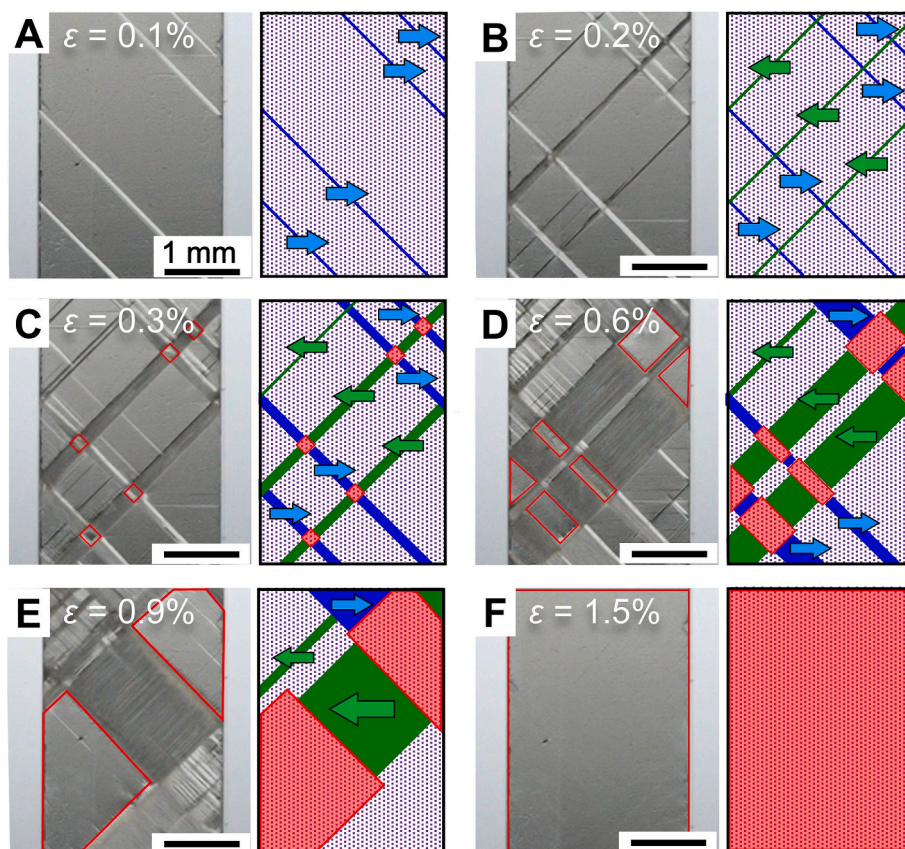


Fig. 2. Detailed in-situ images taken from the center of the F-surface. The strains in (A), (B), (C), (D), (E), and (F) correspond to 0.1%, 0.2%, 0.3%, 0.6%, 0.9%, and 1.5%, respectively. Here, the images on (A), (B), (D), and (F) are the same as those in (I), (II), (III), and (IV) of Fig. 1D.

configuration was mapped after removing the load, indicating that it persists after stress relief. Intriguingly, the polarization axis in the final domain configuration is aligned along a single lateral axis, despite the crystallographic equivalence of the x - and y -axes under uniaxial compression.

Phase-field simulations were further conducted to elucidate the microscopic domain evolution on the F-surface under compressive strain. In image (I) of Fig. 1D and Fig. 2A, 45° line-like contrasts emerge on the F-surface at the very early stage of uniaxial compression, indicating the formation of a_2 domains within the c domains. Phase-field simulations suggest that nanometer-scale surface steps locally perturb the otherwise uniform polarization distribution under the unloaded equilibrium state and thereby act as preferential nucleation sites for a_2 domains under applied compressive stress (Supplementary Text F). In Fig. 3D, one such $+a_2$ domain first nucleates within the c domains. Subsequently, $-a_2$ domains form and collide with the $+a_2$ domain, resulting in the creation of alternating thin $+a_2$ and $-a_2$ domains (Fig. 3E). These domains fold and grow along horizontal 180° domain walls at the intersection points (Fig. 3E), effectively reducing the electrostatic energy of unfavorable head-to-head domain configurations. These 180° folded domain structures gradually expand and eventually spread across the entire sample surface (Figs. 3F and 3G). The final domain configuration obtained from simulations (Fig. 3G) closely matches the PFM image in Fig. 3A. The complete movie for the phase-field simulation is available in the Supplementary Materials (Movie S3). Unlike electric-field-induced domains that align along a single polarization axis, mechanically induced domains in this study form a macroscopically nonpolar state with nanoscale charge mixing.

In ferromagnetic materials, domain width typically scales with the square root of sample thickness, following Kittel's law [38]. A similar scaling law has also been established for ferroelectric materials by

Mitsui and Furuichi [39]. The Kittel–Mitsui–Furuichi law describes equilibrium domain periodicity in unloaded ferroelectrics and remains applicable over a remarkably broad thickness range, from millimeter-scale bulk crystals down to nanometer-thick films [40]. In the present work, however, the application of compressive stress modifies the energetic balance governing domain periodicity, enabling the emergence of nanometer-scale domain structures in millimeter-sized single-domain BaTiO₃ crystals that would otherwise be energetically unfavorable within the classical Kittel–Mitsui–Furuichi framework. The observed nanodomains therefore represents a stress-driven deviation from the equilibrium domain periodicity scaling behavior. Notably, such mechanical nanodomainization can be achieved under compressive stresses of only a few MPa.

Dielectric responses related to nanodomainization

Changes in domain structure can lead to significant alterations in the functional properties of the BaTiO₃ crystals. Dielectric permittivity was measured for samples subjected to different strains. The measurements were performed after stress relief. Fig. 4A shows the stress-strain curve of a representative sample (the same as in Fig. 1C), while Fig. 4B presents the curve obtained from a different sample that exhibited atypical switching behavior. The dielectric permittivity measured after removing the load at selected strain levels is displayed above each stress-strain curve. Measurements were performed along the x -, y -, and z -axes, corresponding to the directions normal to the F-, R-, and T-surfaces in Fig. 1A, respectively. As shown in Fig. 4A, the unloaded single-domain sample exhibits clear anisotropy in permittivity. The permittivity along the x - and y -axes (the [100] and [010] axes, perpendicular to the polarization direction), was measured to be 4449 and 4339, respectively. In contrast, the permittivity along the z -axis (the [001]

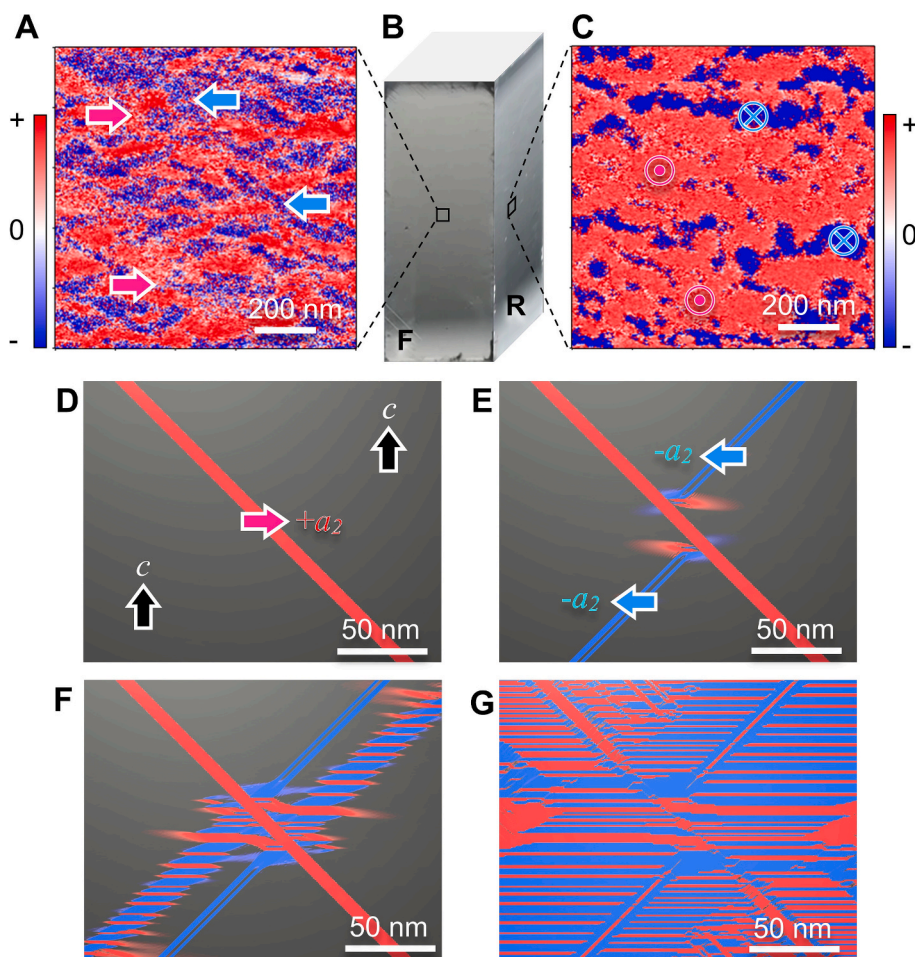


Fig. 3. (A and C) PFM phase images taken from the F-surface and the R-surface at the final state, respectively. (A) In-plane PFM phase image showing the in-plane polarization components. (C) Out-of-plane PFM phase image showing the out-of-plane polarization distribution. (B) Optical image of the loaded sample showing the locations where the PFM images of (A) and (C) were acquired. (D–G) Microscopic 90° domain switching behavior under compressive strain obtained by phase-field simulations on the F-surface.

polarization axis) was significantly lower, at 119. These anisotropic permittivity values are consistent with previous reports [41,42]. With increasing strain, these values changed significantly. In the 1.5% strained sample, the permittivity along the z-axis increased 17-fold to 2047, whereas the permittivity along the y-axis decreased by nearly 97% to 117, a value comparable to the original [001] axis permittivity. Remarkably, the permittivity along the polarization axis was restored to a value close to that of the original single domain, despite the presence of numerous domain walls and nanoscale domains. This highlights the dynamic relationship between strain-induced domain transformations and the functional properties. Detailed permittivity measurement results were shown in [Supplementary Text C](#). It should be noted that the present work focuses on the small-signal dielectric response. The measured permittivity may contain both intrinsic lattice contributions and extrinsic contributions arising from domain-wall vibration, as reported for BaTiO₃ ceramics by Hoshina et al. [43]. Quantitatively distinguishing these contributions would require dedicated broadband dielectric spectroscopy over a much wider frequency range and is beyond the scope of this study.

In most cases, the polarization axis after loading aligned uniformly along either the x-axis or the y-axis. However, in rare instances, the final state exhibited mixed polarization along both the x- and y-axes. [Fig. 4B](#) presents a representative stress–strain curve for such a rare case. Surface observations corresponding to this case are provided in [Supplementary Text D](#). As in most cases, the stress–strain curve deviated from elastic behavior at a low stress of about 3 MPa. However, the subsequent

behavior differed, with a temporary stress increase up to 8 MPa before conventional elastic behavior resumed by 1% strain. The permittivity of the final state in this case is also presented in [Fig. 4B](#). Unlike the results in [Fig. 4A](#), the permittivity along the x- and y-axes decreased to 1456 and 976, respectively, with neither value approaching that of the original z-axis.

The observed differences in the stress–strain curve and permittivity measurements suggest the occurrence of atypical 90° domain switching behavior. To further investigate this, NMR measurements were performed, and the results are shown in [Fig. 4C](#). Two distinct peaks were observed at all rotation angles, indicating the coexistence of a_1 and a_2 domains due to 90° switching along both lateral directions. The integrated intensity ratios in each spectrum reveal that two types of domains with different polarization axes coexisted at an approximate ratio of 49.2:50.8. It is worth noting that, despite applying mechanical loading to a substantial number of BaTiO₃ crystals, the formation of complex nanodomains, as observed in [Fig. 4B](#) and C, was rare. In most cases, the polarization axis aligned with only one of the lateral axes, as demonstrated in [Figs. 1, 2, 3, and 4A](#). To illustrate the contrast, [Figs. 4D](#) and [4E](#) provide schematic illustrations: most samples finally exhibit 90° domain switching along a single lateral axis ([Fig. 4D](#)), whereas rare cases show switching along both lateral axes ([Fig. 4E](#)).

Although the two lateral directions are crystallographically equivalent in an unloaded sample, 90° domain switching under a few MPa of mechanical stress typically occurs along only one lateral axis. One plausible explanation for this behavior is a slight variation in loading

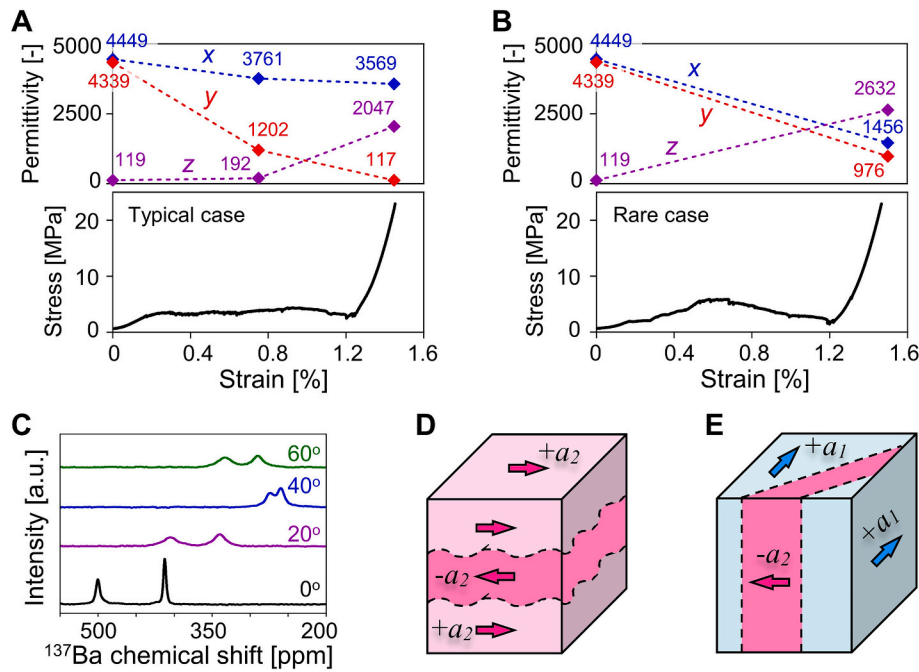


Fig. 4. (A) A representative stress–strain curve (bottom panel), which is the same as in Fig. 1C. (B) A different type of stress–strain curve showing different 90° domain switching behavior (bottom panel). The dielectric permittivity measured after removing the load at selected strain levels is displayed above each stress–strain curve. (C) NMR spectra obtained for the final state of the sample shown in (B). (D) A schematic of the final domain structure after the typical mechanical domain switching occurring in (A). (E) A schematic of the final domain structure when lateral domain switching occurs in two directions.

conditions, likely caused by inevitable deviations in sample orientation. Additionally, large single-domain crystals are inherently more prone to imperfections, including latent nucleation sites for domain walls. These pre-existing features may bias the formation and growth of domains along a more preferred polarization direction. As a result, even low mechanical stress can induce the emergence of a nanodomain configuration with polarization along a specific lateral axis.

Notably, the stress required to induce 90° domain switching in large bulk crystals is much lower than that reported for smaller-scale single crystals. According to Deng et al. [18], 90° polarization reversal in single-crystalline BaTiO_3 nanopillars with a diameter of approximately 130 nm completes under a compressive stress of 280 MPa. Sumigawa et al. reported a critical compressive stress of 210 ± 21 MPa for 90° domain switching in a 1.2- μm -long BaTiO_3 single crystal [31]. In contrast, our study demonstrates that a compressive stress of only 2–4 MPa is sufficient to induce 90° domain switching in millimeter-sized BaTiO_3 crystals. This trend suggests a possible scaling effect in mechanically driven 90° domain switching that resembles the well-established size dependence of electric-field-driven switching in ferroelectrics, where the coercive electric field E_c generally decreases with increasing sample dimensions from nanometer-thick films to bulk single crystals [26,29]. A possible origin of the scaling effect in mechanically driven 90° domain switching is that larger single domains are more likely to contain localized heterogeneities, such as crystal defects [44] and surface imperfections, that act as preferential nucleation sites for switched domains. In addition, the higher surface-to-volume ratio in smaller-scale materials may amplify surface effects, thereby restricting domain wall mobility through stronger interactions between domain walls and surfaces. It is also noteworthy that previous research on tetragonal $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ has shown that 90° domain switching has a lower activation energy than 180° switching [45], suggesting an intrinsically low energy barrier for mechanically driven 90° switching. Systematic experiments using crystals with controlled dimensions (e.g., micropillars with heights ranging from submicrometers to tens of micrometers, and bulk samples with thicknesses ranging from hundreds of micrometers to several millimeters), surface conditions (e.g., polished vs.

chemically etched vs. as-grown surfaces), and defect populations (e.g., nearly dislocation-free crystals versus crystals containing dislocations introduced by high-temperature plastic deformation) will be necessary to better clarify the mechanical threshold for 90° domain switching in ferroelectrics across different length scales.

Conclusion

This study demonstrates that bulk BaTiO_3 single-domain crystals can undergo unique nanodomainization under a few MPa of mechanical stress at room temperature. Millimeter-sized single domains evolved into nanometer-sized domain structures, deviating from the conventional equilibrium behavior set by Kittel's law, indicating a stress-induced modification of the energetic balance governing domain periodicity. Notably, the resulting polarization predominantly aligns along a single lateral axis, with only rare cases exhibiting alignment along both equivalent lateral axes. During the nanodomainization process, domain-wall-related contrasts cascade throughout the entire bulk, while NMR, XRD, and dielectric measurements confirm macroscopic 90° domain switching of the crystal. Yet, the resulting domain configuration visually resembles the initial single-domain state. The stress required to induce such 90° domain switching is remarkably low, only a few MPa, two orders of magnitude below the values typically reported for micrometer- and nanometer-sized single-crystalline BaTiO_3 . The comparison with literature data suggests a possible scaling effect of the mechanical threshold for 90° domain switching in ferroelectrics, which warrants systematic investigation in future work. Permittivity measurements further revealed that these nanodomain structures retain dielectric properties comparable to those of the original single-domain state, despite significant internal restructuring into complex nanodomain structures. Traditionally, domain engineering in ferroelectrics has relied on high electric fields and/or elevated temperatures. In contrast, our findings establish that a few MPa of mechanical loading at room temperature can effectively modulate both nanodomain structure and associated functional properties in bulk crystals. Beyond classic ferroelectric memory applications, this low mechanical threshold for

90° domain switching also points to new opportunities in ferroelastic actuators, elastocaloric cooling technologies, and mechanical tactile ferroelectric memory devices.

Materials and methods

Sample preparation

Bulk BaTiO₃ single-domain crystals provided by Crystalbase Inc., Japan, were used in this study. The sample size is $2.5 \times 2.5 \times 6.0 \text{ mm}^3$, with the long axis along the [001] polarization direction and the side faces perpendicular to the [100] and [010] axes. Prior to mechanical experiments, the BaTiO₃ single crystals were poled along the sample height direction, with the aim of aligning the polarization along the [001] crystallographic direction. The poling was performed at 130 °C under an electric field of 100 V cm^{-1} for 3 h. The achievement of a single-domain state was verified by NMR spectra, XRD, and macroscopic piezoelectric and dielectric measurements. Prior to uniaxial compression, the samples were sequentially cleaned with acetone, ethanol, and ultrapure water to remove surface contaminants.

Uniaxial compression tests

Mechanical loading tests were performed under uniaxial compression using a precision universal testing machine (AGX-V 50 kN, Shimadzu). The load and displacement data were recorded in real time. The test conditions were: initial strain rate of $\dot{\epsilon} = 1.0 \times 10^{-5} \text{ s}^{-1}$, room temperature (24 °C) in air, and the loading axis was oriented along the initial [001] direction, corresponding to the sample's long axis. Sample surfaces during mechanical loading were monitored in situ using digital single-lens reflex cameras ($\alpha 7 \text{S III}$ and $\alpha 7 \text{R III}$, Sony). Macroscopic shape changes of the samples were evaluated by measuring the dimensions of the samples using a digital micrometer before and after the loading tests. All uniaxial compression tests were performed under displacement-controlled mode. In a single continuous test, states (I)–(IV) in Fig. 1C were reached sequentially by increasing the applied displacement. Since the measured strain includes contributions from the jig, system compliance, and cushioning foils, the linear slope in the stress–strain curve does not correspond to the intrinsic Young's modulus of BaTiO₃.

Solid-state nuclear magnetic resonance (NMR) measurements

Solid-state ^{137}Ba NMR spectra were acquired using an Avance III HD spectrometer (Bruker) with a 14.1 T Oxford wide-bore magnet. A single-axis goniometer NMR probe (NMR Service, Erfurt) featuring 0.1° nominal resolution was tuned to 66.71 MHz. Spectra at specific angles were recorded by positioning a (100)/(010)/(001) face. The angle of 0° corresponds to the sample holder's normal vector aligned with the magnetic field B_0 . A 90° – 180° Hahn-echo sequence was applied with $\tau = 30 \mu\text{s}$, a 1 s recycle delay, and 0.05 s acquisition time. The 90° pulse duration was $3.5 \mu\text{s}$, with 1024 scans performed. Chemical shifts were referenced to a 1 mol BaCl₂ solution (0 ppm).

PFM characterization

Before PFM characterization, the samples were mechanically polished using diamond suspensions with particle sizes of $3 \mu\text{m}$ and $0.5 \mu\text{m}$, followed by chemical mechanical polishing with 60 nm colloidal silica to ensure adequate surface flatness. The samples subsequently underwent thorough ultrasonic cleaning. Local piezoelectric response was characterized at room temperature under ambient conditions using piezoresponse force microscopy (PFM, Cypher, Asylum Research, USA) equipped with conductive EFM probes (NanoWorld, Model: EFM, Pt/Ir coating, spring constant of 2.8 N/m, resonance frequency of 75 kHz). The AC drive amplitude was set to 800 mV, and the scan rate was 0.5 Hz. The dual AC resonance tracking (DART) mode was employed to track

the contact resonance frequency for optimal signal sensitivity. The set-point force was carefully adjusted to maintain stable tip-sample contact. Further details of the PFM measurement mechanism and how to distinguish positive and negative signal directions are provided elsewhere [37].

Dielectric permittivity measurements

Dielectric permittivity was measured by applying a 1 V AC electric field to a sample using an impedance analyzer (HP 4192 A, Hewlett Packard). The measurements were conducted on different samples over a broad frequency range from 1 Hz to 10^5 Hz . The dielectric permittivity values at 10^4 Hz were shown in Figs. 4A and 4B. More detailed data were shown in Fig. S7.

Phase-fields simulations

Phase-field simulations with the spontaneous polarization $\mathbf{P} = (P_1, P_2, P_3)$ as an order parameter were performed to simulate the formation and evolution of polarization microstructures in a BaTiO₃ single crystal during the compression loading. The two-dimensional simulation model has a $1900 \text{ nm} \times 2000 \text{ nm}$ BaTiO₃ region with 50 nm thick vacuum regions at both ends along the vertical direction. The entire simulation model is divided into 5 nm square grids, resulting in a total of 400×400 grid points. Periodic boundary conditions are applied to the polarization, electric field, and strain in both directions. As an initial condition, a uniform upward polarization is given to the BaTiO₃, with a 10 nm width horizontal domain (90° domain) introduced to mimic an initial domain wall. The system is first relaxed to a stress-free equilibrium state. Subsequently, vertical compression loading is applied by 0.5% compressive strain increments. The time-dependent Ginzburg-Landau (TDGL) equation is expressed as

$$\frac{\partial P_i}{\partial t} = -L \frac{\delta F}{\delta P_i}$$

where t is time, L is the kinetic coefficient, and F is the total free energy of the system. The TDGL equation was solved numerically [46,47]. The total free energy consists of Landau energy, gradient energy, electrostatic energy, and elastic energy, and is written as

$$F = \int f dV = \int (f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electro}} + f_{\text{elastic}}) dV$$

where f is the total free energy density. The details of each free energy term and material parameters can be found in previous studies [48–51]. In addition, the mechanical equilibrium equation written as

$$\sigma_{ij,j} = \left(\frac{\partial f}{\partial \epsilon_{ij}} \right)_j = 0$$

where ϵ_{ij} is strain, and Maxwell's equation written as

$$D_{i,i} = \left(-\frac{\partial f}{\partial E_i} \right)_i = 0$$

where E_i is an electric field, were solved simultaneously with the TDGL equation. More details are presented in Supplementary Text E.

Funding sources

This work was mainly supported by Japan Society for the Promotion of Science KAKENHI grant numbers JP24H00285, JP24H00032, JP23H00159, and JP24K17169. This project is partly supported by State Key Laboratory of New Ceramic Materials Tsinghua University (No. KF202501). T. Shimada acknowledges Japan Science and Technology Agency FOREST Program grant Number JPMJFR222H. F. Z. acknowledges the German Research Foundation (DFG) (project No. 530438323).

and the start-up grant from the University of Science and Technology of China (Project No. KY2030000260). G. B. acknowledges funding from the DFG under contract Bu-911-28-2. K. W. acknowledges support from the National Natural Science Foundation of China (no. 52325204). T. Shibamoto acknowledges the support for pioneering research initiated by the next generation from Japan Science and Technology Agency (grant number JPMJSP2138).

CRediT authorship contribution statement

Takeshi Shibamoto: Writing – review & editing, Writing – original draft, Visualization, Investigation, Data curation. **Yan Li:** Writing – review & editing, Writing – original draft, Validation, Supervision, Investigation, Funding acquisition, Data curation. **Kohta Kasai:** Visualization, Software, Investigation, Data curation. **Yuqi Jiang:** Investigation, Data curation. **Felix Dietrich:** Investigation, Data curation. **Ze Xu:** Writing – review & editing, Investigation. **Keitaro Horikawa:** Resources, Methodology. **Fangping Zhuo:** Writing – original draft, Validation, Resources, Methodology, Investigation, Funding acquisition, Data curation. **Ke Wang:** Writing – review & editing, Methodology, Funding acquisition. **Gerd Buntkowsky:** Resources, Methodology, Funding acquisition. **Takahiro Shimada:** Writing – original draft, Validation, Supervision, Software, Resources, Methodology, Funding acquisition. **Atsutomu Nakamura:** Writing – original draft, Validation, Supervision, Resources, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors acknowledge helpful discussion with Prof. Jürgen Rödel at Technical University of Darmstadt.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.mattod.2026.103433>. Supplementary material includes details of NMR, PFM, TEM, XRD, optical characterization, dielectric measurements, phase-field simulations and analyses, supplementary figures (Figs. S1–S9), supplementary tables (Tables S1 and S2), and related references. Additional movies supporting the results of this study are provided as Supplementary videos 2–4.

Data availability

Data will be made available on request.

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