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Mechanical tailoring of dislocations in ceramics at room temperature: A perspective

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Abstract

The potential of dislocations (line defects) in ceramics may have been greatly underrated until most recently. Promising proofs-of-concept have been demonstrated for dislocation-tuned functional and mechanical properties, revealing a new research front for dislocations in ceramics for a wide range of potential applications. However, it is commonly known that ceramics are hard (difficult to deform) and brittle (easy to fracture), particularly at room temperature. It remains a great challenge to mechanically tailor dislocations in ceramics. To address this pressing bottleneck, this article discusses the mechanics-based dislocation engineering in ceramics by examining the three fundamental factors of dislocation nucleation, multiplication, and motion. Successful experimental approaches to tune dislocation density and plastic zone size on single-crystal strontium titanate are demonstrated. The dislocation-based competition between plastic deformation and crack formation is discussed. The aspects of coupling external fields to manipulate dislocations are highlighted, which may hold the key to modulating the charged dislocation cores in ceramics and opening new routes for mechanical tailoring of dislocations at room temperature. Some open questions and challenges for engineering dislocations in ceramics are discussed.

KEYWORDS
charged dislocation core, dislocation engineering, dislocations in ceramics, room-temperature plasticity, strontium titanate

1 | INTRODUCTION: ARE DISLOCATIONS (UN)IMPORTANT IN CERAMICS?

Dislocations, one-dimensional or line defects in crystalline solids, are the main carriers of plastic deformation. Dislocations were first conceptualized by Taylor,[1] Orowan,[2] and Polanyi[3] independently in 1934, long before the experimental observation. Nowadays, dislocations are best known and most extensively studied in metals for their importance on the mechanical properties such as strength, hardness, ductility, and toughness. In ceramics, dislocations have long been a research "outlier": putting *dislocations* and *ceramics* together may even seem odd contrasting the
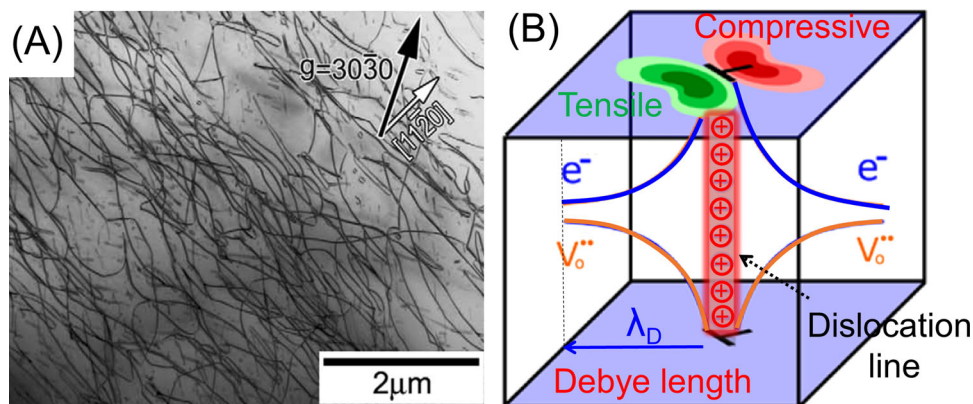
FIGURE 1 (A) Transmission electron microscopy image of dislocation lines in Al_2O_3 after uniaxial compression first at 1400°C and subsequently at 1200°C. Image reprinted and modified with permission from Ref. [17]; (B) schematic representation of charged dislocation core with a surrounding space charge layer, with the strain field and the radius of space charge zone indicated. Note that the schematic illustration is idealized, as the real situation is more complicated.

conventional picture that "ceramics are brittle" and display little plasticity at ambient temperature. Rarely has the topic of *dislocations in ceramics* been addressed in ceramics textbooks. An exception is the one by Carter and Norton,[4] with a chapter entitled "Are dislocations unimportant?" dedicated to ceramics. With such a chapter title, they even (maybe jokingly) wrote in the preface, "Yes, we have tried to be a little provocative at times."

In fact, research on dislocations in ceramics has a long history. One of the earliest direct evidence of dislocations in crystals was reported in 1951 on single-crystal SiC (Carborundum, with the surface growth spirals observed), independently by Verma and Amelinckx.[5] In this sense, the study of dislocations in ceramics can date back by more than 70 years. In the late 1950s and 1960s, Gilman and Johnston systematically investigated dislocation mobility and multiplication[6–10] with their comprehensive and elegant dislocation etching studies on single-crystal LiF. At about the same time, research on dislocations in semiconductors started to rise,[11] marked by the seminal paper by Gallagher in 1952[12] on the high-temperature plastic deformation of germanium and silicon. There were extensive studies on dislocations in semiconductor materials (Si, Ge, GaAs) in the past decades. However, dislocations in semiconductors have long been undesired as they act as scattering and trapping centers, significantly reducing the mobility of charge carriers and degrading the functional properties.[13] Great efforts have been made to avoid dislocations in mainstream semiconductors during the growth of crystals and thin films.

Recent years' research progress in ceramics oxides brings a new perspective, where dislocations are purposefully engineered to harvest both functional properties and mechanical properties. Dislocations in ceramics (Figure 1A) are recognized to bring degrees of free-dom to tune the functional properties owing to their elastic and electrostatic (charged) features. In addition to the local elastic strain field, dislocations in ionic ceramics feature a charged dislocation core, with a surrounding charge-compensation layer described by the Debye-Hückel cloud[14] (Figure 1B). The charge and local strain field offer abundant opportunities to tune the energy barrier for thermal/electrical scattering and provide sites for enhanced ion/electron transport and chemical reaction rate.[15] This opens an exciting new field for dislocation-tuned functionality with proofs-of-concept established for photoconductivity,[13] electrical conductivity,[16–18] superconductivity,[19] and large-signal piezoelectric coefficient ($d_{33}*$).[20] From the mechanical point of view, once engineered into ceramics, dislocations can improve plasticity and increase fracture toughness (resistance to crack propagation).[21–25] Both aspects will be elaborated on in the following section.

The field of dislocation-tuned functionality and mechanical properties has been largely neglected in the past as ceramics are typically sintered at high temperatures, with extremely low dislocation density of ~10^9 to ~10^{10}/m^2 [26,27] retained in the sintered samples. This means one barely finds a dislocation intersecting an area of tens of square micrometers in a ceramic sample. The consequences due to the lack of preexisting dislocations and the strong covalent/ionic bonds in ceramic materials are evident in their mechanical properties, as most ceramics are commonly known to be brittle, with high strength but poor ductility and low fracture toughness.[28] Yet, dislocations with high density (up to ~10^{13}/m^2 and higher) are often required to significantly improve functionality.[27,29,30] The grand challenge of tailoring dislocations of high density in large plastic zones in ceramics without crack formation, particularly at room temperature, leads to limited studies

so far. Although abundant research on high-temperature deformation of ceramics is available, prominently in the 1970–1990s,[31–33] the focus then was completely different as barely any attempt was made to purposely tune the dislocation density, arrangement, and plastic zone size.

Dislocations are undoubtedly important in ceramics. Considering the enormous amount of studies scattered over the last 70 years in various fields (materials science, semiconductor physics, mechanics, defect chemistry, geology, etc.) with a wide range of testing conditions (temperature, pressure, microstructure, sample size, strain rates, etc.) on different materials[34] (see also examples in following sections), no attempt will be made at this stage for an exhaustive review of dislocations in ceramics. Instead, this work aims to sketch a practical framework, with a focus on room-temperature mechanical tailoring of dislocations in ceramics.

The urgent need for dislocation-rich ceramic samples for testing versatile functionality and mechanical properties was the original driving force for the current author and his team to investigate mechanics-based dislocation engineering in ceramics, with a particular focus on room-temperature plastic deformation of ceramics for the sake of simple, fast, and energy-efficient dislocation engineering. Targeting this objective, we have developed different experimental protocols and made promising progress.[15,22,25,35–45] In what follows, the author will first present an overview of dislocation-tuned functional and mechanical properties in ceramics, demonstrating concrete examples to showcase the potential held by dislocations. Various approaches and methods for engineering dislocations into ceramics will be demonstrated, with a focus on a variety of boundary conditions and their impact on plasticity. Then, the fundamental factors of dislocation nucleation, dislocation multiplication, and dislocation motion in ceramics will be examined, with direct experimental visualization. Furthermore, the potential of external physical fields, in particular electric field and light illumination, which have been demonstrated to hold the key to modulating dislocation mobility in certain ceramics even at room temperature, will be discussed based on charged dislocation cores and their potential interaction with charged point defects. In the end, open questions and challenges that need to be tackled for dislocations in ceramics are posed.

2 | DISLOCATION-TUNED PROPERTIES OF CERAMICS

Plenty of proofs-of-concept for dislocation-tuned properties in ceramics have been established in bi-crystals,[46,47] thin films,[48,49] single crystals,[19,20,29] and polycrystalline samples.[27,50,51] Functional-wise, dislocations are engineered to tune the physical properties such as electrical conductivity, thermal conductivity, and photoconductivity, to name a few. Mechanical-wise, the focus has been placed on dislocation-enhanced plasticity, fracture toughness, and damage tolerance. The following section will briefly summarize and discuss some representative progresses.

2.1 | Dislocation-tuned functional properties

Since early 2000, several research milestones for dislocations in ceramic oxides have been made. The concept of using dislocations as conducting nanowires in insulating Al_2O_3 was proposed by Nakamura et al.[16] who engineered dislocations as 1D nanowires by high-temperature bulk deformation, first at 1400°C with a plastic strain up to 5%, followed by further deformation up to 10% plastic strain at 1200°C. Titanium was then diffused through pipe-diffusion along these dislocations with a density up to $\sim 10^{13}/m^2$. Using conducting scanning probe microscope, they found an increase in the conductivity $(1 \times 10^{-1}\,\Omega^{-1}\,cm^{-1})$ by almost 13 orders of magnitude along Ti-decorated dislocations compared with high-purity sapphire. The concept of dislocation nanowire was further explored in other material systems such as single-crystal yttria-stabilized zirconia and bi-crystals.[52] The dislocation engineering technique shifted also from bulk compression of single crystals to well-aligned edge/screw dislocations in bi-crystals fabricated by thermal diffusion bonding.[53] The bi-crystal fabrication technique turns out to be powerful and will be discussed in more detail in Section 3.1.

In single-crystal $SrTiO_3$, Szot et al.[54] reported in 2002 a localized metallic conductivity after thermal reduction treatment. The increased conductivity was attributed to the high concentration of vacancy defects along the dislocations (the extended defects) in the near-surface region. The dislocation density was estimated to be $\sim 10^{13}/m^2$ via chemical etching. Another extensively investigated model system by Szot and colleagues is TiO_2, which can be found in their reviews.[26,55] Most recently, Hameed et al.[19] reported dislocation-tuned superconductivity well above the superconducting transition temperature (T_c) in Nb-doped single-crystal $SrTiO_3$ that was plastically deformed at room temperature. The superconducting correlations appear at temperatures as high as $30-50$ K in the deformed sample,[19] which is suggested to be influenced by the local strain surrounding the dislocations. Höfling et al.[20] mechanically imprinted dislocation networks into bulk single-crystal $BaTiO_3$ by uniaxial compression experiments at 1150°C and increased the large-signal piezoelectric coefficient ($d_{33}*$) by 19-fold. Such a giant increase
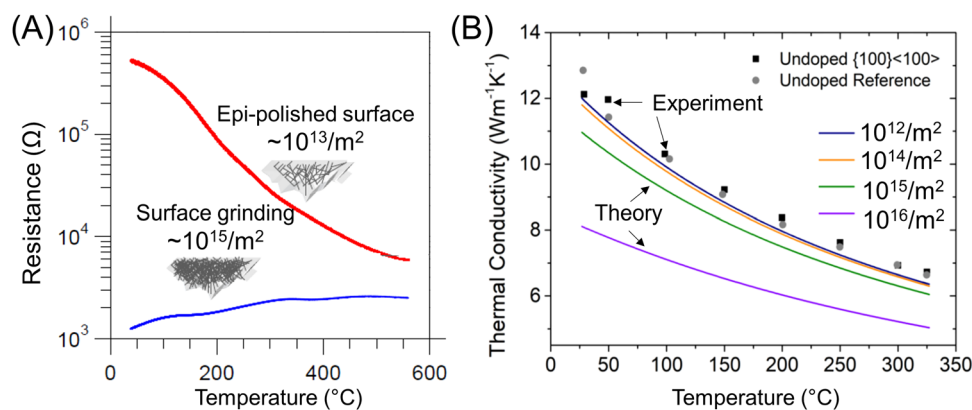
FIGURE 2 Impact of dislocations on functionality in single-crystal SrTiO$_3$: (A) resistance decreases by ~3 orders of magnitude if the density increases by ~2 orders of magnitude at 25°C (Image reprinted and modified with permission from.[26] with source data extracted from Refs. [55] and [57]); (B) thermal conductivity as a function of temperature and dislocation density. Image reprinted and modified with permission from Ref. [27].

is attributed to the ferroelectric domain structures being pinned by the dislocations, which strongly modify the electromechanical forces for dislocation-domain interactions. A similar approach using high-temperature (up to 1050°C) bulk deformation was adopted to plastically deform single-crystal TiO$_2$, in which the electrical conductivity was increased by a factor of 3 with an average increase of dislocation density from ~10^{10}/m^2 in pristine samples to ~10^{13}/m^2 in deformed ones.[29] These promising proofs-of-concept may extend the spectrum of designing tools for dislocation-based functional devices for thermoelectrics, superconductors, and functional ferroics.[20]

Since these functional properties are directly induced by dislocations, it is expected that the functional performance will be further improved with higher dislocation density. For instance, Figure 2A illustrates that an increase in dislocation density from ~10^{13} to ~10^{15}/m^2 leads to a decrease in the electrical resistance by almost 3 orders of magnitude in single-crystal SrTiO$_3$. Furthermore, Figure 2B features the predicted decrease of thermal conductivity (solid lines) of single-crystal SrTiO$_3$ as a function of dislocation density and temperature. A dislocation density of ~10^{16}/m^2 is theoretically predicted to match the length scale of the mean free path (several nm for SrTiO$_3$[56]) as well as the elastic strain field for strong phonon scattering to reduce the thermal conductivity by ~30% at 25°C.

Concerning electrical conductivity, dislocation density itself is not the sole player. A whole range of dislocation mesostructure complexity (kinks, pairs, dipoles, screw/edge/mixed dislocation, etc.) as well as space charge and dislocation core effect has been discussed by Porz et al.[58] and later reviewed by Armstrong et al.[59] A critical examination as well as some open discussions on the impact of dislocations (cores) on the transport properties in SrTiO$_3$ and related perovskite oxides can be found in recent viewpoint works by De Souza.[60,61]

Contrasting the complex mesostructure and impact of dislocations cores,[62] the functional anisotropy is more readily tunable thanks to the intrinsic asymmetry of the strain field and extrinsic structural arrangement of dislocations on different slip planes. The anisotropy of the thermal conductivity[48] and superconductive response[63] parallel and perpendicular to the dislocations are reported in previous studies. For instance, the anisotropy ratio due to the dislocation arrangement can be as high as 10 for thermal conductivity.[48] Another significant example of functionality tuning is highlighted by Zhuo et al.[64] on the dislocation—domain wall interaction in single-crystal BaTiO$_3$ dependent on their spatial arrangement. Such an anisotropy engineering approach is critical for future device design.

2.2 | Dislocation-tuned mechanical properties

Dislocations are the main carriers of plasticity.[14] Engineering dislocations into ceramics can therefore significantly impact the mechanical properties such as yield strength, hardness, and creep. More attractively, dislocations can improve fracture toughness and damage tolerance to combat the brittleness of ceramics. For instance, Li et al.[65] reported ~12% plastic strain at room temperature with micro-pillar compression of flash-sintered TiO$_2$. The large plasticity (six times higher than conventionally sintered TiO$_2$) is proposed to be caused by the enrichment of preexisting dislocations and stacking faults generated during the highly nonequilibrium sintering process. Nakamura et al.[66] demonstrated a tunable room-temperature plasticity in single-crystal SrTiO$_3$ by changing the Sr/Ti ratio in the starting powder and the dislocation density.[43] The samples with a higher dislocation density (~10^{11}/m^2)
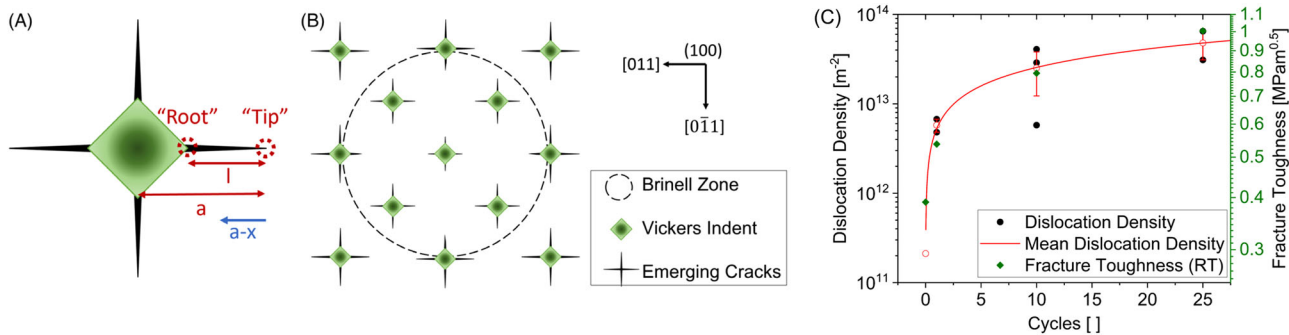
FIGURE 3 (A and B) illustration of the indentation crack length (ICL) method, with Vickers indents performed inside the plastic Brinell zone. (C) Indentation fracture toughness increases with the dislocation density increase. Figures adapted from Ref. [25]. Image used is under Creative Commons CC-BY License.

exhibit lower yield strength (~15% decrease) and much higher fracture strain (~70% increase).[66] Besides tailoring plasticity, Moon et al.[21] reported an increase in fracture toughness from 0.9 MPa m$^{1/2}$ (as received) to 3.7 MPa m$^{1/2}$ in single-crystal Al_2O_3 by shot blasting and then high-temperature annealing at 1400°C to form dislocation sub-boundaries in the sample surface. Porz et al.[22] demonstrated a twofold increase in the crack-tip toughness by engineering surface dislocations in single-crystal $SrTiO_3$.

Most recently, Okafor et al.[67] from the current author's group used the cyclic Brinell indentation technique and achieved a plastic zone size of up to ~200 μm (in diameter and depth) with tunable dislocation density up to ~10^{13}/m^2. Based on this technique, Salem et al.[24] further introduced an additional step of thermal treatment of the Brinell plastic zone and increased the pre-engineered dislocations further by one order of magnitude to ~10^{14}/m^2. With the indentation crack length (ICL) method, we evaluated the fracture toughness and hardness, both are increased by ~20%. A more significant increase of the dislocation-based fracture toughness by a factor of ~3 using the ICL method is demonstrated on single-crystal $KNbO_3$ with a density up to ~10^{14}/m^2 (Figures 3A and B),[25] in line with the experimental protocol by Salem et al.[24] Analogous to the functional properties, the mechanical properties are also found to be dependent on the dislocation density. Take the example of $KNbO_3$, the higher fracture toughness is correlated to higher dislocation density (Figure 3C).

2.3 | A combination of both

Down the road, it is realized that so far there has been a lack of combining both functional and mechanical properties tuned by dislocations. The brittleness of most ceramic materials leads to low fracture toughness. Mechanical reliability can become a critical point for the functional devices made of these materials. As separately discussed above, take the example of $SrTiO_3$, it has been demonstrated that engineering dislocations through mechanical deformation increases not only the electrical conductivity[26] but also enhances the fracture toughness.[22,24] This promising example suggests that a simultaneous enhancement of the mechanical and functional properties is achievable via the single engineering ingredient—dislocations. The question now is, how can we efficiently engineer dislocations into ceramics without breaking them?

3 | DISLOCATION ENGINEERING IN CERAMICS

3.1 | Methods to introduce dislocations into ceramics

The key prerequisite for harvesting the dislocation-tuned properties in ceramics is the successful introduction of dislocations into ceramics. This can be achieved mainly by (1) interface design, (2) processing, (3) irradiation, and (4) mechanical deformation. Although the focus of this work is on the mechanical tailoring of dislocations, the other methods are briefly discussed here for comparison.

3.1.1 | Interface design

Well-aligned dislocations can be produced by bi-crystal interface diffusion bonding[53,68–70] (Figure 4A). By tiling or twisting the angle between the two joining crystals, edge or screw dislocations with controllable spacing can be engineered. This approach allows an elegant control of dislocations at the interface and has been favorable for fundamental studies and atomic structures in various
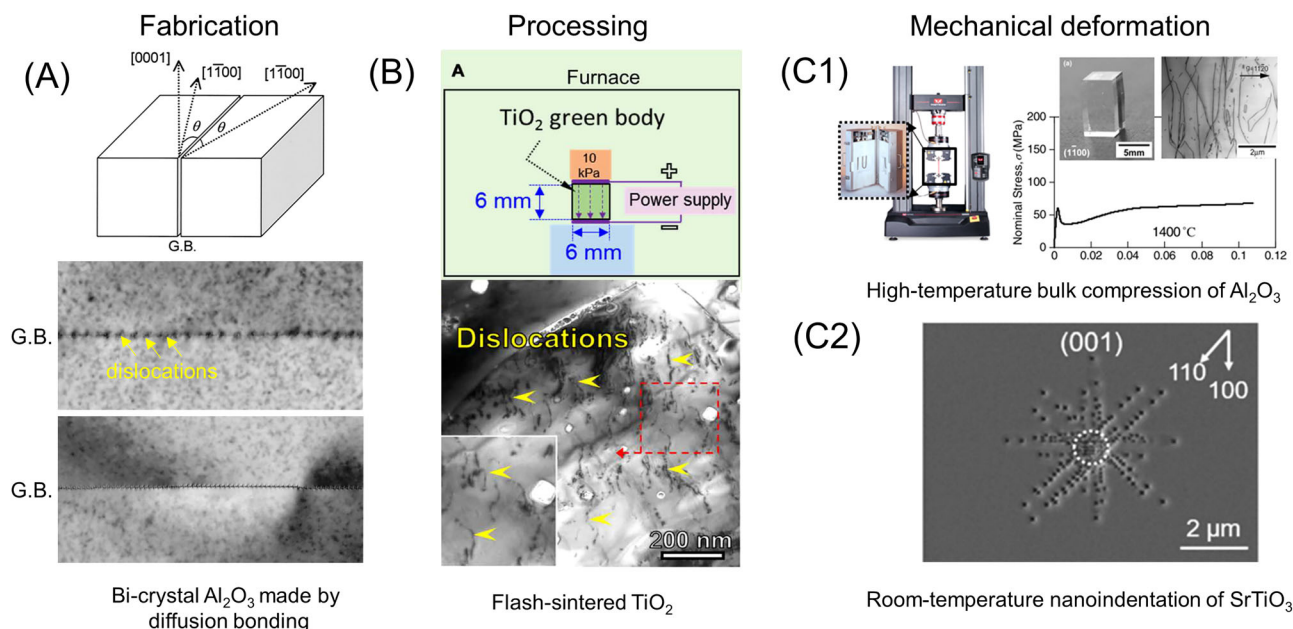
FIGURE 4 Different approaches to engineer dislocations into ceramics: (A) bi-crystal fabrication, image reprinted and modified with permission from Ref. [53]; (B) flash sintering, image reprinted and modified with permission from Ref. [65]; (C1) high-temperature bulk deformation, image reprinted and modified with permission from Ref. [17]]; (C2) room-temperature nanoindentation test image reprinted and modified with permission from Ref. [43].

oxide systems, such as Al_2O_3,[53] ZrO_2,[71] TiO_2,[72] and $SrTiO_3$.[73] Another example of interface design is thin film deposition. By using interface strain caused by atomic mismatch (heteroepitaxial growth), high-density threading dislocations (up to $\sim 10^{14}/m^2$)[48] can be achieved by, for example, using plasma-assisted molecular beam epitaxy. Sun et al.[48] found such a high dislocation density in InN films can effectively reduce the thermal conductivity. A dislocation-density-dependent thermal conductivity was also identified. Both methods require strict growth parameters (temperature, pressure, impurity of the crystals, etc.) to achieve samples of high quality.

3.1.2 | Processing

High-density of dislocations up to $\sim 10^{14}/m^2$ can be generated by using high-pressure torsion[74] and new sintering techniques such as flash sintering of TiO_2[65] (Figure 4B), spark-plasma sintering of $SrTiO_3$,[75] and cold sintering of $(K_{0.5}Na_{0.5})NbO_3$[76] and ZnO.[77] These processing techniques yield polycrystalline samples that are very often rich in pores and micro-cracks, while grain boundaries also add to the structural complexity. A highly heterogeneous structure may be produced (bimodal grain-sized samples, with only a fraction of grains containing dislocations) and it is still not possible to control dislocations via such sintering approaches.

3.1.3 | Irradiation

Dislocation loops can be produced by the ion irradiation approach.[51,78] Using 1.6 MeV protons at 700°C on a polycrystalline cerium dioxide sample, Khafizov et al.[51] observed faulted dislocation loops in the irradiated sample, which also displayed lower thermal conductivity. Ion irradiation may also induce amorphization[79] and very often is limited to the skin region of the samples.

3.1.4 | Mechanical deformation

Aside from the above approaches, dislocation engineering via mechanical deformation offers the opportunity to deform samples from bulk[20] (Figure 4C1) to submicroscales[43,80–82] (Figure 4C2), from single crystals to polycrystalline samples,[27] from room temperature[42,83–85] to high temperatures.[16,52] The given slip systems in the crystals also confine the dislocations on their slip planes,[27,52,83] which allows for functional anisotropy tuning, as briefly discussed above. Yet mechanical deformation can be rather complicated when it comes to tuning plasticity while suppressing crack formation. The mechanical deformation boundary conditions play a crucial role in determining whether or not the deformed sample will survive or just fracture.

3.2 | Mechanical deformation: boundary conditions matter

Plastic deformation in ceramic materials can be achieved given the right deformation conditions. These boundary conditions for mechanical deformation cover a wide spectrum of parameters such as temperature, pressure, microstructure (texture), length scale, and crystal orientation, to name a few. They are briefly discussed here.

3.2.1 | Temperature

Due to the strong ionic and/or covalent bonding in ceramics, which is also responsible for the high Peierls stress (usually several orders of magnitude higher than in FCC metals[86]), plastic deformation of most ceramic materials strongly relies on thermal activation and dislocation dissociation.[87] Dislocations are thus introduced mainly via high-temperature bulk compression above, for example, 1000°C.[16,17,20,27,88] It is not the intention of this work to focus on plastic deformation of ceramics that require thermal activation at high temperatures, which were heavily studied in the past, and comprehensive review papers and book chapters particularly focusing on high-temperature dislocations in ceramics are available.[31,32,87] Thermal activation at temperatures above the brittle-to-ductile transition temperature (BDTT) can activate additional slip systems, which are required for dislocation-mediated plastic deformation in polycrystalline ceramics. This will be explained in greater detail in the next section.

Quite some ceramic materials (single crystals) exhibit large plasticity at temperatures lower than 800°C, which may hold much more engineering potential. For instance, besides the widely studied *ductile* single-crystal MgO[89,90] and alkali halides (e.g., LiF,[10] NaCl,[91] and KCl[92]), other technologically important oxides such as SrTiO$_3$[93] and KNbO$_3$[94] have been identified in recent years to exhibit large plasticity during room-temperature bulk compression.

Using SrTiO$_3$ as a model system (which is often adopted for dislocation-tuned functional properties in Section 2.1), a tunable plastic zone size from micrometers up to centimeters (in length), with dislocation density ranging from ~10^9/m^2 in the pristine samples up to ~10^{15}/m^2, has been achieved. These approaches have been established at room temperature without visible cracks. An overview of the methods and the generated dislocation density as well as plastic zone size is given in Panel A at the end of this article.

3.2.2 | Texture (microstructure)

Here, by texture it refers to the sample being either single crystal, bi-crystal, or polycrystalline. This makes a huge difference in the deformability, most crucially at room temperature. The von Mises criterion[95] and Taylor criterion[96] state that five independent slip systems are required for the general plastic deformation of polycrystalline materials. The insufficient available slip systems in ceramics (for instance, SrTiO$_3$ and most rock salt structured ceramics have six physically distinct {110} <110> slip systems at room temperature, but only two of them are independent[14,97]) do not allow effective slip transmission to take place during room-temperature plastic deformation, or even at high temperature.[98] The consequence is that dislocations would pile up, if any, at grain boundaries and initiate cracks.[99] Another widely encountered challenge for sintered ceramics is that there are abundant pores and/or micro-cracks present. Such defects are the primary sources for crack initiation/propagation. By activating additional high-temperature slip systems above BDTT, an appreciable plastic strain of polycrystalline ceramics can be achieved. For instance, in SrTiO$_3$ the room-temperature slip systems are {110} <110>, while at high-temperature additional slip systems {100} <100> will be activated.[27,100] However, at high temperatures, other active mechanisms such as high-temperature creep, dislocation climb, recovery, and grain boundary sliding[101] will add to the complexity of deformation mechanisms.

Aside from single-crystal deformation, an insightful experiment looking at the dislocation-grain boundary (GB) interactions at room temperature is carried out recently by Kondo et al. on bi-crystal SrTiO$_3$ using the in situ indentation technique in the transmission electron microscope (TEM)[70] (Figure 5). Their finding suggests that certain grain boundaries (in their case, a low-angle grain boundary) in ceramics indeed allow slip transfer (Figure 5B).

3.2.3 | Pressure

In this case, high pressure (usually accompanied by a high degree of confinement) is beneficial for the plastic deformation of even bulk ceramic materials. For instance, Rabier et al.[102] achieved dislocation-mediated plastic deformation of single-crystal Si at the macroscopic scale (with a sample size of $3.5 \times 2 \times 2$ mm^3) at room temperature under a confining pressure of 5 GPa (below the phase transition pressure) in a multi-anvil apparatus. This multi-anvil apparatus was designed by Cordier et al.[103] to generate large deviatoric stress in addition to the very high hydrostatic pressure (due to the high confinement), origi-
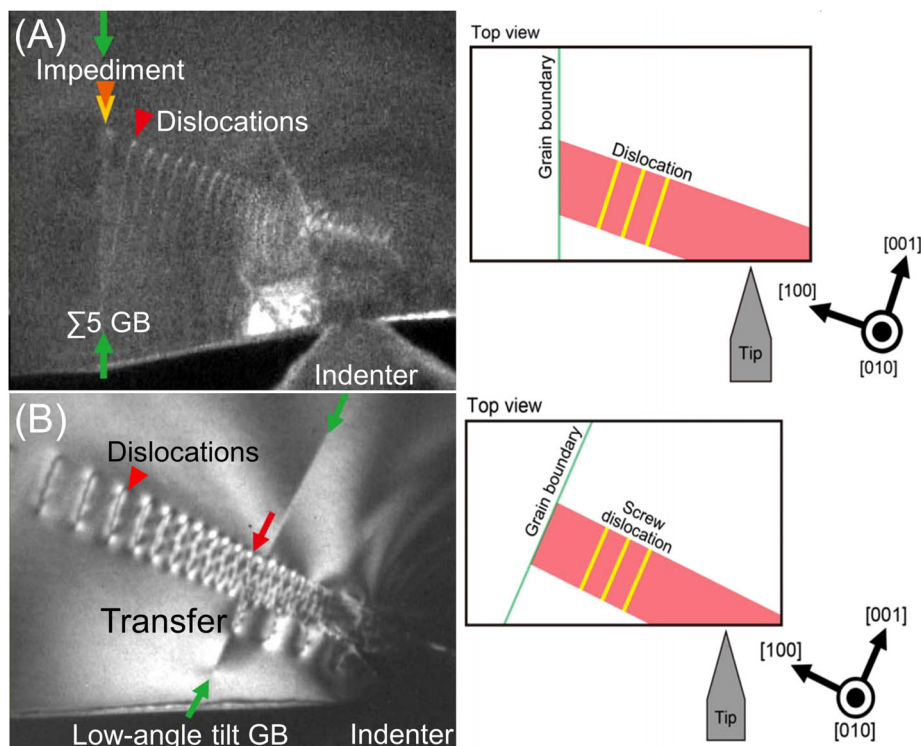
FIGURE 5 Direct observation of dislocation interaction with GBs (bi-crystal SrTiO$_3$) using in situ indentation in TEM, images adapted with permission from Ref. [70].

nally for geology studies. Another typical example would be the rheology study of mineral olivine[104,105] in geoscience, which utilizes a high-pressure cell to mimic the conditions in the earth's mantle. Contrasting the extremely large length scale in geology, the nanoindentation technique serves as another excellent example for generating locally extremely high pressure (high stress), for instance, with the Berkovich diamond indenter. Upon incipient contact, maximum shear stress of up to tens of GPa can be induced in the material being deformed underneath the indenter tip. Such a high level of shear stress is sufficient to break the bonds in most ceramic oxides to nucleate dislocations from an originally dislocation-free volume, as will be discussed in greater detail in the next section.

3.2.4 | Size effect

The size effect (sample size or deformed volume-dependent mechanical properties) in metallic materials has been extensively studied in the past decades, primarily focusing on the strength (plasticity) as a function of intrinsic microstructure or extrinsic sample size. The latter is further correlated to, in most cases, the population of defects such as dislocations in the stressed volume. For instance, the "smaller is stronger" effect is most evidently demonstrated in pillar compression,[106,107]

the nanoindentation size effect showcasing increased hardness with decreased indentation depth,[108,109] and the nanoindentation pop-in size effect[110] in metallic materials.

In contrast, the size effect in ceramics is slightly more complicated than merely yield strength and plasticity, but also the competition between plasticity and cracking. The advancement of the focused ion beam coupled with in situ testing platforms (in both SEM and TEM) has opened up a large number of micro-/nanoscale studies of plastic deformation of various ceramics at ambient temperatures. Typically, nanoindentation tests[41] and micro-/nanopillar tests[82] are employed to understand the plastic behavior (very often accompanied by cracking). For instance, quite some studies are published in the last two decades on micro-/nanopillar compression, addressing the plastic deformation of carbides,[111] oxides,[112,113] and mainstream semiconductors such as Si[114] and GaAs.[115] A common observation is that, once the diameter of the pillars decreases below a critical value (e.g., ~300 nm for Si[116] and ~150 nm for dislocation-free SrTiO$_3$[117]), plasticity sets in without fracturing the pillars and a large plastic strain can be achieved. Upon further deformation, dislocations within the intersecting slip planes start to strongly interact and/or react with each other, ultimately leading to crack initiation and failure of the samples.[116,118] This can be coined as size-dependent brittle to ductile transition[118] for materials that are usually considered brittle at the
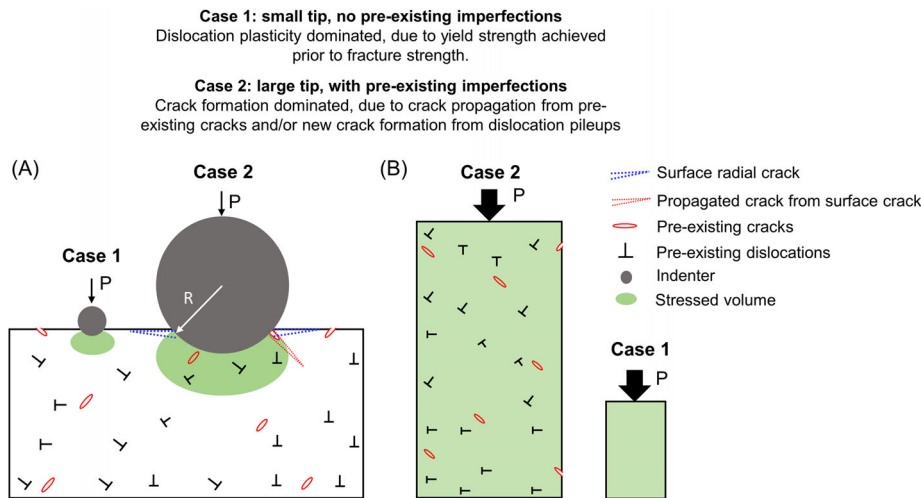
FIGURE 6 Effect of defect population on the competition between plasticity and fracture: (A) indentation tests; (B) uniaxial compression tests. Two ideal cases are illustrated: Case 1: small indenter tip or small-scale pillar compression, no preexisting imperfections exist in the stressed volume, where dislocation plasticity dominates due to yield strength achieved before fracture strength. Case 2: large indenter tip or bulk compression, with preexisting imperfections, where crack formation dominates due to crack propagation from preexisting cracks and/or new crack formation from dislocation pileups. Figure (A) adopted from Ref. [41] is under Creative Commons CC-BY License.

macroscopic scale. An extreme example is shown in diamond, commonly known as the hardest material in nature, that dislocation motion and multiplication are observed during in situ testing of nanopillar (∼200 nm in diameter) compression.[119]

In short, both plasticity and fracture in ceramics are scale-dependent[41,120,121] (Figure 6). At nano-/microscales, the resolved shear stress in the deformed volume may easily exceed the shear strength before the maximum tensile stress reaches the fracture strength, thus the dislocation plasticity is promoted while the fracture is suppressed. This is demonstrated in oxides by the current author and coworkers.[41] We examined the nanoindentation pop-in events on different oxides ($SrTiO_3$, $BaTiO_3$, TiO_2, and Al_2O_3) using different nominal indenter tip radii (from 100 nm up to 25 μm), and found an indenter-tip-size-dependent competition between purely dislocation-dominated plastic deformation (small tip radius) and simultaneous dislocation activation and cracking upon pop-in (large tip radius). On the safe side, a sharp Bekovich tip would facilitate the generation of dislocations while suppressing crack formation thanks to the high local shear stress and hydrostatic compressive stress.

3.2.5 | Orientation

The importance of crystal orientation manifests itself particularly in uniaxial compression due to the limited slip systems in ceramic materials, even for high-temperature deformation. For instance, $SrTiO_3$ deformed at high temperatures (between 1200−1500 K) shows a strong depen-dence on the compression axis, with samples compressed in the <100> axis more prone to fracture while other orientations such as <110> , <211> , and <543> exhibiting mainly plastic yielding.[100] For macroscale deformation, the critically resolved shear stress (CRSS) can be computed using the Schmid law, which resolves the compressive stress to the shear stress responsible for dislocation gliding along the slip plane. By properly choosing loading directions concerning the orientation of the slip plane, a maximum Schmid factor of 0.5 can be achieved. Such loading directions are usually referred to as "soft" deformation directions in, for example, cubic $SrTiO_3$ and MgO. On the other hand, loading directions with a Schmid factor of 0 are tagged as "hard" deformation directions, which are to be avoided to minimize the chance of fracturing the samples.

Contrasting macroscale compression, for micro-/nanopillar compression, the requirement for aligning the "soft" deformation directions becomes less critical as the deformed volume is most likely crack-free, which can sustain much higher compressive stress and a higher CRSS for activating dislocation glide on the slip planes. For indentation tests, the stress distribution underneath the indenter becomes much too complicated, plus extremely high local shear stress can be induced depending on the indenter tip radius, and the effect of orientations becomes less prominent.

3.2.6 | Other factors

For room-temperature deformation, the strain rate effect on ceramics deformation has long been studied dating

back to LiF.[122] A recent study with strain rates of 10^{-1} and 10^{-5}/s on bulk compression of SrTiO$_3$ by Nakamura et al.[66] can be found. A majority of studies are oriented in the direction of small-scale testing, for instance, nanoindentation tests. It has been most recently reported that for single-crystal SrTiO$_3$,[123] ZnO,[124] and ZnS,[125] no significant impact of the strain rates was observed on the dislocation generation during pop-in. On the other hand, the light illumination was found to exhibit a great impact on the creep strain rates on bulk compression of ZnS, which can be changed by up to 3 orders of magnitude with the light switched on/off.[126] It is clear evidence that light illumination significantly reduces dislocation mobility.

In short, there are abundant approaches to engineer dislocations in ceramics, ranging over a wide length scale and materials with different microstructures. For mechanical deformation, the various examples presented above suggest that it is critical to specify the boundary conditions for deforming ceramics when plasticity is the focus. Changes in temperature, pressure, sample size, microstructure as well as external fields (see Section 5) may easily "overrule" the conventional statement "ceramics are brittle."

4 | DISLOCATION MECHANICS IN CERAMICS: AN OVERARCHING FRAMEWORK

Following the above *extrinsic* deformation boundary conditions, here we focus more on the *intrinsic* dislocation mechanisms for promoting dislocation plasticity. With the competition between plasticity and fracture outlined in ceramic materials, we aim to promote dislocation plasticity while suppressing crack formation (crack initiation and propagation[40]). To facilitate the generation of increased plastic zone size and tunable high-density dislocations, it is helpful to examine the three key factors for dislocation plasticity, namely, dislocation nucleation, multiplication, and motion, which was investigated and discussed by Gilman and Johnston on LiF,[10] although their focus back then was not the same as now. The current author briefly discussed these factors while attempting to bridge the gap between bulk compression and nano-/microscale indentation tests on single-crystal SrTiO$_3$ at room temperature.[42] Needless to say, the correlation and competition between these three factors are equally important.

If a sample is capable of plastic deformation, according to Orowan's equation, there is $\dot{\varepsilon} = bv\rho_{\text{mobile}}$. Note it is written in rate form, with $\dot{\varepsilon}$ the plastic strain rate, b Burgers vector, and ρ_{mobile} mobile dislocation density. This governing equation suggests that mobile dislocations with a high density are desirable for plastic deformation. For room-temperature plastic deformation of ceramics, par-

ticularly for bulk deformation, the starting point shall be selecting the right material that has room-temperature mobile dislocations, which is the case for the model system SrTiO$_3$. In what follows, the author presents examples of dislocation nucleation and multiplication in this material.

4.1 | Dislocation nucleation

Dislocations are very often considered rare in polycrystalline ceramics due to the conventional high sintering temperature that may well anneal them out.[27] The growth of single crystals usually retains grown-in dislocations with a density of ~10^9 to ~10^{10}/m^2.[22] In other words, a dislocation can be barely found within a cross-section area of hundreds of μm^2 in ceramic materials. In contrast, metals, as fabricated, usually have a dislocation density of several orders of magnitude higher than in ceramics.

For dislocation-scarce ceramic materials, dislocation nucleation becomes the first bottleneck for plasticity. Dislocation nucleation is often categorized into homogeneous nucleation and heterogeneous nucleation.[14] Homogeneous dislocation nucleation in perfect crystal lattices requires a maximum shear stress approaching the theoretical shear stress (i.e., ~G/10 or ~G/2π, G being the shear modulus) in metals[127,128] and ceramics.[41,129] This mainly occurs in a locally dislocation-free region. An example of single-crystal SrTiO$_3$ using nanoindentation is showcased in Figure 7. The low dislocation density (Figure 7A) allows placing the sharp Berkovich nanoindenter tip (with an effective tip radius of ~90 nm) in regions that are dislocation-free (Figure 7A). The pop-in events (Figure 7B) occur at a displacement of ~15 nm, indicating dislocation nucleation from dislocation-free regions and the transition from Hertzian elastic contact to elastic-plastic deformation. The newly generated dislocations are revealed by the etch pits (dark dots in the inset figure of Figure 7A). Note that the majority of the maximum shear stress (derived from the Hertzian contact theory[127]) corresponding to the onset of the first pop-in events lies in the range of 14–18 GPa (Figure 7C), although the corresponding load was extremely small (about 0.1 mN; Figure 7B). The scattered maximum shear stress distribution (Figure 7C) indicates there shall be defects (point defects such as oxygen vacancies or surface terraces from both the sample and the indenter) involved during the pop-in. Therefore, strictly speaking, it may not fully conform with the definition of a perfect crystal lattice for homogeneous dislocation nucleation.[130] More examples with quantified maximum shear stress approaching G/2π for dislocation nucleation in other ceramics like TiO$_2$,[131] ZnO,[45] and ZnS[38] are most recently reported.
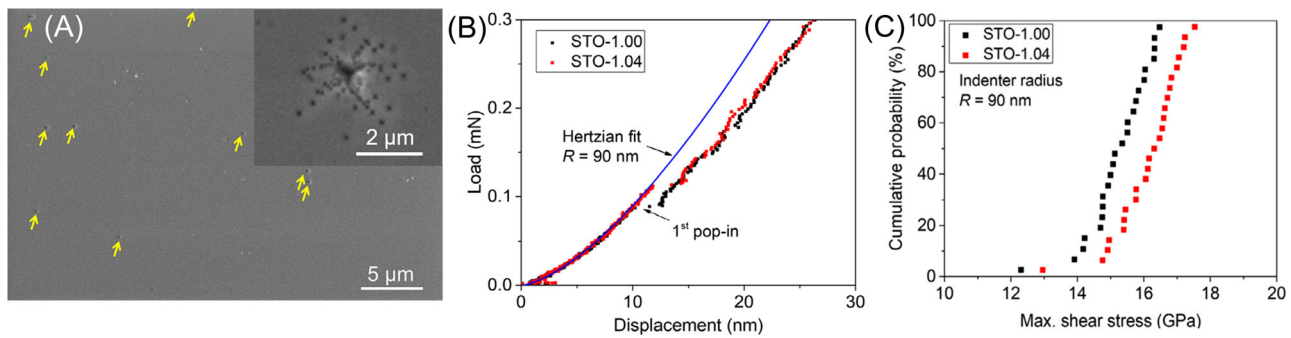
FIGURE 7 Dislocation nucleation from dislocation-free regions in single-crystal $SrTiO_3$: (A) chemical etching reveals the dislocation etch pits before (yellow arrows) and after (insert, dark dots) nanoindentation test; (B) nanoindentation load-displacement curves exhibiting pop-in events, which correspond to dislocation nucleation; (C) statistical distribution of the calculated maximum shear stress for the 1st pop-in events, and each data point is obtained from one nanoindentation test. STO stands for single-crystal $SrTiO_3$, and 1.00 and 1.04 are the Sr/Ti ratios of the starting powder for growing such single crystals. Images reproduced and modified with permission from Ref. [43].

Many studies on dislocation nucleation exist in metals using the nanoindentation pop-in method.[132] Worth mentioning is, the knowledge and skillset transfer from metals to ceramics may not be as straightforward as it seems. In ceramic materials, the pop-in events can also be caused by cracking (see discussion in Section 3.2.4) or phase transition. Therefore, a careful postmortem examination of the deformation volume is necessary to rule out such factors.

Contrasting homogeneous dislocation nucleation, heterogeneous dislocation nucleation[133] from preexisting defects requires lower stress. Sources for heterogeneous dislocation nucleation can be abundant:[7] surface terraces, vacancy clusters, voids, grain boundaries, precipices and inclusions, and grown-in dislocations, to name a few. Yet not all of them are equally effective to promote sufficient dislocation nucleation, especially in ceramic materials at room temperature. Some of them (for instance, voids and grain boundaries) also serve as sites that induce stress concentration for nucleating and propagating cracks. Some of them (e.g., higher oxygen vacancy concentration[35]) do not drastically decrease the stress level for heterogeneous nucleation to a practical level for bulk deformation. A logical reasoning would be to increase the *effective dislocation source* (preexisting dislocations) as well as its density during materials processing to facilitate dislocation multiplication.

To fully make use of the multiplication mechanisms to enhance dislocation densities at room temperature, the author and coworkers have employed the cyclic Brinell indentation[67] and the cyclic scratching method.[44] The essence of cyclic deformation is that the dislocations induced from the previous deformation cycle serve as effective sources to further multiply in the upcoming cycle. For instance, with cyclic Brinell indentation up to 10 cycles, the dislocation density in $SrTiO_3$ was increased by 3 orders of magnitude from $\sim 10^{10}$ up to $\sim 10^{13}/m^2$.[67] By using scratching, the density was further increased up to $\sim 10^{14}/m^2$.[44] These methods are simple but powerful and have been applied to various materials at room temperature[44] without inducing visible cracks. These methods are capable of increasing the plastic zone size up to millimeters/centimeters in length/width and hundreds of micrometers in depth.

There are, however, many remaining open questions, such as whether cyclic testing (in the sense of fatigue) would induce dislocation-based fatigue in those plastically deformable ceramics, or if persistent slip bands would form as in the case of metals, or how cracks would initiate and propagate upon high cycles. Studies dependent on frequency, temperature, stress, lubricant, microstructure, and so on are currently being carried out in the author's group.

4.2 | Dislocation multiplication

The most effective way to increase the mobile dislocation density is by dislocation multiplication, such as Frank-Read sources (in-plane multiplication) and cross-slip (out-of-plane multiplication). A classic example of LiF was demonstrated by Johnston and Gilman,[8] which is adopted in dislocation textbooks.[130]

4.3 | Correlation and competition

Strictly speaking, the dislocation nucleation during mechanical deformation is simultaneously accompanied by dislocation motion and often also dislocation multiplication. In real practice, it is rare that these three factors are completely isolated, except when only a few dislocations were focused on for their motion, multiplication, or

interaction with other defects, as in the case of in situ TEM studies.[134].

Although a collaborative and interactive operation of all three factors is needed for the plastic deformation of ceramic materials, each factor may play a dominant role at different deformation stages. Take bulk deformation of single-crystal SrTiO$_3$ as an example, at the onset of yield, dislocation multiplication and motion are most likely the responsible mechanisms. The yield strength during bulk compression is ~100 MPa along the <001> direction, far below the (homogeneous) nucleation stress level of 14–18 GPa. Same for single-crystal ZnS,[85] which exhibits a bulk compression yield strength of ~30 MPa (in darkness), which is much lower than the nucleation stress level of an average of 4 GPa.[38] This strongly indicates that the lattice friction stress for dislocation motion, as well as multiplication, is very low, almost comparable to some FCC metals. It was suggested that engineering dislocation sources to circumvent dislocation nucleation may be a feasible way to promote dislocation plasticity in ceramics.[34] An example of such is, as discussed before, flash-sintered TiO$_2$ that contains a high density of dislocations and stacking faults and can exhibit plastic strain up to ~12% during room-temperature pillar compression,[65] while conventionally sintered, dislocation-free TiO$_2$ samples have a fracture strain of ~2%. In this sense, the previously presented cyclic Brinell indentation and cyclic scratching methods show consistent results: during the first cycle of indentation/scratching, fresh dislocations with a medium density are produced, which serve as effective sources for upcoming deformation cycles. Ultimately, the dislocation densities are increased by 4–5 orders of magnitude compared with those in the undeformed samples.

The competition between the dislocation mechanisms sets in with the increase of dislocation density and accumulated plastic strain inside the deformed volume. A fraction of the dislocations become less mobile or even immobile due to dislocation interactions, as indicated by the work hardening in the stress-strain curves during bulk compression of single-crystal SrTiO$_3$.[93] Moreover, the activated different slip planes intersect each other, dislocations start to pile up at such intersecting points and eventually would lead to crack nucleation, as observed and discussed in single-crystal MgO.[135,136] Another direct example of dislocation pileup at larger plastic strain is demonstrated in SrTiO$_3$ using the nanoindentation test.[40] We demonstrated that edge dislocations pileup induced at the sample/indenter interface leads to crack initiation, which is discussed in the framework of Zener-Stroh crack formation.[137]

To summarize, examining dislocation nucleation, multiplication, and motion individually is helpful to understand the basic factors for the mechanical engineering of dislocations. In particular, for room-temperature dislocation engineering, it is suggested to avoid homogeneous dislocation nucleation,[22] choose materials that exhibit good room-temperature dislocation mobility (low lattice friction stress, as in the case of aforementioned SrTiO$_3$, MgO, and ZnS), and use cyclic deformation to promote dislocation multiplication. A practical case study is presented in Panel A. Ultimately, the competition between these factors needs to be taken into consideration by avoiding dislocation pileup (which happens more readily at high dislocation density and lower dislocation mobility) that may cause crack formation.

5 | INFLUENCE OF EXTERNAL FIELDS ON DISLOCATION-GOVERNED MECHANICS IN CERAMICS

5.1 | Charged dislocation cores in ceramics

Unlike in metals, dislocations with charged cores in ionic crystals feature both elastic and electrostatic characteristics,[138,139] which are subject to modifications in physical fields such as light irradiation and electric field. This offers new opportunities to tune the dislocation-mediated plastic deformation[38,85,139] and fracture toughness.[36] Dislocations with charged features were elucidated in the seminal paper by Eshelby et al.[138] in 1958. In ionic crystals, excess ions with one sign (+/−) could arrange along the dislocation cores.[138] In thermal equilibrium, the charged cores are surrounded by charge-compensating defects (forming the Debye-Hückel cloud with a radius of λ_D; Figure 1B).[140] In the case of charged jogs, provided that the spacing between jogs along the dislocation is small compared with λ_D, there is $\lambda_D^2 = \varepsilon kT / 8ne^2\pi\alpha$,[14] where ε is the dielectric constant, n is the number of available sites for either cations or anions, α is a constant related to the free energy of a cation and anion vacancy, the rest of the symbols have the normal meaning.

To date, the majority of studies on charged dislocations focused on alkali halides (rock salt structure),[139] which are difficult to be adopted for modern electrical and optical applications due to their large band gap. In contrast, the new horizon of dislocation-based functionality finds its playground mainly in functional oxides and semiconductors. Lately, the charged cores in oxides have been simulated[140,141] and quantified by using aberration-corrected scanning transmission electron microscopy in various functional ceramics such as SrTiO$_3$[73] (Figure 8) and Al$_2$O$_3$.[142]

The electrostatic interaction due to charged features of dislocations has been found to offer room for tuning plasticity under specific conditions such as external physical
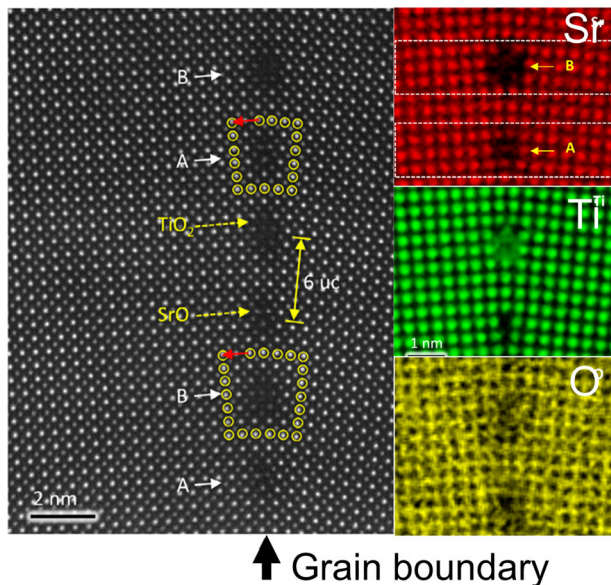
FIGURE 8 Atomic resolution TEM images of two types of dislocation cores in bi-crystal SrTiO$_3$ with a 10° tilt grain boundary, with inserts showing energy dispersive X-ray spectra (EDS) of the elemental distribution (Sr, Ti, and O). Images were reprinted and adapted with permission from Ref. [73].

fields. In what follows, examples of electroplasticity (with electric field) and photoplasticity (with light illumination) are presented.

5.2 | Chemoplasticity

This section elucidates the modification of defect chemistry for tuning the plasticity of ceramics, or chemoplasticity, which will be critical in correlation to the impact of external fields such as electric field and light illumination, detailed in the following sections. The theory of charged dislocations (cores) implies that the Debye-Hückel radius λ_D can be modified by the valence and concentration of the charged point defects, regulating the elastic and electrostatic forces around the dislocations to promote (heterogeneous) dislocation nucleation, multiplication, and motion. To this end, defect chemistry[60,143,144] engineering (e.g., reduction treatment, stoichiometry change, and doping) on plasticity is discussed here. Evidence of such is rich in alkali halides.[139] An interesting example is the temperature-dependent fluctuation (double maxima and double minima) of the yield stress (σ_Y) of sodium chloride doped with divalent impurities presented by Eshelby et al.[138] They proposed the mechanisms to be the temperature-dependent charge characters of the dislocations (concerning the mobility of the charged impurities): At lower temperatures, the thermal equilibrium is usually not established as the mobile dislocations interact with the charged point defects, resulting in a higher σ_Y. With the temperature increasing to a critical value, that is,

the isoelectric temperature (T_{iso}), the dislocations become uncharged and σ_Y drops to a minimum. Further increase in temperature reverses the charge[138] and increases σ_Y again. At sufficiently high temperatures, the thermal activation breaks out the charge cloud drag of the dislocations leading to a drop of σ_Y again, and no qualitative modification on charged dislocations would occur anymore, as suggested by Koehler et al.[145] The isoelectric point is estimated by $T_{iso} = -F_a/k \ln v_a c_i$,[14] with v_a the volume per ion pair, F_a the free energy of an anion vacancy, and c_i the charged impurity concentration. T_{iso} is closely related to the defect chemistry of the target materials.

Evidence of defect chemistry engineering in functional ceramics is rare but traceable. Ashbee et al.[146] deformed nonstoichiometric rutile TiO$_2$ as a function of temperature and showed an analogous variation of σ_Y (double maxima and double minima, resembling those in sodium chloride). No such effect was found in the stoichiometric samples. Nakamura et al.[66] modified the Sr/Ti ratio in SrTiO$_3$ and observed at room temperature a ~15% lower σ_Y and ~60% higher fracture strain in Sr/Ti = 1.04 samples in comparison with Sr/Ti = 1.00 samples. Hirel et al.[140] simulated dislocation climb at high temperatures by investigating the interaction of vacancies with a charged edge dislocation core using atomic-scale simulation in perovskite MgSiO$_3$ and concluded that dislocations favor a nonstoichiometric, oxygen-deficient configuration associated with a positive charge, with electrostatic force dominating the dislocation-vacancy interaction. Such chemoplastic effect with respect to charged dislocations deserves much more attention.

Following the aforementioned work by Nakamura et al.[66] and by using nanoindentation pop-in tests, we recently demonstrated that dislocation nucleation (under sharp indenter to suppress crack formation) is facilitated in samples rich with point defects (O$_O$ + Sr$_{Sr}$ ↔ V$''_{Sr}$ + V$^{\cdot\cdot}_O$ + SrO), while the dislocation motion (under larger indenter and bulk) is impeded owing to the 'solute drag' by the point defects.[43] The indentation study underpinned the mechanisms in the bulk deformation by Nakamura et al.[66] The same phenomena are also observed on reduced SrTiO$_3$,[35] which exhibits easier dislocation nucleation due to the increased oxygen vacancy concentration but lower dislocation motion is observed most likely due to the drag effect caused by the charged oxygen vacancies interacting with the dislocations. These experimental observations were supported by recent molecular dynamics (MD) simulations by Li et al.[147]

5.3 | Electroplasticity

The earlier experiments demonstrated that moving dislocations that carry charges would produce an electrostatic field.[139] The reciprocal effect, namely, an

external electric field enhancing the dislocation mobility (electroplasticity[148]), also exists in ceramic materials (e.g., alkali halides[149]). Conrad et al.[148] systematically investigated bulk compression tests of ceramic oxides with an applied DC electric field and reported a significant reduction of the flow stress of fine-grained Al_2O_3 and ZrO_2 at high temperature ($T > 0.5$ T_m) with a field <1 kV/cm. In addition to the possible Joule heating effect, which may greatly facilitate the dislocation mobility, two mechanisms were proposed[148] for the enhancement of dislocation mobility under the electric field based on the studies of halide crystals: (i) the electric field exerts a driving force (electron wind force) on the charged dislocations; (ii) the electric field reorients the charged impurity-vacancy dipoles and lowers the barriers for dislocation motion. However, these mechanisms on dislocation motion find difficulty being directly transferred to fine-grained oxides deformed at high temperatures as in Ref. [148] because the sliding of charged GBs and interaction of GBs with charged point defects complicate the scenario. Such complexities could be minimized using model single crystals to discern the effect of an electric field on charged dislocations (cores), as elegantly demonstrated most recently by Li et al.[150] on single-crystal ZnS using in situ TEM coupled with an electric field (by contacting a tungsten tip with the sample), which not only provides direct evidence for modulating dislocation motion with an electric field, but also brings new perspectives to revisit and understand electroplasticity.[151]

5.4 | Photoplasticity

The almost-forgotten research in the 1960s by Osipťyan et al. on CdS[152] and in the 1970s on ZnO by Carlsson et al.[153] showed that light illumination increased yield strength by decreasing dislocation mobility. Such an effect was coined as photoplasticity[154] and the impact of light is most significant near the absorption edge (bandgap).

Recently, Oshima et al.[85] demonstrated a striking example of photoplasticity on bulk single-crystal sphalerite ZnS (II–VI group semiconductor): an ultimate plastic strain of ~45% was observed in complete darkness contrasting the almost immediate fracture (~2% strain) under UV light (365 nm, near the bandgap). The decreased plasticity of ZnS in UV light was attributed to the suppressed dislocation mobility caused by the reconstruction of charged dislocation cores due to the electron/hole pairs excited by light. This is further supported by density functional theory (DFT) simulation by Matsunaga et al.[155] Most recently, Nakamura et al.[38] further advanced the understanding of dislocation nucleation in photo-

plasticity using nanoindentation coupled with accurate control of light illumination (*photoindentation*[38,45]), where the light illumination is found to mildly affect dislocation nucleation but dramatically changed the dislocation mobility at the nanoscale. Since the dislocation mobility and dislocation nucleation are more favorable in darkness, more ductile deformation of ZnS is observed in darkness. We further found the fracture toughness of single-crystal ZnS increased from 0.18 MPa m$^{1/2}$ under UV light (365 nm) to 0.26 MPa m$^{1/2}$ in complete darkness.[36] The increase in the absolute values of fracture toughness is marginal, but the underlying physics and mechanics (crack tip-dislocation interaction under light illumination and in complete darkness) may be worth further investigation.

In summary, the study of mechanics-based engineering of dislocations in functional ceramics assisted by different external fields is craving for growth and more research efforts. In single crystals, dislocation nucleation, multiplication, and motion concerning charged point defects need to be addressed to better understand dislocation plasticity. In polycrystals, it is most critical to understand the slip transfer (charged dislocation–GB interaction) and identify transferrable GBs. Although the number of slip systems is limited in ceramics at room temperature, the dislocation density (by easier nucleation and multiplication) and mobility (by an electric field and defect chemistry engineering) can be tuned in ceramics. Coupled with the concept of surface deformation and texturing of ceramics to circumvent von Mises and Taylor criterion, we may ultimately achieve room-temperature plastically deformable polycrystalline ceramics mediated by dislocations.

6 | OPEN QUESTIONS AND CHALLENGES

Accompanied by the progress being made in the mechanical tailoring of dislocations in ceramics, numerous open questions and challenges emerge. In resonance with the previous sections, some challenges are highlighted and discussed here.

6.1 | Integration of mechanical and functional properties

Dislocation-based simultaneous tuning of the mechanical and functional properties is desirable for application purposes. This has been demonstrated feasible in $SrTiO_3$, in which higher dislocation density leads to higher photoconductivity[13] and higher fracture

toughness.[24] Hence, it may be practical to ask, what is the upper limit of dislocation density that can be introduced for the selected ceramic materials? On the other hand, extremely high-density dislocations can potentially lead to severe work hardening, dislocation pileup, hence crack formation. Is there a limit also on the plastic zone size that can sustain such a high dislocation density (e.g., $\sim 10^{15}/m^2$) without forming cracks?

Considering such a twofold impact of dislocations, to achieve the integration of both mechanical and functional properties, a balance in the dislocation density needs to be sought. Is it possible to construct a guiding map to correlate dislocation density with functionality (e.g., electrical conductivity, thermal conductivity, and photoconductivity) as well as mechanical properties (plastic strain, fracture toughness, and damage tolerance)? However, this question may need to be taken more critically, as solely dislocation density may not be sufficient. Dislocation type, spatial arrangement, and so on may well increase the complexity.

6.2 | Dislocation imprinting

Bearing in mind the boundary conditions for mechanical deformation of ceramics, it seems no longer a big hurdle to engineer dislocations in some ceramics at room temperature (see Panel A for $SrTiO_3$). However, the true challenge lies not only in increasing the density and plastic zone size but also in the precise control of dislocation types (edge, screw, mixed), dislocation mesostructure (kinks, jogs, dipoles, loops),[58] and dislocation spatial arrangement. Challenges remain to precisely imprint dislocations, which may hold the potential to pinpoint the fundamental mechanisms for electrical conductivity, which is still under debate when it comes to the exact role of dislocations. In the sense of electrical conductivity, it is unclear yet whether or not mechanical deformation would be the best approach to deconvolute the mechanisms. On the other side, for functional properties such as thermal conductivity and surface reactivity, and mechanical properties such as damage tolerance, the "messy" dislocation structures engineered through mechanical deformation turn out to be beneficial.

For tuning functional and mechanical anisotropy, previous attempts such as using notched bulk samples to induce local stress concentration and to activate particular slip planes in $SrTiO_3$,[58] as well as using different compression directions on $BaTiO_3$[20,64] (though at high temperature only) based on Schmid factor calculation have been made. Other possibilities remain to be explored.

6.3 | Dislocation mechanics

The previously discussed dislocation nucleation, multiplication, and motion are, at the engineering level, probably the most straightforward approach to follow. However, the deformation mechanisms down to the fundamental level at the dislocation/atomic scale are much more complicated. Aside from the kinks, jogs, and dipole formation, other factors such as dislocation dissociation,[87] twinning,[17] and stacking faults[156] need further examination. For instance, it was reported that a faulted dipole structure could exist in $SrTiO_3$ after room-temperature grinding.[57] Further systematic examination of the dislocation types (via Burgers vector and dislocation line vector analysis) is required, as different dislocation types are found to be affecting the dislocation motion and in particular multiplication mechanisms significantly.

Once dislocations are successfully engineered into ceramics, their interactions with various other types of defects will open many more windows of questions. For instance, point defects, such as oxygen vacancies, in ceramic oxides can carry charges. Considering the charged dislocation cores, how would the charged point defects interact with the charged dislocation cores, thus potentially impact the mechanical and functional stability over a long-time span? In particular, how would the external stimuli affect such interactions? Defect chemistry engineering for dislocation plasticity requires interdisciplinary knowledge and skillsets across chemistry, materials science, mechanics, and physics.

Coming to dislocation–GB interaction, as GBs are well known for hindering dislocation motion, a detailed study on dislocation slip transfer (dislocation–GB interaction) is helpful. Regardless of the abundant literature on slip transfer in metals,[157] so far a thorough and quantitative understanding of slip transfer has been lacking,[158] which is even more scarce in ceramics at room temperature. The insufficient independent slip systems in ceramics below the BDTT do not favor slip transfer at most GBs, consequently, dislocation pileup and cracking at such GBs are induced.[97,159] On the other hand, there are specific "soft," penetrable GBs (e.g. low-angle GB as shown by Kondo et al.[70]) allowing for slip transfer, so that appreciable plastic strain is achievable. As the established knowledge rarely considered the charge effect of dislocations, it is thus pertinent to ask, by considering the interaction between charged GBs[160] and dislocations, in comparison with neutral ones, whether the energy landscape will be changed to facilitate or impede slip transfer. Could we identify more "soft" GBs that favor slip transfer? Using the crystallographic tailoring for textured ceramics,[161] such "soft" and transferrable GBs can be textured through templated grain

growth[161] for polycrystalline ceramics. In this sense, the polycrystalline samples shall be deformable along certain loading directions to allow slip transmission to achieve appreciable plasticity.

An alternative approach to circumvent von Mises and Taylor criterion is by deforming polycrystalline samples in the surface regions. This may eventually be feasible thanks to the relaxation at free surfaces, yet systematic analysis is still lacking. On the other hand, high-temperature extrusion was adopted by Skrotzki et al.[162] to induce elongated grains in polycrystalline KCl for deformation anisotropy, but no consideration of slip transfer was made back then.

6.4 | External fields

As initial dislocation density in ceramics is normally low and dislocation nucleation is difficult, it is logical to assume that the starting dislocation density may be a limiting factor to exploit the potential of external fields to harvest dislocation plasticity. So far such fields have been found to most dramatically impact the dislocation motion, except for a couple of recent works using photoindentation pop-in tests[38,45,125] to look into the dislocation nucleation under light illumination. Considering that external fields need dislocations as active agents to exhibit their impact on plasticity, it is plausible to argue that: with a much higher dislocation density, the impact of external fields on the sample's plasticity shall be more dramatic. With the new playground developed for dislocation engineering in $SrTiO_3$, namely with a large plastic zone and high dislocation density (see Panel A), we are currently investigating the influence of such preengineered dislocations with different densities and their impact on the photoplasticity and electroplasticity. In addition, the dislocation type and core structure in concert with the external field remains challenging, which requires not only advanced TEM characterization but also novel simulation toolboxes, which will be briefly discussed at the end of this section.

6.5 | Operational stability

When it comes to prospective applications, the most critical issue is operational stability. This concerns both functional stability and mechanical reliability. Considering possible operational scenarios, the thermal, mechanical, electric, optical, and magnetic fields are highly relevant for advanced ceramics. The alternating loading with different frequencies of such fields can also be critical (e.g., in the case of cyclic mechanical deformation, dislocation structure evolves drastically). A practical question would be, is there a threshold value for the external fields

(such as threshold temperature, light or electric field intensity), under which the dislocation structure as well as the dislocation-tuned functional and mechanical properties would remain stable up to a long-operational duration (e.g., 1000 h or longer with statistics, which will be challenging in most laboratories)? So far, the coupling effect among some of these fields on the dislocation mechanics has been attempted, yet the almost infinite choice of the combination of different fields as well as deformation conditions makes it not feasible to perform tests only experimentally. Analogous to other new research trajectories, many new experimental/simulation protocols for this field are foreseen to be developed.

6.6 | Room-temperature ductile ceramics

It may be an ultimate but also naive question for the field of dislocations in ceramics: what are the fundamental mechanisms for controlling bulk plasticity in single-crystal ceramics under ambient conditions? Why are some ceramics (mostly discussed in previous sections) plastically deformable at room temperature and ambient pressure in bulk shape while most other ceramics are not, regardless of their same crystal structure and almost identical lattice parameters? When it comes to discovering more room-temperature ductile ceramics for macroscale deformation, no clear route yet exists. So far, many discoveries of such materials have been rather empirical or by accident. Considering the rapid rise of machine learning (ML) in materials science, resorting to ML someday for answering this question may not be that far away. Before that, it is still critical to decide which physical parameters and what (numerous and sensible) experimental data should be used as input for the learning model.

Although the above open questions discussed in Sections 6.1–6 are separated for simplicity, these aspects and influencing factors may all come together at one point for the true realization of dislocation-based applications.

6.7 | Simulation

Last but not least, simulation has been barely touched so far till this section. However, compared with the vast amount of experimental data on dislocations in ceramics exploring both mechanical and functional properties, it may be seen that simulation works in this field are much fewer. An increasing number of simulation studies on dislocation mechanics are gradually coming into sight. MD simulations in $SrTiO_3$ on dislocation-point defect interaction,[147] dislocation nucleation,[163] mobility,[164] and comprehensive analysis by Klomp et al.[165] on dislocation dissociation,

motion, and multiplication are available. Another intensively modeled system is MgO.[166–168] DFT simulation on dislocation core structure reconstruction was most recently conducted on ZnS to reveal the light illumination effect on dislocation mobility.[155] More recent DFT efforts in the direction to address photoplasticity in various semiconductors are being made.[169–171] In addition, several simulation works on dislocation-tuned functionality can be found for thermal conductivity[172] and electrical conductivity.[59]

Challenges remain such as determining the (1) accurate potential for each material; (2) charge feature for different types of dislocations (charged edge type and neutral screw type[165]; (3) nonstoichiometry at dislocation cores, for which even detailed experimental observations and quantifications are still lacking except few examples such as $SrTiO_3$ (Figure 8) and Al_2O_3.[142] Except for point (1) on obtaining the accurate potential, which is a general challenge also for simulation in metallic materials, points (2)–(3) are more relevant for ceramics with ionic/covalent bonding. In the best scenario, simulation shall provide predictions for dislocation engineering, dislocation–GB interaction, and dislocation-crack tip interaction, which further requires simulation across the length scales. This, however, may not be so straightforward within a short time. Nevertheless, joining hands between simulation and experimental evaluation to provide an overarching framework shall bring in many more new insights for dislocation engineering in ceramics.

7 | CONCLUSIONS

The potential of dislocations may have been underappreciated in advanced ceramics, which are predominantly engineered by point defects or interfaces.[173] The field of dislocations in ceramics is now embracing a wide range of exciting opportunities, with the dislocation-tuned functional and mechanical properties bringing a new perspective of (re)thinking ceramics engineering. Emerging concepts in concert with experimental proofs are gradually bringing this field to the spotlight. However, pressing challenges stand before the new horizon can be truly revealed for dislocation technology in ceramics.[52]

Despite abundant approaches being readily available to evaluate the dislocation-tuned functionality, grand challenges remain to tame dislocations in ceramics without crack formation through mechanical deformation. The strong atomic bonds, limited available slip systems, and thermochemical stability make the majority of ceramics brittle and exhibit little or no dislocation activity except at very high temperatures.[31,32,87] Hence, this new field urgently calls for novel approaches coupling external physical stimuli to introduce and control dislocations (density, type, spatial arrangement) with large plastic zones in ceramics without crack formation. The available model material system, in particular $SrTiO_3$, suggests that generating and stabilizing dislocations in large plastic volumes with high density up to $\sim 10^{15}/m^2$ is readily achievable even at room temperature, which allows for macroscale preparation for assessment of both functional and mechanical properties. Yet the transfer to a much broader range of functional ceramic materials at technologically relevant temperatures (e.g., below 800°C) is pending. Considering the multifaceted and interdisciplinary nature of the mechanical tailoring of dislocations and the accompanying functionalities, joint forces from different fields are highly desired to boost the research on _dislocations in ceramics_.

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PANEL A: ROOM-TEMPERATURE TOOLBOX FOR MECHANICAL ENGINEERING OF DISLOCATIONS IN SRTIO₃

Following the practical guideline (Figure A1), here the author demonstrates a case study on room-temperature dislocation engineering in single-crystal $SrTiO_3$, a prototypical perovskite with a cubic structure at room temperature. In 2001, Brunner et al.[93] found that single-crystal $SrTiO_3$ can yield plastic deformation up to ~7% in uniaxial bulk compression at room temperature. Later, a few studies on plastic deformation of this material were carried out over a wide temperature range, from 77K (liquid nitrogen) up to 1800K from bulk deformation[100,174] to Vickers indentation[175] further down to nanoindentation testing.[163] Considering the desirable high dislocation density and continuous large plastic zone, bulk deformation is not efficient as the highest dislocation density is ~10^{13}/m^2 inside the slip bands, which are very often discrete and confined in localized regions, with a large volume of the deformed sample remaining dislocation free. On the other hand, nanoindentation yields too small a plastic zone for use. To overcome these drawbacks, the author and coworkers adopted the cyclic Brinell ball indentation[67] and cyclic scratching method[44] at room temperature, and successfully pushed the boundaries of the dislocation density up to ~10^{14}/m^2 with the plastic zone up to millimeter/centimeter in length/width and hundreds of micrometers in depth, without inducing visible cracks. An overview of the various deformation methods is presented in Figure A2, and the corresponding dislocation density and plastic zone size are outlined in Figure A3.
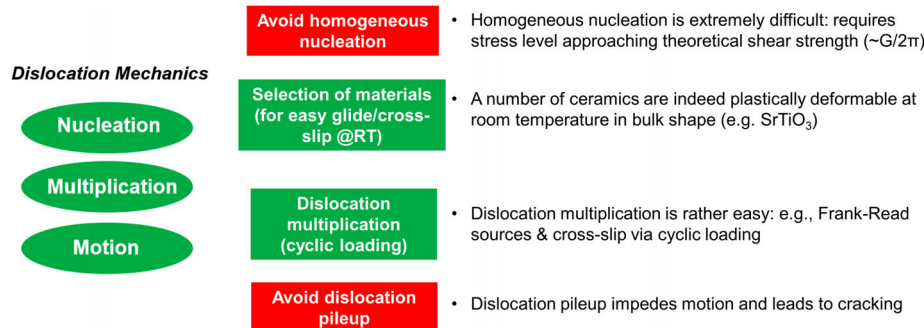
FIGURE A1   A practical guideline for room-temperature (RT) mechanical tailoring of dislocations in ceramics.
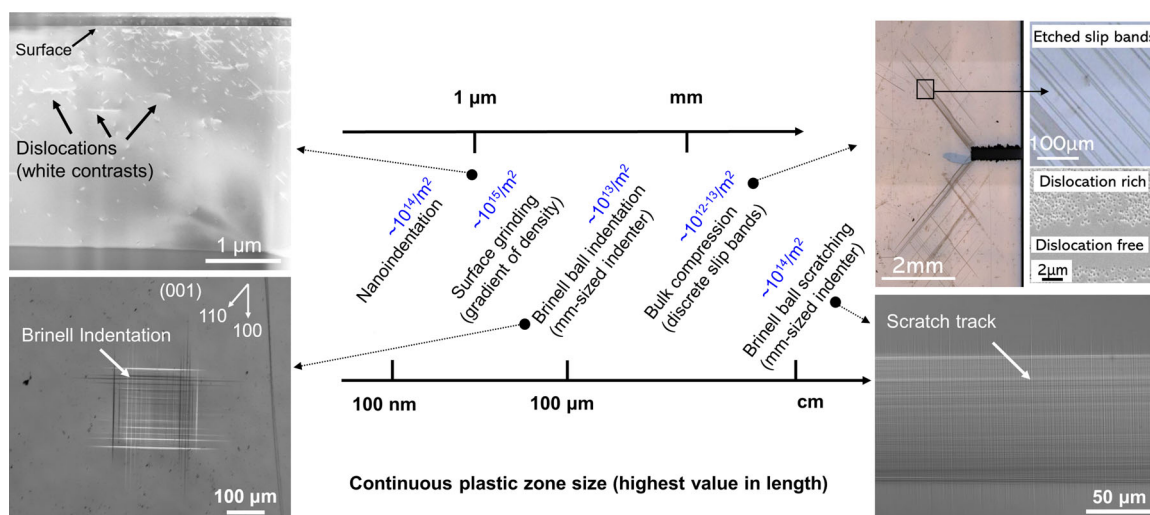

FIGURE A2   Summary of currently available methods to mechanically engineer dislocations into single-crystal SrTiO$_3$ at room temperature. Image reprinted and modified with permission from Refs. [13] and [15] and Ref. [58]. TEM image for dislocations induced by surface grinding is provided by W. Lu.
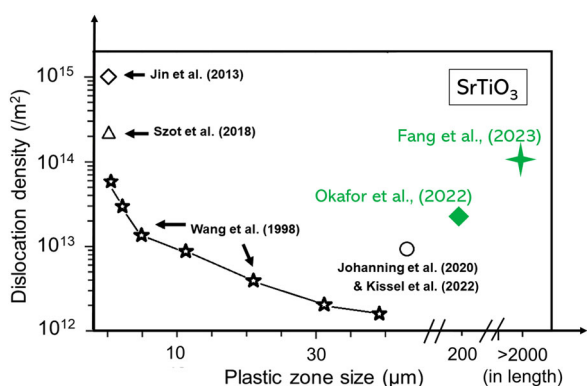

FIGURE A3   Overview of room-temperature dislocation engineering in SrTiO$_3$, displaying the capability of achieving dislocation densities from ~10^9/m^2 (in undeformed reference samples) up to ~10^{15}/m^2, with a continuous plastic zone size from hundreds of nanometers up to mm- or cm-sized regions (in length) without crack formation. The data are extracted from references listed in the figure: Jin et al.[57]; Szot et al.[26]; Wang et al.[176]; Johanning et al.[27]; Kissel et al.[13]; Okafor et al.[67]; Fang et al.[44] The author's works are highlighted in green.