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Published in:
APL Materials

Link to article, DOI:
[10.1063/5.0176307](https://doi.org/10.1063/5.0176307)

Publication date:
2024

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Pryds, N., Park, D.-S., Jespersen, T. S., & Yun, S. (2024). Twisted oxide membranes: A perspective. *APL Materials*, 12(1), Article 010901. <https://doi.org/10.1063/5.0176307>

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Twisted oxide membranes: A perspective

Cite as: APL Mater. 12, 010901 (2024); doi: 10.1063/5.0176307
Submitted: 12 September 2023 • Accepted: 8 December 2023 •
Published Online: 2 January 2024



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ABSTRACT

Freestanding oxide membranes have recently emerged as a promising platform, offering new opportunities for experimentalists to design materials with novel properties. These membranes, typically consisting of transition metal oxides, can be synthesized as freestanding ultra-thin quasi-2D layers and, subsequently, reassembled into multilayer stacks with controlled relative twist angles. In these heterostructures, controlling the moiré superlattices pattern of the two lattices can play a crucial role in creating unique and new electronic, optical, magnetic, and mechanical properties that do not exist in the individual constituent materials or in traditional non-twisted epitaxial heterostructures. Here, we explore the role of stacking and twisting of freestanding oxide membranes, including the underlying principles and potential applications. We also discuss the current challenges and provide a perspective on some of the possible future directions of the field. Our aim is to highlight the opportunities for creating new functional materials by stacking and twisting freestanding oxide membranes.

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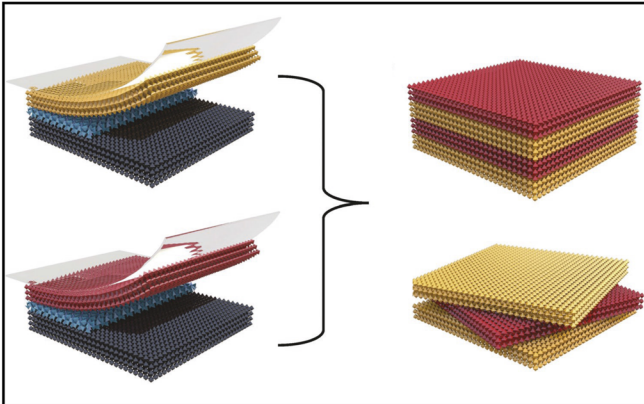
INTRODUCTION

Growing dissimilar materials into epitaxial heterostructures is a cornerstone of modern materials science and technology. The developments in the field of 2D van der Waals (vdWs) materials expanded the concept to mechanically assembled heterostructures formed by layered materials, such as graphene and transition-metal dichalcogenide.^{1–3} Complex metal oxides offer a wide range of functionalities not accessible in 2D materials. These typically arise due to strong correlations between the electrons in the *d* orbitals, which give rise to a complex interplay between the charge, spin, orbital, and lattice degrees of freedom,⁴ generating a rich spectrum of competing phases and physical responses, including high temperature superconductivity,⁵ metal-insulator transitions,⁶ colossal magnetoresistance,⁷ ferromagnetism,⁸ ferroelectricity,⁹ piezoelectricity,¹⁰ and multiferroic¹¹ properties. Traditionally, complex oxides are grown epitaxially with a well-regulated growth scheme and atomic-level control over the material interfaces and substrate surfaces.¹² However, these methods have fundamental limitations that restrict their manipulation, integration, and applications. For example: (i) Traditional epitaxy is only applicable within limit sets of compounds having similar lattice parameters; and (ii) epitaxial films are clamped by the rigid substrate, which can dampen important properties.

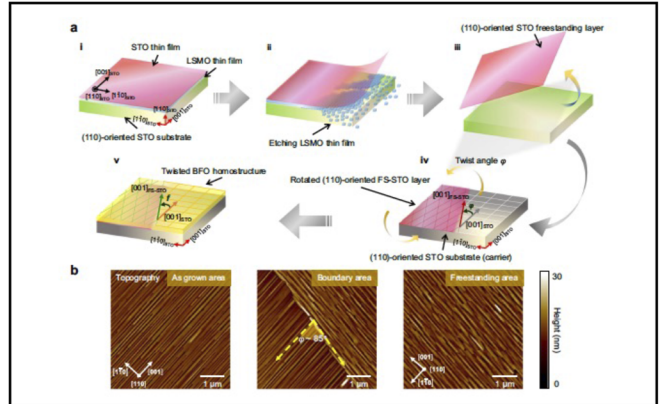
Furthermore, the high temperature growth of epitaxial films both (iii) facilitates atomic intermixing, which prevents the formation of atomically abrupt interfaces; and (iv) hinders the integration of materials that are thermodynamically unstable when in contact with each other.

Recently, new possibilities have appeared by merging epitaxial complex oxides with the successful methodology of 2D vdW heterostructures.¹³ To deal with the strong 3D bonding of oxides, novel methods for exfoliating ultra-thin freestanding complex oxide crystals from their growth substrate have been established based on (i) water-soluble sacrificial epitaxial layers,^{13,14} (ii) controllable weakening of epitaxial crystals by inserting 2D layers,^{15,16} or (iii) self-formed freestanding membranes^{17–19} (see our recent review paper on this topic).²⁰ By mechanically reassembling such complex oxide thin films, it is, in principle, possible to bypass the epitaxial roadblocks outlined above.²¹ For example, we showed in Ref. 21 that it is possible to assemble stacks with atomically sharp interfaces and controlled twist angles exhibiting well-defined moiré patterns in transmission electron microscopy (TEM). The ability to modulate the interfacial properties of the stack through layer selectivity and twisting introduces the potential for growing twisted oxide lateral homostructures with controllable in-plane conjunctions²² [Figs. 1(a) and 1(b)]. This method also enables the synthesis of freestanding

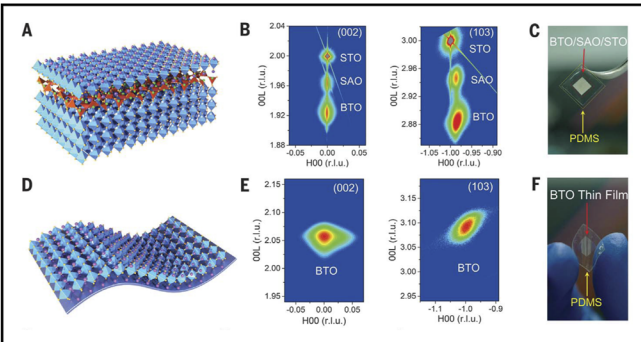
[A] ASSEMBLY OF FREESTANDING OXIDE MEMBRANES



[B] MEMBRANES FOR AS TWISTED EPITAXIAL GROWTH²²



[C] SUPER-ELASTIC FERROELECTRIC MEMBRANE²³



[D] ARTIFICIAL OXIDE TWISTING²¹

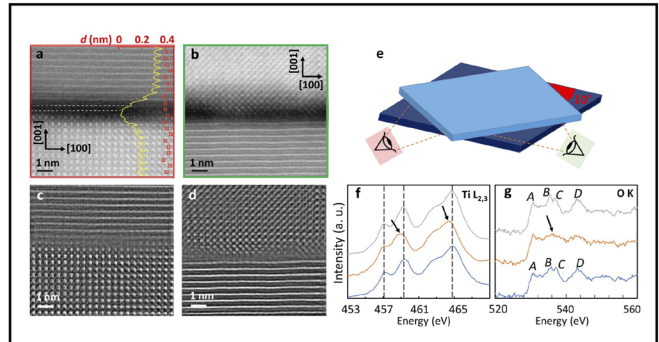


FIG. 1. (a) Schematic illustration of the release, stacking, and twisting; (b) the potential for growing twisted oxide lateral homostructures with controllable in-plane junctions;²² (c) freestanding single-crystalline ferroelectric barium titanate (BaTiO₃) membranes with super-elasticity and ultraflexibility;²³ and (d) control over the twisting process, ensuring atomically sharp interfaces.²¹

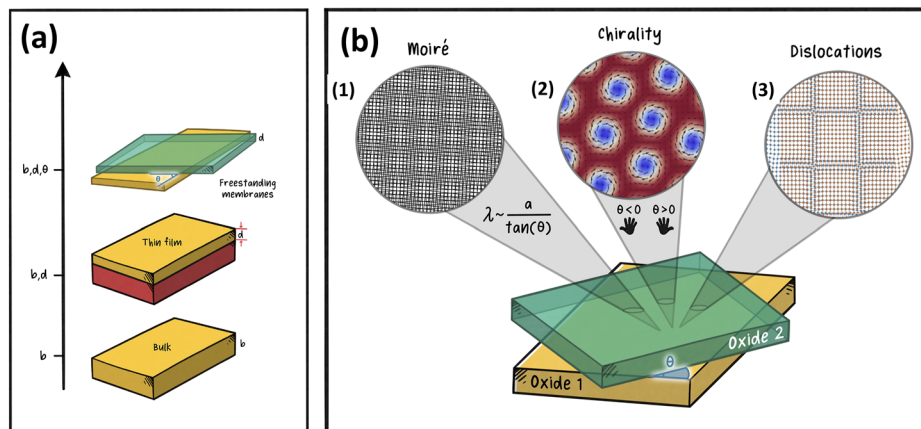


FIG. 2. (a) Schematic illustration of moiré, chirality, and dislocation in a freestanding oxide membrane with the degrees of freedom, bulk (b), thickness (d), and twist angle (θ). (b) Schematic illustration of the possible interactions that happen at the interfaces when two layers are rotated with respect to each other: (1) moiré, (2) dislocation, and (3) chirality. We define a length scale, λ , which depends on the lattice parameter (a) and twist angle (θ).

single-crystalline ferroelectric barium titanate (BaTiO_3) membranes with super-elasticity and ultraflexibility²³ [Fig. 1(c)]. In addition, it provides complete control over the twisting process, ensuring atomically sharp interfaces²¹ [Fig. 1(d)].

While these initial demonstrations suggest the assembly of freestanding oxide membranes as a promising new direction, many questions still remain. What are the physical and chemical properties of freestanding oxide membranes that make them unique? Do the freestanding oxide membranes properties differ from their counterpart bulk structure? To what degree are the physical properties of the freestanding oxide membranes influenced by stacking and twisting? What is the nature of the bonding across the interface when two freestanding oxide membranes are stacked? Will the concept of freestanding membranes be scalable to the level, where it can become technologically relevant? Below, we delve into the present challenges and offer a perspective on potential future trajectories in the field. Our primary objective is to underscore the vast potential of crafting innovative functional materials through the twisting of freestanding oxide membranes. In addition, we emphasize that the reduction of dimensionality from 3D bulk to 2D thin films followed by freestanding membranes (quasi-2D) finely tuned by precise twisting of the freestanding oxides can lead to moiré patterns, chirality, and dislocation with unexplored properties, as schematically represented in Fig. 2(a).

FUTURE DIRECTION IN FREESTANDING TWISTED MEMBRANES

Moiré engineering

A key barrier to the systematic study of correlated electron physics is the lack of tunability of conventional chemical compounds. The experimental parameters, which can usually be varied to understand the properties of correlated phases in the solid state, are pressure, mechanical strain, and chemical doping. Thus, extending this list will lead to the experimental exploration of a wider phase diagram. This expansion is essential to guide the discovery of novel quantum phases, or realizations of exotic topological states of matter that have applications in emerging quantum technologies, such as quantum computing and simulation.

What happens when bilayers of a freestanding material are rotated with a finite twist angle (θ) with respect to each other? Artificial moiré superlattices are created with a length scale (λ), which is larger than the lattice constant (a) of the individual layers, given by $\lambda \approx a/\tan(\theta)$ [Fig. 2(b1)]. The angle of rotation between the layers will play an important role in determining the properties of the re-assembled oxide materials. This approach has been highly successful in manipulating the properties of bilayer graphene and 2D materials showing flat bands,^{24,25} moiré excitons,²⁶ surface plasmon polaritons,²⁷ surface phonon polaritons,²⁸ surface exciton polaritons,²⁹ interlayer magnetism,³⁰ and 2D ferroelectricity,³¹ where the Moiré superstructure can also act as a powerful tool for revealing the consequences of the out-of-plane stacking sequence at the interface.³²

Recently, we demonstrated moiré superlattices with different periodicity in twisted perovskite oxide membranes.²¹ In 2D van der Waals materials, covalent bonds allow the atoms within the plane to remain stable, while the comparatively weaker van der Waals forces

in between the layers hold the layers to be stacked vertically. Remarkably, the observations in oxide membranes show that atomic bonds based on covalent or ionic bonds are formed at the interfaces without the need to apply high pressure and high temperature,³³ but only by “placing” the layers on top of each other in the right termination order and post-annealing at 650 °C.²¹

The weak interaction between the 2D layers in van der Waals materials allows the rotation of the layers at a certain angle with respect to one another, either by rotational alignment during assembly or *in situ* rotation of the layers.²² In contrast, the strong interlayer bonding between two oxide membranes may not allow *in situ* rotation of the layers. It is believed that the rotation of oxide membranes cannot evolve continuously with twist angle due to the nature of their bonds. Consequently, the ability to control the interfacial properties with the twist angle can be a challenge.

If we can control the interfacial coupling using the twist angle, we anticipate it will be pivotal in revealing properties distinct from those of 2D vdW materials, paving the way for an entirely new realm of material exploration. This naturally leads to the following open questions: Is there new physics that can derive from the electron's correlations behind a twisted oxide layer? What is the role of defects and strain in twisted oxide superlattices? Does twisted oxide exhibit discrete twisting angles, or can the angle be continuously adjusted? With dominating interlayer coupling, small twist angles may result in the formation of networks of localized screw dislocations to accommodate for the miss-match across the interface rather than a large period of smooth moiré beating. Similar to what was observed in graphene bilayers when they are rotated with respect to each other, a distinct shape and periodicity of a moiré pattern, which resembles grain boundaries, were observed.³⁴ This may enable new opportunities to tune the ionic conductivity, which is attributed to the high concentration of strain-induced defects at grain boundaries,³⁵ which is crucial for a wide range of energy technologies, such as fuel cells, batteries, and catalysis.

Chirality of twisted interfaces

Another fascinating and intriguing characteristic is chirality, which can be observed at the interfaces of twisted-and-stacked oxide layers. Chirality is one of the most fundamental asymmetries in nature. It is the geometrical properties of an object that are physically distinguishable by the original object from its nonsuperimposable mirror image, such as the spread fingers of a human's left and right hands [Fig. 2(b2)]. The lattice structure of chiral materials possesses a well-defined handedness, characterized by an absence of inversion, mirror, or roto-inversion symmetries. For the last decade, the electronic and magnetic properties of chiral materials in both the real-space and reciprocal space, e.g., chirality of molecular enantiomers in stereoisomers, continuous or discrete chiral magnetic textures,^{36–39} and chiral topological matters,^{40,41} have gained attention with the discoveries of exotic phenomena, such as chirality-induced spin selectivity (CISS) effects, magneto-chiral anisotropy, large anomalous Hall effects, and quantum transport for chiral spintronics.³⁶ For example, electron spins can interact with the potential of chiral objects, such as specific enantiomers, experiencing a force similar to the Lorentz force created by a perpendicular magnetic field to their velocity. The dipole moment of chiral helical molecules can be altered using external electric fields,

guiding electrons along a helical path. This induces an effective magnetic field, influencing the electron's magnetic moment and favoring a particular spin current based on momentum and spin orientation. Consequently, a chiral object's handedness dictates the spin's path relative to the electric field, causing temporary spin polarization. This phenomenon, known as CISS, has been observed in chiral molecules over short distances (around 1 nm),⁴² generating significant spin currents in various chiral materials, proving beneficial for spin polarizers and filters.^{41,43}

Another notable phenomenon is the creation of chiral magnetic textures influenced by spin-orbit coupling (SOC). SOC is a relativistic interaction between an electron's spin and its orbital movement in an internal electric field, often produced when there is a disruption in space-inversion symmetry at the boundary of two different materials. This leads to the Dzyaloshinskii-Moriya interaction (DMI), a chiral asymmetric exchange interaction. The DMI can result in two-dimensional spin vortices called skyrmions, made up of spiraled spins with a unique arrangement protected from atomic irregularities and temperature changes. Historically, producing skyrmions in metallic layers required combining a heavy metal layer with magnetic layers to ensure a strong SOC at their interface. This approach was later applied to 5*d*-transition metal oxide heterostructure systems (e.g., ferromagnetic SrRuO₃/paramagnetic SrIrO₃), where the topological Hall effect (THE) is observed. The observed THE is associated with the interfacial DMI due to the inversion asymmetry.⁴⁴ Recent findings show that, without the inherent strong SOC ("heavy metal" layer), as the skyrmions can be formed at complex oxide heterointerfaces between ferromagnetic and ferroelectric layers (SrRuO₃/BaTiO₃), also confirmed by measuring a THE. The results indicate that the lattice distortion and subsequent inversion symmetry breaking occur in the ferromagnetic SrRuO₃ layer adjacent to the interface, driven by a ferroelectric proximity effect. These chiral spin textures can exhibit particle-like behaviors when exposed to external factors, such as spin current injection. The motion of skyrmions and chiral domain walls (DWs) can be influenced by current, driven by the DMI's internal magnetic field and the spin-orbit torque (SOT) effect. This chiral effect differs from the conventional spin transfer torque effect, which has a minimal impact on DW dynamics. The DMI-induced chiral effects are promising for applications, such as SOT devices and high-speed, high-density logic/memory devices.

In analogy to the topological magnetic systems (e.g., skyrmions), the intricate interactions between spin, lattice, orbital, and charge degrees of freedom in ferroic systems can lead to the formation of nontrivial topological structures, such as clockwise/counterclockwise polar vortices, domain walls, and skyrmions. The formation of spontaneous toroidal dipole moments in polar structures has been found in epitaxially grown non-polar/polar oxide superlattices, e.g., (SrTiO₃)₁₀/(PbTiO₃)₁₀ with a 10-nm thick superlattice period.⁴⁵ These superlattices also exhibit chirality owing to the formation of a chiral array of polar vortices. The origin of the polar vortices in low-dimensional ferroelectrics has been attributed to the interplay of elastic (/the effect of constrained layers), electrostatic (the energy minimization for the DW configurations), and gradient (long-range interactions in the superlattices) energies in the polar material in order to maintain the system charge neutral.⁴⁶ In contrast to conventional ferroelectric materials that do not possess chiral textures/vortices, the chirality of a vortex-antivortex

pair can be locally created by a simple line scan with the DC-biased conducting AFM tip.⁴⁶ This indicates the tunability of polar chiral textures, and thus, it is a promising route to design polarization topologies for high-density non-volatile memory device applications. Chiral materials can effectively transfer a chirality response to the sublattice of adjacent layers^{48,49} and break the intrinsic centrosymmetric structure of the neighboring layers.⁴⁹⁻⁵¹

In oxide freestanding membranes, chiral lattices and spin textures can arise by stacking one complex transition metal oxide layer onto another with a lattice mismatch or a slight twist angle. These moiré oxide membranes, formed by stacking or twisting oxide layers (e.g., ferromagnetic/paramagnetic heavy metal membranes and ferromagnetic/ferroelectric membranes), offer opportunities to create new super-periodic lattice patterns, especially when inversion or mirror symmetry is broken. Here, we define a modulation period (λ) of the chiral pattern that depends on the twist angle, θ [Fig. 2(b2)]. Twisting the oxide layers introduces geometrical handedness, bringing forth chirality. Hence, the control of stacking/twisting distinct freestanding membranes will create new interfacial properties (e.g., Moiré superlattices, SOC, interlayer magnetic exchange, and dipolar interactions). This can lead to novel chiral effects, such as the CISS effects, topological skyrmion lattices, non-collinear spin textures, emergent magnetic phase/couplings, THE, and polar vortices. These twisted oxide membranes have potential applications, from spin-based devices to data storage. With careful engineering, they can be tailored for specific chiral properties, paving the way for the next generation of oxide-based devices.

Dislocation engineering

In general, the functionalities of transition metal oxides (TMOs) are strongly related to the partially filled *d*-orbitals of the transition metal (TM) atoms. Their properties are significantly influenced by local lattice distortions that can arise in the presence of defects, such as point defects, dislocations, grain boundaries, and interstitials. This has motivated significant activity both theoretically and experimentally toward understanding and controlling defects,⁵³ and dislocations have been shown to provide an intriguing method for realizing nanoscale channels of enhanced electronic and ionic transport.

Screw-dislocation networks are commonly observed when wafer-bonding with a relative twist angle. For the twisted oxide membranes, such dislocations can provide a new route for engineering dislocations and their associated functionalities in oxides. As an illustration, we consider two identical membranes stacked at a small twist angle. By local rotations, the two crystals may locally form regions of perfectly coherent epitaxial crystals, with the twist-induced lattice mismatch focused into highly strained boundaries by the formation of screw dislocations. This leads to networks of screw dislocations, ideally following the periodicity of the moiré lattice, which is controllable by the imposed twist angle and the symmetry of the crystal surface [see Fig. 2(b3)]. The twisted interface can be defined either by stacking a membrane on the parent bulk crystal; however, the full potential appears when assembling multi-membrane stacks.

Dislocation networks have previously been studied in direct wafer-bonding of bulk rotated semiconductors,⁵⁴ where dislocations have a marked impact on the electrical and optical properties of

silicon, and supermetallic conductivity (one-dimensional electron gas) has been observed along single screw dislocations. Large scale dislocation networks were also recently observed in BiFeO₃/DyScO₃ heterostructures, and accelerated ionic transport was observed along dislocations in epitaxial La_{0.8}Sr_{0.2}MnO₃ thin films.³⁴ Generally, engineering the lattice distortions associated with screw dislocation can be used as a very effective method for modifying the local and global functionalities of oxides. The freedom of stacking multiple layers generates a large parameter space for experimental exploration for controlling and characterizing layer materials, layer thickness, lattice constants, symmetries, and twist angles.³⁵

As an important feature, we note that the platform of twisted oxide membranes provides new possibilities for characterization of defects and dislocations and, thus, for accelerating our understanding of the correlation between twisting angle, defects, and functionalities. For example, while the imaging of dislocations is a challenge for bulk heterostructures, the membrane geometry holds the added benefit of being readily compatible with TEM studies or investigations by scanning probe techniques, thereby obviating the complicated processing needed for mechanical or chemical thinning of conventional bulk samples.

OUTLOOK

The ability to realize high-quality twisted oxide heterostructures has the potential to impact a wide range of research areas and technologies. Here, we have addressed only a few of the many exciting aspects focusing on future possibilities within moiré, dislocation engineering, and the design of novel chiral structures. Our aim is to give one perspective on future directions and to motivate further exploration. Many challenges, however, lie ahead for the community to address.

Interface engineering

The properties of twisted oxide membranes strongly depend on the interfaces between the stacked layers. Controlling and manipulating these interfaces to achieve desired functionalities can be challenging. Understanding the interfacial phenomena, such as charge transfer, orbital hybridization, and strain effects, is essential for tailoring the electronic, magnetic, and optical properties of the system.

Characterization techniques

Characterizing the twisted oxide membranes and probing their properties at the atomic scale can be demanding. Traditional characterization techniques, such as electron microscopy and x-ray diffraction, may face challenges in resolving the twisted structures and interfaces. Developing advanced characterization techniques specifically tailored for twisted oxide membranes is necessary for deeper insights into their properties.

Synthesis and fabrication

One of the primary challenges in working with twisted oxide membranes is the synthesis and fabrication process. Achieving oxide membranes that are flat and free of defects (e.g., cracks and wrinkles) is a challenging task. Furthermore, precise control over the twist

angle and ensuring high-quality interfaces between the layers can be technically challenging. Achieving a sharp interface between the freestanding layers remains a significant challenge. This process requires meticulous control of contaminants during the release and transfer of the membranes. In addition, the synthesis of the stack must occur in a protective atmosphere to ensure quality and precision.

Material compatibility

Twisted oxide membranes often involve combining different oxide materials with different crystal structures and lattice parameters. Ensuring the compatibility of these materials is crucial to avoid strain, defects, or even chemical reactions at the interfaces. It requires a deep understanding of the materials' properties and careful selection of suitable combinations.

Stability and scalability

Ensuring the stability and reliability of twisted oxide membranes is crucial for practical applications. The twisting of the layers can introduce strain and defects, which may affect the long-term stability of the material. Understanding the stability of the twisted structures under different environmental conditions and developing strategies to mitigate degradation is a significant challenge. Developing scalable fabrication techniques and methods for transferring and integrating twisted oxide membranes into device architectures is a significant challenge.

Theoretical understanding

Developing theoretical models and computational tools to understand and predict their properties is a challenging task. The interplay between different factors, such as lattice mismatch, electronic correlations, and interfacial effects, requires advanced theoretical frameworks to unravel the underlying physics.

Addressing these challenges will pave the way for harnessing the full potential of twisted oxide membranes and unlocking their applications. For example, by integrating different layers with distinct functionalities, such as ferroelectric, ferromagnetic, and superconducting properties, it becomes possible to realize devices with multiple functionalities on a single platform. The unique properties of these membranes offer opportunities for technological breakthroughs and advancements in various fields in areas, such as electronics, spintronics, catalysis, and energy storage.

ACKNOWLEDGMENTS

N.P. acknowledges the support of Novo Nordisk Foundation Challenge Programme 2021: Smart nanomaterials for applications in life sciences, BIOMAG Grant No. NNF21OC0066526, the support from the ERC Advanced "NEXUS" Grant No. 101054572, and the Danish Council for Independent Research Technology and Production Sciences for the DFF-Research Project 3 (Grant No. 00069B). D.-S.P. acknowledges the funding from Villum Fonden (SYMTEC, Grant No. 111814).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

N. Pryds: Conceptualization (lead); Project administration (lead); Supervision (lead); Writing – original draft (lead); Writing – review & editing (equal). **D.-S. Park:** Writing – review & editing (equal). **T. S. Jespersen:** Writing – review & editing (equal). **S. Yun:** Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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