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Abstract

The ability to rationally design materials with specific properties is a major motivator in materials science and condensed matter physics. Driven by this motivation and by the potential to open new technological avenues, researchers are continually pushing the boundaries of established materials while pioneering new ones. This push has led to newly developed techniques for fabricating, handling, and transferring freestanding oxide membranes (FOMs), providing a new avenue to overcome traditional oxide epitaxial growth limitations, which relies on specific substrates and synthesis conditions. These advances enable the design and creation of novel materials with previously inaccessible interface symmetries, stacking configurations, and atomic reconstructions, opening exciting possibilities for exploring new physics and developing innovative device architectures using atomically engineered complex oxides. At the same time, these advancements present new challenges in the synthesis, membrane transfer, characterization, and final functionalization of FOMs. The field is still in its early stages and necessitates further scientific and technological progress. This roadmap offers an overview of the current state of the field and perspectives on its future, highlighting opportunities, challenges, and guiding principles for the research community.

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1. Introduction

Nini Pryds, Felix Gunkel, Felix Trier, Zoran Jovanović and Lior Kornblum

The successful isolation of graphene from graphite two decades ago can be regarded as a ‘big bang’ in materials science. This breakthrough propelled the development of two-dimensional (2D) van der Waals (vdW) compounds and has become a focal point of research for many scientists around the world. This development has opened up opportunities for exploring, tailoring, and controlling novel physics and emerging semiconductor materials. These materials exhibit unprecedented new properties, often dramatically influenced by factors such as stacking order, thickness, twist, and interlayer spacing. Recently, advances in synthesis of transition metal oxides (TMOs) have led to the creation of the so-called, free-standing oxide membranes (FOMs), which have gained significant attention. This development has the potential to unlock new extrinsic phenomena in TMOs, and enrich the already complex phase diagram of their intrinsic functional properties. Unlike 2D vdW materials, which derive their physical properties from weak interactions between electrons residing in *s*- and *p*-orbitals, TMOs exhibit a range of unique characteristics driven by stronger electron-electron interactions present in their occupied *d*-orbitals. TMOs have been a fascinating area of study in physics, electronics, and materials science for decades, thanks to their diverse functional properties such as superconductivity, ferroelectricity, magnetic orders, and mixed ionic-electronic behavior, all of which can be widely tuned through lattice, orbital, and spin degrees of freedom.

While TMO thin films, heterostructures, and interfaces have been studied in great detail, the application of atomically designed TMOs has been widely limited due to their fundamental epitaxial restriction to single crystalline substrates and synthesis processes. Such epitaxial schemes pose challenges, for example, regarding compatibility with silicon, CMOS-based process flows, and existing technological platforms.

Recent advances in the fabrication of FOMs via growth-and-release techniques have opened a new pathway to overcome these fundamental limitations. These developments allow freestanding oxides to be handled as nanometer-thin membranes down to the unit-cell level. This new approach to materials design includes transferring layers onto new platforms and combining, twisting, and engineering stacked oxide heterostructures using direct-transfer techniques. This approach may produce new interfaces that are structurally impossible to achieve using conventional thin-film synthesis. It can break symmetry constraints and enable the intentional design of structural chirality through specific twist angles. Moreover, direct combinations of oxides with 2D materials that are typically unstable under oxide synthesis conditions are now possible, potentially bridging the gap between the previously disparate fields of complex oxide electronics and 2D materials.

The potential for discovering new physics is one of the key advantages of TMOs. Additionally, developing new methodologies and applications is another exciting outcome of this emerging field, as single crystalline TMOs can now be fabricated in arbitrary configurations. This flexibility allows their transfer into operando cells for microscopy and spectroscopy, onto MEMS chips for *in-situ* manipulation, or onto flexible substrates, opening up a wide range of novel device architectures. Such capability is particularly important for phenomena such as ferroelectrics and magnetoelectronics, where single-crystallinity and atomic-level control of oxide materials can lead to new properties that can now be integrated into advanced electronic systems across various platforms. Oxide twistronics is a new field still in its infancy. Insights from 2D materials can guide the field but must be adapted to account for the distinct bonding and structural relaxation mechanisms in twisted oxide heterostructures, likely requiring the development of a dedicated experimental and theoretical framework of understanding twisted oxide interfaces and their atomic reconstructions in detail.

Integrating FOMs into diverse environments, such as silicon and CMOS platforms, while exploiting new degrees of freedom, including strain and vibrational modes, promises major advances in physics, electronics, and materials science. Despite growing interest in the field, several important challenges remain. The processes for handling FOMs, including synthesis, transfer, and stacking, require further development to ensure reproducibility. At this stage, understanding and controlling defect structures, interface sharpness, strain relaxation, and the long-term stability of delaminated membranes are essential for advancing the field of oxide membranes. Material yields need improvement, and new characterization and application tools must be advanced and developed. Drawing on existing experiences with 2D materials could foster valuable synergies and offer potential useful guidance for overcoming these challenges.

In this roadmap, we have brought together experts in the field to highlight their perspectives on the scientific and technological future of FOMs. Starting with a perspective on *the status of 2D materials and*

respective lessons to be applied to oxides, this compilation will address state-of-the-art and future perspectives in the ***synthesis and processing of FOMs***, the ***dedicated characterization of their properties***, the ***current status and challenges of twisted and stacked FOMs***, the ***co-integration of oxides with 2D materials***, and finally the resulting *Properties And Applications Of FOMs*.

This roadmap, with a primary emphasis on future directions, aims to tighten the bonds between the different research communities and ultimately bridge the gap between neighboring fields of complex oxides and 2D materials, by illustrating the interdisciplinary nature and the intersections of these rapidly growing research areas at the interface of chemistry, physics, materials science, and device engineering.

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2. Overview

2.1. Heterostructures with freestanding oxides and 2D materials

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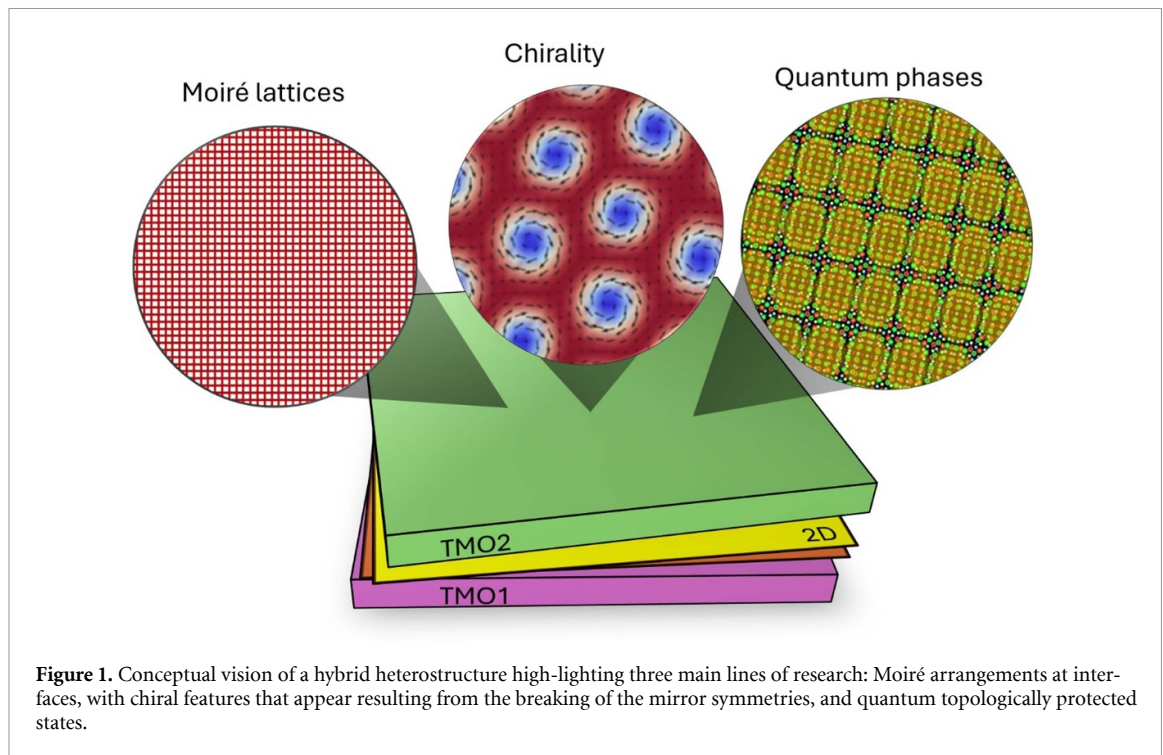
Status

vdW heterostructures consist of ultrathin materials that adhere only by vdW forces. This architecture eliminates lattice match requirements, allowing countless combinations of materials to be combined with atomic precision while maintaining interface quality. For the extensive class of atomically flat and thin 2D materials, the combination of extreme proximity and control of scattering and screening, has led to an explosion of fundamental physics and applications. The most striking feature is the twist degree of freedom, where sub-degree control of layer-to-layer angle produces surprising physics, including unconventional superconductivity. Up to recently, moiré engineering has been limited to 2D materials. Now, with freestanding oxides available in nanometer thicknesses, complex TMOs homo and heterostructures can be assembled like 2D materials. Note that we refer to a freestanding oxides as TMO membranes free from the growth substrate and transferred to a final substrate or layered structure of our interest. While integrating freestanding TMOs into heterostructures is compelling, due to their robust properties from ferromagnets to superconductors, these structures face challenges. TMOs grown epitaxially on substrates may lack atomic flatness, with interfacial defects and substrate interactions potentially compromising the resultant properties and limiting the possibilities that twist-based heterostructures otherwise offer. The combination of freestanding TMOs with superlubric 2D materials could chemically and mechanically decouple oxides, allowing dynamically twisted oxides analogous to all-2D heterointerfaces, where the 2D materials provide a mechanical and chemical slip-layer function that is distinct from, and does not preclude, electronic coupling such as proximity effects or interfacial hybridization across the interface. The combination of these dissimilar materials opens new possibilities for emergent interfacial physics. One can pair a magnetic or ferroelectric oxide with a 2D conductor, potentially inducing magnetism or ferroelectric order in the 2D layer via proximity effects. These freestanding TMO/2D heterostructures provide an excellent platform for realizing phenomena unattainable in conventional epitaxial systems, with potential impact on electronic, spintronic, and energy devices.

Current and future challenges

Recently experimental demonstrations [1, 2] and first principles theoretical simulations [3, 4] have showed that TMO crystals can be assembled into homo- and heterostructures with twist angle control. However, significant differences exist between conventional vdW layers and TMO membranes. Unlike vdW materials, TMOs are characterized by strong ionic and covalent bonding, interface reconstructions, oxygen stoichiometry variations, and termination-dependent chemistry. As a result, key questions remain regarding whether and how the conventional moiré physics paradigm emerges when such fundamentally different layers are twisted relative to one another: What unique physical and chemical properties do TMO membranes possess? How do TMO stacking, interfacial orientation, and twist angle influence these properties? Will TMO membranes exhibit ‘magic angles’ like those in 2D heterostructures? Can physical phenomena such as flatband superconductivity, moiré-induced charge or spin ordering, interfacial ferroelectric textures, or orbital-selective electronic phases be ‘dialed in’? What type of bonding occurs at the interface of two FOMs?

Recent reports [5] on chiral arrangements of electrical polarization in twisted MoS₂ bilayers linked to moiré patterns suggest that topologically protected interfacial states may not be exclusive to twisted 2D materials and flat band scenarios and even ‘a magic angle’ have been predicted for BaTiO₃ twisted layers [6]. The combination of TMO homostructures with conventional 2D materials atop or between freestanding TMO layers, as in figure 1, presents new options for engineering and reading out emergent properties [7, 8], while the chiral and topological features sketched remain to be experimentally realized, and involve substantial fabrication challenges. The close proximity of the layers, variety of physical properties of individual layers, and interface differences contribute to the complexity of these structures. The loss of atomic periodicity at twisted interfaces complicates theoretical descriptions and predictions require large numerical models which are computationally very demanding.



While techniques for stacking 2D heterostructures like the hot-pickup method [9] can be modified for assembling 2D/oxide stacks with twist angle control, developing reproducible, scalable processes for continuous control of twist angles and interface integrity will remain a significant challenge. Methods to grow epitaxial large area complex oxides on sacrificial layers enabling efficient detachment and transfer of oxide membranes have evolved using molecular beam epitaxy (MBE), sputtering and pulsed laser deposition (PLD) techniques [10, 11]. However, if 2D layers serve as lubricants between oxide membranes for continuous rotation, remote epitaxy [12] offers a path to avoid imperfections from successive transfers in hybrid heterostructures. Due to the partial oxygen pressure needed for the synthesis of TMOs, the challenge is reconciling the high temperature needed for defect-free oxides with the chemical stability of the 2D layer.

It is necessary to explore the symmetry breaking at twisted TMO/2D interfaces and its effects on spin, charge, structural degrees of freedom, and proximity effects. While continuous twisting of TMO layers is not feasible yet, using 2D materials to reduce inter-layer bonding could enable tunable emergent quantum states, where ionic potentials in TMO unit cells could enable operation at higher temperatures than all-2D heterostructures. In our view, the introduction of 2D slip-layers between FOMs is the most promising route to dynamically twistable oxide moiré systems and should be prioritized as a near-term experimental target. Control of quantum states with external stimuli like strain, electric/magnetic fields and light requires increased focus on interfacial engineering research.

Theoretical modeling using an atomistic approach will play a crucial role in understanding these complex interfaces and phenomena. Density Functional Theory (DFT) provides insights into atomic structure, vibrational/dynamical, and mechanical properties, and serves as a starting point for electronic structure and electron transport in oxides [3], ultra-thin oxides [4], and 2D materials [13]. DFT has recently studied twisted TMO interfaces of BTO and STO, showing how polarization patterns can be tuned and, as for MATBG, flat band conditions [6, 14]. However, for interfaces the Å-scale unit-cell periodicity is lost, and moiré-scale atomic relaxations can play an important role, while standard DFT is typically limited to unit-cells with a few thousand atoms. Moreover, the TMO-2D interface involves multiple species and types of bonding, covalent and vdW, and predictive computational studies of twisted TMO/2D systems requires computationally efficient approximations. To this end, DFT-trained machine learning force fields (MLFF) can now be applied to reach realistic moiré cells of TMOs, including the long-ranged Coulomb forces [15]. While MLFFs may describe structural and anharmonic vibrational properties, for the electronic structure of TMOs and TMDs the d-electrons can play a crucial role, requiring extensions beyond DFT + U such as dynamical mean field theory, and possibly including spin-orbit interaction. This direction still awaits methodological breakthroughs, possibly involving machine learning (ML), to reach the moiré scale.

Advances in science and technology to meet challenges

Realizing scalable fabrication of hybrid twisted TMO/2D heterostructures with precise twist-angle control requires technological advances in transfer methods. Manual pick-and-place methods used for stacking small-scale heterostructures must evolve into automated, wafer-scale processes capable of aligning and stacking large-area membranes. Advanced robotic systems with high-resolution optical or electron-based alignment techniques are needed to ensure precise twist-angle registry across interfaces. Roll-to-roll or continuous transfer methods could enhance scalability and uniformity, especially when combined with *in-situ* monitoring and feedback-controlled alignment. Critical to these advances is refining techniques for maintaining interfacial cleanliness during transfer through integrated vacuum or inert atmosphere systems, to prevent contaminants and defects, ensuring consistent high-quality interfaces for exploring twisted heterostructures at scale. Among these requirements, we identify automated wafer-scale stacking with sub-degree twist registry as the principal near-term enabling technology: without it, twisted oxide heterostructures will remain confined to single-flake demonstrations and the field will not transition from proof-of-concept to systematic exploration.

Additionally, metrology capable of detecting and exploiting subtle electronic and structural changes at buried interfaces must be advanced. Such capabilities are critical for validating theoretical predictions and guiding material optimization, ultimately enabling practical applications of twisted oxide-based heterostructures, where buried 2D or TMO layers can serve multiple roles as sensor layers, dielectric spacers, mechanical slip-layers or active functional components, such as electrostatic gates.

Emergent quantum phases as a function of twist angle have been reported in 2D materials using various techniques where heterointerfaces are twisted at a fixed angle [16]. If interfacial friction between TMO membranes can be reduced by introducing 2D lubricating layers, a quantum twisting microscope adapted to TMO materials, could enable continuous, efficient screening of electronic, optical and phononic properties of twisted 2D materials [17].

The exploration of opportunities with twisted TMO/2D heterostructures requires not only experimental property screening but also improvements in predictive modeling. Advancing predictive modeling needs better multiscale computational approaches that bridge first-principles calculations with computationally efficient modeling. ML and neural network methods [18] are revolutionizing and extending DFT capabilities for materials screening. Training sets of smaller DFT systems generate MLFF or ML interatomic potentials [19–21] enabling simulations of equilibrium atomic structure and molecular dynamics for systems beyond 100,000 atoms. For electronic structure, DFT-parametrized tight-binding (TB), e.g. obtained from DFT via the Wannier orbitals [22], has successfully described important aspects of 2D moiré electronic structure [4, 13]. However, predictive modeling of the correlated electronic structure of twisted TMO/2D interfaces beyond mean-field DFT represent a longer-term goal that may also benefit from machine-learning advances.

Concluding remarks

The integration of freestanding TMO membranes into heterostructures offers unique opportunities for creating hybrid heterostructures that circumvent inherent limitations of conventional epitaxial systems. Precise strain control is essential, as uncontrolled lattice relaxation can disrupt targeted electronic configurations such as direct bandgaps or flat bands; this can possibly be alleviated by integrating 2D materials within TMO heterostructures as slip layers, with an additional perspective of expanding the functionality. Achieving wafer-scale, ultraclean transfer protocols remains critical to prevent contamination, and this is expected to remain a serious challenge. The recent progress in ML techniques enable efficient multi-scale modeling of TMO/2D interfaces the moiré scale at the level of DFT.

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2.2. Growth of oxide membranes, vector substrates and heterointegration

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Status

Fabricating heterostructures via epitaxial growth enables materials with different physical properties to be combined into composite materials, which are often characterized by clean interfaces between the constituent layers [23, 24]. However, single-crystalline heterostructures must be grown epitaxially on single-crystalline substrates. This requirement restricts the functionality of oxide heterostructures. However, novel methods of decoupling films from their substrates and transferring them cleanly onto a different material platform allow us to overcome these limitations. These advances open up new possibilities for material synthesis and device functionality.

Current and future challenges

The world of thin films and thin film devices grown by heteroepitaxy is constrained by the constraints of lattice size and symmetry match on the surface of the substrate. Even with the most advanced heteroepitaxy methods, no platform exists for strain- and domain-engineering in highly lattice-mismatched materials, which makes monolithic integration of different functional materials difficult. An effective solution to this problem is to use epitaxial lift-off techniques. This approach allows for either lifting off the full device stack for heterointegration [25–27] or combining lift-off, stacking, and further growth [28–31] to build functional heterostructures that are not limited by the constraints of heteroepitaxy. Stacking of different materials takes place at room temperature and thus reduces disturbing the thermodynamic stability of the chemical compositions and the interfacial lattice structures which may be unstable at elevated temperatures. Interdiffusion is usually minimal at room temperature, which contributes to the advantages of heterostacking. Possible sample fabrication at low temperatures, as opposed to the high temperatures typically required for epitaxial growth, opens up new fields of application. For example, they incorporate functional oxides into the thermal budget of CMOS technology (<450 °C by heterointegrating materials at room temperature rather than at typical growth temperatures of complex oxides of >600 °C. Thus, membrane stacking for heterointegration opens the door to combining biochips and oxide electronics as well, thereby advancing bio- and chemical sensing capabilities. Stacking thin films also far extends the phase space of materials-by-design, offering a versatility that cannot be achieved by state-of-the-art techniques [32, 33]. This opportunity is highlighted in the concept of vector substrates, where a membrane transferred onto an appropriate carrier substrate can be then used as a substrate for epitaxy itself. This concept significantly widens the horizons of heterointegration (figure 2) [31].

There are two families of methods available for lifting thin films from the substrates on which they were grown in order to obtain membranes [34]. The first family is based on wet etching, whereby a sacrificial layer is selectively etched to release the membrane. The second set of methods uses dry lift-off processes. These include laser lift-off, spalling and remote epitaxy. Dry methods are quick and can easily be extended to wafer scale. In contrast, wet methods tend to be slow and tedious. They also limit the choice of sacrificial layers, which must be compatible with the selective process. Despite these limitations, wet methods offer a broad range of materials and the ability to produce grown films with low defect densities. The ultimate goal is to develop a universal method for membrane lift-off and high-yield transfer that can withstand a large number of substrate reprocessing cycles (see figure 2).

Advances in science and technology to meet challenges

The field of freestanding single-crystalline membranes has grown rapidly over the past decade. The community has made advances in discovering new phenomena and making use of the new experimental possibilities enabled by these membranes. These efforts have led to significant progress in overcoming many of the aforementioned challenges. A variety of sacrificial layers spanning a significant proportion of the available lattice constants of single-crystalline substrates have now been optimized [35]. It has been found that matching the lattice and minimizing inbuilt strain during epitaxial growth is important for minimizing crack formation during lift-off [31, 36, 37]. Optimizing the lift-off procedure further enhances the fabrication yield of large-area, high-quality membranes [38, 39]. Wafer scale lift-off and control of defects in the membrane caused by strain relaxation and dissolution will be critical for the integration of oxide membranes into the future generations of electronics [40].

The described challenges are short-term and urgent, primarily concerning membrane production for heterointegration. Furthermore, the platforms onto which the membranes are transferred are highly

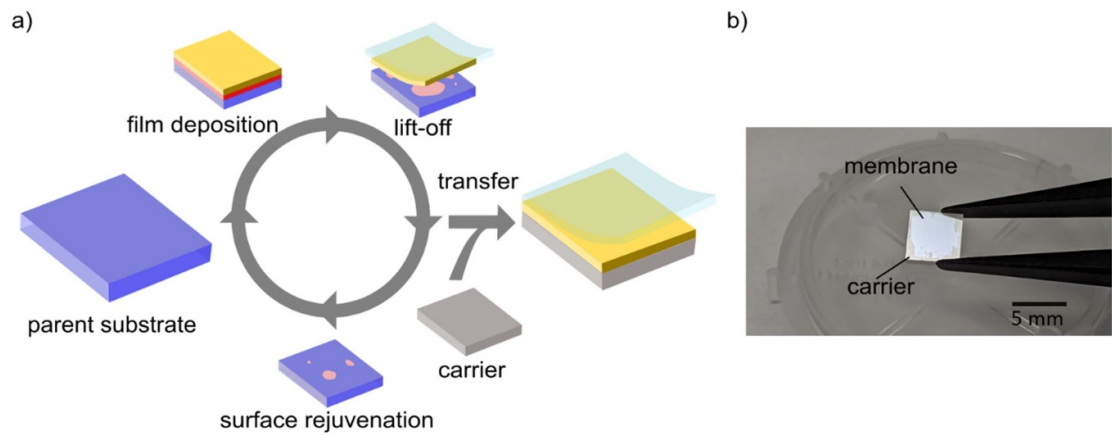


Figure 2. (a) Idealized workflow for the creation of a vertical heterostructure using wet membrane transfer. An epitaxial lift-off on a parent single crystal substrate, followed by transfer onto a carrier, results in a stacked heterostructure that can be used to define the architecture of the device through further growth or patterning. After appropriate surface treatment, the parent substrate is rejuvenated for reuse, making the process self-sustaining. This is further enhanced by the fact that the transferred membrane can be used as a parent substrate for further processing. (b) Realization of a vector substrate obtained by transferring a SrTiO_3 membrane onto a sapphire carrier. The vector substrate can be handled like a standard substrate for thin film—if harsh chemical etching steps are avoided.

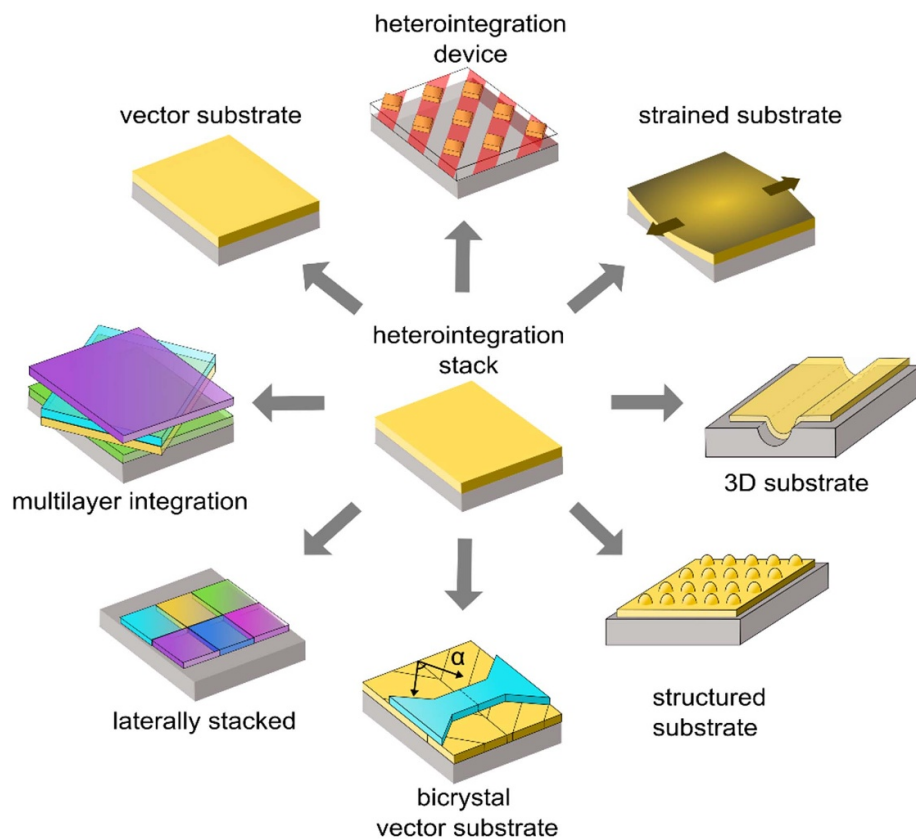


Figure 3. Multiple use cases of heterointegration of materials by stacking. Moving beyond the limitations of traditional epitaxial growth opens up a variety of avenues, enabling not only monolithic out-of-plane heterointegration, but also the lateral definition of heterostack properties. Exciting new directions to explore via membrane stacking-enabled heterostructuring include processes such as micro- and nanostructuring, twisting-driven moiré, lateral stacking of transferred membranes, and macroscopic 3D geometry on rigid or flexible carriers. Vector substrates, heterointegrated bicrystal vector substrates and 3D substrates have already been demonstrated.

relevant to future membrane applications and are therefore the subject of intense research. In the future, analog and digital electronics may benefit from vector substrate technology by integrating the functionality of complex materials such as functional oxides or nitrides onto standard chips fabricated on standard substrates. Sapphire, for instance, is an ideal substrate for many high-frequency devices. High- T_c

bicrystal Josephson junctions can now be easily integrated onto such substrates [41]. There are numerous further possibilities for what a vector substrate, i.e. a membrane on top of a pre-defined carrier, can look like. These include 3D pre-patterned or nanostructured substrates onto which membranes can be transferred, thus enabling local and in-plane manipulation of functional properties. In addition, epitaxial growth will be possible on curved surfaces. This key advance will open new paths for applications of epitaxial growth, for example in the fields of optical and electro-optical coatings (figure 3). The fabrication of such heterostructures requires the close co-optimization of the substrate surface, the transfer and the post-processing to ensure a clean carrier-membrane interface and membrane conformity with the carrier surface [11, 41, 42]. This also involves preparing the transferred membrane for further growth and stacking.

Another area that will require significant effort in the future is developing a framework for reproducibly transferring large area membranes with high angular and spatial precision. This capability is essential if heterostacking is to progress beyond the R&D stage and enter the world of applied electronics. The next step will then be to automate the entire process, from epitaxial growth and lift-off to transfer.

Concluding remarks

The realization of epitaxially grown, freestanding membranes, and their subsequent emergence as a fertile field of research, has created new opportunities in electronics. This progress has been achieved by incorporating transferred membranes ex-situ onto new platforms. Device stack transfer, heterointegration, and large-scale membrane production are possible, as is the use of membranes for further growth. Future advances are already apparent. Improvements in yield, scale, replicability and consistency demonstrate that the concept of producing high-quality devices by combining the growth, release, and stacking of a wide variety of materials is a realistic prospect.

2.3. TMO with a twist

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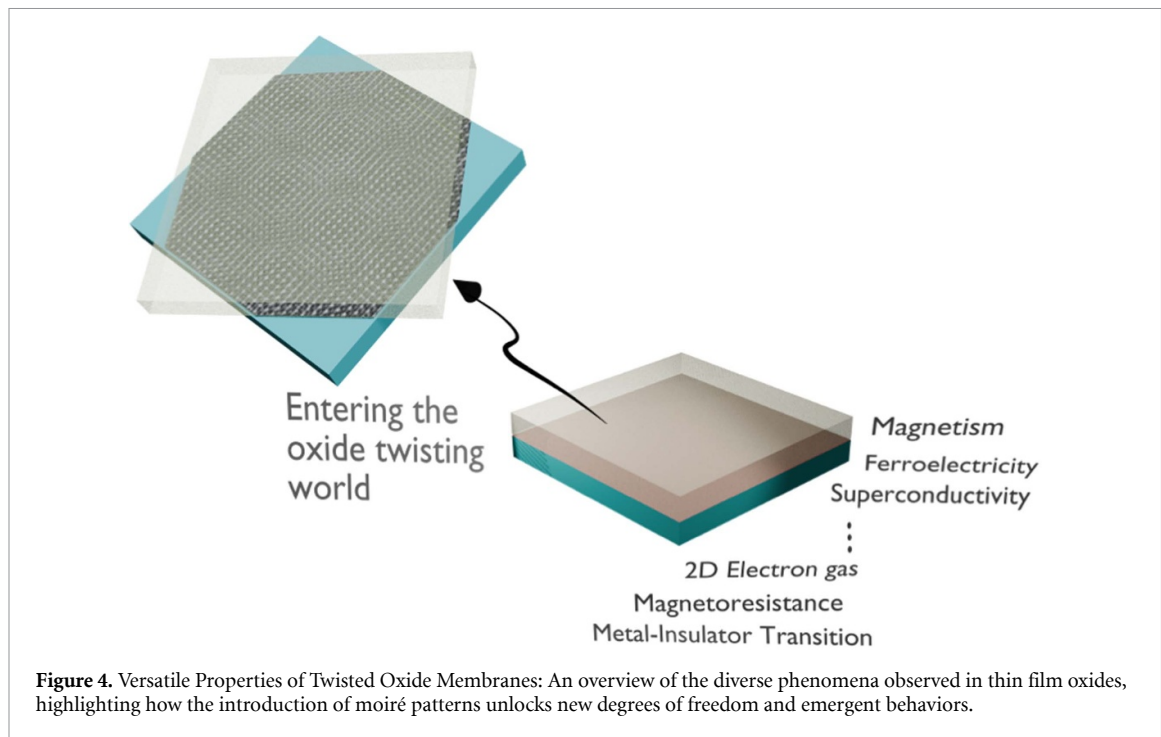
Status

When a sheet of 2D material is placed over another and slightly rotated, the resulting twist can significantly alter the bilayer's properties and lead to exotic physical behaviors. A notable example is the discovery of a flat band in twisted bilayer graphene, which laid the foundation for the field of twistrionics or Moiré materials. These atomically thin crystals have revolutionized our understanding of materials, enabling the tuning of their properties without altering chemical composition through methods such as strain, electric fields, or twisting. This capability to induce new states of matter gave rise to the emerging field of twistrionics, where twist and electronics are intricately combined.

Building on this foundation, twisted TMOs are now emerging as a promising class of nanostructures. They offer unique opportunities due to their rich electronic, catalytic, and ferroelectric properties, which can be engineered via controlled twisting. However, challenges remain in scalable fabrication, precise control of twist angles, and understanding the complex interplay between mechanical deformation and material functionalities. This marks just the beginning of exploring twisted TMOs, which hold vast potential for future applications. Progress in synthesis techniques, *in situ* characterisation, and theoretical modeling will be crucial to fully harness these structures. This perspective for the roadmap outlines the key scientific challenges and future directions for advancing twisted oxides from experimental curiosities to practical technologies. Given the rich history and rapid evolution of moiré materials, further discoveries and surprises can be expected, inspiring a new frontier at the intersection of twist engineering and TMOs.

Current and future challenges

A twisted structure is created by stacking two or more adjacent layers at specific angles, resulting in a moiré pattern. The emerging field of moiré quantum matter gathers new collective phenomena and interactions driven by unexpected band reconstructions [43–45]. The electronic properties of moiré materials created by twisting vdW layered materials are largely determined by the bonding at the partially incommensurate interface between the twisted crystalline surfaces [46]. In the presence of the weak vdW interlayer interaction, structural relaxation is relevant mostly in the limit of small twist angles, where nearly coincidental atomic registry of both layers occurs over large areas. In fact, for large enough moiré periodicities, interfaces are known to relax into commensurate domains separated by relaxed domain walls, which can be characterized as networks of dislocation lines [47, 48]. In twisted structures based on TMO membranes [34, 49–51], with strong mixed ionic/ covalent bonding between the layers, structural reconstructions controlled by the activation energy to form bonds at the interface between the layers may occur in a wider range of twist angles and stacking conditions. However, bonding strength at the twisted interface may depend critically on the termination of the oxide layers and the presence of interface adsorbates or interlayer disorder driven by ripples or cracks. Although some works have shown that a degree interlayer interaction survives across 0.5–1 nm disordered interface widths [52], other works have demonstrated that oxygen annealing enables substantial interface reconstruction [32, 53], which may occur even under electron irradiation in the electronic microscope [42], evidencing that as-twisted oxide interfaces are in highly metastable states. In fact, recent x-ray work on oxygen-annealed, high-temperature homo-interfaces in twisted SrTiO₃ (STO) has suggested the presence of a periodic lateral superlattice, consistent with the formation of dislocation networks [53], indicating strong bonding and significant strain relaxation. Moreover, highly reconstructed interfaces may even form between materials with different crystalline structures under high-temperature oxygen annealing [11], signaling interesting new directions for the design of otherwise ‘impossible’ interfaces. Optimizing the twisted interface may require controlling surface termination, cleanliness and morphology to avoid adsorbates or ripples, possibly by ‘brushing and ironing’ the layers prior to or after twisting. On the other hand, the atomic reconstructions and strain relaxations may depend sensitively on surface termination through its effect on the relative weights of ionic and covalent bonding channels [6]. For example, in the case of twisted BaTiO₃ (BTO) bilayers, while BaO–BaO interfaces foster a pure ionic bonding, TiO₂–TiO₂ or mixed TiO₂–BaO terminations have larger participation of covalent bonding, which, as demonstrated by first principles calculations, determines stronger interlayer interaction and smaller interface widths with dramatic consequences on the electronic structures [6]. In addition, in the covalent bonded interfaces, Ti



under-coordination may give rise to modified Ti doping [14, 55]. Controlling the surface terminations of twisted interfaces may be another important challenge to meet towards the control of their atomic reconstructions and consequently their electronic properties. Yet, it becomes clear that the much stronger interlayer interactions prompted by the stronger bonds of oxides, as compared to layered vdW materials, may be responsible for deeper electronic reconstructions driven by twisting [14]. For instance, flatband scenarios appear in twisted oxides at much larger twist angles approaching 20° than in layered materials ($1\text{--}5^\circ$) as recently reported in twisted BTO [6] and twisted STO [6, 14, 54]. Flatband scenarios in twisted transition metal membranes, as shown theoretically for BTO and STO [6, 14, 54], may trigger unexpected responses driven by the already strongly interacting d electrons. Although the analogy with twisted 2D materials is appealing, strong ionic–covalent bonding, interfacial reconstruction, oxygen non-stoichiometry and termination-dependent chemistry may fundamentally limit its extension to oxide membranes. Beyond changes prompted by the modified electronic structure, the effect of twisting of oxide membranes hosting ferroic orders (ferroelectrics, ferromagnets, multiferroics) may result in unexpected polar topologies driven by (chiral) symmetry breaking [1, 52, 55]. Reducing the dimensionality from 3D bulk materials to 2D thin films and then to freestanding membranes (quasi-2D), with precise twisting of the oxide layers, can generate moiré patterns, chirality, and dislocations—features that are absent in conventional 2D materials—and result in novel, largely unexplored properties [49, 54]

Advances in science and technology to meet challenges

Moiré superlattices in layered TMOs can fundamentally alter their electronic properties. This shift from traditional thin film fabrication to moiré engineering in oxides paves the way for innovative applications and new functionalities (see figure 4). Controlling these moiré patterns is, therefore, crucial for tailoring material behavior. To study phenomena that depend on the twist angle between layers, it is essential to produce high-quality, large-area twisted materials with accurate angle precision.

Current research efforts mainly focus on developing advanced fabrication techniques to accurately manipulate and control twist angles in oxide membranes. However, creating these membranes remains challenging due to the difficulty of maintaining precise control over layer orientation and ensuring clean, defect-free interfaces. Producing defect-free, high-quality membranes demands highly precise and sensitive manufacturing processes, which require accurate selection of the sacrificial layer [56] as well as innovations at the nanoscale to minimize imperfections and eventually interface reconstruction strategies. Indeed, recent high-resolution imaging of unreconstructed interfaces has revealed substantial structural disorder and nanoscale regions of incomplete contact between the layers [57]. Moreover, the inherent fragility of these membranes makes maintaining their structural integrity during twisting operations particularly difficult. New handling and processing methods are needed to prevent damage and ensure consistent quality. Moreover, managing mechanical strain is critical, as excessive or uneven strain can

introduce defects, distort the lattice, or cause membrane failure. Developing strategies for real-time strain monitoring and control is thus vital. Achieving uniform twist angles [58] and consistent material properties across multiple membranes remains a significant challenge, necessitating improved control over the fabrication process and greater reproducibility. Integrating oxide membranes with other 2D layered materials offers exciting opportunities [59] but requires careful interface engineering to preserve desirable properties and ensure seamless operation in composite systems. To fully understand these complex structures at the nanoscale, advanced analytical techniques, including high-resolution microscopy, spectroscopy, and electron ptychography [57] and tomography, must be further developed to probe their structural, electronic, and vibrational characteristics in detail. Additionally, tailoring membrane properties for specific applications involves overcoming hurdles related to chemical and mechanical stability, scalability for mass production, and integration into existing electronic, energy, and sensing devices. On the theoretical front, developing new models [60] and methods [1] capable of accurately describing larger and more complex structures is essential for predicting behavior, optimizing designs, and guiding experimental efforts. Collectively, these scientific and technological advancements are critical for overcoming current limitations, unlocking the full potential of twisted oxide membranes, and enabling their deployment in next-generation technologies.

Undoubtedly, the ability to produce high-quality, large-area twisted materials is a fundamental step toward exploring twist-angle-dependent physical phenomena and advancing applications across electronics, energy, and sensing fields.

Concluding remarks

This perspective article highlights the potential of twisting techniques to influence the physical properties of FOMs. Twisted oxide heterostructures present exciting opportunities in condensed matter physics and materials science, owing to their tunable and diverse properties. However, challenges persist in precisely controlling twist angles, creating high-quality interfaces, and advancing fabrication methods for practical applications. Non-idealities currently limit reproducibility and complicate interpretation, but they do not invalidate twisted oxide membranes. Instead, they define the key materials challenges that must be solved to make the platform reliable. The exploration of the twist angle-dependent physical phenomena at the new frontier of twisted oxides will require future efforts on the preparation of high-quality, well-defined twisted angle materials.

Acknowledgements

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3. Synthesis and realising: Status and future

3.1. Oxide freestanding membranes fabricated with PLD

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Status

Oxide moiré heterostructures offer unprecedented control over electronic, magnetic, and structural properties through the interplay between intrinsic oxide functionalities and engineered moiré periodicity [2, 14, 33, 54, 55]. The foundation of oxide moiré engineering lies in the ability to synthesize high-quality epitaxial membranes and assemble them into twisted heterostructures with atomic-level precision. Epitaxial thin-film growth is achieved via PLD [61], with *in situ* high-pressure reflection high-energy electron diffraction (RHEED) providing real-time monitoring and feedback to ensure precise layer-by-layer control [62].

To produce FOMs, the sacrificial layer method has become the standard approach. Typically, epitaxial layers of SrTiO₃ (STO) and Sr₂CaAl₂O₆ (SCAO) are grown by PLD [63]. However, the synthesis of effective sacrificial layers presents notable challenges that demand careful material selection. For example, while SCAO preserves the perovskite step-terrace morphology, precise control of surface termination [64] remains difficult. In many cases, alternative sacrificial layers are required to achieve or preserve the desired interfacial termination [65].

A prominent example involves LaAlO₃/SrTiO₃ bilayers [66–68], where a SrRuO₃ sacrificial layer is inserted between the PLD-grown film and the substrate to establish the correct interface termination. SrRuO₃ facilitates the fabrication of FOMs with modified interfacial terminations—such as AO- instead of BO₂—due to termination layer conversion that occurs during its growth on TiO₂-terminated SrTiO₃ substrates [69].

These advances have enabled one of the most significant breakthroughs to date—direct observation of moiré charge localization in twisted SrTiO₃ bilayers fabricated by PLD [54]. Using depth-resolved scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS), researchers visualized 2D spatial charge modulation within moiré supercells. This charge modulation arises from local variations in atomic coordination at different coincidence site lattice (CSL) points within the moiré pattern (see figure 5).

Current and future challenges

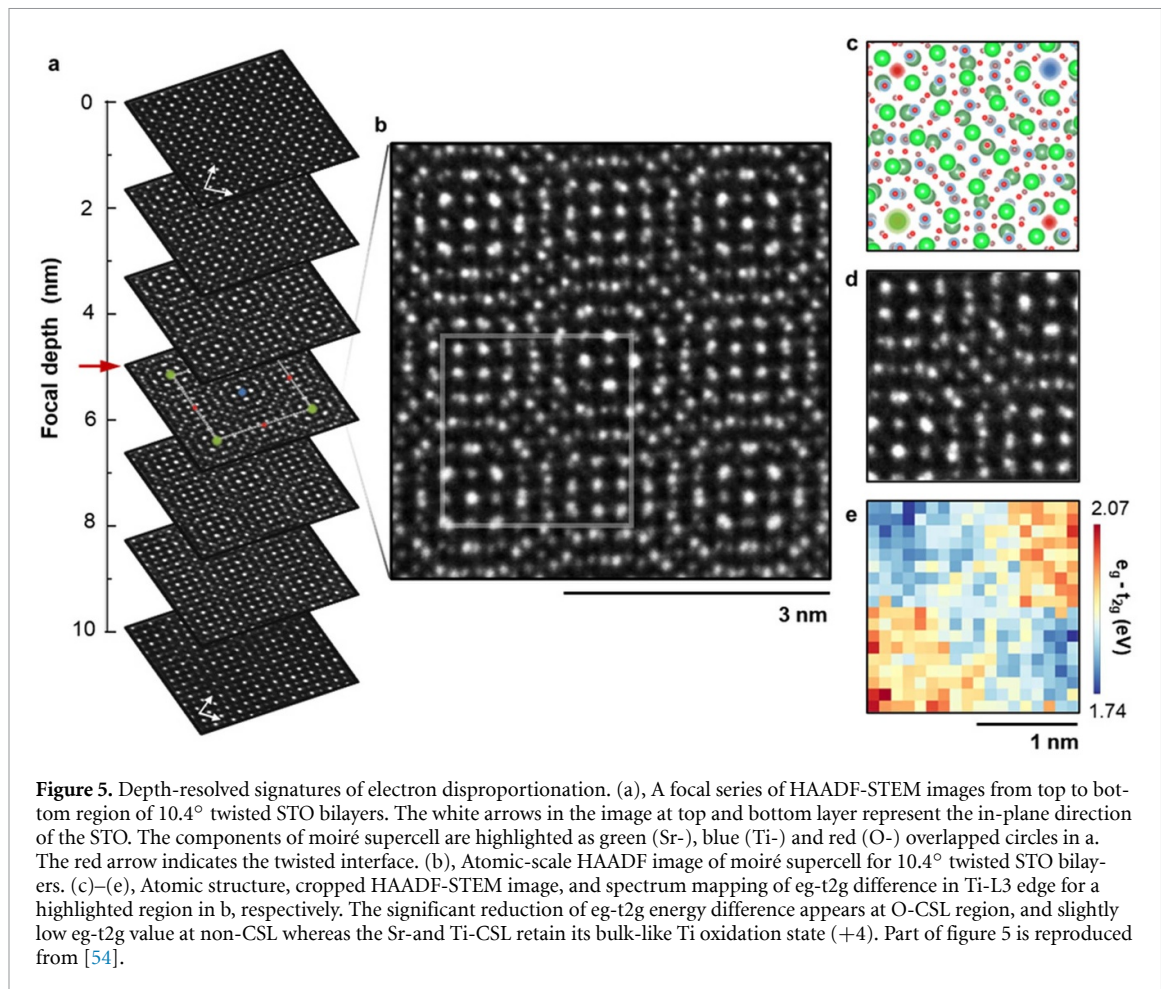
FOMs are positioned to mitigate the inherent sensitivity of modern quantum materials to point defects [70]. This sensitivity demands new synthesis approaches capable of producing single crystal thin film materials and heterostructures clean enough to probe fundamental quantum phenomena [71, 72]. In complex oxide thin films, reducing point defects remains a major challenge, primarily due to their high density arising from the non-stoichiometry of multicomponent systems. Achieving precise stoichiometry and controlling point defect concentrations is particularly difficult in PLD when using a single compound target, which limits independent control over individual elemental fluxes.

Freestanding membranes offer a promising alternative by enabling the heterogeneous integration of complex oxides onto virtually any host platform without sacrificing their intrinsic functionality. Recent advances have demonstrated the fabrication of freestanding LaAlO₃/SrTiO₃ (LAO/STO) membranes that retain key functional properties, paving the way for integration with hybrid electronic systems [65]—especially those requiring reprogrammable control of charge and spin states at the nanoscale.

However, two critical challenges must be addressed to fully realize the potential of freestanding membranes:

1. Precise alignment and transfer of the membranes onto designated regions of the host substrate.
2. Flipping of the heterostructure stack during integration.

The ability to flip oxide heterostructures is particularly valuable, as it enables engineering of terminations and interfacial properties that are otherwise inaccessible via bottom-up epitaxial growth. This is especially beneficial for backside processing and hybrid integration. Moreover, certain flipped configurations of complex oxide heterostructures are difficult or impossible to achieve using conventional thin-film deposition methods, due to kinetic limitations, such as cation interdiffusion or thermal expansion mismatch. Therefore, developing reliable methods for producing high-quality, transferable, flipped



complex oxide heterostructures presents significant opportunities for multifunctional integration in next-generation nanoelectronic devices.

The assembly of twisted oxide heterostructures further requires precise control of the twist angle, which is achieved using a rotational stage with 0.1° accuracy. One membrane is rotated to the target angle, while the second is positioned using a micromanipulator, allowing the creation of twisted bilayers with atomically sharp and clean interfaces.

Advances in science and technology to meet challenges

• Electronic-grade oxides via metal-organic PLD (MOPLD) and hybrid PLD

We have developed two novel synthesis methods to meet the challenges of oxide membranes and heterostructures. The first, MOPLD [73], is designed for precise control of point defects in complex oxide thin-film quantum membranes and heterostructures. These defects can significantly degrade or alter the fragile quantum phenomena these structures are intended to investigate. Using the $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) interface as a prototype 2D electron gas (2DEG) system, we observed a substantial enhancement in low-temperature electron mobility and clear Shubnikov–de Haas oscillations. The MOPLD process resulted in a dramatically reduced concentration of point defects in SrTiO_3 thin films, confirming the method's capability for producing electronic-grade oxide materials.

Hybrid PLD addresses a different challenge: the synthesis of quantum materials that involve elements with large vapor pressure mismatches—beyond the reach of conventional PLD techniques. Recently, we demonstrated high-quality epitaxial KTaO_3 (111) thin films using an adsorption-controlled hybrid PLD process, enabling the fabrication of superconducting heterostructures. Structural and electrical characterization revealed that the heterointerface between amorphous LaAlO_3 and KTaO_3 supports a 2DEG with significantly enhanced electron mobility, superconducting transition temperature, and critical current density compared to bulk single-crystal KTaO_3 -based systems [74]. This hybrid approach opens pathways to epitaxial growth of other alkali metal-based oxides previously considered inaccessible by conventional methods.

● Heterogeneous integration of flipped oxide heterostructure membranes

We demonstrated that the LAO/STO heterostructure, when fabricated as a freestanding membrane, can be flipped and precisely integrated onto silicon and other host platforms using polymer-free micromanipulation techniques [75]. The transferred membranes retain atomically smooth surfaces, high crystallinity, and their essential electronic properties.

Notably, ultra-low-voltage electron-beam lithography enables the writing of conductive nanostructures through the 44 nm insulating STO layer, targeting the now-exposed STO/LAO interface. These nanostructures function as programmable local gates, modulating charge carriers in the underlying silicon. In this configuration, the LAO layer serves as an ultrathin gate dielectric, while the reconfigurable interface acts as a nanoscale gate electrode.

This platform introduces a versatile strategy for integrating complex oxide heterostructures with semiconductors, quantum materials, and flexible substrates—paving the way for reprogrammable nanoelectronic device architectures.

Concluding remarks

Oxide moiré heterostructures represent a paradigm shift in quantum materials research, with new electronic, magnetic, and structural functionalities arising from the interplay of intrinsic oxide functionalities and engineered moiré periodicity. The demonstrated charge localization, predicted flat-band formation, and dynamic strain control capabilities firmly establish this field as a frontier for next-generation quantum technologies.

The atomic-level control enabled by PLD and *in situ* high-pressure RHEED has proven highly effective for fabricating high-quality freestanding membranes, including both single-layer and heterostructured epitaxial oxide thin films. Recent advancements in the fabrication and heterogeneous integration of $\text{LaAlO}_3/\text{SrTiO}_3$ flipped freestanding complex oxide heterostructures onto diverse host platforms open new pathways for reconfigurable nanoelectronic architectures.

Synthesis of electronic grade complex oxide thin films such as SrTiO_3 and KTaO_3 by recently developed MOPLD and Hybrid PLD allows us to produce oxide free standing membranes and interfaces clean enough to probe fundamental quantum phenomena.

The roadmap outlined here provides a framework for systematic progress—from fundamental understanding to practical device applications. Achieving this vision will require coordinated efforts in synthesis, characterization, theory, and device engineering, ultimately driving transformative advances in quantum computing, spintronics, and correlated electron systems.

Acknowledgements

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3.2. MBE synthesis of free-standing oxide membranes

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Status

MBE is a thin-film growth technique with decades of development spanning a wide range of material systems. Its application to complex oxide membranes, however, represents a more recent frontier [50]: over the last decade, oxide MBE has opened new research frontiers by enabling the heterogeneous integration of single crystalline oxides on state-of-the-art semiconductor platform (e.g. Si) and the realization of unit-cell-thick oxide membranes [10], allowing for the exploration of exotic functionalities (e.g. ferroelectricity [76–78], flexoelectricity [79], and piezoelectricity [80,81]) in correlated oxides; thus, these advances have further led to emerging device applications, such as ferroelectric field-effect transistors [82] and neuromorphic computing architectures [83]. These frontiers are enabled by the unique capabilities of FOMs, which facilitate the study of intrinsic material properties by decoupling functional materials from the growth substrate and offering a versatile platform for designing symmetry-mismatched, and artificially-engineered heterostructures [31, 35, 80].

MBE is a thin-film growth technique that employs molecular beams from high-purity elemental sources, often combined with reactive gases, to achieve atomically precise growth under ultra-high vacuum conditions (figure 6(a)). In conventional MBE, pure metals are sublimated or evaporated in inert crucibles to generate beam fluxes (corresponding vapor pressures in the range 10^{-3} – 10^{-6} Torr). Oxide MBE further supplies oxidizing species (e.g. radio frequency (RF) oxygen plasma, electron cyclotron resonance oxygen plasma, distilled ozone, or high-purity oxygen gas) to stabilize oxide phases. Growth proceeds by co-deposition or shuttered deposition at elevated temperatures, where thermodynamically self-regulated, adsorption-controlled growth may occur—allowing stoichiometric incorporation of species while excess flux desorbs and is pumped away, providing exceptional batch-to-batch reproducibility [84]. In other cases, careful flux and temperature control is required to achieve the desired stoichiometry. Furthermore, MBE has demonstrated uniform epitaxial oxide growth on substrates up to 4-in scale [85].

High-quality epitaxial heterostructures synthesized via MBE can be subsequently processed to form the freestanding membranes using a chemical-lift off (sacrificial layer) or 2D-material assisted lift-off (remote or vdW epitaxy) [84, 85]. The chemical-lift off approach has been widely used for *in-situ* growth of epitaxial heterostructures with atomic-level precision, where a sacrificial layer is inserted between the functional film and the growth substrate. The sacrificial layer can then be selectively etched in solvents, allowing the freestanding functional membrane to be released from the growth template. This approach offers several advantages, including lattice parameter tunability of the sacrificial layer to match the functional thin films, fast and selective solubility in water or mild acids, and reusability of the growth substrate. Various sacrificial layers have been employed such as (Ca, Sr, Ba)₃Al₂O₆, (Ca, Sr, Ba)₄Al₂O₇, (Ca, Sr, Ba)O, La_{0.7}Sr_{0.3}MnO₃, SrVO₃, SrRuO₃, YBa₂Cu₃O₇, and SrCoO_{2.5}, providing a wide range of tunable lattice parameters (2.98–5.12 Å) (figure 7). Most recently, BaAl₂O₄ sacrificial layer has been introduced offering 6- or 3-fold structural symmetry, extending the membrane platform beyond perovskite oxides to hexagonal oxide systems [86]. Among them, the binary oxide sacrificial layers based on (Ca, Sr, Ba)O are particularly favorable for MBE growth owing to their simple stoichiometry and the ease of synthesis.

Current and future challenges

Recent progress in oxide membrane research has opened new opportunities for heterostructure engineering, yet several fundamental challenges remain. In the sacrificial-layer approach, a central limitation is the absence of a universal sacrificial material that can form structurally and chemically compatible interfaces with diverse functional oxides. A lattice mismatch between the sacrificial and functional layers often induces extended defects during epitaxial growth, which later manifest as catastrophic cracks upon membrane release. Even when lattice matching is achieved, high-temperature growth can drive interdiffusion across the interface, producing compositional gradients that degrade structural and chemical uniformity. As indicated by the limited number of MBE-based reports (figure 6(b)), complex oxide sacrificial layers are particularly difficult to synthesize by MBE. This difficulty stems from the presence of ‘B-site’ elements (e.g. Ru, V, Ti, Co, Al, Mn), which require high temperatures to generate sufficient

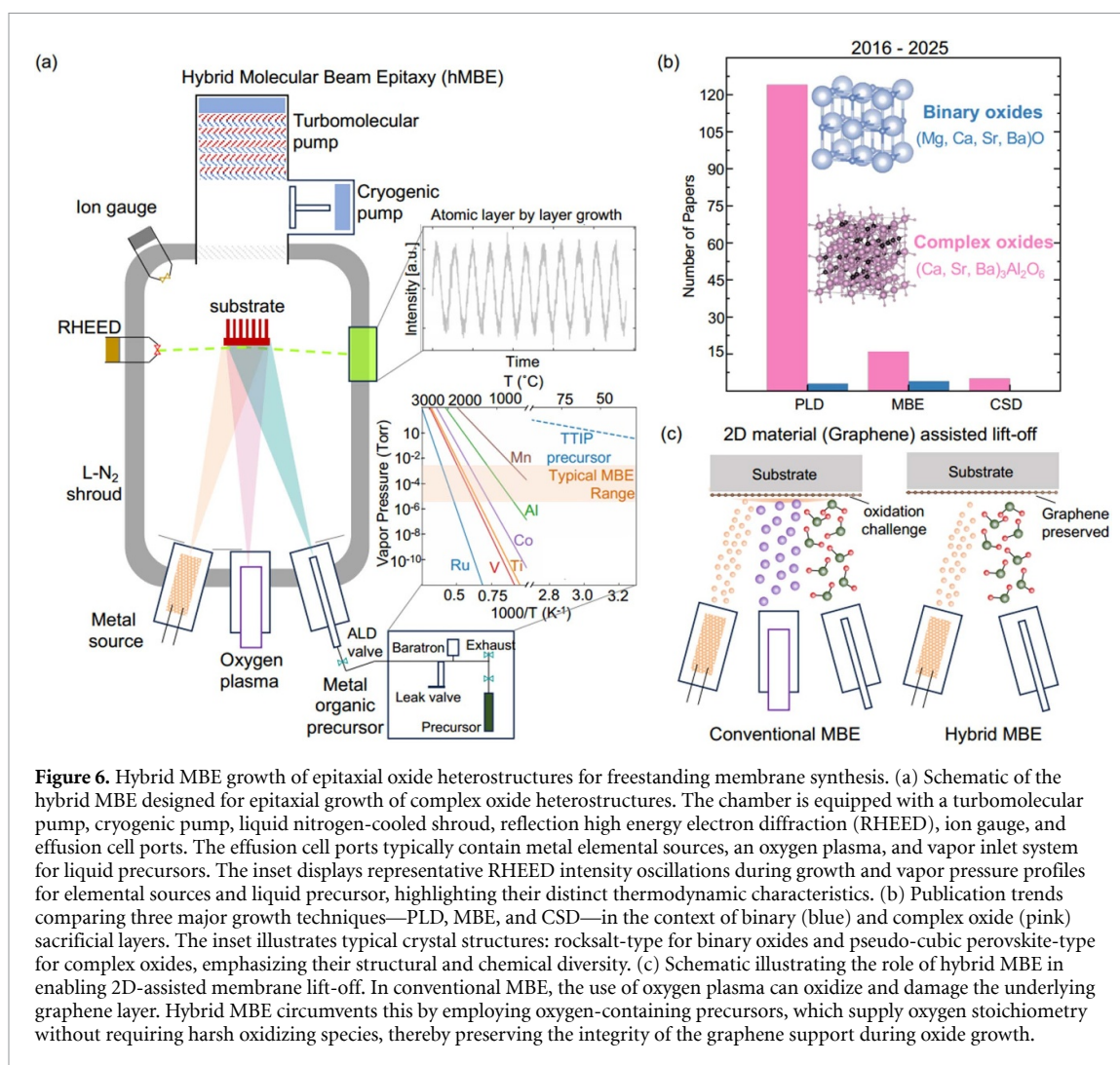
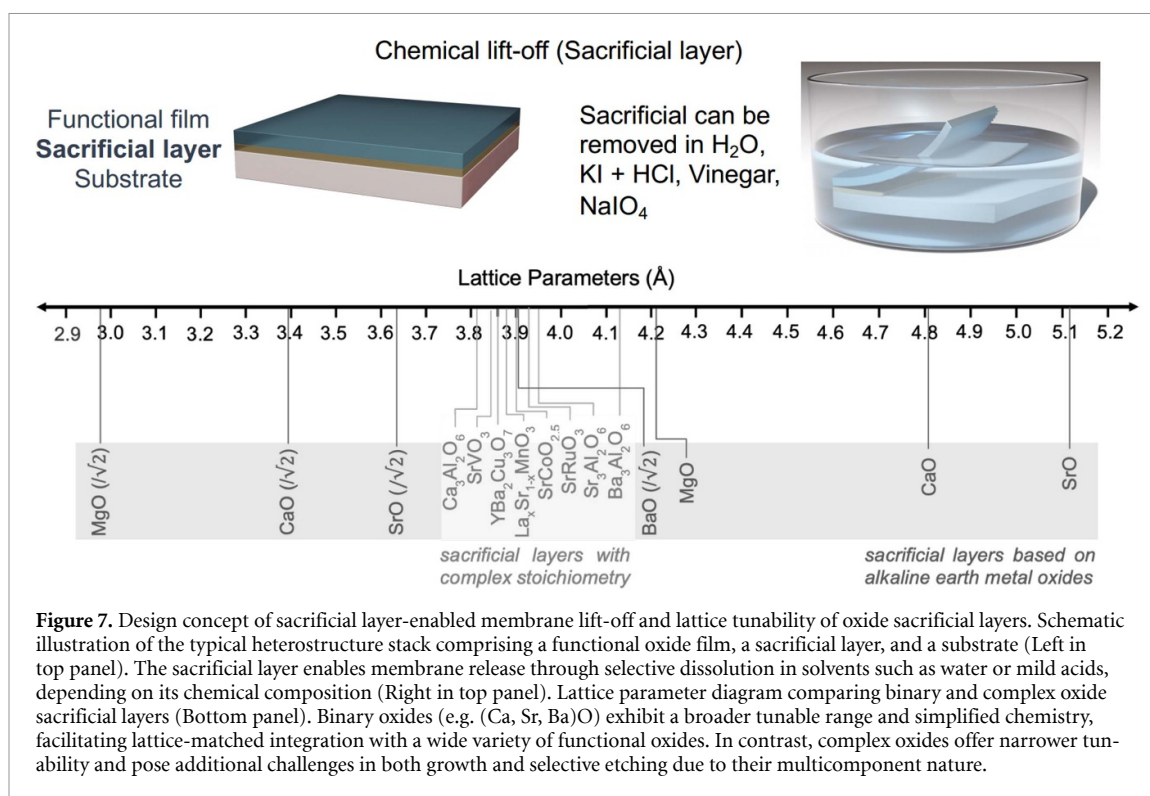


Figure 6. Hybrid MBE growth of epitaxial oxide heterostructures for freestanding membrane synthesis. (a) Schematic of the hybrid MBE designed for epitaxial growth of complex oxide heterostructures. The chamber is equipped with a turbomolecular pump, cryogenic pump, liquid nitrogen-cooled shroud, reflection high energy electron diffraction (RHEED), ion gauge, and effusion cell ports. The effusion cell ports typically contain metal elemental sources, an oxygen plasma, and vapor inlet system for liquid precursors. The inset displays representative RHEED intensity oscillations during growth and vapor pressure profiles for elemental sources and liquid precursor, highlighting their distinct thermodynamic characteristics. (b) Publication trends comparing three major growth techniques—PLD, MBE, and CSD—in the context of binary (blue) and complex oxide (pink) sacrificial layers. The inset illustrates typical crystal structures: rocksalt-type for binary oxides and pseudo-cubic perovskite-type for complex oxides, emphasizing their structural and chemical diversity. (c) Schematic illustrating the role of hybrid MBE in enabling 2D-assisted membrane lift-off. In conventional MBE, the use of oxygen plasma can oxidize and damage the underlying graphene layer. Hybrid MBE circumvents this by employing oxygen-containing precursors, which supply oxygen stoichiometry without requiring harsh oxidizing species, thereby preserving the integrity of the graphene support during oxide growth.

vapor pressure (inset, figure 6(a)) and often show limited oxidation under standard MBE growth conditions. While electron-beam evaporators for refractory elements or distilled ozone for enhanced oxidation are often used, these approaches remain hazardous and costly, as local heating in an oxygen environment can cause flux instability, and the use of ozone can potentially damage hot filaments and may oxidize source materials. Moreover, controlling such complex stoichiometry only to sacrifice the layer after growth is inefficient and impractical.

Post-transfer processing introduces additional challenges, particularly in achieving atomically smooth, well-terminated interfaces between the released membrane and the host substrate. Residues from the release or transfer process often require oxygen annealing or solvent treatments for effective removal. More fundamentally, transfer outcomes are governed in large part by the structural compatibility between the functional film and the sacrificial layer. As discussed above, the difficulty of synthesizing complex sacrificial layers by MBE significantly restricts the available combinations of functional film and sacrificial layer accessible in MBE. This constraint limits the ability to achieve optimal lattice matching, and the resulting mismatch directly manifests as geometric defects (e.g. wrinkles, cracks, and tears) upon release, making large-area, uniform transfer technically demanding and directly degrading overall membrane yield. Thus, in addition to reliably create, on demand, flat and smooth single-crystalline, atomically-precise membranes of the desired oxide with controlled thickness, the key challenges also include developing methods to tailored doping, and defined surface terminations; achieving precise stacking of such membranes; performing nanoscale characterization of interfacial structures and strain profiles across the bicrystal thickness; probing both local and extended properties; and advancing predictive modeling that links structure to properties. Among these, developing MBE-compatible sacrificial layers with broad lattice tunability is the most pressing priorities for the reliable integration of oxide membranes into next-generation devices.



Advances in science and technology to meet challenges

To circumvent these issues, hybrid MBE has been adopted, where ex-situ metal-organic precursors are introduced via vapor inlet systems. These precursors typically require only moderate heating ($\sim 100\text{ }^\circ\text{C}$ – $200\text{ }^\circ\text{C}$) to achieve sufficient vapor pressure (inset, figure 6(a)). A stable precursor flow is maintained using a Baratron controller linked to a linear leak valve, enabling atomic layer-by-layer growth within an adsorption-controlled growth window [84]. For solid metal-organic precursor, an effusion cell is used [12].

The challenge of complex sacrificial layer stoichiometry has recently been addressed through the development of binary oxide sacrificial layers (figure 7). When used in combination with complex layers, they offer broader lattice tunability and greater thermodynamic accessibility. In particular, alkaline earth metals can be readily sublimated at moderate temperatures ($300\text{ }^\circ\text{C}$ – $500\text{ }^\circ\text{C}$) and easily oxidized to form the desired oxide phases [86]. Their simple chemistry further allows straightforward alloying—for example, through (Ca, Sr, Ba) substitution to continuously tune the lattice parameter across a wide range, enabling closer matching to diverse functional oxides without requiring complex stoichiometric control. Furthermore, the hygroscopic nature of these binary oxides facilitates rapid and selective dissolution, minimizing membrane deterioration during release and transfer, while maintaining compatibility with other growth techniques such as PLD using commercially available targets [87, 88]. In parallel, complex sacrificial layers have also been integrated into MBE workflows by ex-situ deposition via chemical solution deposition (CSD), followed by recrystallization for epitaxial growth [89].

An alternative strategy leverages 2D materials as release layers. Graphene, with its weak out-of-plane bonding, enables membrane lift-off but is highly vulnerable to oxidation during oxide deposition (figure 6(c)). Recent work has demonstrated hybrid MBE growth of SrTiO_3 on graphene without supplying additional oxygen, relying instead on oxygen carried by metal-organic precursors [90]. This ‘softer’ approach achieves stoichiometric oxide films while preserving graphene integrity, establishing a viable pathway for scalable 2D-assisted oxide membrane fabrication [90].

Beyond single-layer membranes, emerging efforts on bilayer and stacked heterostructures highlight the need for precise control of interfacial reconstruction [91]. In 3D/3D epitaxial stacks, strain is locally confined to the interface, where atomic registry and lattice mismatch impose strong geometric and energetic constraints [92, 93]. These confined regions act as well-defined environments that drive spontaneous atomic rearrangements into energetically favorable configurations, which can in turn alter the functional properties of the entire stack [50]. Harnessing such controlled interfacial reconstruction offers a powerful strategy to engineer emergent phenomena in membrane-based oxide heterostructures.

Concluding remarks

The sensitivity of oxide membranes to interfacial structure highlights the need for growth techniques with atomic-level precision and chemical sharpness—conditions uniquely fulfilled by oxide MBE. Its low-energy, adsorption-controlled environment enables the creation of abrupt, epitaxial, single-crystalline interfaces even between chemically dissimilar layers, while minimizing intermixing and providing deterministic control over termination, reconstruction, and bonding. These capabilities establish MBE-grown freestanding membranes as a powerful platform to probe and exploit interfacial reconstructions in the absence of substrate clamping. Looking forward, such membranes offer unprecedented opportunities to design lateral heterointerfaces, engineer symmetry-breaking environments, design artificial grain boundaries, and program spatially heterogeneous strain fields—paving the way for emergent functionalities in next-generation oxide heterostructures.

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3.3. Advances and prospects of FOMs *via* sputtering deposition techniques

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Status

RF magnetron sputtering is a well-established physical vapor deposition (PVD) technique that has been successfully used for over three decades to grow high-quality epitaxial thin films and heterostructures of complex oxides. Among the various deposition techniques explored for free-standing oxide membranes, such as PLD or MBE, sputtering offers a scalable and cost-effective platform for the precise deposition of functional oxide materials, which makes it particularly well positioned to bridge the gap between laboratory demonstrations and industrial-scale manufacturing owing to its established use in large-area thin-film processing, high throughput, and compatibility with semiconductor production lines. For readers interested in the development and achievements of sputtering for epitaxial oxide growth, we refer to the comprehensive review by Dawber [94].

Despite its demonstrated capability for the growth of complex oxide films, sputtering remains remarkably underexplored in the context of FOMs, a field where most progress to date has relied on PLD or MBE [(see sections 3.1 and 3.2 in this roadmap). In table 1, we compile the limited number of reports employing sputtering for the fabrication of freestanding oxide films. Notably, there are only a few studies combining sputtering film growth with water-soluble sacrificial layers based in $\text{Sr}_3\text{Al}_2\text{O}_6$ and related compounds—a strategy that has proven effective and clean for releasing ultrathin membranes from their substrates.

This lack of exploration represents a missed opportunity. Given sputtering's advantages in scalability, process compatibility, and material versatility, advancing sputtering-based approaches for FOMs is essential to unlock their full potential in real-world applications. Further development in this direction could significantly enhance the prospects of transitioning freestanding oxide technologies from laboratory demonstrations to industrial-scale production, opening pathways for integration into flexible electronics and beyond-CMOS devices [109]

Current and future challenges

Efforts to produce FOMs using sputtering deposition have followed two main methodologies:

1-Epitaxial lift-off. This is the most widely used approach, where the film is separated from the substrate through selective chemical etching (figure 8(a)). Over the years an increasing number of groups have been using this approach. Interestingly, some of the first demonstrations were done in thin films deposited by rf-sputtering, by the group of C.B.Eom in the late nineties [99]. The approach was however not very practical, since it involved etching the SrTiO_3 substrate in a strong acidic solution to release a film of SrRuO_3 (a chemically very stable oxide), a process that very few oxides could withstand.

A more practical strategy involves inserting a chemically etchable sacrificial layer between the substrate and the functional oxide. Sputter-grown $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ and ZnO , both etchable in diluted HCl , have enabled freestanding membranes of ferroelectric (BaTiO_3 , $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$) and correlated oxide (VO_2) films [7, 99, 100]. More recently, following seminal work by H.Y. Hwang's group in 2016 [63], the field has largely shifted toward water-soluble $\text{A}_3\text{Al}_2\text{O}_6$ ($\text{A}=\text{Ba}, \text{Sr}, \text{Ca}$) compounds as sacrificial layers. These offer excellent epitaxial compatibility and allow clean lift-off in water without compromising the functional film—except in specific water-sensitive materials.

Despite the increasing use of $\text{A}_3\text{Al}_2\text{O}_6$ in freestanding membrane fabrication, there are only a few reports of high quality epitaxial $\text{Sr}_3\text{Al}_2\text{O}_6$ growth via off-axis RF sputtering [104–107]. Another adopted strategy is to deposit sputtered films on top of $\text{Sr}_3\text{Al}_2\text{O}_6$ templates grown by PLD or MBE [101–103]. While this hybrid approach works, it undermines the scalability advantage of sputtering if the sacrificial layer requires non-scalable methods.

2-Atomic Lift-Off. This dry, chemical-free alternative is inspired by exfoliation techniques used for vdW materials. While direct exfoliation is generally not feasible for epitaxial oxides due to strong interfacial bonding, J. Kim and coworkers have shown that stress-induced exfoliation is possible when a stressor layer forms a stronger bond with the oxide film than the film does with the substrate (figure 8(b))

Table 1. Reference works using sputtering technique to grow films for freestanding membrane fabrication, etching a substrate, etching a sacrificial layer with acidic solutions, etching a water-soluble sacrificial layer deposited ex-situ or *in-situ* by only sputtering steps, and via mechanical exfoliation.

Etching a substrate	BiFeO ₃	Si (substrate)	dry etching	[95]
	Pb(Mg _{1/3} Nb _{2/3})O ₃ –PbTiO ₃	Si (substrate)	HF + HNO ₃	[96]
	SrRuO ₃	SrTiO ₃ (substrate)	HF + HNO ₃	[97, 98]
Etching a sacrificial layer with acidic solutions	PZT	ZnO	Acetic acid	[99]
	VO ₂	ZnO	HCl	[100]
	BaTiO ₃	La _{0.7} Sr _{0.3} MnO ₃	HCl + KI	[7]
Etching a water-soluble sacrificial layer deposited ex-situ	Pb(Mg _{1/3} Nb _{2/3})O ₃ –PbTiO ₃	Sr ₃ Al ₂ O ₆ (PLD-grown)	Water	[101]
	PbTiO ₃ /SrTiO ₃	Sr ₃ Al ₂ O ₆ (PLD-grown)	Water	[102]
	PbTiO ₃	Sr ₃ Al ₂ O ₆ (PLD-grown)	Water	[103]
<i>In-situ</i> by only sputtering steps	GBCO	Sr ₃ Al ₂ O ₆ (<i>in-situ</i>)	Water	[104]
	PbZr _{0.2} Ti _{0.8} O ₃ & PbZr _{0.2} Ti _{0.8} O ₃ /La _{0.67} Sr _{0.33} MnO	Sr ₃ Al ₂ O ₆ (<i>in-situ</i>)	Water	[105, 106]
	NiCo ₂ O ₄	Sr ₃ Al ₂ O ₆ (<i>in-situ</i>)	Water	[107]
	NdNiO ₃ /SrTiO ₃	Ca ₃ Al ₂ O ₆ (<i>in-situ</i>)	Water	our work
Via mechanical exfoliation	Pb(Mg _{1/3} Nb _{2/3})O ₃ –PbTiO ₃	none	Mechanical release	[108]

[108]. So far, this method has only been demonstrated for Pb-based oxides (PbTiO₃, Pb(Mg_{1/3}Nb_{2/3})O₃–PbTiO₃) grown by sputtering, but it offers a promising route to produce freestanding membranes without sacrificial layers or wet etching.

Advances in science and technology to meet challenges

Achieving high-quality oxide films via sputtering is key to making FOM technology scalable and industrially viable. As outlined above, a major challenge is the realization of water-soluble sacrificial layers grown entirely by sputtering for use in epitaxial lift-off.

The group from Prof. X. Hong in Nebraska, in several papers, has successfully demonstrated the growth of high quality sacrificial Sr₃Al₂O₆ by using off-axis RF sputtering in a highly oxidizing 10 mTorr atmosphere Ar:O₂ (1:8) and temperatures from 650 °C–700 °C on different substrates. Free-standing epitaxial membranes of ferroelectric PbZr_{0.2}Ti_{0.8}O₃ (PZT) [105], as well as PZT/metal La_{0.67}Sr_{0.33}MnO₃ bilayers [106] have been fabricated after water dissolution and membrane transfer has been achieved on MoS₂ 2D semiconductor to demonstrate a reliable top-gated FET device. The same group has successfully applied the same full sputtering deposition approach to fabricate free-standing layers of ferrimagnetic NiCo₂O₄ spinel to explore its anisotropic magnetoresistance [107].

In the same direction, our group has taken steps in this direction by synthesizing Ca₃Al₂O₆ thin films via off-axis RF magnetron sputtering. We find that growth at 1 mTorr in an Ar/O₂ (8:2) atmosphere, with 28 W RF power (1.5-inch target) at 700 °C, yields films with good crystallinity and smooth surfaces (RMS roughness ~450 pm for a 20 nm film). These layers are stable for over 16 h in ambient conditions and fully dissolve in water within 4 h, as confirmed by the disappearance of the Ca₃Al₂O₆ (008) XRD peak and reappearance of substrate terraces in the topography (figure 8(a), middle panel), thus allowing to reuse the substrate for subsequent growths.

Importantly, we demonstrate that these sputter-grown Ca₃Al₂O₆ films function as effective sacrificial layers for releasing functional freestanding membranes, such as NdNiO₃. After 1–2 d in water, NdNiO₃ membranes can be successfully lifted off while retaining their characteristic metal–insulator transition (figure 8(a), bottom panel).

These results establish a clear pathway for the integration of water-soluble layers into sputtering-based workflows.

From a technological perspective, implementing lift-off processes entirely by sputtering could enable the production of wafer-scale, single-crystal oxide membranes, which are compatible with silicon transfer and wafer bonding techniques already used in the semiconductor industry [110, 111]. This would mark a crucial step toward the long-sought CMOS integration of complex oxides and their functionalities. However, it should be noted that the implementation of sputtering-based fabrication routes for free-standing oxide membranes is still at an early stage of development and is primarily focused on demonstrating feasibility and functionality.

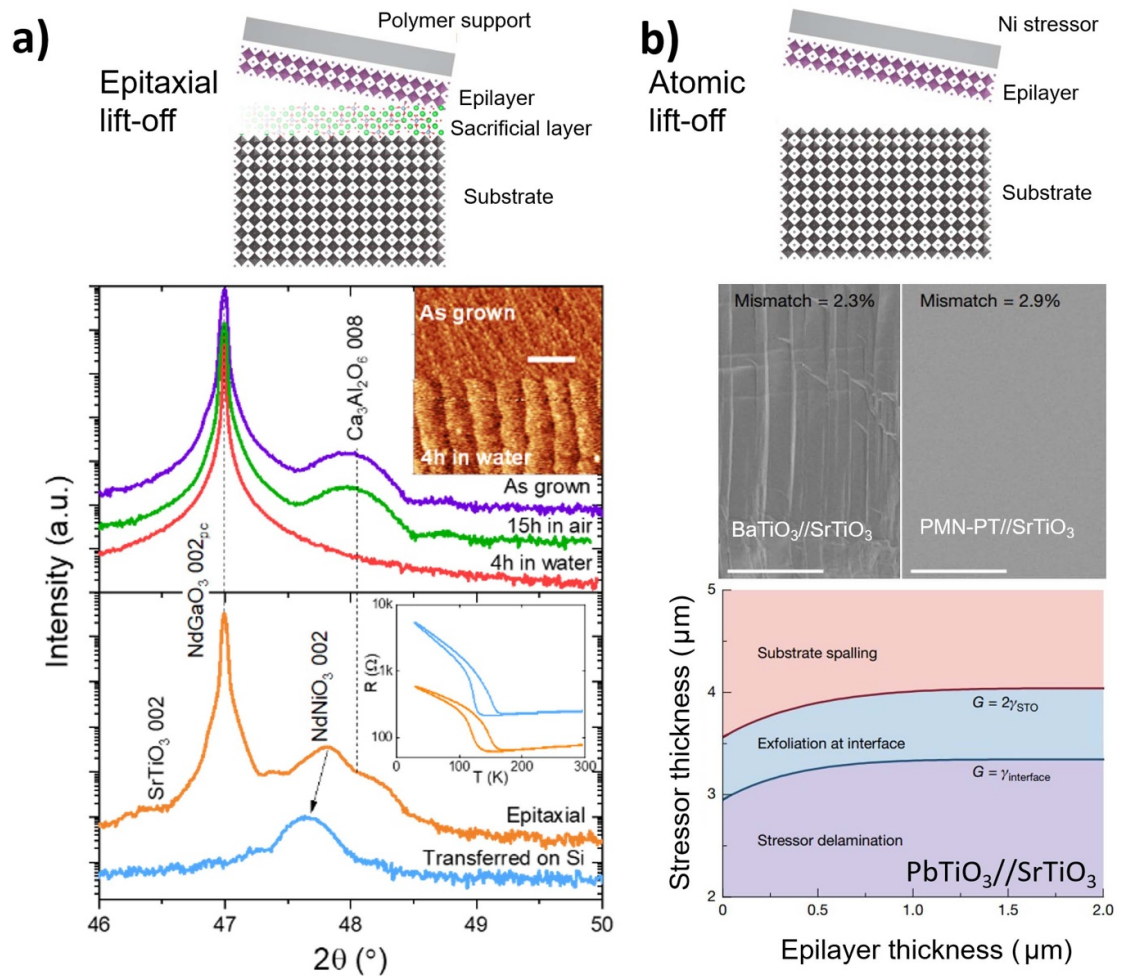


Figure 8. Freestanding oxide membranes prepared via RF-sputtering: (a) Top: schematic of epitaxial lift-off process; Middle: x-ray diffraction scans of $\text{Ca}_3\text{Al}_2\text{O}_6$ grown via RF-sputtering on NdGaO_3 substrates, as grown, after 15 h in air and after 4 h in water (film etched), inset shows film topography of as-grown film, and after 4 h in water, scale bar is 500 nm; Bottom: XRD scans of $\text{NdNiO}_3/\text{SrTiO}_3$ film grown on $\text{Ca}_3\text{Al}_2\text{O}_6/\text{NdGaO}_3$ via sputtering and $\text{NdNiO}_3/\text{SrTiO}_3$ film transferred to silicon after etching $\text{Ca}_3\text{Al}_2\text{O}_6$, inset shows temperature dependent resistance of NdNiO_3 film before (orange) and after transfer (blue). (b) Top: Schematic of exfoliation process; Middle: Plan-view scanning electron microscopy images showing exfoliated film surfaces of BaTiO_3 (left) - in which cracks propagated into the SrTiO_3 substrate leading to spalling of the epilayer together with part of the substrate - and $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ (right) - that is precisely exfoliated from the interface of the SrTiO_3 substrate, scale bar is 5 μm ; Bottom: Peeling modes for a $\text{PbTiO}_3/\text{SrTiO}_3$ heterostructure as a function of stressor thickness at the Ni stress level of 600 MPa. For Pb-containing films it is found that for certain stressor thickness range, interfacial energy can be lower than the surface energy of both films and substrates, leading to atomic lift off at the film/surface interface. (b) Reproduced from [108], with permission from Springer Nature.

Concluding remarks

FOMs offer a compelling platform for integrating complex oxides into flexible and CMOS-compatible technologies. To transition from laboratory-scale demonstrations to industrial relevance, it is essential to develop scalable deposition routes. Developing reliable protocols for growing water-soluble sacrificial layers entirely by sputtering remains a critical bottleneck in this process. Progress in this direction would enable fully sputtered lift-off architectures, compatible with large-area manufacturing and heterogeneous integration schemes. Related metrics such as fabrication yield, membrane cracking and wrinkling, wafer-scale thickness and property uniformity, batch-to-batch reproducibility, defect density, transfer yield, process throughput, and long-term mechanical and functional reliability have not yet been systematically evaluated in the literature. Addressing these aspects will be essential for assessing the scalability of the technology and its potential adoption in industrial environments. With its alleged scalability and materials versatility, sputtering is uniquely positioned to meet the processing demands of next-generation oxide electronics.

Acknowledgements

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3.4. Oxide freestanding membranes fabricated *via* metal-organic chemical vapor deposition (MOCVD)

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Status

Materials research on the deposition of thin oxide layers focuses primarily on PVD techniques, such as PLD and MBE. PLD and MBE operate under (ultra) high vacuum conditions and utilize a directed particle beam from the source to the substrates resulting in limited substrate sizes and low conformal coverage of patterned or non-planar surfaces. Low oxygen partial pressure results in the formation of oxygen vacancies. MBE offers the highest degree of layer-by-layer thickness control and the lowest defect densities for simple oxides, but suffers from low growth rates, and challenges in incorporating volatile multi-component species at the required fluxes. PLD provides good stoichiometry transfer for multicomponent targets and is currently the dominant technique for sacrificial-layer growth, but particulates and droplets often act as defect sources. Magnetron sputtering is a PVD method that enables thin-film deposition over large areas and is widely used in industry. However, it has certain drawbacks, such as a tendency for the deposited film's stoichiometry to deviate from that of the target—particularly with multi-component films—as well as the formation of oxygen vacancies and plasma-induced damage to the film surfaces caused by impact of high-energy particles.

In contrast, chemical vapor deposition (CVD) of thin oxide layers has been employed comparatively rarely. This discrepancy is particularly pronounced for FOMs: to the best of our knowledge, there are no reports in the scientific literature to date regarding CVD processes for such membranes. This lack is attributed to the inherent challenges of the CVD process for complex oxides and compatible sacrificial layers suitable for chemical lift-off—such as the thermal instability and low vapor pressure of many metal-organic (MO) precursors [112–114]. Combining MOCVD with alternative fabrication schemes for freestanding membranes, such as remote epitaxy and mechanical lift-off assisted by 2D materials, have so far been unsuccessful. The reason for this is the incompatibility between typical MOCVD-growth conditions for oxides with 2D materials such as graphene [29].

Nevertheless, MOCVD (or more specifically for epitaxial films: metal-organic vapor phase epitaxy—MOVPE) offers distinct advantages over PVD methods (like MBE and PLD) with respect to scalability, large-area uniformity, and industrial relevance [115]; it is a well-established technique in semiconductor industry. In contrast to the sputtering technique, MOCVD operates at relatively high pressure in a continuous gas flow. This enables uniform deposition over large-areas (up to 8 inches for semiconductor and up to 6 inches for oxide films) and three-dimensional substrate geometries.

Optimization of process parameters and careful tuning of MO precursor chemistry enable the fabrication of complex, multicomponent, high-quality crystalline oxide films on large areas with clean surfaces and minimal defect densities. MOCVD has been successfully applied to a broad range of perovskite and perovskite-like oxide films exhibiting diverse functionalities, including superconductivity, colossal magnetoresistance, high- and giant-k dielectric behavior, electrical conductivity, piezoelectricity, and ferroelectricity (see e.g. [114, 116, 117]).

The main features of MOCVD relevant to freestanding membrane fabrication are as follows: (i) High partial pressure of reactant gases during growth significantly mitigates the formation of oxygen and cation vacancies, especially for volatile elements such as K, Bi, and Pb. This enhances chemical stability and stoichiometric control. (ii) Near-equilibrium growth conditions help to avoid kinetically induced defects and enable the growth of epitaxial films with high crystalline quality, low defect density, smooth surfaces, and sharp interfaces. (iii) Excellent conformality: CVD techniques provide uniform film thickness and composition even for complex topographies. (iv) Industrial compatibility, since MOCVD does not require ultra-high vacuum conditions. It is readily scalable to large-area substrates, making it suitable for industrial applications. To minimize impact of the precursors on environment, strict regulations governing handling protocols, gas purification systems, and protective equipment in production facilities will ensure safe production.

Current and future challenges

Controlling layer thickness with high reproducibility at the unit-cell scale remains more challenging in MOCVD than in MBE, which inherently enables control over layer-by-layer growth. However, the use of *in-situ* characterization techniques (such as spectroscopic ellipsometry or reflectometry) allows for more precise control of layer thickness and surface quality. Reproducible sub-10 nm films with smooth surfaces have been demonstrated for several perovskite oxides so far. However, ensuring layer-thickness

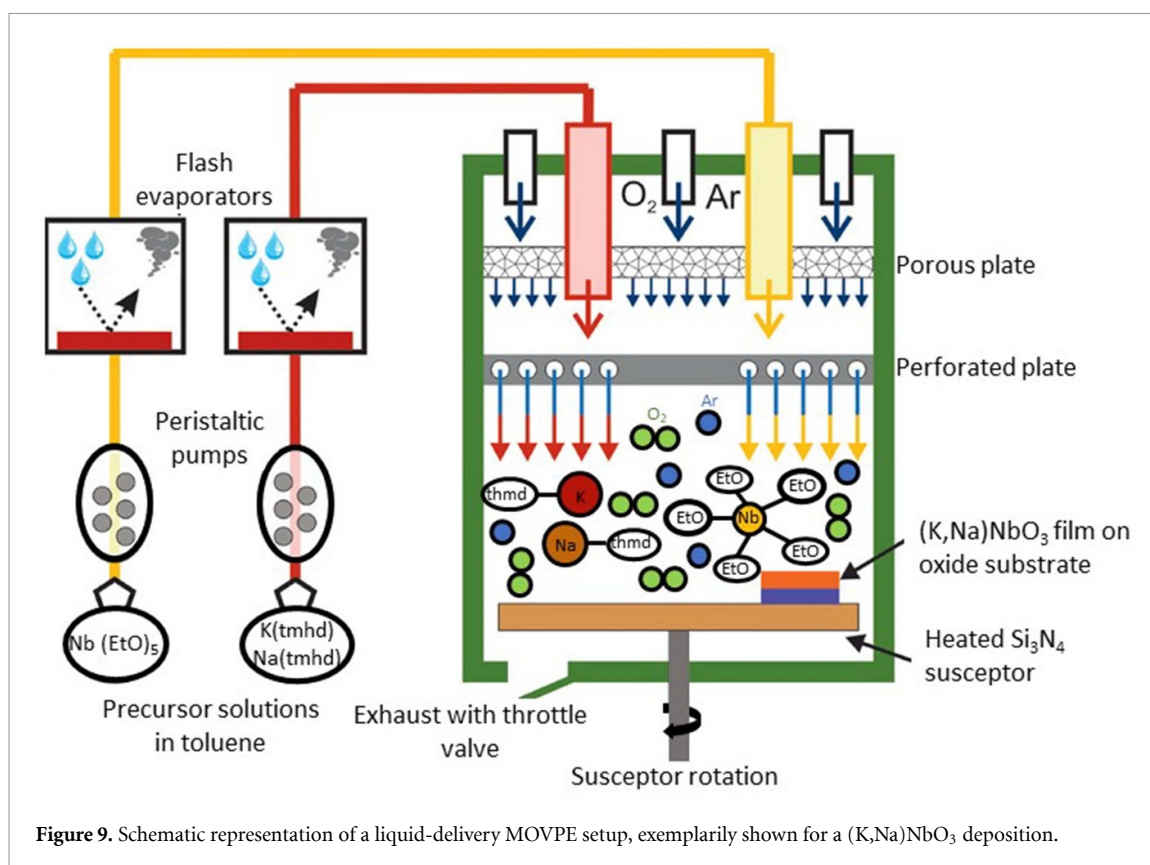


Figure 9. Schematic representation of a liquid-delivery MOVPE setup, exemplarily shown for a $(\text{K},\text{Na})\text{NbO}_3$ deposition.

reproducibility at the unit-cell level across process runs for multi-component systems remains an unresolved challenge.

A critical prerequisite for a robust and reproducible MOCVD process is the availability of high-purity MO precursors with sufficient vapor pressure and thermal stability. This requirement is generally met for gaseous precursors; however, for many complex oxides the MO precursors are liquid or solid materials with low volatility. To address this problem, a liquid-assisted MOCVD process was developed in which dissolved MO compounds are transferred into the gas phase by flash evaporation [113, 115, 117], the schematic setup of a liquid-delivery MOVPE is shown in figure 9.

Precursor chemistry remains one of the central challenges for MOCVD of multicomponent oxides. The development of new precursors aims to address the issue of precursor instability [112, 118]. Different precursors typically exhibit varying volatilities, evaporation rates, and decomposition temperatures. Achieving precise cation stoichiometry requires careful calibration of the individual precursor fluxes, identification of a common growth temperature window in which all precursors decompose efficiently, and the use of self-regulating growth regimes—whereby excess volatile species re-evaporate from the growing surface [112, 113, 119]. An alternative is the development of single-source precursors (e.g. $\text{Dy}(\text{dsml})_3\text{bipy}$ for DySixOy films) [113, 120]. In addition, incomplete decomposition of organic ligands can lead to residual carbon incorporation into the film, which may degrade crystallinity, increase leakage currents, and affect the electrical and optical properties relevant to membrane applications. Continued development of precursors with cleaner decomposition pathways is therefore an important direction for improving both stoichiometry control and film purity in multicomponent oxide MOCVD [113, 116].

Despite the advances in oxide MOCVD in last years, no reports are currently available on the application of MOCVD for the growth of sacrificial layers or the fabrication of FOMs. A major obstacle for the sacrificial-layer method is the lack of an established CVD process for the epitaxial growth of sacrificial layers, such as the widely used water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$. Even in combined processes, in which, for example, the sacrificial layer is deposited by PLD [50] and the functional oxide is subsequently grown by MOCVD, several challenges arise. This includes the degradation of the $\text{Sr}_3\text{Al}_2\text{O}_6$ surface upon exposure to ambient conditions during transfer through air, caused by its high moisture sensitivity. The near-equilibrium, high-pressure regime that favors good stoichiometry control in MOCVD, poses a problem for the sacrificial layer, especially $\text{Sr}_3\text{Al}_2\text{O}_6$ with its high surface reactivity. Since this compound can be regarded as a Sr- and O-deficient structure, it is inherently susceptible to structural and chemical changes under typical MOCVD conditions. Unintended reactions during MOCVD process, such as

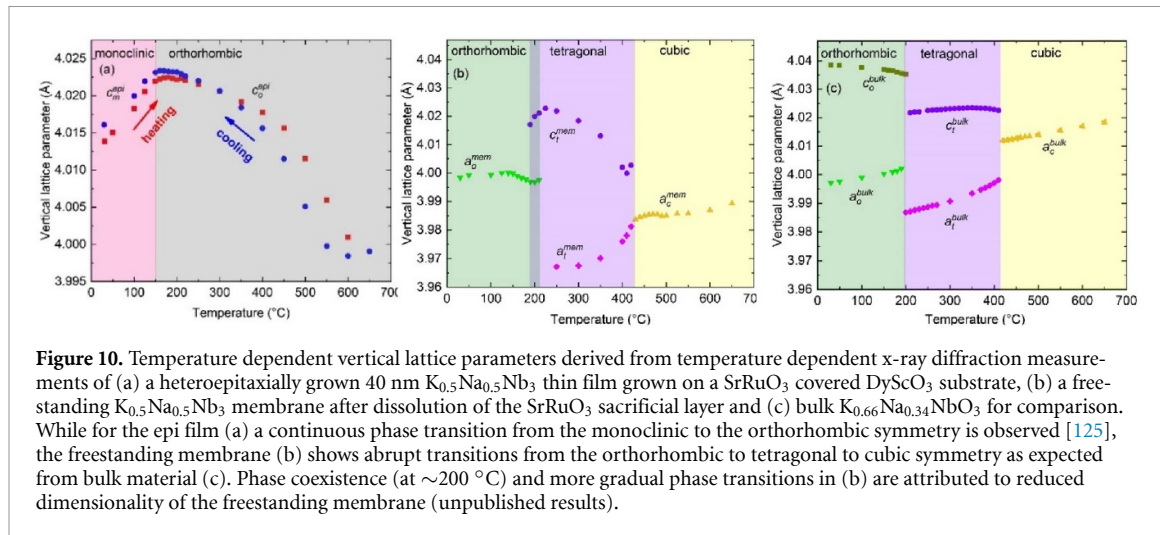


Figure 10. Temperature dependent vertical lattice parameters derived from temperature dependent x-ray diffraction measurements of (a) a heteroepitaxially grown 40 nm $\text{K}_{0.5}\text{Na}_{0.5}\text{Nb}_3$ thin film grown on a SrRuO_3 covered DyScO_3 substrate, (b) a freestanding $\text{K}_{0.5}\text{Na}_{0.5}\text{Nb}_3$ membrane after dissolution of the SrRuO_3 sacrificial layer and (c) bulk $\text{K}_{0.66}\text{Na}_{0.34}\text{NbO}_3$ for comparison. While for the epi film (a) a continuous phase transition from the monoclinic to the orthorhombic symmetry is observed [125], the freestanding membrane (b) shows abrupt transitions from the orthorhombic to tetragonal to cubic symmetry as expected from bulk material (c). Phase coexistence (at ~ 200 °C) and more gradual phase transitions in (b) are attributed to reduced dimensionality of the freestanding membrane (unpublished results).

hydrolysis or carbonate formation at the $\text{Sr}_3\text{Al}_2\text{O}_6$ interface, can either degrade its solubility (through the formation of SrCO_3 or other water-insoluble by-products) or attack the surface, thereby compromising the epitaxial template for the functional layer. High substrate temperatures enhance the problem, because interdiffusion of metal ions and oxygen across the $\text{Sr}_3\text{Al}_2\text{O}_6$ /film interface into the sacrificial layer is increased [121]. This alters the structural and chemical properties of the sacrificial layer, rendering it insoluble in water [122, 123]. Similarly, remote epitaxy and 2D-assisted mechanical lift-off techniques are currently incompatible with oxide MOCVD, as 2D materials such as graphene are thermally and chemically unstable in high-temperature, oxygen-rich environments. 2D materials are known to oxidize and etch away under O_2 exposure at temperatures of 600 °C–900 °C, which destroys the release mechanism and the clean vdW interface for the remote epitaxy [29, 34, 35, 124].

A potential pathway forward involves the identification and implementation of alternative sacrificial materials, such as SrRuO_3 , $(\text{La,Sr})\text{MnO}_3$, $(\text{Mg,Ca,Sr,Ba})\text{O}$, or VO_2 [118, 121, 123]. At IKZ, a notable advancement was achieved with the fabrication of $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ freestanding membranes by liquid-delivery spin MOVPE. The $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ films were grown at 730 °C at a gas pressure of 25 mbar with an $\text{Ar}:\text{O}_2$ ratio of 5. $\text{K}(\text{tmhd})$, $\text{Na}(\text{tmhd})$ (thd = 2, 2, 6,6-tetramethyl-3,5-heptanedione), and $\text{Nb}(\text{EtO})_5$ [$(\text{EtO})_5$ = pentaethoxide] dissolved in dry toluene were used as the metal-organic source materials. The $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ films were epitaxially grown on 20 nm SrRuO_3 serving as sacrificial layer deposited by PLD on a (110) DyScO_3 at a substrate at 750 °C, an oxygen partial pressure of 0.15 mbar, a KrF laser (248 nm) fluence and frequency of 50 mJ and 5 Hz, respectively, and a target-to-substrate distance of 55 mm. The SrRuO_3 layer was subsequently removed by selective etching in an aqueous NaIO_4 solution during 8 h. With the aid of a polymer stamp, $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ membranes with different thicknesses were transferred onto SiO_2/Si substrates. This proof-of-concept enables direct evaluation of the intrinsic properties of nanometer-scale ferroelectric films in their freestanding form, facilitating comparison with their bulk counterparts (e.g. in terms of phase transitions, see figure 10). It represents a first milestone toward the realization of MOCVD-based FOMs. The combination of different techniques, for example, PLD-grown sacrificial layers and MOCVD-grown functional films, may therefore represent a practical near-term strategy.

Advances in science and technology to meet challenges

MOCVD offers a unique approach for the fabrication of high-quality FOMs on large areas with high conformality and excellent compositional control. However, to enable the fabrication and integration MOCVD-grown FOMs in both basic research and industrial applications, several key challenges must be addressed in future:

Sacrificial layers compatible with MOCVD conditions, or directly grown with MOCVD: The goal is sacrificial layers with high structural quality and compatibility to the subsequent MOCVD deposition of functional layers without surface degradation, interfacial diffusion, or cross-contamination [35]. This is particularly challenging for $\text{Sr}_3\text{Al}_2\text{O}_6$ -based films, but alternative, non-water-soluble sacrificial layers (e.g. SrRuO_3) may enable hybrid approaches combining different growth methods for the sacrificial layer and the complex oxide film. Ultimately, a dedicated MOCVD-process for the sacrificial layer itself is necessary. The demand of chemical resistance against MOCVD conditions may result in harsher etching

conditions for the sacrificial layer [50]. This in turn may challenge compatibility of a suitable polymer carrier during the release process.

In-situ monitoring of the growth process: To effectively and reliably control the growth rate, the evolution of surface and interface roughness, and the uniformity of composition and thickness of both the functional film and the sacrificial layer during the growth process, an *in-situ* monitoring system is highly desirable. Optical techniques based on reflectance spectroscopy or spectroscopic ellipsometry can provide valuable insights into interface and phase formation during MOCVD. However, owing to the large number of elements involved, these techniques are often not sensitive enough for complex multicomponent oxides [119]. Development of monitoring approaches with sufficient elemental and compositional sensitivity remains an important goal for achieving reproducible unit-cell-level thickness control.

Development of optimized MO precursors [113, 116]: The next generation of MO precursors must be (i) soluble and chemically stable in compatible solvents without precipitate formation over extended periods; (ii) volatile at moderate evaporation temperatures (150 °C–250 °C), with a wide thermal window between evaporation and decomposition; and (iii) suitable for clean decomposition, producing the desired oxide phase with minimal carbon contamination. Precursor design is closely linked to the goal of lowering growth temperatures (e.g. by using e.g. alkylamide- and guanidinate-based precursors) [113], since processes at reduced temperatures would also broaden the range of compatible sacrificial layers and substrates. Large-area growth and defect scalability trade-off: While MOVPE is inherently scalable for oxide film growth over large substrates, growth conditions optimized for high throughput, high growth rates and large-area coverage may cause increased defect densities, including point defects, threading dislocations, and compositional inhomogeneity across the wafer. Such defects can compromise membrane integrity during the release and transfer steps, where mechanical stresses are highest. Balancing growth rate and area against defect density and membrane yield is therefore a key consideration that must be addressed explicitly when scaling MOCVD-based membrane fabrication toward larger substrate sizes. One step toward scalability would be to use STO/Si as a growth substrate instead of typical small-scale oxide substrates.

Concluding remarks

The versatility and scalability of MOCVD offer a compelling platform for tailoring structural, electrical, and optical properties of FOMs for integration into CMOS platforms and advanced device architectures. Key challenges for the field include the development of low-temperature growth processes for better compatibility with sacrificial layers and flexible substrates, advanced precursor chemistries with improved volatility and stoichiometry control as well as cleaner decomposition, robust *in-situ* monitoring methods for complex multicomponent oxides, and growth and transfer strategies that maintain low defect densities at large area. Overcoming these challenges through innovations in precursor design, interface engineering, and sacrificial layer processing is crucial for unlocking the full potential of MOCVD-based membrane fabrication, for example, for information technology (see chapter 6.5) or varactors (see chapter 6.4). Addressing these challenges requires intense collaboration and exchange between the growing oxide membrane community and the more applied MOCVD community, working together toward scalable growth and transfer techniques.

Acknowledgements

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3.5. FOMs fabricated by cost-efficient chemical methods, atomic layer deposition (ALD) and CSD

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Status

The development of freestanding single crystalline TMO membranes has revolutionized the field of oxide electronics offering new playgrounds to control and understand their properties, uncover emergent phenomena and facilitate integration with technological substrates [33, 80]. Together, these advancements may empower a real opportunity to apply single crystalline complex oxides in practical electronic and energy devices.

The synthesis process plays a pivotal role. The use of a sacrificial layer that serves as a template to transfer the epitaxy from the single crystal substrate to the functional oxide and be selectively removed, has been widely used to fabricate crystalline membranes [63]. Most investigations have focused on single-crystalline complex oxide membranes prepared by high vacuum thin film deposition methods, such as PLD, MBE and sputtering [50]. For example, membranes with a perovskite structure such as SrTiO_3 (STO) and BaTiO_3 (BTO) have attracted a great deal of attention showing exciting altered properties [126, 127]. Additionally, membranes with other crystalline structures, diverse compositions, superlattices and nanostructures have also been explored to further tune their physico-chemical properties [128]. However, when evaluating the suitability of a functional oxide material it is important to consider cost-effective and sustainable processing techniques that can offer complementary features.

Over the past few years, CSD and ALD have consolidated as attractive alternatives to produce crystalline thin film oxides with smooth surfaces. CSD is well known for being an ambient-pressure deposition technique that permits fine stoichiometry control in multicomponent materials. It enables the preparation of films from a few nm to micron-thick nanostructures and patterns, figure 11(a) [129, 130]. On the other hand, ALD is also a budget friendly technique that uses low temperature and low-vacuum conditions. The self-limiting surface reaction mechanism ensures high conformality and thickness control over large areas, especially in binary compounds. Additionally, epitaxy can be achieved at relatively low deposition temperatures compared to CSD and high vacuum deposition techniques, figure 11(b) [131, 132]. These characteristics make these techniques very appealing to be adopted for preparing singular single crystalline membranes. In fact, the first results have not taken long to appear. Sacrificial layers with composition $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_6$ (SCAO) have been successfully prepared by CSD providing a wide range of lattice parameters and tuned stability (H_2O , ambient humidity) which offers versatility in terms of membrane composition [133] but also on the possibility to combine ex-situ deposition techniques, figure 12(a). In fact, it has been demonstrated that (flexible) PLD- $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) and Hybrid-MBE STO single crystal membranes can be obtained from vacuum annealed CSD-SCAO sacrificial layers, figure 12(b) [91, 122]. Then, these architectures can be used as building blocks to prepare more elaborated architectures such as CSD- BiFeO_3 /PLD-LSMO crystalline bilayer membranes, figure 12(c) [134].

Typical membrane thicknesses spans from few tens of nanometers up to 150 nm. On the other hand, the first steps on combining ALD with as-deposit CSD- $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) demonstrated that bendable and polycrystalline membranes including ALD- CoFe_2O_4 (CFO) and ALD-BFO with thicknesses ranging from 10 to 60 nm can be delivered with no interface reaction, enabled by the gas-phase characteristics of the ALD technique, figure 12(d) [135].

The reproducibility and transfer yield of chemically deposited membranes prepared on water-soluble sacrificial layers are governed largely by the sacrificial-layer quality. Stringent control of precursor handling, solution deposition and sample storage under an inert atmosphere ensure good batch-to-batch reproducibility, achieving transfer yields exceeding 80% in $5\text{ mm} \times 5\text{ mm}$ membranes. Alternatively, a recently developed approach based on the electrodynamic etching of conductive sacrificial layers offers a compelling pathway toward 12 inch wafer scale membrane fabrication fully compatible with chemical deposition approaches, eliminating dependence on ambient exposure and markedly reducing the presence of morphological defects [136]. Note that the formation of morphological defects during the membrane release, including cracking and wrinkling, can be effectively controlled by engineering the system lattice mismatch, the thickness of the functional oxide, and the mechanical properties of both the oxide film and the host substrate [137]. Thus, the use of ambient or low-vacuum chemical-deposited techniques to prepare freestanding membranes has vast potential to be exploited.

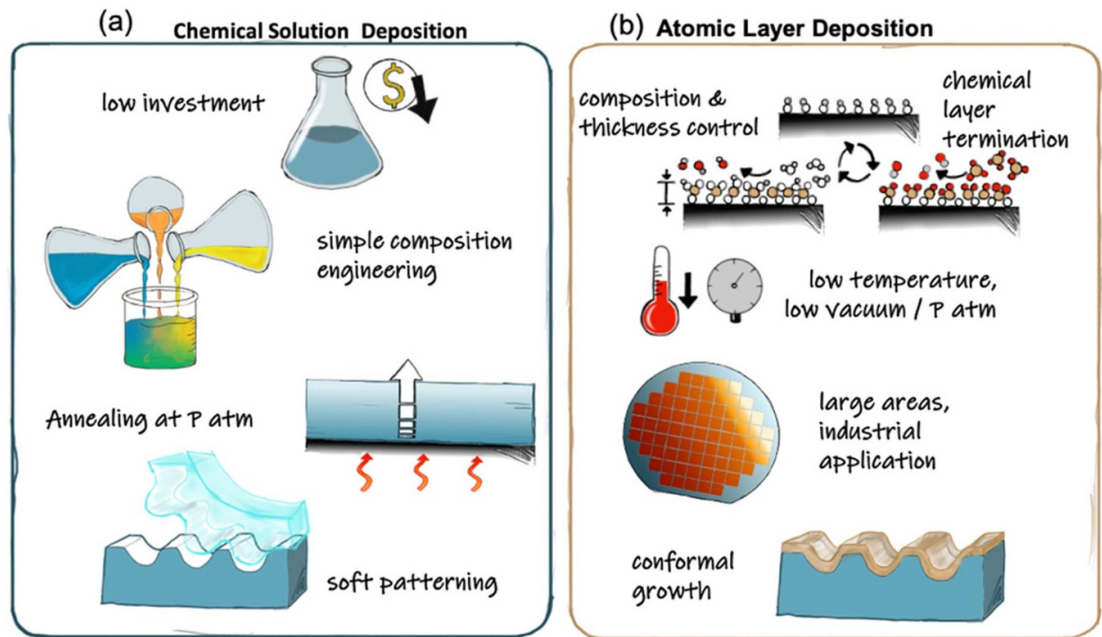


Figure 11. Snapshot of the characteristic features of chemical solution deposition and atomic layer deposition.

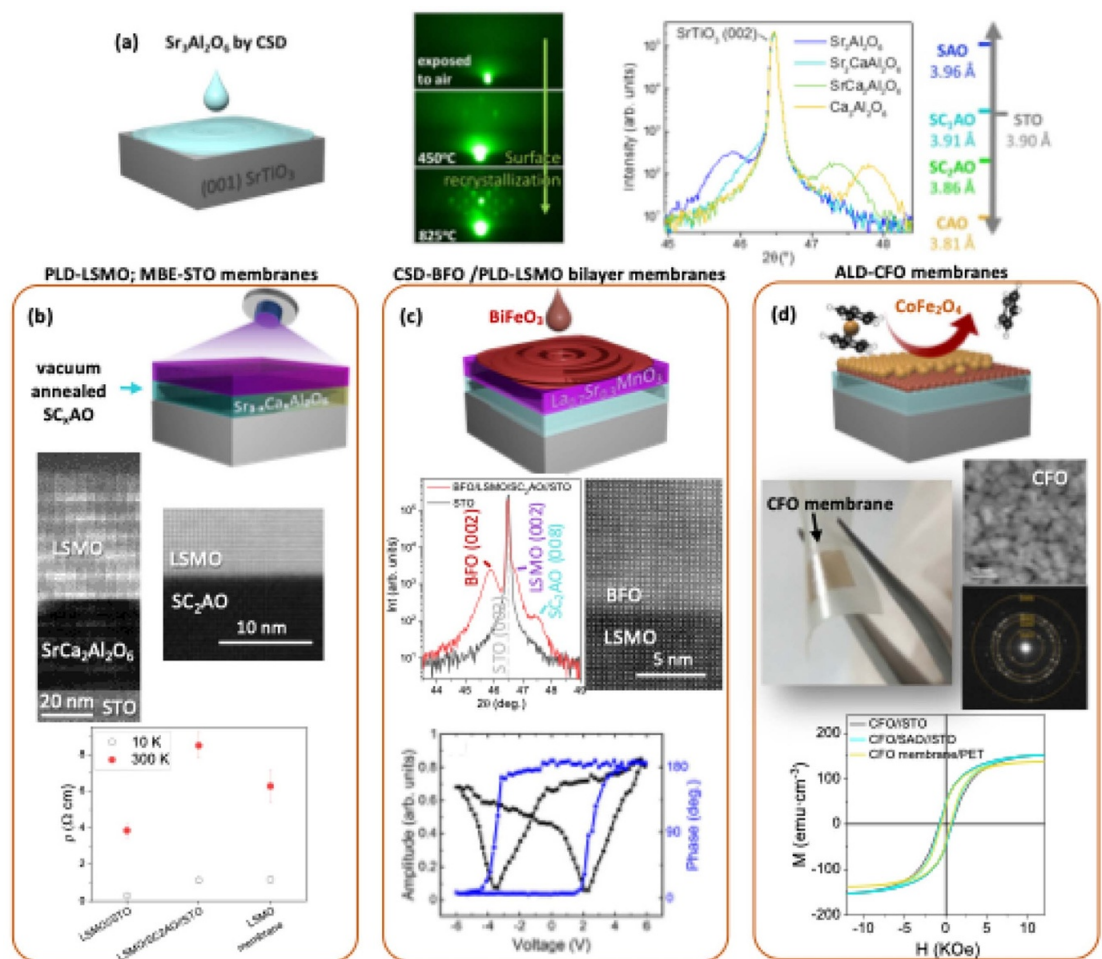


Figure 12. Summary of the transition metal oxide membranes prepared from (a) CSD-SCAO sacrificial layer which includes (b) PLD-LSMO, (c) CSD-BFO/PLD-LSMO bilayers and (d) ALD-CFO. Reproduced from [134]. CC BY 4.0. Reproduced from [135]. CC BY 4.0.

Current challenges

Nowadays, an all-CSD approach based on water-soluble sacrificial layers is not fully developed for single crystalline membranes. The chemistry of a specific oxide that makes it optimal to selectively react with a solvent and be used as sacrificial layer can also make it suitable to react with a precursor solution of the functional oxide and impede the obtention of a single crystalline membrane [134]. Judicious engineering of the precursor solution chemistry could help providing a more stable approach where the identification of compatible solvents, the use of chemical stabilizers and the control of the pH environment with acid, bases or buffering the solutions can be pivotal. Also, accurate control of the working atmosphere during the synthesis and storage will prevent undesired reactivity, degradation and surface contamination. *In-situ* monitoring of the conversion process from the deposited gel to the crystalline oxide will also help understanding the reaction mechanism to prevent interface reactivity and be able to release highly crystalline membranes. Solution deposition can introduce edge effects arising from the inhomogeneous solution accumulation during deposition. Consequently, the resulting membranes exhibit smaller uniform areas compared to those prepared by conventional vacuum-deposited techniques. Solution rheology, deposition and thermal conditions offers room for improvement. Then, the CSD-growth mechanism typically leads to rougher surfaces compared to high vacuum techniques which can influence the resulting membrane texture [122]. The development of ALD single crystalline complex oxide membranes is less advanced than the CSD-derived counterparts, and it is partially subjected to the identification of thermodynamically compatible metal-precursors and the appropriate selection of the co-reactant (H_2O , O_2 , O_3) [131], being specially relevant in water-soluble sacrificial layers. Additionally, in some scenarios, achieving epitaxy at the typically low deposition temperature of ALD may require long processing times, however, ex-situ post annealing and plasma-assisted deposition can overcome this issue [129]. It should be noted that the distinctive high conformality earned in the ALD deposits can impede a rapid edge-etching of the underlying sacrificial beneath the functional oxide. Masking the sacrificial prior the subsequent ALD or scratching the heterostructure before solvent immersion can solve this aspect. Finally, the number of reported ALD membranes are rather limited and in most of the cases are polycrystalline or amorphous, guided by the amorphous surface nature of the air-exposed SAO sacrificial [135]. The use of environmentally stable sacrificial layers (i.e. LSMO, CAO) and the electrodynamic etching approach are foreseen to help delivering single crystalline ALD- membranes.

Future opportunities

The consolidation of CSD and ALD as alternative deposition techniques to prepare oxide membranes is not only feasible but it opens up exciting opportunities that differentiate them from conventional vacuum-based techniques. Inspired by the research done in thin films, here we identify some of these potential scenarios.

First, they could enable the crafting of unique nanostructured membranes. The fact that the CSD process begins with a metal-organic solution it allows for the embedding of pre-designed nanocrystals. Following deposition, the film can undergo a thermal treatment to further enhance crystallization, if necessary [138]. Moreover, this method permits patterning of the deposited gels using cost-effective lithography techniques such as soft-lithography. Pre-defined nanostructures can be fabricated to guide the epitaxial growth [139]. Additionally, these pre-defined structures serve as templates for ALD-conformal growth, enabling precise nanometer-scale thickness control, figures 11(a) and (b) bottom panels.

Second, the ALD technique's capability for low-temperature, conformal deposition -even on corrugated or complex surfaces- can be transformative for integrating membranes with atomically smooth surfaces onto temperature-sensitive devices. In parallel, there are CSD approaches to lower the processing temperature using photosensitive precursors that decompose under light exposure rather than heat, thereby, reducing the processing temperature below 400 °C or using solution combustion synthesis, where a self-sustained reaction is initiated by a fuel incorporated into the solution, enabling more energy-efficient processing. Then, adopting artificial intelligence (AI) tools capable of accelerating the screening of compatible chemistries will be of great benefit for both techniques, alongside AI-driven interfaces for autonomous synthesis processes in ALD [140].

Finally, CSD and ALD present viable pathways for the scalable production of oxide membranes- an essential consideration for technological applications. Although membrane fabrication using CSD has so far primarily relied on spin-coating, other deposition techniques such as dip-coating and slot-die coating are readily adaptable for large-area roll-to-roll deposition although the level of precision and uniformity will depend on the material system.

On the other hand, ALD is becoming mainstream in many industries including micro-electro-mechanical systems, optics, photonics, image sensors and green energy. Emerging ALD variants such

as spatial ALD (s-ALD) enable higher throughput by spatially separating the precursors, rather than cycling them temporally, thereby, making continuous roll-to-roll processing feasible. Additionally, there are reactors that include multiple processing modules being extremely appealing for the fabrication of high-grade heterostructures with perfect interfaces which are very convenient to deliver single crystalline membranes with exceptional surface quality. It is important to mention that to scale up the fabrication of single crystalline complex oxides there are bottlenecks that need to be solved related to the material itself rather than the deposition techniques. For example, the limited availability of large-area single-crystal substrates required for the epitaxial growth of the sacrificial layers and the inherently slow etching process of the sacrificial which proceeds laterally from the edges. Addressing these limitations by developing faster etchants, further exploring electrodynamic etching or introducing patterned release paths will be crucial for enabling wafer-scale, and potentially roll-to-roll, production of freestanding oxides.

Concluding remarks

The development of single crystalline complex oxide membranes represents a transformative advancement in oxide electronics, enabling new opportunities for property control, heterointegration and study emergent phenomena. This work highlights the potential of combining CSD and ALD to complement or surpass conventional high-vacuum techniques.

CSD offers unique advantages such as low-cost processing and compatibility with nanostructure and patterning approaches, while ALD provides exceptional conformality, atomic-level thickness control, low-temperature and large-area processing. The development of CSD-derived membranes are in a more advanced stage where recent demonstrations have shown the feasibility of using CSD-S(C)AO sacrificial layers to enable the transfer of functional oxide membranes prepared via PLD, MBE, ALD and a combination of CSD/PLD, establishing the groundwork for hybrid architectures.

Nonetheless, some challenges remain in fully realizing the potential of these chemical routes. In CSD, issues related to chemical and thermodynamic compatibility between sacrificial and functional layers must be addressed through solution engineering, the use of buffer layers and *in-situ* monitoring. In ALD, the main constrain is probably the limited precursor availability to develop new compositions which requires further innovation from the synthetic inorganic chemists and the design and performance of AI agents. However, it represents a highly promising technique toward wafer-scale uniform fabrication of high-grade heterostructures.

Looking forward, the consolidation of CSD and ALD for membrane synthesis could open new arenas of research from a fundamental standpoint envisaging new applications in flexible electronics and energy devices. Overcoming current limitations—making the most of the chemistry control, precursor design, and etching strategies—will be pivotal in the manufacturing of complex oxide membranes.

Acknowledgements

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4. Fabrication and growth of thin films with 2D materials

4.1. 2D materials as interlayer for direct, vdW and remote epitaxy of functional oxides

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Status

Conventional heteroepitaxy (**direct epitaxy**) of oxide materials relies on strong chemical bonding between the film and substrate, which tightly clamps the film material to the substrate lattice. This mechanism can produce high-quality single-crystal films, but it also imposes a strict lattice matching requirement. An example of the complexity of direct epitaxy represents integration of functional oxides onto a silicon platform. The large lattice and thermal expansion mismatches cause most film materials to crack, and chemical reactions at the interface (e.g. silicate/silicide formation) can occur due to the lack of a diffusion barrier. Despite these challenges, the ability to combine such dissimilar materials remains highly desirable, driving the search for new epitaxial strategies.

VdW and remote epitaxy have recently emerged as key innovations to overcome the lattice-matching limitations of conventional epitaxy. Both methods introduce an ultra-thin **2D material interlayer** between the substrate and the film, avoiding the usual film–substrate direct chemical bonds. The film–substrate bonds are replaced by either chemical bonds between film and the 2D-layer or by much weaker vdW interactions. This decoupling has two significant benefits:

- (1) It decreases the lattice constraint, allowing single-crystal films to grow with minimal strain even on highly mismatched or amorphous substrates. In essence, vdW and remote epitaxy effectively ‘de-clamp’ the epitaxial layer from the substrate by eliminating strong interfacial bonds. The ability to produce high-quality, strain-free films of materials that were previously incompatible opens up a range of novel applications and at the same time offers ‘universality’ in materials choice.
- (2) The as-prepared epilayer can be exfoliated from the substrate with relative ease, yielding a freestanding membrane and leaving the substrate intact for reuse [141]. This is possible because the epilayer is only loosely bound to the substrate through vdW forces, so it can be detached without damaging either the film or the substrate.

To distinguish the local true epitaxial growth on each individual oxide nanosheet flake to that of using graphene, where there is no true epitaxial relationship between the film material and each graphene flake, we introduce the term quasi-direct epitaxy in the context of using 2D materials as seed-layers. Schematics for all four types of epitaxis are represented in figure 13.

Current and future challenges

Across all three approaches—(quasi-)direct, vdW, and remote epitaxy—the overarching challenge is to surmount the traditional limits of lattice matching and interface compatibility, while maintaining material quality (see table 2). Direct epitaxy demands solutions for strain management and chemical compatibility; vdW epitaxy requires mastering growth on inert, unconstrained surfaces; and remote epitaxy calls for verification and control of a subtle long-range interaction.

With the focus on vdW and quasi-direct epitaxy we briefly review two types of 2D material interlayers, **Graphene And Oxide Nanosheets**.

The deoxidation and passivation of a silicon surface represent one of the most important steps in the successful integration of functional oxides with silicon. We have shown that application of Sr and SrO may result in oxide-free and atomically flat silicon surface [143, 144]. In the context of functional oxide-silicon integration with the help of 2D materials, this approach proved to be very useful, since it allowed oxides integration by entailing co-existence of direct and vdW epitaxy. The quality of the resulting films was comparable to contemporary integration approaches and with functional properties on the level of the most sophisticated growth methods [145]. This proved that discontinuity of the 2D material, i.e. its nanosheet form of finite lateral size, is not a limiting factor for integration of oxides with silicon (figure 14).

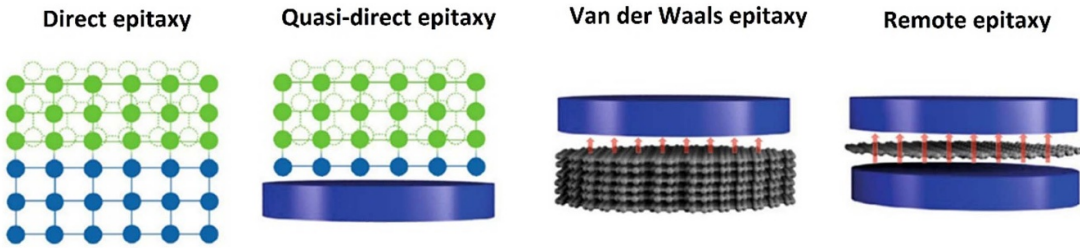


Figure 13. Schematics of direct, quasi-direct, vdW and remote epitaxy. Reproduced from [142]. CC BY 4.0.

Table 2. Comparative analysis of epitaxial growth in 2D material–assisted heterointegration.

Feature	(Quasi-)direct epitaxy	van der Waals epitaxy	Remote epitaxy
Interface bonding and strain	Strong chemical bonds at the interface result in film’s clamping and strain formation.	Interface is bonded only by weak vdW forces. Strain from lattice mismatch is negligible.	Film binds via 2D interlayer with only vdW forces, reducing interface stress.
Lattice matching requirement	Requires close lattice matching between film and substrate (nanosheets).	No lattice matching needed.	2D interlayer relaxes the requirement for a lattice match.
Interaction at interface	High—the substrate’s dangling bonds form strong chemical bonds with the film.	Minimal—only weak physical interaction occurs.	Minimal—only long-range electrostatic fields from the substrate interact with the film.
Substrate reusability	No—epilayer is chemically bonded to the substrate.	Yes—epilayer can be mechanically exfoliated.	Yes—the epitaxial film peels off from the 2D-coated substrate.
Nucleation and orientation control	Excellent lattice-guided nucleation and film orientation (locally for nanosheets).	Challenging—inert 2D surface results in delayed nucleation and multiple in-plane orientations.	The substrate’s electrostatic potential guides nucleation and enforces oriented growth of the film.
Growth techniques	Grown by standard epitaxy techniques (MBE, CVD, etc).	Uses 2D-material–coated surfaces as substrates; conventional growth methods apply.	Involves a 2D mask on a crystalline substrate; grown with conventional methods.

In case of the oxide nanosheets, single layers of oxide nanosheets, are used as a seed for epitaxial or textured growth of a functional oxides on either a non-crystalline substrate (e.g. glass) or on a chemically incompatible substrate (e.g. Si). An example by Nijland *et al* [146], where such an approach was used, is depicted in figure 15. Here, a CaNb_2O_6 (CNO) or TiO_x nanosheets served as a template for the (001) or (011) oriented growth, respectively, of ferromagnetic SrRuO_3 perovskite. CNO nanosheet layers were deposited on Gallium Nitride (GaN)-on-Si substrates by the Langmuir-Blodgett method to obtain (001)-oriented growth of perovskite oxide thin films using PLD [147]. Other studies showed that it is possible to extend this approach to fully amorphous substrates, such as fused silica [148]. A nice review on this topic is given by Shaheen *et al* [149] and Chen *et al* [150] and present a comparative study of the nanosheets to other template layer methods.

Advances in science and technology to meet challenges

To **control nucleation on inert vdW surfaces**, researchers have developed seeding and defect-engineering strategies. One approach is *pinhole epitaxy*, where engineered nanoscale defects or pores in the 2D layer intentionally expose spots of the underlying substrate to promote localized nucleation [151]. Adatoms selectively anchor at these defect sites and then coalesce laterally, leading to oriented growth despite the otherwise inert 2D surface. Such controlled nucleation techniques improve orientation uniformity and film continuity. Additionally, surface treatments (e.g. gentle plasma or thermal pretreatments) can create chemically active sites on graphene without destroying its continuity.

Another critical advance is the **validation and refinement of the remote epitaxy mechanism**. It has been confirmed that for polar substrates, long-range electrostatic fields can indeed traverse a monolayer 2D interlayer and guide epitaxial registry. For instance, monolayer hexagonal BN (h-BN) was shown to

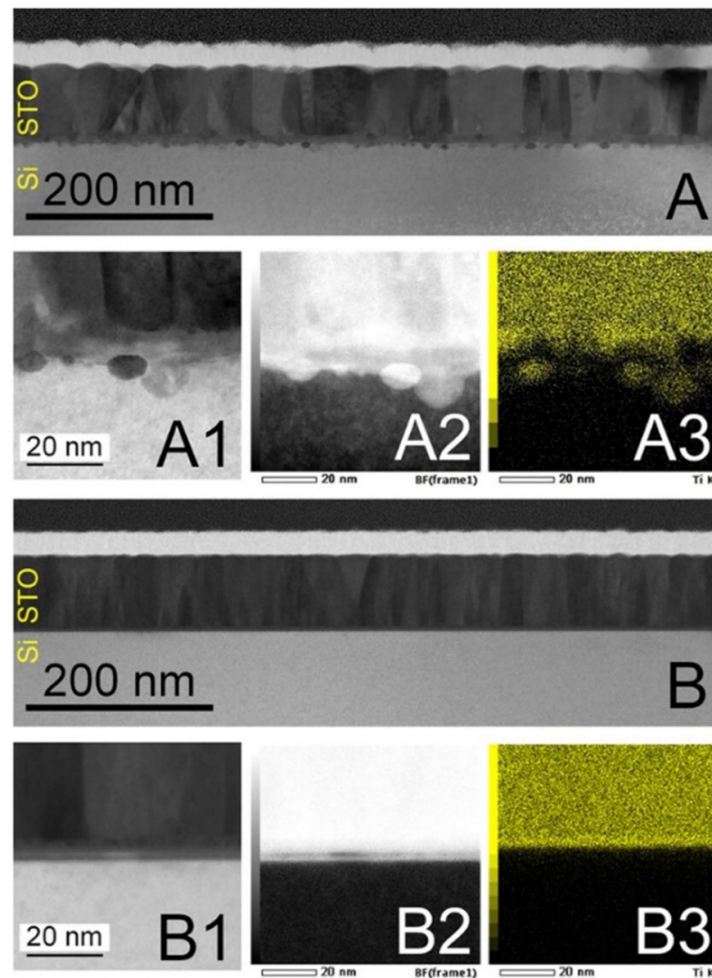


Figure 14. Interface after the growth of 85 nm thick STO layer on (A) bare and (B) rGO-buffered SiO_2/Si surface. The STO was grown after SrO-assisted deoxidation. (A1, B1), (A2, B2), and (A3, B3) represent the dark-field, bright-field, and Ti-mapping obtained by TEM analysis, respectively. Reproduced from [145]. CC BY 4.0.

be sufficiently ‘transparent’ for the sapphire substrate’s potential, enabling remote epitaxial growth of aligned ZrS_2 , whereas a bilayer h-BN attenuated the field and led to random orientation (vdW growth) [152]. Such findings bolster the understanding that true remote epitaxy (as opposed to growth through accidental pinholes) is achievable under the right conditions.

Next, researchers are exploring **alternative 2D materials beyond graphene** to improve stability and epitaxial transparency. h-BN, in particular, has emerged as a robust insulating buffer that withstands high-temperature oxidizing environments far better than graphene [153]. Its wide bandgap means it induces minimal charge screening, allowing more of the substrate’s field to reach adatoms. This property was leveraged in the aforementioned ZrS_2 remote epitaxy example, and h-BN can be further explored for integrating perovskite oxides that require oxygen-rich growth conditions.

On the materials side, there has been **significant progress in large-area 2D interlayer synthesis**.

Wafer-scale single-crystal graphene and h-BN layers can now be produced by optimized CVD on metal or sapphire substrates [154]. These uniform 2D films provide consistent surfaces for epitaxy across entire wafers, addressing scalability and alignment. In terms of device integration, full wafer demonstrations of vdW and remote epitaxy are emerging. A notable example is the fabrication of wafer-scale GaN-based LEDs on amorphous substrates via graphene buffers, yielding flexible optoelectronic devices with high efficiency [155]. In fact, recent research in remote epitaxy has transitioned from fundamental proof-of-concept studies to solving critical bottlenecks in template integrity, growth competition, and material universality. The recent literature identified the competition between remote, pinhole-seeded, and vdW epitaxy as primary problem. Researchers are moving beyond simply assuming ‘remote’ interaction and are now using advanced diagnostics to prove it. Studies on Heusler films

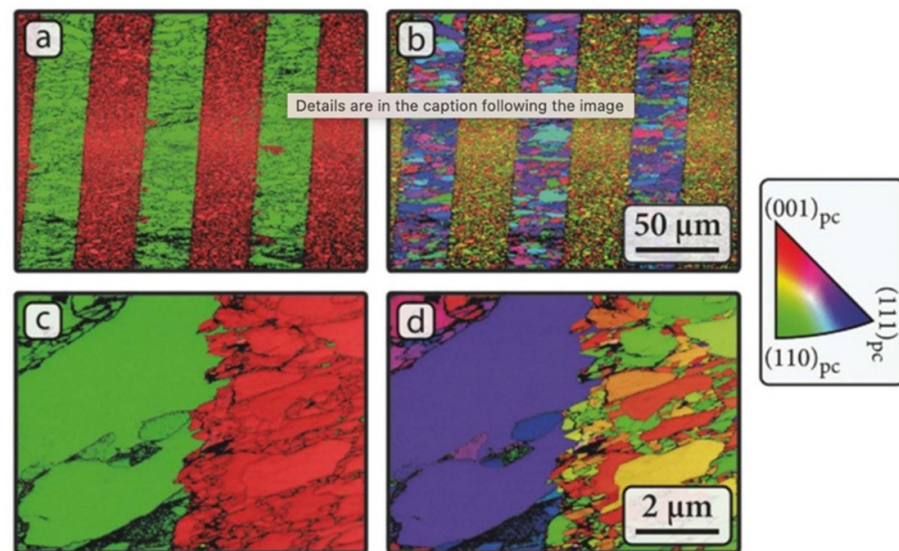


Figure 15. SrRuO₃ thin films on CNO and TiO_x nanosheets on Si. Inverse pole figure maps at (a), (b) low (step size of 300 nm) and (c), (d) high (step size of 10 nm) magnification measured by electron backscatter diffraction (EBSD). For both magnifications, the orientation of the film is displayed from (a), (c) the out-of-plane and (b), (d) an in-plane perspective. All maps are viewed as measured, without application of any form of noise reduction [146]. John Wiley & Sons. © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

and GaN have identified specific ‘fingerprints,’ such as a 30-degree lattice rotation, to distinguish genuine remote epitaxy from pinhole-seeded growth [156]. In line with this, a progress has been made in long-distance (at distances of 2–7 nm) and defect-mediated epitaxy in which defects in the substrate project their strain fields through thick 2D or amorphous membranes, allowing for epitaxial alignment [157]. Another tendency is development of universal amorphous templates, such as 2D amorphous carbon films fabricated at low temperatures (300 °C), suitable for the growth of high-quality GaAs and InGaAs on various substrates, bypassing the lattice-matching constraints of conventional epitaxy [158]. Also, development of alternative 2D materials, such as hexagonal boron nitride (hBN) and amorphous BN, has been beneficial for strain relaxation of InGaN and wide-bandgap semiconductor integration [159]. The ultimate goal of these efforts is the creation of freestanding nano-membranes for heterogeneous integration.

So far, the graphene dominates the field, enabling remote epitaxy of complex oxides (BaTiO₃, SrTiO₃, HfO₂) [160–162], vdW epitaxy of layered materials (GaTe, topological insulators) [163], and thermal management in wide-bandgap semiconductors (β -Ga₂O₃) [164]. The primary challenge of graphene degradation during harsh environments is being addressed through argon background gas, controlled plume kinetics, and transition to gentler MBE processes [165].

For **oxide nanosheets**, two important challenges remain for applications in our view: First, the in-plane registry control. It was shown that an improved (local) registry can be achieved by controlling the size and shape of the original crystallites (pre-exfoliation) [166]. Second, as with the vdW materials, large-area oxide interlayer synthesis has been achieved for laboratory purposes but hardly qualifies for large-scale fabrication. Scientifically, one important challenge remains and that is to understand the bonding between the nanosheet interlayer and the substrate after the functional oxide layer has been grown at high temperature and increased oxygen pressures. This in turn beckons to investigate the effect of the precise processing conditions on said bonding and stability of the nanosheet inter layer. Finally, it is yet to be established to what extent the finite lateral size of nanosheets of 2D materials (rGO, CNO, TiO_x, etc) may present opportunities in the context of FOMs.

Concluding remarks

2D-material-assisted epitaxy represents the current state-of-the-art solution for integrating materials that were previously thought incompatible, revitalizing research in oxide–semiconductor hetero-integration and other advanced material systems. Successfully addressing the exposed challenges will enable ‘materials-by-design’ heterostructures, where each method may carve out its niche depending on

the final application. However, the continued exploration of these challenges is not just about overcoming current bottlenecks, but also about discovering unforeseen phenomena at these hybrid interfaces, which could lead to entirely new directions in the fields of electronics, energy materials, and medicine.

Acknowledgements

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4.2. Remote Epitaxy: status, challenges, and future directions

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Status

Remote epitaxy was first introduced in 2017 as a promising strategy to produce freestanding single-crystalline membranes that preserve their structural quality after separation from the host wafer [167]. The technique employs a substrate coated with a vdW material, typically graphene. During epitaxial growth, the overlayer film inherits the lattice structure of the underlying substrate due to electrostatic potential fields from the substrate's ionic bonds that penetrate through the atomically thin 2D layer. Because the bonding across the 2D interface is governed by weak vdW interactions, the exfoliation of the epilayer can be precisely achieved, often assisted by stress from a deposited Ni layer (figure 16(a)).

Compared to other release methods such as chemical lift-off (slow and etch-based) or mechanical spalling (non-uniform), remote epitaxy enables instantaneous and uniform delamination of epilayer. This results in uniform film thicknesses, intact substrates that can be directly reused without repolishing, and high throughput. This technique is broadly applicable across diverse material systems. Complex oxides are of great interest due to their inherently high ionic character and multifunctional properties. In 2020, the first oxide demonstrations were reported, including perovskite SrTiO₃ (STO) and BaTiO₃ (BTO), spinel CoFe₂O₄ (CFO) and garnet Y₃Fe₅O₁₂ (YIG), which are grown on graphene-coated STO, MgAl₂O₄ (MAO) and Gd₃Ga₅O₁₂ (GGG) substrates, respectively, followed by mechanical exfoliation [29] (figures 16(b) and (c)). Since then, remote epitaxy has been extended to other oxides such as VO₂, LiNbO₃ and K_{0.5}Na_{0.5}NbO₃ [169–171].

Since the initial reports, experimental evidence supporting remote epitaxial interaction through 2D interlayers has accumulated. Systematic studies using GaN showed a transition from single-crystalline to polycrystalline growth as the thickness of the graphene increased, demonstrating progressive screening of substrate potentials [172]. Later, the observation of a 30° rotated superstructure in Heusler compounds grown on graphene/sapphire has been proposed as an experimental fingerprint of remote epitaxy, as it cannot be explained by alternative mechanisms [156]. More recently, direct TEM imaging of BTO nuclei on graphene-coated STO confirmed atomic registry with the underlying substrate in the absence of pinholes under the entire nucleation site, providing direct evidence for remote epitaxy [173].

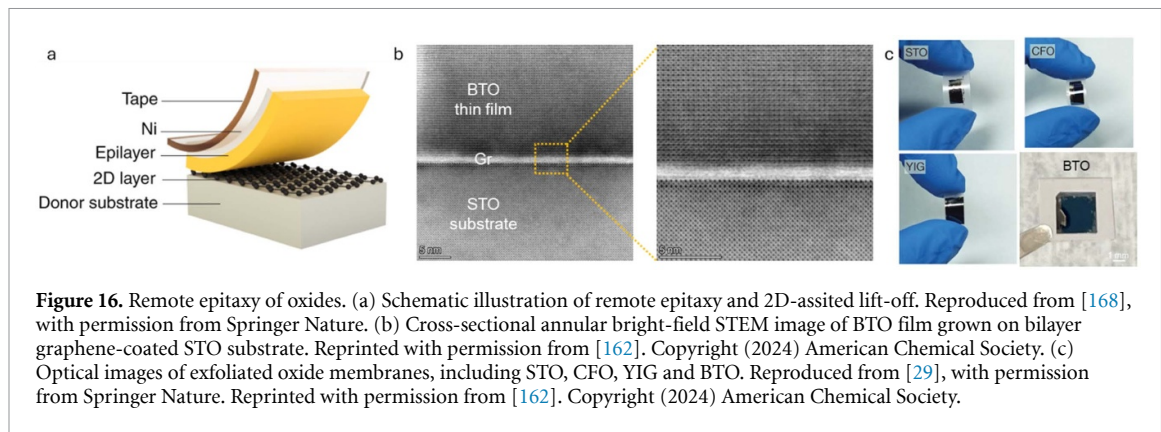
Beyond the intrinsic ferroic, pyroelectric, piezoelectric, and magnetostrictive functionalities of FOMs, remote epitaxy allows dissimilar materials to be integrated into artificial heterostructures that are unattainable with conventional epitaxy. Examples include magnetoelectrically coupled heterostructures with enhanced responses free from substrate clamping [29], and tunable diode characteristics achieved in 2D/3D/2D stacks [169]. These advances position remote epitaxy as a versatile platform for both fundamental investigations and next-generation device architectures.

Current and future challenges

By enabling access to new interfacial coupling phenomena and expanding functional possibilities for devices, remote epitaxy opens an exciting frontier. However, several major challenges remain and continue to drive intensive research.

A primary difficulty lies in creating a suitable growth template for complex oxides with an atomically clean 2D surface. Graphene is the most common choice because its polarity is sufficiently low to avoid screening while allowing substrate potentials to penetrate through. Yet, direct synthesis of uniform graphene on oxides is challenging [174]. Consequently, transfer methods are more practical, but inevitably introduce defects such as wrinkles, cracks, metallic residues, and organic contamination [175] (figure 17(a)). Such imperfections disturb the remote interaction between substrate and epilayer, degrading crystalline quality and reducing exfoliation yield, which are key barriers to industrialization in oxide-based microelectronics.

Another major challenge is the preservation of 2D interlayer during epitaxy process, which typically requires harsh growth conditions. PLD, the archetypal technique for complex oxide growth, produces a high-energy plasma plume that makes graphene highly vulnerable to damage. Elevated growth temperatures (>750 °C) and oxygen pressures exacerbate oxidation (>5 × 10^{−2} Torr) and decomposition of



graphene. This correlation is evident in systems ranging from spinel CFO to garnet YIG and perovskite STO, increasingly aggressive growth conditions reduce exfoliation yields from over 90% to only a small fraction (figure 17(a)). Protective strategies, such as depositing a 5–10 nm buffer layer under high vacuum (10^{-6} Torr) at the initial growth stage, can mitigate oxidation, but cannot fully shield graphene from kinetic plasma bombardment.

A further complication arises from controlling growth mode on 2D interlayers. Due to the large surface energy mismatch between 3D oxides and 2D layers, nucleation often proceeds via Volmer–Weber growth [171]. This makes it difficult to obtain uniform ultrathin membranes only a few unit cells thick. However, such ultrathin oxide membranes are highly desirable, which can break fundamental limit in the as-grown films, enabling exploration of moiré superlattices for new physics and delivering enhanced performance in device applications.

Advances in science and technology to meet challenges

To address these challenges, advances in both materials engineering and growth techniques have been made. Considerable effort has been devoted to improving graphene quality and its integration on oxide substrates. For example, direct growth of thick graphene on STO followed by atomic-layer etching has yielded pristine mono- to bilayer graphene suitable for high-yield remote epitaxy [176]. In parallel, optimization of transfer methods, such as using single-crystalline graphene grown on SiC combined with dry transfer, has reduced interfacial defects. As a result, the crystalline quality of remote epitaxial BTO films improved, with the XRD rocking-curve full width at half maximum decreasing from 0.84° to 0.57° [162].

Growth conditions have also been finely tuned to protect the 2D interlayer. Introducing argon as an inert background gas during PLD reduces the kinetic energy of the plasma plume and thereby mitigates graphene damage. Increasing the argon pressure from 1×10^{-5} mbar to 1×10^{-1} mbar increases the graphene Raman 2D/G intensity ratio to above 0.5, indicating a lowered defect concentration inside the graphene interlayer. Adjusting laser spot size has further mitigated damage to graphene [160, 177]. Beyond PLD, MBE is known as a milder growth environment, and its lower incident energy flux preserves graphene even at growth temperature as high as 970°C [162]. Hybrid MBE techniques take this further by removing oxygen source, and replacing the elemental source with a metal-organic source, such as titanium tetraisopropoxide (TTIP) for STO growth, which provide self-regulation of cation stoichiometry as well as sufficient oxygen supply [92].

Beyond graphene, alternative interlayers have been explored. MoS_2 , though less transparent to substrate potentials, can be synthesized more readily on various oxide substrates and still enables remote epitaxy in highly ionic systems such as ZnO [178]. Additionally, sub-nanometer amorphous layer has been demonstrated as an interlayer for remote epitaxy of VO_2 . These layers withstand growth temperature of 550°C while still permitting potential penetration from the substrate [179]. More recently, long-distance remote epitaxy has been reported with epilayer-substrate distance of 2–7 nm using amorphous carbon buffer layers. Remote epitaxial growth of CsPbBr_3 film, KCl film and ZnO microrods was achieved, providing new insights into the range of substrate-mediated interactions and suggesting alternative strategies for protecting ultrathin 2D interlayers from growth-induced damage [157].

Recently, to avoid the initial island growth mode on 2D surface, an innovative approach called atomic lift-off has been introduced, which eliminate the 2D interlayer by utilizing the intrinsic weakened bonding between epilayer and substrate, thus manufacturing ultrathin FOMs [108].

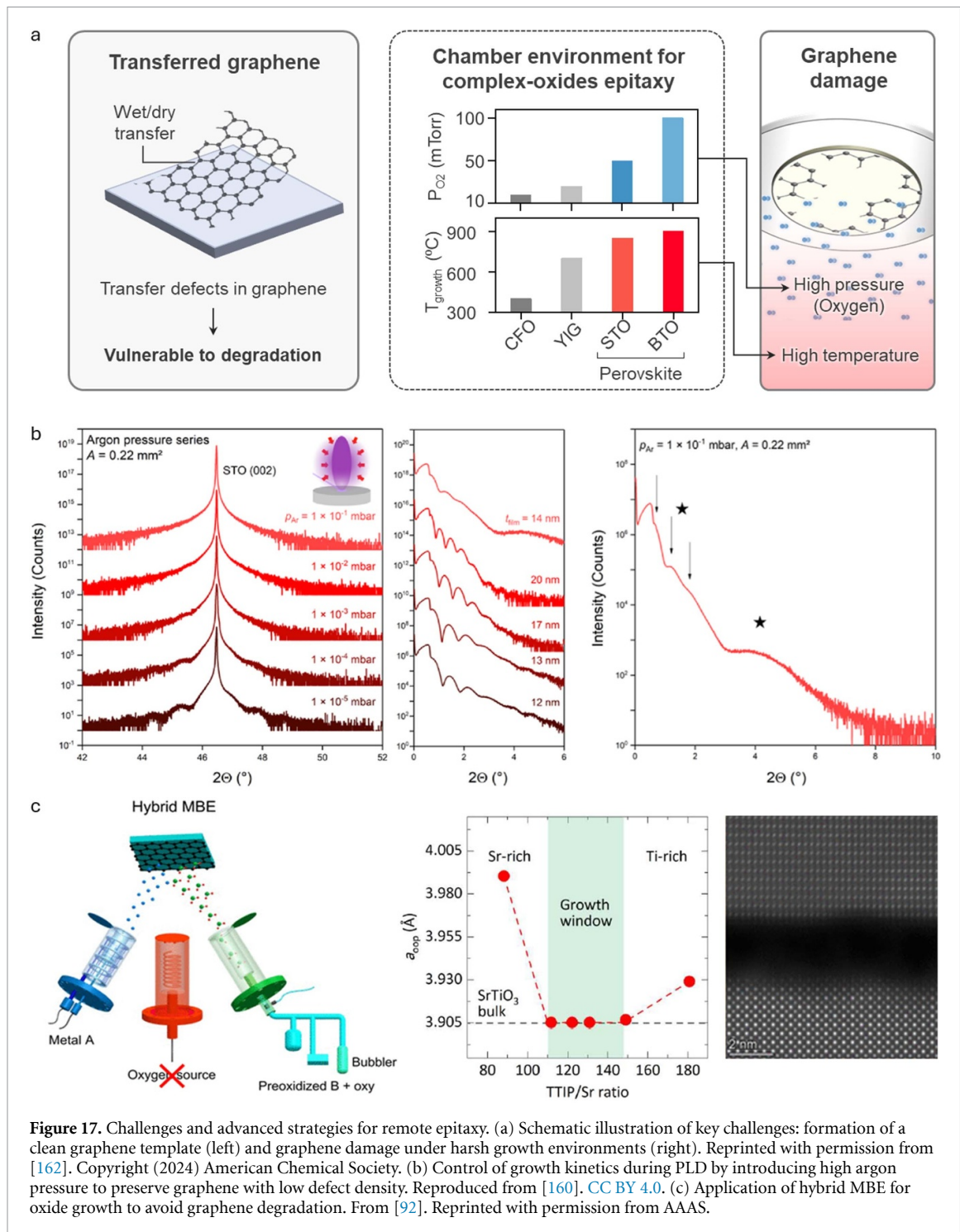


Figure 17. Challenges and advanced strategies for remote epitaxy. (a) Schematic illustration of key challenges: formation of a clean graphene template (left) and graphene damage under harsh growth environments (right). Reprinted with permission from [162]. Copyright (2024) American Chemical Society. (b) Control of growth kinetics during PLD by introducing high argon pressure to preserve graphene with low defect density. Reproduced from [160]. CC BY 4.0. (c) Application of hybrid MBE for oxide growth to avoid graphene degradation. From [92]. Reprinted with permission from AAAS.

On the other hand, advances in *in situ* characterization with growth chambers have further promoted understanding of oxide remote epitaxy, such as including synchrotron x-ray [180] and spectroscopic ellipsometry [162], providing real-time insights into graphene preservation and oxide nucleation mechanisms.

These studies are beginning to address the challenges of oxide remote epitaxy by optimizing 2D-coated templates, improving interlayer robustness, and refining growth dynamics, which are critical steps toward achieving wafer-scale, reproducible and industrially viable processes.

Concluding remarks

Remote epitaxy of oxides combines the advantages of substrate reusability, uniform film release and high throughput, establishing it as a promising technique for FOMs. The next phase of development should focus on two challenges: wafer-scale growth with consistently high crystalline quality and reliable

integration of released membranes onto arbitrary target platforms. Addressing the former will require scalable synthesis of vdW interlayers on oxide substrates that can withstand the demanding conditions of oxide epitaxy. Progress in the latter will depend on transfer and bonding strategies that preserve membrane integrity.

Solving these challenges would establish a practical manufacturing route for single-crystalline oxide membranes. Remote epitaxy provides a direct pathway toward twisted and stacked oxide heterostructures with independently engineered lattice, strain, and rotational degrees of freedom. Such structures could serve as a new platform for investigating correlated electron phenomena, and moiré-driven quantum states in complex oxides. In parallel, remote epitaxy will accelerate the integration of oxide functionalities with silicon and emerging semiconductor platforms. The ability to decouple oxide growth from the final device substrate removes long-standing constraints associated with lattice matching and thermal budget, enabling heterogeneous integration of ferroelectric, piezoelectric, pyroelectric, magnetic, and photonic materials. This capability could drive the development of negative-capacitance electronics beyond conventional scaling limits, highly sensitive sensing, neuromorphic memories, and integrated photonic devices. As these advances converge, remote epitaxy is positioned to become a key enabling technology for three-dimensional heterogeneous systems that combine the functionality of complex oxides with the scalability of modern semiconductor manufacturing.

5. Characterization of oxide membranes

5.1. Probing TMO membranes with advanced techniques at large-scale facilities: challenges and perspectives

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Status

TMOs exhibit a wide spectrum of correlated-electron phenomena, including metal–insulator transition, superconductivity, magnetism, and multiferroicity [181]. While lab-based methods provide fundamental properties and signatures of these effects, large-scale facility-based techniques offer significant advantages, owing to their higher probe intensity and tunability, in uncovering the underlying microscopic mechanisms. A broad suite of advanced characterization methods has proven exceptionally powerful for probing the structural, electronic, magnetic, and dynamical properties of TMO heterostructures, ranging from ultrathin films only a few nanometers thick to highly complex multilayer architectures. These capabilities include x-ray photoelectron spectroscopy (XPS) [182, 183], x-ray absorption spectroscopy (XAS) and its polarization-dependent variant x-ray linear dichroism (XLD) [184], x-ray magnetic circular dichroism (XMCD) [185], angle-resolved photoemission spectroscopy (ARPES) [186], resonant inelastic x-ray scattering (RIXS) [187], x-ray diffraction/reflectometry (XRD/XRR) [188], polarized neutron reflectometry (PNR) [189, 190], and low-energy muon spin rotation/relaxation (LE- μ SR) [191]. At modern synchrotron, neutron, and free-electron laser facilities, these techniques enable element-specific, depth-resolved, and momentum-resolved insights under operando conditions [192].

Freestanding TMO membranes and related heterostructures have emerged as a transformative platform for probing novel intrinsic material properties free from substrate-induced constraints [80]. In addition to eliminating substrate-induced constraints, freestanding geometries introduce fundamentally new mechanical boundary conditions, including bending, curvature, dynamic strain modulation, and topological deformations. These unique capabilities allow access to regions of phase space that are fundamentally inaccessible in conventional epitaxial heterostructures, providing unprecedented opportunities to manipulate and investigate coupled lattice, charge, orbital, and spin degrees of freedom. Large-scale facility-based investigations are already underway, with SrTiO₃ (STO) serving as a prototypical model system. For example, synchrotron XRD studies on freestanding STO membranes have revealed an enhanced antiferrodistortive (AFD) phase, highlighting the profound influence of substrate-free boundary conditions on structural instabilities and phase behavior [193]. The XLD and diffraction measurements have shown that TMO membranes, including STO, La_{0.7}Ca_{0.3}MnO₃, and LaNiO₃, can sustain large tensile strains, enabling fully reversible strain tuning [194]. XLD has also proven to be a valuable technique for probing and confirming antiferromagnetic ordering in three-dimensional oxide membranes under different strain regimes [195]. Although ARPES requires ultra-clean surfaces, it has been successfully applied to freestanding STO and related systems, demonstrating the feasibility of combining freestanding membrane platforms with state-of-the-art photoemission experiments at large-scale research facilities [186]. At neutron-based large-scale facilities, PNR has been successfully employed to investigate substrate-driven modulation of magnetism in manganite membranes [196].

Table 3 summarizes the key physical properties each technique probes, their depth sensitivity, operando capabilities, and the specific experimental challenges posed by the mechanical fragility and low signal levels of freestanding membranes. These techniques are ideally suited for probing thin films and heterostructures, where strain, interfacial coupling, and reduced dimensionality can be exploited to access emergent quantum phases. Their application to freestanding membranes has already uncovered unique strain states, orbital reconstructions, magnetic anisotropies, and correlated-electron phenomena inaccessible in substrate-bound thin films.

Current and future challenges

While freestanding TMO membranes provide a rich platform for exploring emergent physics, whether as individual membranes, membrane-based heterostructures, or hybrid architectures formed by integrating freestanding membranes with other 2D materials, their investigation using large-scale facility-based techniques remains both experimentally and conceptually challenging. Employing characterization techniques at large-scale x-ray and neutron scattering facilities, typically accessed through peer reviewed user

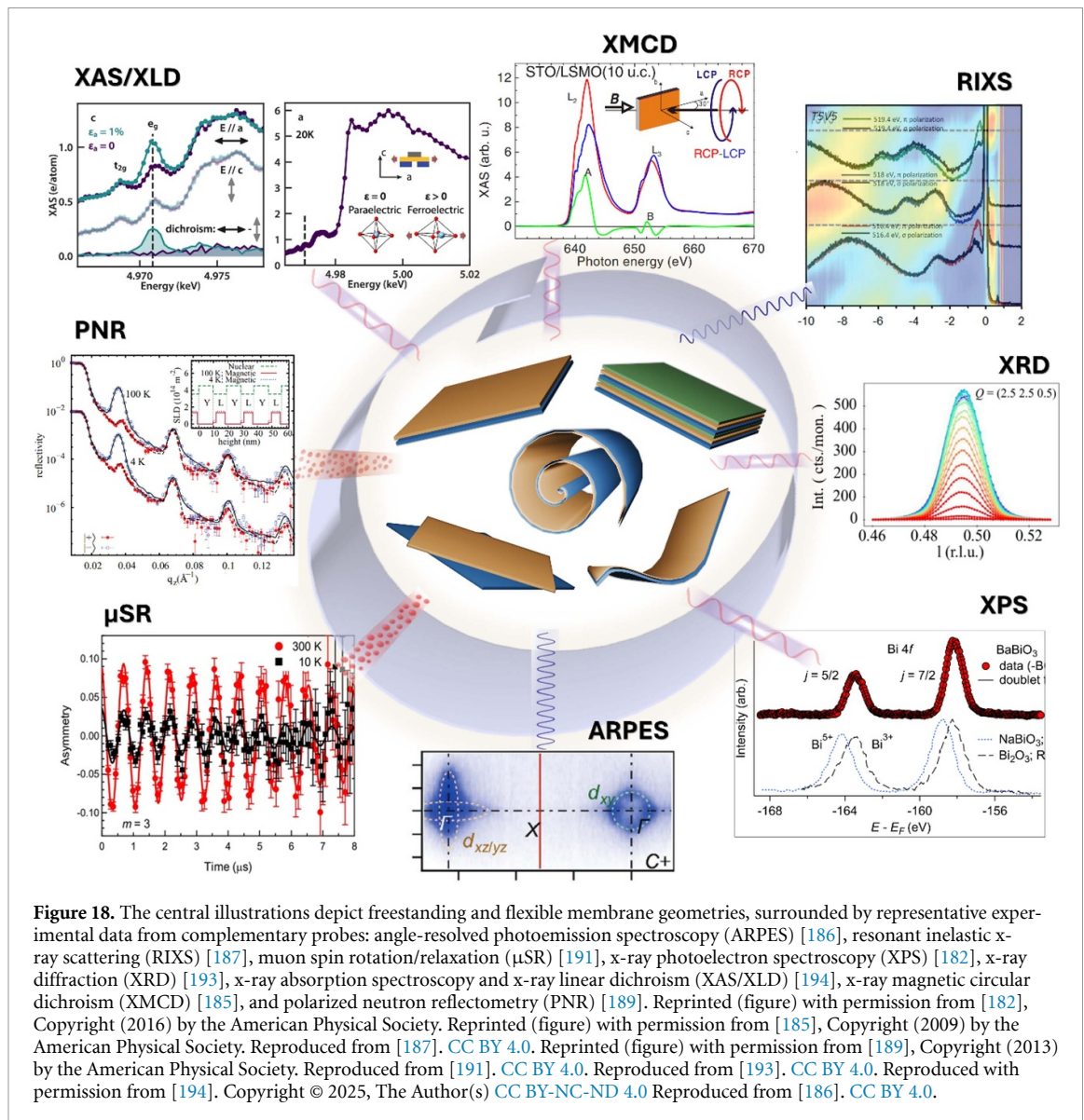
Table 3. Key large-scale facility-based experimental techniques for investigating TMOs, highlighting the type of information each method provides on structural, stoichiometric, electronic, and magnetic properties, as well as their *in situ* and *operando* compatibility.

Technique	Probed properties	Depth sensitivity	Operando capabilities	Challenges specific to freestanding membranes
XPS	Chemical composition, oxidation state	Surface to bulk (10 nm via increasing photon energies)	Yes: Temperature, variable pressure environments, pump probe Limited: magnetic fields, gating	Charging effects, fragile under ambient pressure
XAS/XLD	Local structure, orbital occupancy anisotropy, crystal-field symmetry, orbital ordering	Surface to bulk (≈ 10 nm)	Yes: Temperature, variable pressure environments, magnetic fields and gating (in FY mode)	Requires precise alignment with respect to the x-ray beam (polarization)
XMCD	Element-specific magnetic moments (spin and orbital), magnetic ordering	Surface to bulk (≈ 10 nm)	Yes: Temperature, high magnetic fields, gating, strain control	Requires precise alignment with respect to the x-ray beam (polarization) and magnetic field
ARPES	Electronic structure, band dispersion	Surface to bulk (10 nm via increasing photon energies)	Yes: Temperature Limited: gating, pump probe	Alignment issues, charging effects
RIXS	Elementary excitations (spin, orbital, phonon)	Surface to bulk (10–100 nm)	Yes: Temperature, variable pressure environments, magnetic field, gating	Low signal in ultra-thin membranes, long acquisition times
XRD	Lattice parameters, strain, crystal quality, ordering	Surface to bulk	Yes: Temperature, variable pressure environments, magnetic field, gating, pump probe	Weak diffraction from ultra-thin membranes
PNR	Depth-resolved magnetism and structure profile	Variable (few—100 nm)	Yes: Temperature, variable pressure environments, magnetic field, gating	Requires Large-area samples
LE- μ SR	Local magnetism, phase transitions	Variable (few—100 nm)	Yes: Temperature, variable pressure environments, magnetic field, gating	Requires large-area and flat membranes

programs, requires careful logistical coordination. Beamlines often lack adjacent sample preparation laboratories and the specialized tools needed to handle such delicate materials and systems. As a result, one of the central difficulties lies in transferring freestanding samples from the fabrication site to beamline endstations, where ultra-clean surfaces are often required for advanced characterization experiments. While specially designed vacuum or inert-gas transport cases can be used for this purpose, this is only the first step.

In the following section, we outline some of the most prominent obstacles encountered during advanced characterization under these conditions.

- 1. Mechanical stability.** Preparing stable yet sizable samples based on TMO membranes is challenging, as they typically only a few tens of nanometers thick, and inherently fragile, especially when mounted onto sample holders or left freestanding over open windows (frames). Maintaining their structural integrity during transfers (often under ultra-high-vacuum (UHV) conditions), mechanical handling, or in operando experiments involving external stimuli (e.g. electric fields, bending, or temperature gradients) is nontrivial. Warping, rupture, or delamination can severely compromise data quality or prevent meaningful comparison across experiments.
- 2. Sample alignment.** Due to their freestanding and ultrathin nature, TMO membranes are significantly more prone to misalignment than conventional substrate-supported films. Furthermore, mechanical deformations such as bending, curvature, or wrinkling may arise during handling, transfer, and mounting, posing additional challenges for characterization techniques that require precise sample geometry and alignment, including ARPES, RIXS, PNR and XRD/XRR.
- 3. Reduced scattering volume.** For scattering-based experimental methods such as XRD, RIXS, and PNR, the small available volume of ultrathin membranes results in weaker signals, requiring longer acquisition times or the use of ultra-bright beamlines with highly optimized optics.



4. **Surface sensitivity.** Techniques such as RIXS, XAS, XPS, and especially ARPES require ultra-clean surfaces for high-resolution measurements. Preserving surface integrity demands meticulous vacuum-transfer procedures from the sample preparation site to the end station, along with stringent handling protocols to prevent contamination. In this context, hard x-ray photoelectron spectroscopy (HAXPES) offers a complementary approach, as its significantly larger probing depth extends well beyond the near-surface region [197, 198]. This enhanced bulk sensitivity makes HAXPES particularly attractive for investigating freestanding membranes, buried interfaces, and bulk electronic and chemical structure while reducing sensitivity to surface contamination. In addition, HARPES at synchrotron facilities can be employed for operando studies under near-ambient-pressure conditions. The higher photon energies increase the electron mean free path in the gas phase while maintaining surface sensitivity when used in grazing-incidence geometry [199]
5. **Quantitative strain control.** While the intrinsic flexibility of membranes enables strain-tuning experiments, the *in-situ* application and measurement of well-defined strain states, particularly under thermal gradients or bending, remains challenging. However, laser-based curvature measurement techniques can provide real-time input on membrane deformation, which, when combined with modeling, can offer insights into the local strain distribution.

Given the challenges outlined above, it is important to recognize that while freestanding TMO membranes open exciting new scientific opportunities, fully exploiting their potential will require advances

in beamline instrumentation, endstation design, and sample environments tailored to their unique requirements.

The next section outlines the key technological developments needed to address these challenges.

Advances in science and technology to meet challenges

Freestanding TMO membranes and related heterostructure, despite their nanoscale thickness, mechanical fragility, and inherently low signal levels, require tightly coordinated experimental workflows integrating advanced synthesis, transfer, and state-of-the-art characterization. Establishing such interconnected workflows is a key prerequisite for fully exploiting large-scale user facilities in membrane-based research. Rather than co-locating all capabilities, a synergistic approach linking complementary infrastructures is most effective. Bridging materials growth with large-scale facility-based techniques that are highly sensitive to surface contamination, such as ARPES, XPS, XAS, and RIXS, requires UHV compatibility, including ultra-clean transfer and intermediate exchange chambers for mounting samples onto technique-specific holders (see figure 18). Solutions include vacuum suitcase systems, glovebox-integrated load-locks, and inert-gas-compatible capping layers (e.g. amorphous Al_2O_3), complemented by *in situ* surface preparation tools such as mild annealing, ozone cleaning, or low-energy sputtering, all adapted to the mechanical limits of freestanding membranes.

Accurate characterization further demands beamline-compatible platforms that minimize mechanical stress while enabling cryogenic operation, high magnetic and electric fields, and precise strain control. Key components include UHV-compatible holders with microfabricated frames, integrated strain gauges, low-background supports, piezo-driven stages, and active-feedback alignment systems using optical interferometry or capacitance sensors for sub-micron precision.

Low scattering volumes and photoemission yields in ultrathin membranes pose challenges for RIXS, XRD, and PNR, while muon spectroscopy requires large-area membranes due to macroscopic beam spots. Diffraction-limited storage rings, equipped with new types of undulators and modern optics, can deliver higher flux to the sample while improving energy resolution. In addition, when combined with nanofocusing optics (e.g. Kirkpatrick–Baez mirrors, capillary optics, or Fresnel zone plates), they can achieve the high spatial resolution essential for resolving inhomogeneities, nanoscale variations in membrane properties, and functional regions in micro-devices while still preserving flux. Regarding PNR, efforts are already underway to develop a next-generation polarized neutron reflectometer designed to probe magnetism and dynamics in nanostructures and complex multilayers and limited-volume samples. For example, the magnetism second target advanced reflectometer, under development as part of the second target station (STS) at Oak Ridge National Laboratory, is designed to enable new classes of experiments on weakly scattering materials and small-volume systems [200]. The enhanced brightness and optimized instrumentation planned for STS are expected to significantly expand opportunities for neutron studies of FOMs, particularly for investigations of emergent magnetism, interfacial phenomena, and dynamical processes that are currently constrained by sample size and signal strength.

At the Paul Scherrer Institute (PSI), ongoing upgrades of the Swiss muon source ($S\mu S$) are increasing the sensitivity of μSR experiments and extending their applicability to increasingly smaller sample volumes [201], opening new opportunities for studies of FOMs and membrane-based heterostructures. Furthermore, investigating ultrafast lattice, electronic and magnetic dynamics in FOMs will provide unique insights into nonequilibrium phenomena and transient states of matter. X-ray free-electron lasers (XFELs) offer unique opportunities to study such phenomena, and their femtosecond pulse duration enables direct observation of nonequilibrium phase transitions, transient strain states, and photoinduced emergent phenomena that cannot be accessed by static measurements.

While freestanding geometries enable dynamic strain tuning, achieving quantitative strain mapping remains a significant challenge. Laser-based curvature measurement [202] and integrated Raman strain mapping [203] offer valuable in-operando capabilities, enabling high-spatial-resolution characterization in techniques such as micro(nano)-ARPES, micro(nano)-RIXS/XAS, and scanning x-ray diffraction, where correlating local strain with electronic or structural properties is essential.

Finally, theoretical modeling under realistic experimental boundary conditions is essential for interpreting experimental observations and extracting meaningful insights from freestanding systems.

Concluding remarks

Heterostructures based on freestanding TMO membranes are redefining how new correlated-electron phenomena are explored, opening a unique route to manipulating intrinsic lattice, orbital, charge, and spin degrees of freedom, while enabling unprecedented control through reversible strain and integration into flexible device platforms and formation of interfaces that are not constrained by conventional structural or chemical coherence. Large-scale facility techniques have already demonstrated exceptional power

in revealing complex emergent or even hidden quantum phenomena in conventional TMO thin films and heterostructures. At the same time, fully exploiting the scientific opportunities offered by freestanding membranes will require adapting existing experimental methodologies to the unique characteristics of these systems. Achieving this goal demands a concerted effort to develop dedicated infrastructures that seamlessly integrate synthesis, ultra-clean transfer, and advanced characterization. This includes UHV-compatible transfer pathways, adaptive sample environments capable of applying and controlling a wide range of strain states, embedded real-time metrology for quantitative in operando control, and experimental protocols specifically tailored to freestanding architectures. Equally critical is the development of theoretical frameworks capable of accurately describing the mechanical, thermal, and boundary conditions unique to freestanding systems.

By combining the unique opportunities offered by freestanding membranes with the unparalleled capabilities of synchrotron, neutron, muon and XFEL facilities, it will become possible to unveil the structural, electronic, magnetic, orbital, and dynamical characteristics of complex oxides with unprecedented sensitivity and precision. Such facility-enabled insights will be essential for revealing hidden or emergent quantum phenomena, thereby accelerating the discovery and design of next generation functional oxide materials.

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5.2. Structural and functional property characterization of FOMs with transmission electron microscopy

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Status

Transmission electron microscopy (TEM) is a powerful technique for characterizing materials. This includes the crystal structure through electron diffraction, chemical composition and electronic structure through methods such as EELS [107], and functional properties like magnetic fields through Scanning TEM—differential phase contrast (STEM-DPC) [204].

A challenge for TEM is restrictions to specimen thickness that should ideally be less than 100 nm, and below 50 nm for high-end atomic resolution imaging. Most material deposition strategies for oxide thin films utilize thick substrates, and needs to be thinned before examination in TEM. A common sample preparation method is the use of the focused ion beam (FIB) with the lift-out process [205]. With this method it is easy to get a uniform sample thickness across the film structure in cross section geometry, with a region of interest of about $2 \times 2 \mu\text{m}$. Having uniform thickness across a sample is highly advantageous for TEM experiments. In addition, the region of interest is protected by a FIB deposited layer, resulting in minimal damage to the film. The vast majority of TEM studies on perovskites oxides have been in the cross section geometry.

However, there is a lack of information about in-plane properties, as preparing TEM-samples for the in-plane geometry is a much greater challenge. Here, the region of interest should be $10 \times 10 \mu\text{m}$ or larger, as the domains and feature sizes are typically 100 nm or larger. Using FIB for thinning with uniform thickness over this area without altering the material is prohibitively difficult.

FOMs, on the other hand, are inherently thin, and can be detached from their substrate and transferred onto a standard TEM Si_3N_4 grid. These Si_3N_4 grids have 9 electron transparent windows which are $100 \times 100 \mu\text{m}$ size and 20 nm thick. Here, we achieve sufficiently large regions to study, uniform thickness, and avoid alterations with an ion beam. Notable examples are Shao *et al* which observed polar skyrmions in perovskite oxide multilayers, utilizing scanning electron diffraction [206]. Another example is using twisted layers of freestanding ferroelectric membranes, where the TEM imaging revealed vortex patterns which changed as a function of the twisting angle (figures 19(a) and (b)) [33]. Dagenborg *et al* used a freestanding magnetic perovskite oxide membrane to study the effects of crystallinity on magnetic STEM-DPC imaging, as shown in figure 19(c) [207].

Current and future challenges

An important aspect of functional materials is how they respond to external stimuli. For example, how a ferroelectric material responds to electrical biasing, or a ferromagnetic material to cooling. One way of studying a ferroelectric freestanding membrane in a TEM, could be to apply electric biasing to the material outside the TEM. Then insert it into the TEM and image the polarization, followed by removing the sample from the TEM, doing another biasing and imaging the polarization in the TEM again. Such an *ex situ* experiment would give valuable information about the material, by providing snapshots of the switching process. However, being able to characterize the material during the switching is favored, as it would yield more detailed information about the ferroelectric properties.

There are several issues doing *in situ* experiments in the TEM: the lack of available space around the sample, the very high vacuum, and any modifications interfering with the electro-optics reducing the resolution. However it is possible in the TEM to apply a range of different external stimuli. The easiest is to apply an external magnetic field, as this can be done with the electromagnetic TEM lenses. *In situ* heating, cooling, straining, gas and liquids is also possible, using specialized TEM sample holders [208].

For materials for use in novel device concepts, the most interesting external stimuli are those related to the operation of the device itself. For example, if the device concept relies on the movement of ferromagnetic domain walls as a function of electrical current pulses, being able to image the domains while the current is applied is highly advantageous. Furthermore, the experiments should be done as similarly as possible to the device concepts, utilizing the same electrodes, size, geometry and in sufficient numbers to get good experimental statistics. Such *operando* experiments are a subset of *in situ* experiments, and electrical *operando* experiments are possible to do in the TEM, by using advanced MEMS based ‘chips’ with electrical contacts as specimen holders and *in situ* TEM holders with integrated electrical feedthrough.

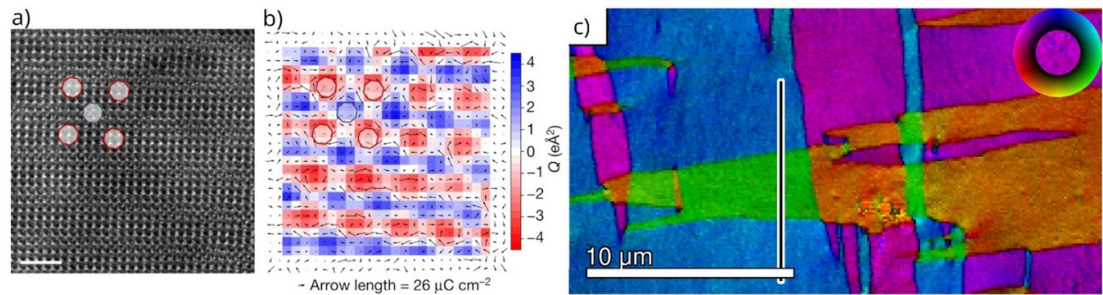


Figure 19. (a) Atomic resolution STEM-HAADF of twisted ferroelectric bilayer stack. Reproduced from [33]. CC BY 4.0. (b) Ti-displacement, showing a network of ferroelectric vortices. Reproduced from [33]. CC BY 4.0. (c) STEM-DPC image of free-standing ferromagnetic oxide membrane, showing the magnetic domain structure. The color wheel shows the direction and magnitude of the magnetization. Reprinted (figure) with permission from [207], Copyright (2025) by the American Physical Society.

However, MEMS based chips are supplied by the commercial vendor of the TEM holders, and are designed for use with the FIB lift-out process. Thus, they work very well for some types of experiments, but they are not well-suited for the FOMs due to the lack of electrical contacting between the chip and the membrane. Thus, doing electrical *operando* TEM experiments on FOMs is currently not feasible.

Advances in science and technology to meet challenges

The main unresolved issue for doing electrical *operando* TEM experiments on FOMs is the limitations of the available commercial MEMS-based chips. However, customized chips for the specific experiments can be fabricated. Spies *et al* demonstrated the preparation and use of customized chips for the TEM-characterization of nanowires [209]. Here, they used clean room microfabrication techniques like lithography and chemical etching to produce full 4 inch wafers, yielding 384 customized TEM chips compatible with an *in situ* TEM holder. This allowed for several types of *in situ* and *operando* TEM experiments, including biasing of individual nanowires to extract dopant concentrations, and visualizing a solid state metal-semiconductor reaction through Joule heating.

Similar customized TEM chips could be made for FOMs, enabling electrical *operando* experiments. Making the chips themselves would be similar to Spies *et al*, but with a new type of contacting due to the larger size of the oxide membranes. One possibility could be to use a Pt electrode array combined with wire-bonding to the chip electrodes, as recently reported by Lun *et al* [55]. However, a potential challenge could be wire bond's height being too high due to the limited space within the TEM, and thorough process development and optimization is needed to validate this approach.

By resolving the electrical contacting process for the FOMs with a MEMS-chip, the actual device characterized in the TEM would in functional behavior be much closer to the actual device, compared to specimen prepared using FIB. The method will then unlock detailed analysis under more representative conditions, arguably a key capability for guiding development of novel device concepts.

With the contacting method in place for doing *operando* studies, the powers of TEM can be fully utilized: combining structural, crystal, electric and functional characterization at the nanoscale as a function of external stimuli. Thanks to recent advances in automation of TEM such experiments would be almost fully automated, yielding a wealth of information on the dynamic coupling of crystal structure, chemical composition and functional properties like polarization or magnetization. The combination of identical TEM chips combined with automation, will enable a large amount of similar samples to be studied in a standardized and reproducible way, the only variation being the freestanding oxide itself. The next challenge would then be how to handle such large amounts of complex data, often in the 10+ TB per experimental session.

Concluding remarks

Due to their uniform thinness and ability to be placed directly onto TEM grids, FOMs are very suitable for characterization in the TEM. This has not only made the TEM a useful tool for studying both the membrane's structural and functional properties, but also made the membranes very useful model systems for developing TEM methodology. While some *in situ* TEM studies have been performed, there is a lack of the experiments most relevant for future devices like electrical biasing. By making customized *in situ* MEMS-based TEM chips and advancing methods for electrical contacting, such *operando*

experiments of novel device concepts can be performed in the TEM, yielding important knowledge about the connection between structure and function at the nanoscale.

Acknowledgements

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5.3. Probing and controlling oxide membranes with light and electron beams

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Status

In this section, we focused on the optical study of FOMs using spectroscopy techniques. Among the various characterization tools, such as electrical transport and magnetic property measurement, optical techniques are ideally suited for oxide membrane studies due to their unique ability to monitor intrinsic material properties without physical contact or invasive methodologies. Moreover, optical techniques provide sensitive, symmetry-specific access to intrinsic phenomena, such as electronic correlations, ferroelectricity, and magnetism. Due to these advantages, optical techniques have been widely adopted to study other categories of low-dimensional materials, including vdW materials. As interest in free-standing oxide films has grown in recent years, optical techniques are also being employed in the study of free-standing oxide films (see figure 20).

Optical Second harmonic generation (SHG)

SHG, a particular case of sum frequency generation, plays a crucial role in studying FOMs. SHG is highly sensitive to inversion symmetry breaking, as it is the primary source of the second-order non-linear process [210], making it an effective probe of broken symmetry, such as ferroelectricity and surface/interface states [211]. By employing SHG, recent studies have demonstrated ferroelectric properties in freestanding PbTiO_3 membranes [212], the emergence of ferroelectricity in strained SrTiO_3 [213], and the modulation of optical properties by varying the stacking sequence between $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ membranes and MoS_2 flakes [106]. Moreover, it has been shown recently that ferroelectric PbTiO_3 membranes exhibit ripples that can self-organize into periodic and crystallographic-oriented patterns [214].

Raman spectroscopy

Raman scattering is a light-matter interaction where incident photons exchange energy with vibrational modes of the crystal lattice (phonons), resulting in a characteristic frequency shift between the incident and scattered light. The inelastic scattering process provides insight into lattice dynamics, symmetry, and structural changes. In FOMs, Raman scattering can be a useful approach for probing strain relaxation and local structural variations without interference from the substrate. Raman scattering has demonstrated that the structural phase transitions are accompanied by the metal-insulator transition in free-standing VO_2 membranes [215], and locally modulated ferroelectricity in BaTiO_3 and $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ membranes [216].

Magneto-optic effects

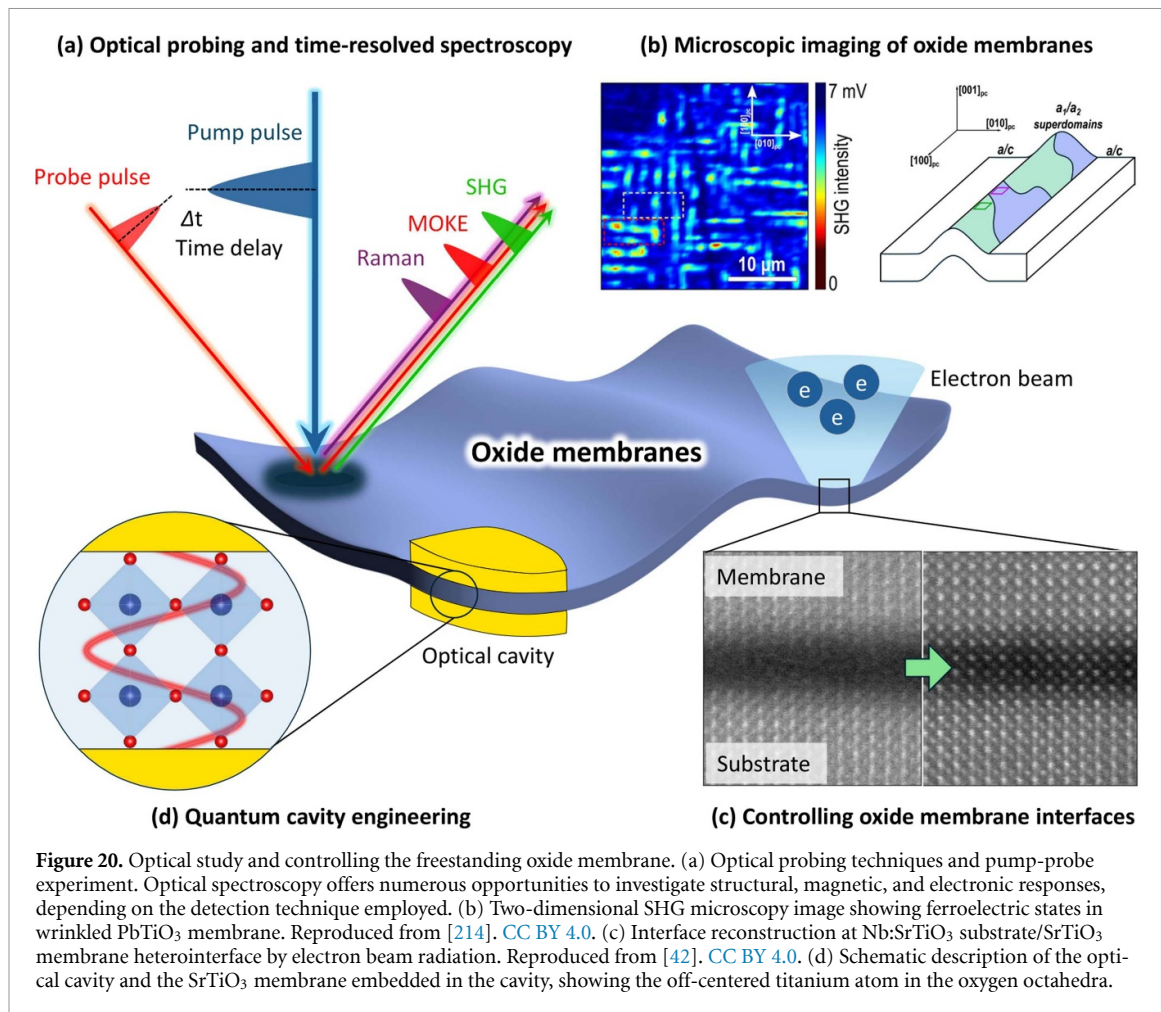
Although magnetic oxide membranes have been relatively rare to date, magneto-optical effects, including magneto-optical Kerr and magnetic birefringence, can be adopted to investigate magnetic properties in oxide membranes with high sensitivity and microscopic probing. For example, magneto-optical probing could serve as a valuable technique to monitor magnetic property variations depending on local curvatures in operando, specifically those arising from local strain gradients during the propagation of spin waves in magnetic oxide membranes.

Scattering-type scanning near-field optical microscope (s-SNOM)

Despite the advantages of conventional optical techniques, the probing resolution is limited to the sub-micrometer scale due to the diffraction limit in the far-field region. Recent advances in optical techniques have enabled probing of local properties with nanometer-scale resolution by combining optics with scanning probe microscopy, thereby enabling nanoscale imaging and spectroscopy [217], namely s-SNOM. Such nanoprobe measurements provide an opportunity to explore nanophotonic phenomena, such as surface phonon polaritons. For example, the localized near field at a conducting tip can induce nanoscale polaritons that propagate through the sample and subsequently reflect from boundaries or defects, thereby realizing polariton interferometry [218].

Current and future challenges

Despite the vast advantages of optical probing for studying FOMs, several issues and challenges must be addressed to fully leverage their potential for both fundamental discoveries and practical applications. Optical approaches enable the high-sensitivity measurement of the optical properties of condensed matter, but may also unintentionally measure unintended regions due to diffraction limits or light propagation within the material. From this perspective, synthesizing an oxide membrane with high crystalline



homogeneity is a crucial element for optical experiments. For instance, nanometer-scale crystal defects, such as misfit dislocations and stacking faults, can induce unexpected structural modulation and correlated electronic band structures, thereby interfering with the measurement of intrinsic material properties. The micrometer-sized structural modulation, such as rippling and bending, can complicate optical alignment and degrade spatial resolution.

In addressing these challenges, we anticipate that nanoscale control of the heterogeneous integration of membranes will play a key role. In contrast to conventional layered vdW heterostructures, oxide membrane heterostructures involve various types of bonding, including vdW, ionic, and covalent interactions, which can serve as additional control parameters. Accordingly, understanding and manipulating the surface and interfacial properties of oxide membrane heterostructures could provide a springboard for further advances in the field, including the development of high-quality, highly homogeneous oxide membranes and the exploration of related phenomena and applications, such as optoelectronic devices, ultrafast dynamics, and strong light–matter interactions.

Interface quality improvements

One of the most critical challenges is controlling the quality of the interface between the membrane and the substrate onto which it is transferred. This level of control is particularly important for enabling the use of oxide membranes in device applications. On one hand, the ability to synthesize membranes of functional oxides opens new opportunities for their integration into CMOS-compatible platforms [219]. On the other hand, precise control over the interface properties is fundamental for their successful integration. For this reason, recent research has focused on developing methods to optimize these interfaces and to control the degree of bonding between the membrane and the substrate.

Exploring and characterizing oxide membrane interfaces

Characterizing the physical properties at the interface of FOM heterostructures remains a significant experimental challenge. In conventional heterostructures, such as complex oxide thin films, proximity

effects resulting from various inter-layer interactions between different materials can lead to emergent phenomena driven by the correlation between lattice, spin, charge, and orbital degrees of freedom [220]. However, isolating the interface optical responses from the bulk response is challenging due to the volume effect. This often results in weak optical signals, which can limit the range of applicable spectroscopic techniques and further complicate the accuracy of precise measurements. Despite these challenges, a deeper understanding of interface-specific optical responses is crucial, as it may provide key insights into novel quantum phenomena arising from interfacial interactions. Advancing optical studies in these systems is therefore essential for uncovering new functionalities in FOM heterostructures, such as twistronics [221].

Advances in science and technology to meet challenges

Controlling oxide membrane interface with an electron beam

Recent studies have demonstrated that atomically flat interfaces can be achieved through various techniques, including electron beam irradiation using a scanning transmission electron microscope (STEM) or high-temperature annealing [11, 42]. These approaches have demonstrated that strong chemical bonds can form between the membrane and the underlying substrate, even when the substrate has a different crystal symmetry or lattice parameter. The formation of such bonds can induce distortions in the first atomic planes at the interface [11, 42], and this interfacial reconstruction may, in turn, lead to local structural modifications that enable the emergence of novel physical properties.

Ultrafast dynamics study in a non-equilibrium state

A significant advantage of optical probing over electronic excitation is the ability to access and explore ultrafast phenomena using time-resolved optical pump-probe spectroscopy. This technique enables time-resolved investigation of ultrafast electronic responses, nonlinear phononics, and magnetic spin dynamics by inducing photoexcited out-of-equilibrium states and employing a proper observation [222]. Recent studies on the interface between perovskite oxide membranes and SiO₂ have demonstrated that ultrafast pump-probe techniques can be effectively used to investigate interfacial properties. By photoexcitation, a gigahertz (GHz) acoustic wave is generated at the interface between the SiO₂ and oxide membrane, which reflects information about the interfacial adhesion depending on the annealing effect [223]. We anticipate that such optical pump-probe spectroscopy will be widely extended and adopted to study structural dynamics in oxide membranes, including acoustic wave propagation through spatially confined structures [224].

Cavity materials engineering

Instead of relying on high-intensity laser fields to drive materials into nonequilibrium states, strong light-matter coupling can be realized by embedding the material within an optical cavity. This method offers the key advantage of enabling strong interactions under equilibrium conditions, thereby providing new avenues for tuning and controlling material properties [225]. FOMs can be a promising cavity-integrable platform due to their lattice-matched interfaces and minimal substrate interference. These membranes enhance photon-phonon and photon-electron coupling efficiency, making them well-suited for strong-coupling regimes within plasmonic or dielectric cavities. For example, emergent phases and quantum functionalities may be dynamically tuned by controlling the interface between the Fabry-Pérot cavities and the target systems [226]. It is anticipated that the combination of oxide membrane functionalities with photonic engineering will offer a powerful playground for manipulating quantum phenomena in equilibrium states.

Concluding remarks

In conclusion, while optical probing techniques are already playing a significant role in the study of FOMs, given that this field is still in its early stages, there is considerable potential and promising prospects for the application of advanced spectroscopic experimental methods in this field. As the membrane synthesis method advances, a better understanding of the intrinsic properties and their controllability under various conditions, including surface/interface states, is anticipated. In particular, interface control of oxide membrane heterostructures will be a springboard in this field, due to the importance of the correlation effect between degrees of freedom in quantum materials. Collaboration with advanced spectroscopic experiments will provide a promising opportunity for key milestones. Time-resolved pump-probe spectroscopy will provide fundamental insights into an in-depth understanding of the interactions between light and materials across various energy scales, spanning the terahertz to

visible range, including various phenomena such as ferroelectricity and magnetism. Moreover, a strong light–matter interaction inside the optical cavity is expected to further expand our knowledge.

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6. Properties and applications of oxide membranes

6.1. Magnetic FOMs

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Status

Magnetic quantum oxides have become a centrepiece of condensed matter research over the last three decades, following landmark discoveries of colossal magnetoresistance and multiferroicity. Oxides exhibit strong spin-charge-orbital-lattice correlations, resulting in a rich variety of magnetic ground-states—ferromagnetic, antiferromagnetic, cycloidal, helical, spin glass and topological states—that are externally tunable. This tunability opens exciting opportunities for reconfigurable spintronic, magnonic, and multi-ferroic platforms that surpass the capabilities of conventional magnets.

Progress in the field has been driven by advanced thin-film synthesis enabling unit-cell-level precision over composition, structure, and strain. This led to the precise engineering of exchange, spin-orbit, and interfacial interactions, and the realization of metastable magnetic phases and textures. However, these growth techniques also introduced a key bottleneck: the reliance on lattice-matched crystalline substrates, making it challenging to integrate magnetic oxides with dissimilar, flexible or silicon-compatible materials—thus limiting their application despite exceptional potential.

Recent developments in fabricating freestanding magnetic membranes represent a breakthrough, as epitaxially-grown membranes preserve the structural and ferroic order [29, 63, 195, 227–229], even under extreme strains and deformations [195, 227, 228], whilst enabling transfer to arbitrary supports [195, 227, 229–233]. This unlocks fundamentally new pathways to tailor the magnetic energy landscape, including:

Strain effects: Extreme mechanical strain can tune the ground-state of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ membranes from a ferromagnetic metal to an antiferromagnetic insulator by modifying the exchange [227] (figure 21(a)). In $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, strain shifts the magnetic easy axis, and when integrated with ferroelectric PMN-PT substrates, enables repeatable, non-volatile electric-field switching of magnetic anisotropy [234] (figure 21(b)). In SrRuO_3 membranes, anisotropy can be tuned via orbital occupancy redistribution [230].

In $\alpha\text{-Fe}_2\text{O}_3$, strain reconfigures axial and basal anisotropies, inducing a Kibble–Zurek topological phase transition and enabling isothermal nucleation of topological antiferromagnetic textures (figure 21(c)). Under uniaxial strain, the system can be driven toward an aligned AFM state [235]. In BiFeO_3 , strain enables manipulation of antiferromagnetic cycloids [235].

Flexure and Ripple effects: Mechanical deformation introduces spatially varying strains and strain gradients that couple to magnetism. In flexed $\alpha\text{-Fe}_2\text{O}_3$ membranes, flexure leads to spatially-selective nucleation of AFM textures [195]. Moreover, ripples in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ modulate local exchange interactions and transition temperature [236].

Outlook: These findings establish magnetic oxide membranes as an outstanding platform for uncovering new physics and enabling novel functionalities. On the fundamental front, membranes facilitate access to spatiotemporal control of strain, allowing precise tuning of anisotropy, exchange, and interfacial interactions, in static and dynamical regimes. They also enable exploration into flexo-magnetic/magnetoelectric effects [237], where strain gradients can be used to control spin distribution. Finally, membranes can be designed in three dimensions through curvature, stacking, and twisting, opening new possibilities for configuring magnetism via geometric curvilinear effects [238], and moiré physics [14]. From an applied perspective, membranes offer a promising new route to magnetic oxide-based applications, including flexible, wearable, strain-engineered and silicon-based spintronic technologies [101, 231, 239, 240].

Current and future challenges

While FOMs offer a transformative platform for designing and exploiting magnetic quantum oxides, the field is in its infancy, and several challenges need to be addressed.

Fabrication: Commonly used pseudo-cubic sacrificial buffer layers, which enable post-growth release and transfer of membranes, introduce their own lattice and symmetry constraints. [63, 227]. This limits the fabrication of high-quality epitaxial membranes for magnetic oxides with large mismatch or

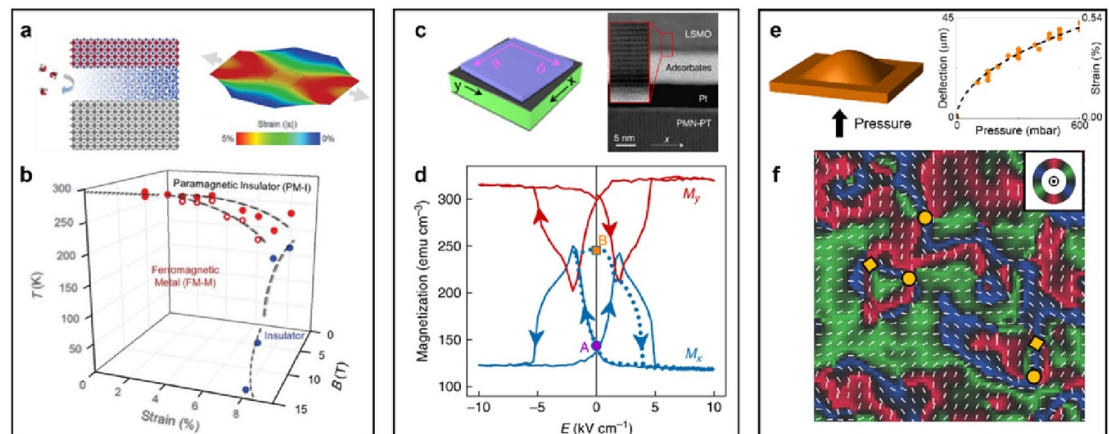


Figure 21. Magnetic control in oxide membranes. (a), (b) Tuning exchange in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$: (a) Fabrication of an epitaxial freestanding LCMO membrane using a sacrificial underlayer. (b) Extreme strains re-engineer the strength and sign of the magnetic exchange. From [227]. Reprinted with permission from AAAS. (c), (d) Tuning anisotropy in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$: (c) Detached LSMO membrane transferred onto an electroactive ferroelectric platform PMN-PT. (d) Electrically driven structural phase transition alters the in-plane lattice constants, triggering a non-volatile rotation of the magnetic anisotropy. Reproduced from [234]. CC BY 4.0. (e), (f) Creating topological textures in $\alpha\text{-Fe}_2\text{O}_3$: (e) Tensile-strained $\alpha\text{-Fe}_2\text{O}_3$ membrane on a flexible Si_3N_4 support. (f) Strain modifies the axial antiferromagnetic anisotropy, resulting in the isothermal nucleation of antiferromagnetic topological textures at room temperature. Reproduced from [195]. CC BY 4.0.

non-perovskite symmetries (e.g. sesquioxides, ilmenites, rutiles, spinels, garnets, pyrochlores) via standard transfer strategies. Remote epitaxy (discussed in section 6) has emerged as a promising route to address this limitation [29]. However, the widespread use of this method is hindered by the degradation of intermediate layer, particularly under high-temperature and oxygen-rich conditions required for oxides.

Transfer: Next, the transfer process creates new interfaces with dangling bonds, surface reconstruction, adsorbates, mechanically induced defects or chemical residues, introducing uncontrolled variables that directly affect spin-based functionalities, including spin transmission, spin/spinwave scattering, interfacial symmetry-breaking, interfacial exchange, exchange bias or electric-field driven effects.

Precise Control: Finally, while strain is a powerful control knob, it can also be inadvertently introduced or modulated spatially during transfer, leading to non-uniform ground states, suppressed ordering, or an inhomogeneous magnetic landscape. These effects would be especially pronounced in materials with strong magnetostrictive or magnetoelastic coupling, low ordering temperatures, or closely competing magnetic phases and ground-states.

Application bottlenecks: Functional spintronic devices rely on complex multi-layer heterostructures, where each layer serves a distinctive role (e.g. free/pinned magnet, spacer, barrier). Unlike metallic systems, quantum oxides demand crystalline coherence, making heterostructure fabrication considerably more challenging. Hence, most membrane studies have focused on single or few-layer oxides, with transferable superlattices or complex stacks remaining largely unexplored. A key challenge lies in achieving epitaxial growth across dissimilar material systems, while preserving structural coherence, interfacial sharpness, and ferroic order throughout the entire stack. Additionally, magnetic and electric dead layers—interfacial regions that become electrically/magnetically inactive due to charge transfer, atomic reconstruction or amorphization—remain a well-known issue in oxide films.

Advances in science and technology to meet challenges

Fabrication and Transfer: A near-term goal is to fabricate high-quality crystalline magnetic oxides with ultra-clean interfaces and minimal defects, enabling the integration of freestanding heterostructures and superlattices where each layer retains functional order. The long-term objective is to achieve deterministic wafer-scale membrane transfer for device applications.

Firstly, to broaden material compatibility, it is essential to engineer and diversify sacrificial layers beyond currently employed $\text{A}_3\text{Al}_2\text{O}_6$ and $\text{A}_4\text{Al}_2\text{O}_7$ systems ($\text{A}=\text{Ca}, \text{Sr}, \text{Ba}$) [56, 63]. Non-perovskite oxides may be grown on symmetry-matched facets (e.g. (111), (110)), though significant mismatch will necessitate buffer layers to suppress structural defects [101, 195, 241]. In parallel, advances in vdW and remote epitaxy are crucial for fabricating oxides incompatible with sacrificial layers [29, 242]. Strategies to

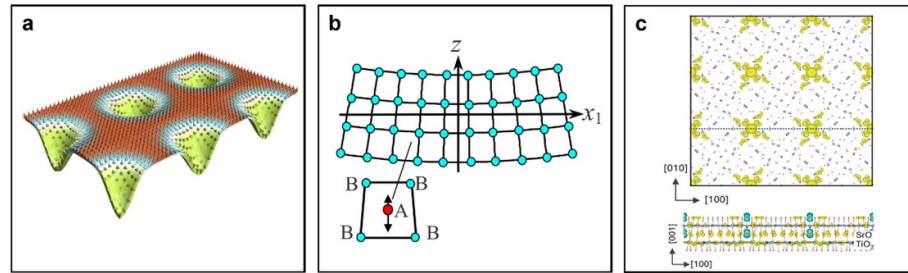


Figure 22. New pathways of tuning oxide magnetism. (a) Geometric curvature can stabilize chiral topological ground-states in a curvilinear membrane. Reprinted (figure) with permission from [238], Copyright (2018) by the American Physical Society. (b) Strain and strain gradients, shown at the unit cell level, can result in flexo-magnetic/magnetoelectric effects. Reprinted (figure) with permission from [237], Copyright (2009) by the American Physical Society. (c) Moiré effects can lead to the emergence of local magnetism, shown as iso-surfaces of charge density (top-view) and spin density (side-view), in twisted and stacked layers of SrTiO₃. Reprinted (figure) with permission from [14], Copyright (2025) by the American Physical Society.

minimize oxygen-rich plasma exposure and to introduce O-deficiency-tolerant buffer layers may improve the viability of remote epitaxy.

For post-growth processing, clean transfer techniques that avoid intermediate polymeric supports are critical to maintaining interfacial atomic structure and avoiding surface contamination. Recent SiN_x-based deterministic transfer methods [243], developed for 2D materials, may be adaptable for oxides. Subsequently, improved membrane-substrate adhesion is crucial for multiple post-processing/nano-fabrication steps, and for homogenizing residual strains.

In-situ Characterisation: Conventional magnetic techniques provide spatially averaged signals and may fail to detect localized magnetic phenomena arising from non-uniform strain, flexure, or curvature. To uncover new physics and test device functionalities, it is vital to perform spatially resolved *in-situ* mapping of magnetic, multiferroic, or spintronic properties. Promising approaches include optical [236], x-ray [195, 229, 233], electron or scanning probe microscopy integrated with *in-situ* strain, curvature or electric/magnetic impulses. Moreover, systematic characterisation of as-grown and transferred membranes, with attention to interface structure, magnetism, and quality, is essential to identify and mitigate growth- and transfer-induced degradation.

Precise Control: Building upon aforementioned studies of large-scale strain or flexure, future efforts must target micro/nanoscale strain engineering, using pre-patterned release geometries or curved substrates to define strain profiles during and after transfer. Additionally, electrically-driven strain platforms, e.g. MEMS/NEMS or piezoelectrics/ferroelectrics [29, 101], offer a compelling route toward reconfiguring magnetism.

Exciting new directions enabled by flexible oxides include curvilinear magnetism [238] and flexo-magnetic/magnetoelectric effects [237], where local curvature, geometry, strain, and strain gradients can be used to discover and tailor the dimensions, topology, and chirality of novel magnetic states.

Moreover, twist and stacking of oxide membranes could enable spatially modulated magnetic exchange and frustration through the moiré potential, leading to nanoscale chiral and topological textures inaccessible in the host material. Exploiting these opportunities will require deterministic geometry and twist-angle control together with high-resolution nanoscale imaging [14]. Translating these ideas into oxides could lead to the discovery of hitherto undiscovered magnetic states and phenomena (figures 22(a)–(c)).

Applications: Another major opportunity lies in leveraging oxide membranes to devise novel reconfigurable magnetic technologies, thanks to their mechanical compliance, chemical phase stability, tunability and access to diverse ferroic phases. Specifically, they are attractive for adaptive magnetic sensing platforms, including magnetic e-skins, contactless sensors and flexible bio-integrated devices, beyond what is possible in metallic magnets [239, 240, 244]. Their strain and curvature-tunable magnetic energy landscape also opens opportunities for reconfigurable RF/microwave components and magnonic architectures, including tunable spin-wave waveguides and filters [240, 245]. Silicon-integrated membranes hosting direct multiferroic coupling [228, 235] or indirect magnetoelectric coupling, enabled through ferroelectric/piezoelectric supports [101, 234, 240], could serve as key components in low-power spin-based logic and memory elements. Looking further ahead, twisted and curved membranes may provide

programmable platforms for engineering functional spin textures and magnonic networks through the combined control of geometry, strain and moiré interactions.

Realizing these applications will require further advances (i) defect-free, high-yield, wafer-scale fabrication and deterministic transfer, (ii) integration into semiconductor process flows, including heterostructure growth and device nano-fabrication, and (iii) reproducible long-term performance and endurance under thermal, electrical, and mechanical cycling.

Concluding remarks

Freestanding magnetic oxide membranes represent a disruptive advance that can push the design, control and applications of quantum oxides. By removing substrate constraints, these membranes unlock exciting new opportunities to leverage flexibility, strain, twisting and curvature that were previously inaccessible in substrate-bound geometries. Early studies have already demonstrated the reconfiguration of magnetic ground states, anisotropy, exchange, and topology in response to mechanical stimuli, establishing oxide membranes as a promising platform for fundamental discoveries and new spintronic and magnetoelectric applications.

Looking ahead, the roadmap for oxide membranes will require interdisciplinary advances in materials fabrication, interface engineering, deterministic transfer, targeted structural and geometric engineering post-growth and *in-situ* characterisation. Membranes offer a route to integrate magnetic oxides with flexible, wearable, and silicon-based technologies. The convergence of mechanical flexibility and chemical stability with tunable spin-based functionalities opens the door to reconfigurable, low-power, and adaptive devices that could surpass conventional magnetic platforms.

Acknowledgements

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6.2. Freestanding ferroelectric-oxide membranes

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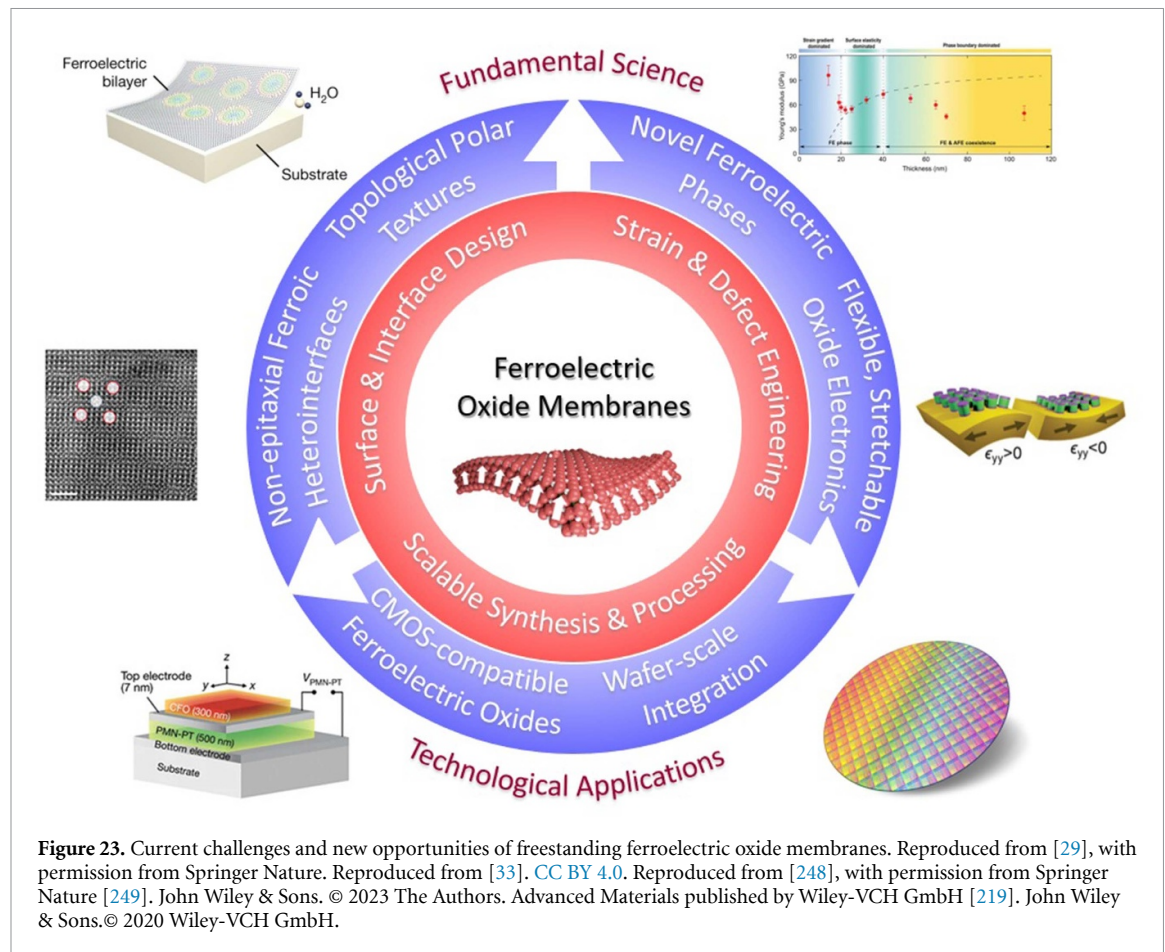
Status

Over the past two decades, there have been tremendous advances in the synthesis and control of ferroelectricity in epitaxial oxide thin films. Among numerous exciting new directions, the emergence of FOMs offers a powerful alternative to conventional thin-film epitaxy, offering new opportunities beyond the limitations of rigid substrates and potential for impact in next-generation heterogeneous integration of these materials [34, 80]. Enabled by selective chemical etching of sacrificial buffer layers, these membranes retain crystallinity and functional properties of epitaxial films, while introducing mechanical flexibility and emergent substrate-decoupled phenomena and properties. The ability to dynamically tune strain, integrate with dissimilar materials, and engineer novel interfaces positions oxide membranes as a compelling platform for both fundamental exploration and device innovation. Since their introduction, the field has rapidly evolved, establishing freestanding-oxide membranes as a versatile platform for uncovering emergent ferroelectric phenomena and enabling next-generation electronic and functional devices (see figure 23). Looking ahead, continued development of FOMs is expected to unlock several transformative research directions:

- *Dissimilar material integration*: The potential for heterostructure construction via membrane transfers circumvents traditional lattice-matching constraints and thermodynamic stability requirements [29, 82]. A wide range of material families, including vdW 2D materials, nitrides, polymers, and oxides can thus be incorporated as hybrid heterostructures—something not feasible previously. Such versatility in heterostructure designs enables new 2D interfaces where ferroelectric order can interact with semiconducting, magnetic, or metallic systems.
- *Unconventional strain engineering*: Unlike static, uniform strain states posed by epitaxial thin films, freestanding membranes offer access to extreme strain states and dynamic strain control [227]. This capability enables strain-induced ferroelectricity, providing access to stabilize ferroelectric states beyond those accessible through epitaxial growth [213]. Stretching and bending membranes also allow to produce inhomogeneous and/or anisotropic strain states along a specific crystallographic orientation, breaking lattice symmetries by design. Moreover, local curvature, ripples and wrinkles can generate large strain gradients, which can enhance flexoelectric responses or stabilize nanoscale-domain structures [127, 246].
- *Moiré lattice design*: Non-epitaxial interfaces between two membranes can create moiré superlattices through long-range periodic modulation in atomic registry [32, 33]. Precise control of twist angle and interfacial registry offers a new degree of freedom for engineering polarization, stacking order, and strain landscapes [247]. We anticipate that moiré engineering will become a new frontier in ferroelectric oxides, enabling the realization of emergent topological polarization textures and collective states that are inaccessible through conventional epitaxial approaches.
- *Substrate-decoupled phenomena and properties*: Freestanding-oxide membranes decouple the mechanical and electrostatic boundary conditions imposed by the growth substrate, enabling access to intrinsic ferroic behaviors that are often obscured in conventional epitaxial thin films. The release of substrate constraints can stabilize the formation of emergent topological polarization textures [206, 248], tailor ferroelectric switching behavior [105], and facilitate antiferroelectric-to-ferroelectric phase transitions [249].

Current and future challenges

Among the scientific challenges facing the field, understanding the scaling limits of ferroelectricity and controlling membrane surfaces and interfaces will likely be critical for realizing robust nanoscale ferroelectric devices. Size effects become increasingly pronounced in oxide membranes as thickness decreases



and surface and interface contributions dominate. A longstanding challenge in ferroelectric materials is size effects, where ferroelectricity is suppressed below a critical thickness. This issue becomes even more pronounced in oxide membranes, where in the ultrathin limit, interface-induced bond breaking can deteriorate crystallinity of membranes, as in the ultrathin limit due to interface-induced bond breaking, potentially leading to partial amorphization and a collapse of lattice coherence, an effect reminiscent of the 2D Berezinskii–Kosterlitz–Thouless transition [126]. While some studies suggest ferroelectricity can persist in the 2D limit of membranes without a critical thickness [10], it remains unclear whether this is a universal phenomenon. Exposed surfaces introduce additional chemical and structural complexity, such as dangling bonds, reconstructions, and interactions with ambient species such as water, hydroxyl groups, and organic species, which can influence interfacial bonding when coupling membranes in moiré systems or integrating them with dissimilar materials [11, 57]. Yet, these features also offer a rich parameter space to control these materials. A systematic reassessment of both intrinsic limits and processing-induced extrinsic effects, together with atomic-scale understanding and control of surface and interface structure, chemistry, crystallinity and defect populations is essential for establishing the ultimate scaling limits and enabling nanoscale ferroelectricity.

Crack formation is another major challenge in fabricating high-quality ferroelectric membranes due to the inherent brittleness of oxides. Strain relaxation upon substrate release can trigger domain reorientation or phase transitions, often accompanied by abrupt lattice distortions that compromise mechanical stability and increase the risk of cracking. Strategies such as using lattice-matched substrates/electrodes [219], minimizing residual stress, and optimizing transfer processes are needed to mitigate cracking [250]. Moreover, mechanical fracture during membrane deformation limits access to extreme strain states needed to explore exotic phase transitions in ferroelectrics. Developing reliable strategies to induce and control such strain states is essential for unlocking extreme strain-induced exotic phases and functionalities in ferroelectrics.

Finally, translating ferroelectric membrane research from laboratory demonstrations to industrial applications also remains challenging. With growing interest in complex oxides for logic, memory, and RF devices, freestanding membrane approaches offer a promising pathway for integrating epitaxial functional materials with CMOS-compatible architectures. Current membrane fabrication methods that rely

on sacrificial-layer etching are, however, often constrained by the scalability and throughput of epitaxial growth techniques such as pulsed-laser deposition (PLD). Although emerging methods such as remote epitaxy have shown considerable promise for wafer-scale membrane manufacturing in III–V semiconductors, its extension to ferroelectric oxides remains challenging due to graphene instability under oxidizing growth conditions, stringent stoichiometric requirements, and the difficulty of maintaining wafer-scale, defect-free graphene interfaces [124]. In addition, achieving wafer-scale manufacturing requires improvements in membrane release yield, transfer reliability, alignment accuracy, and wafer-scale uniformity. Transfer-induced defects, residual strain, interfacial contamination, and membrane damage can introduce substantial device-to-device variability and degrade functional performance. Long-term reliability, including thermal, electrical, and mechanical stability during device operation and packaging, also remains largely unexplored. Furthermore, integration into CMOS process flows requires compatibility with back-end-of-line (BEOL) thermal budgets, contamination-control standards, and large-scale manufacturing infrastructure.

Advances in science and technology to meet challenges

Recent advances in ferroelectric-oxide membranes have opened new avenues for integration beyond conventional epitaxial thin films, but significant scientific and technological challenges remain to translate these materials into scalable, industry-relevant platforms. One major thrust involves efforts to integrate functional oxide membranes with existing CMOS technologies. There is increasing interest in short-circuiting BEOL compatibility issues in semiconductor device fabrication that limit growths to $<400\text{ }^{\circ}\text{C}$ and the materials that can be introduced into CMOS fabrication processes by leveraging 3D heterogeneous integration of membranes [251]—thus offering fundamentally different pathway. By enabling low-temperature transfer of crystalline oxide layers, freestanding membranes circumvent many of the thermal budget limitations and lattice-matching requirements of traditional oxide deposition techniques.

Beyond CMOS, oxide membranes can be heterogeneously integrated with a wide range of dissimilar platforms—including III-nitrides, SiC, and polymers, enabling multifunctional device architectures that combine ferroelectricity with power electronics, optoelectronics, or flexible electronics. For example, the combination of oxide ferroelectrics with GaN-based high-electron-mobility transistors or SiC-based power devices can introduce new avenues for gate tunability, memory-in-logic, or adaptive electronics. In parallel, the mechanical flexibility of oxide membranes supports conformal integration on bendable or stretchable substrates, opening opportunities for wearable sensors and skin electronics. These capabilities open a broader design space for next-generation logic, memory, and RF devices.

Scalable manufacturing is likely to become the primary bottleneck determining the commercial viability of ferroelectric-oxide membranes. Unlike traditional oxide thin films, which rely on small, costly single-crystal substrates, membrane-based approaches enable large-area integration by enabling the transfer of epitaxial oxide layers with preserved crystallinity and functional performance while potentially allowing substrate reuse and compatibility with silicon-based industrial semiconductor manufacturing infrastructure. Key to realizing this vision are ongoing innovations in scalable synthesis and processing techniques. Notably, Lam Research has introduced Pulsus, a wafer-scale PLD platform designed to bridge the scalability gap between research-scale PLD and high-volume industrial manufacturing, with compatibility for 200 and 300 mm wafer processing. The platform builds upon technology originally developed by Solmates, a spin-off from the University of Twente that was later acquired by Lam Research. In parallel, advances in membrane release processes, low-temperature growth, transfer reliability, and wafer-scale bonding technologies are essential for transitioning oxide membranes from proof-of-concept demonstrations to manufacturable components. For example, CSD and non-destructive membrane release processes based on directional electrodynamic decomposition of sacrificial layers have enabled the fabrication of freestanding ferroelectric membranes up to 3 inches for epitaxial PbZrO_3 and 12 inches for polycrystalline films [136]. Furthermore, remote epitaxy has expanded beyond graphene-based interlayers, with amorphous interlayers enabling the production of 4-inch freestanding VO_2 membranes [179]. Together, these developments form a critical foundation for the scalable fabrication of ferroelectric oxide devices compatible with existing semiconductor platforms.

Concluding remarks

In summary, freestanding ferroelectric-oxide membranes are poised to play a pivotal role in the next generation fundamental research and device fabrication of complex electronic materials, offering an unprecedented combination of tunable lattice structure, reconfigurable functionality, and integration versatility. Addressing the remaining scientific and technological challenges will not only advance our fundamental understanding of ferroelectrics but also accelerate the translation of oxide-based materials into a broad range of emerging applications. Realizing this vision will require sustained innovation and

close collaboration across disciplines spanning materials science, applied physics, electrical engineering, and semiconductor manufacturing to fully harness the potential of these transformative systems.

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6.3. Oxide membranes in energy applications

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Status

Oxide materials are central for energy storage, conversion, and coupling of renewable energy to chemical and industrial processes, thus contributing to sustainability goals. They are attractive electrodes for various energy conversion and storage systems, e.g. oxygen evolution and reduction reaction (OER and ORR) in water electrolysis and fuel cells, selective oxidation reactions, supercapacitors, and batteries. Novel synthesis pathways to fabricate, tune, and integrate oxide membranes into such applications may therefore yield new opportunities to improve performances and to generate new electrode and device concepts.

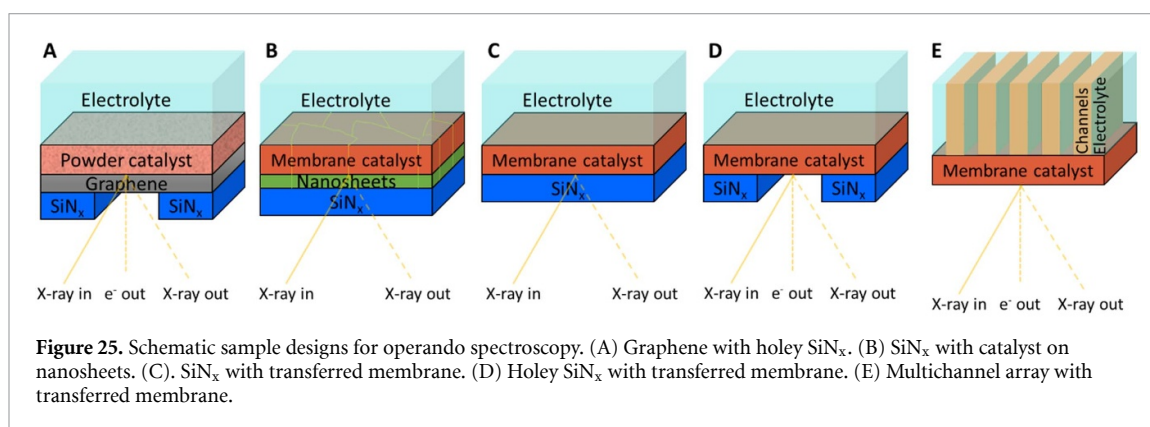
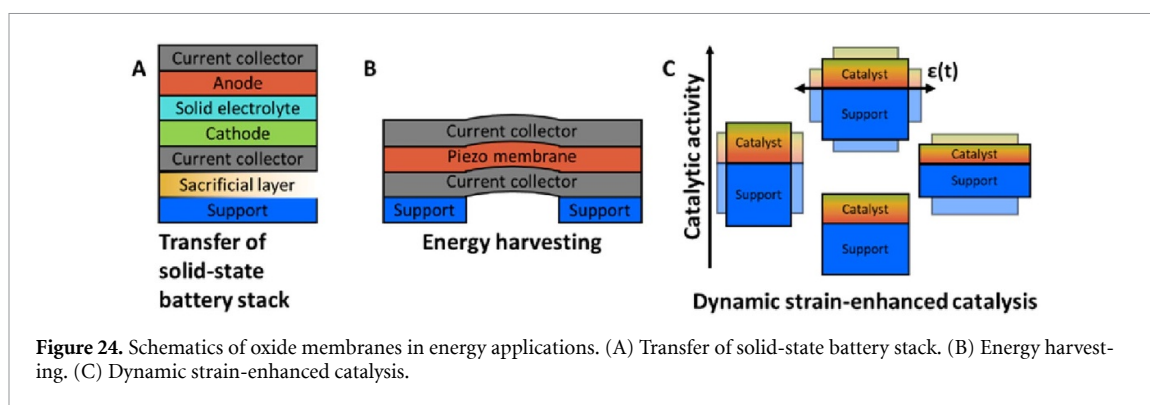
Both nanosheets with a thickness up to a nanometer and lateral size of several microns [252] (cf section 4.1) and membranes fabricated from a transfer with a sacrificial layer with a thickness of several nanometers and a lateral size of several millimeters [253] offer improved functionality in energy applications. These oxide membranes have unique 2D properties (high surface-to-volume ratios, tunable electronic and ionic properties, and mechanical flexibility) leveraging atomic-scale control. For example, metal oxyhydroxide nanosheets improve OER activity because of their high electrochemical surface area, enabling current densities of 1 A cm^{-2} at a cell voltage of only 1.62 V [254]. Similarly, ultrathin $\text{Ti}_2\text{Nb}_2\text{O}_9$ nanosheets arranged in an interconnected porous framework deliver a high sodium storage capacity of 250 mAh g^{-1} at 50 mA g^{-1} , due to ultrahigh surface area and fast diffusion within the expanded interlayer spacing [255]. Building on this concept, membranes can be used to design hybrid electrodes by stacking different materials like HTiNbO_5 and $\text{H-Ti}_3\text{C}_2\text{T}_x$ and/or twisting multilayers, which promise synergistic properties with optimized performance. These structures exploit synergistic interactions between the constituent layers, achieving capacities of 111.5 mAh g^{-1} at 5 A g^{-1} and maintaining 121.7 mAh g^{-1} at 2 A g^{-1} after 1000 cycles [32, 256].

Furthermore, membrane oxides may play a role in emerging applications like thin film microbatteries, enabling longer-lasting, miniaturized power sources for next-generation connected devices [257], using multilayer stacks and flexible substrates (figure 24(a)). For wearables, membranes also enable energy harvesting, e.g. via integrated piezoelectric MEMS (Microelectromechanical Systems, figure 24(b)) [258]. In novel applications modes, membrane oxides may even enable dynamic strain in electrocatalysts, where the adsorption energies of different intermediates, and therefore the catalytic activity are optimized by quickly varying strain (figure 24(c)) [259].

Besides directly improving performance in energy applications, membrane oxides enable advances in fundamental understanding of energy materials and processes, as they provide unique sample geometries for advanced experimental characterization, e.g. offering transparency for x-rays and electrons. In electrochemical reactions, there are dynamic changes in oxidation states, adsorbate coverage and electronic structure of the electrode or catalyst, and these dynamic properties determine the functionality. Accordingly, these properties must be measured under reaction conditions to derive fundamental structure-property-performance relationship [260]. A multitude of complementary scattering, scanning probe and spectroscopic operando techniques may be enabled by the use of oxide membranes. Here, we will focus on XAS, XPS and photoemission electron microscopy (PEEM). These allow probing both the chemical and electronic states under reaction conditions and they will benefit from the emergence of oxide membranes as detailed below. Importantly, oxide membranes can represent epitaxial model systems (atomically smooth, single-oriented surface and termination). These enable fine-tuning of physical and catalytic properties through strain, facetting and precise heterostructure engineering [261], and they are needed as ideal and well-defined starting points for the fundamental understanding using operando spectroscopy [262].

Current and future challenges

Despite the promises for energy applications, general challenges in the synthesis still play a huge role for future developments and actual implementation. High-quality, reproducible, scalable oxide membranes are required; but to date, membrane fracturing is still a major challenge. The effects of contamination and chemical changes occurring during the transfer process have not yet been investigated sufficiently,



and undesired leakage pathways for gases/liquids caused by crack formation have to be addressed. After synthesis, the interface quality and degradation on repeated stimulus (electrochemical cycles) is unknown and for hybrid electronic materials control over the synergistic interactions between mixed phases is required. At the same time, membrane transfer techniques may offer to integrate electrocatalytic oxides into novel environments such as industrial electrode supports (typically Ti or Ni supports), where sufficient adhesion, surface coverage and electrical contact has to be achieved. Overall, further developments in the transfer process and potential degradation pathways during application are required for both fundamental studies of and implementation in electrochemical systems.

The synthesis is particularly challenging for microbattery applications (figure 24(a)). These generally open the pathway for crystalline, highly ordered battery architectures using epitaxial or highly-oriented thin-film stacks, which promise better interface-control and therefore improved long-term stability. But transfer of multilayer stacks is generally more challenging due to built-in strain effects, and the compatibility of various battery-materials with the aqueous liquids used for dissolution of the sacrificial layer is questionable.

Regarding strain-tuning of catalytic activity, epitaxy-imposed static strain already revealed performance enhancement [263], but achievable strain is substantially larger in oxide membranes, and these promise to render dynamically varying strain states achievable (figure 24(c)) [35, 213]. Challenges lie in adhesion to the flexible substrate and understanding, controlling and optimizing the amount of (variable) strain applied during catalysis.

A promising direction lies in delamveraging oxide membranes to overcome limitations in operando spectroscopy. Conventional x-ray operando cell geometries are limited by the inability to probe through thick electrodes and require complex cell designs. Available cell designs are schematically depicted in figure 25, using the general approach to probe electrochemical interfaces from the backside of an x-ray transparent membrane. For XAS, electrodes supported by x-ray transparent SiN_x membranes (~ 100 nm thickness) offer operando capabilities due to relatively large probing depth of the x-rays, but the short inelastic mean free path of photoelectrons requires graphene as photoelectron-transparent membrane for XPS and PEEM of the solid/liquid interface [264, 265]. Here graphene functions as current collector and as a seal between liquid and high vacuum, enabled by its high mechanical stability.

Generally, such measurements are to date limited to feasibility studies with the electrochemically inert graphene itself or particle-based catalyst supported by the SiN_x or graphene membranes (figure 25(a)). We hypothesize that membrane oxides of a few nm thickness can add new insights when placed on SiN_x for XAS or replacing the graphene for XPS. In this manner, oxide membranes will simultaneously serve as catalyst, electrode and x-ray/photoelectron transparent window, decreasing system complexity and enabling the study of model-system, single-crystalline-like catalysts at the solid/liquid interfaces (figures 25(b)–(d)).

Advances in science and technology to meet challenges

A better understanding of the fundamental properties of oxide membranes is prerequisite for their integration into energy devices. This particularly implies the discovery new properties that may arise from finite-size and confinement of the membranes, the specific environment and interfaces forming between membranes and their support. To gain better control over membrane oxide fabrication, recent work has shown advanced understanding of the transfer process, by development of tailored sacrificial layers and fine-tuning the selection of all involved materials, their strain state and the involved electrolytes. For example, non-water based etching agents may be attractive for production and transfer of solid-state battery stacks. Furthermore, for many applications like operando spectroscopy and dynamic strain catalysis, sticking of the membrane to a new substrate plays an important role. To thoroughly investigate all relevant interfaces, cross-sectional scanning and TEM are highly effective techniques.

For current state-of-the-art operando spectroscopy there is a lot of development of new and improved cell designs at synchrotron facilities. Such cells are ideally suitable (with adaptations where necessary) for various electrochemical applications, including electrocatalysis of various reactions and charge storage. Cells used at different facilities should be developed to be interchangeable, calling for a community standard, and should be multi-modal (i.e. other spectroscopies or scattering possible in the same cell). Similarly, laboratory-based instruments increasingly also offer operando capabilities [266], and should therefore make use of the same community standard cell designs. All of this suggests benefits from integration or at least close communication between oxide membrane and characterization communities.

With smart development of cells also smart sample designs are required. As we recently introduced, highly oriented oxide membranes grown using the nanosheet-template approach discussed before [267] or transferred oxide membranes epitaxially grown on sacrificial layers are attractive as single-oriented model systems for operando characterization (figures 25(b) and (c)). Similarly, improved cell and sample design enabled scanning transmission x-ray microscopy on single-crystalline and freestanding thin film battery electrodes [268].

For operando XPS and PEEM, further developments are required. For example, oxide membranes can be transferred to holey SiN_x membranes, similar to what is already achieved with graphene (figure 25(d)) [269]. The bottleneck is the mechanical stability of the oxide membranes; they should be crack-free and leak tight to form a barrier between the liquid and vacuum. One potential approach is to transfer two oxide membranes avoiding overlaps of cracks from both layers, thereby minimizing evaporation. A combination with graphene (or other photoelectron transparent materials) is another option, since only few oxides are effective current collectors. In such a geometry, electrical contact between oxide membrane and graphene is very important. An alternative approach is the use of microchannel arrays, which have been applied successfully to limit the amount of electrolyte spillage and leakage in graphene-based cells (figure 25(e)) [264]. All in all, such dedicated cell and sample designs will enable understanding and further development of new and better energy materials.

Concluding remarks

Oxide membranes and membrane-derived energy devices reflect a promising field for future research, as the flexibility of materials selection, their combination and transfer to new environments, such as battery devices, electrochemical cells and experiment-specific sample holders, enables new opportunities to integrate more complex functional materials into existing and emerging applications. The unique 2D properties hold great potential for enhanced performances, and even to introduce fundamentally new approaches or applications. Advanced research methodology enabled by oxide membranes promises to foster a more detailed understanding of nanoscale phenomena in electrochemical environments. As such, the progress in the rational design of beyond state-of-the-art energy devices may highly profit from the advances in oxide membrane technologies.

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6.4. Tunable devices based on oxide membranes

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Status

All electronic/optoelectronic devices are composed of a multitude of layers with specific functionalities required for device performance. These layers need to be fabricated in a given stacking series by available semiconductor technology which limits the materials and phases that can be used to build a device. The most well-known limitation is the so-called CMOS (complementary metal oxide semiconductor) temperature budget which allows only growth temperatures of about 450 °C to 500 °C with decreasing tendency after the deposition of the semiconductor circuit BEOL. This fact is particularly important for crystalline perovskite oxides that harbor a huge variety of functional properties, going well beyond those attainable with classic semiconductors, e.g. ferroelectricity, ferromagnetism, metallic conductivity, as well as insulating, and semiconducting materials, all in one platform. These properties not only allow for pure electronic devices but give also access to a large range of optical components, including lasers. Unfortunately, they typically require growth temperatures well above the CMOS budget. As an example, LiNbO₃ plays an important role in modern communication technology as electro-optic material. Due to its large $\chi^{(2)}$ susceptibility [270] and transparency at 1550 nm, LiNbO₃ is the standard material for electro-optic phase shifters [271] and Mach-Zehnder Modulators [272]. Another example is the well-known ferro- and piezoelectric perovskite lead zirconate titanate (PZT) that has interesting properties as ferroelectric layer with switchable and tunable polarization [273]. Ferroelectricity and $\chi^{(2)}$ optical nonlinearities require a non-centrosymmetric crystal structure that can only be stabilized at elevated temperatures in perovskites. Novel device concepts like ferroelectric random access memories [274] or ferroelectric field-effect transistors [275] have been commercially realized but never made it out of niche areas due to the incompatibility of semiconducting and perovskite oxide materials. In this case, the unexpected discovery of a new material, ferroelectric HfO₂, that could be fabricated within the CMOS budget and was in other structural phases, amorphous and monoclinic, already part of CMOS technology, changed the focus of the field [276]. However, ferroelectric HfO₂ is not a prototypical ferroelectric material but most likely in a metastable phase that is difficult to reproduce on large scales. Another important example of a perovskite material is the tunable dielectric barium strontium titanate (BST), the material of choice for tunable capacitors (varactors), e.g. in antenna filters in mobile phones. Tunable para- and ferroelectric behavior relies on the possibility to displace negatively and positively charged ions with respect to each other by a DC or RF field, thereby changing the polarization of the materials. A tunable device can make use of the change of permittivity induced in both the paraelectric and ferroelectric state or a sign reversal of the ferroelectric polarization in a switching device. Further, ferroelectricity is always accompanied by pyroelectricity as well as a noteworthy optical $\chi^{(2)}$ non-linearity that can be utilized for infrared and optical device concepts as in shown in [271, 272].

While commercially ceramic BST thick film crystallites are used [277], it has been shown that highly crystalline thin films of BST can considerably increase the performance/quality factor of varactors [278], but the challenges of material integration are still the main obstacle towards application. Oxide membranes are currently under intense exploration to overcome the hurdle of compatibility between different process technologies by separating synthesis and device assembly. First results have shown that individual oxide functional layers of BST and oxide electrodes can be transferred without loss of functionality [279]. Even full device heterostructures of perovskites with well-defined, as-grown interfaces can be transferred in membranes while key device performance parameters remain unchanged [280]. If oxide membrane (layer transfer) technology can be established in combination with semiconductor devices as for example p-type oxides [281, 282], the vast family of (perovskite) oxides in combination with other 2D materials [8, 283] will enrich future oxide electronics making use of their unique functional properties.

Current and future challenges

The current challenge is to establish a reliable and reproducible layer transfer process that conserves the desired as-grown film properties or even alters its properties in a desired way, e.g. by stress or deliberate surface modification. Fundamental studies on how layer properties in individual thin layers of one or a few unit cells in height will be changed, as well as investigation of artificial heterostructures of layers from different materials that do not exist in nature and cannot be synthesized in any other way. Another

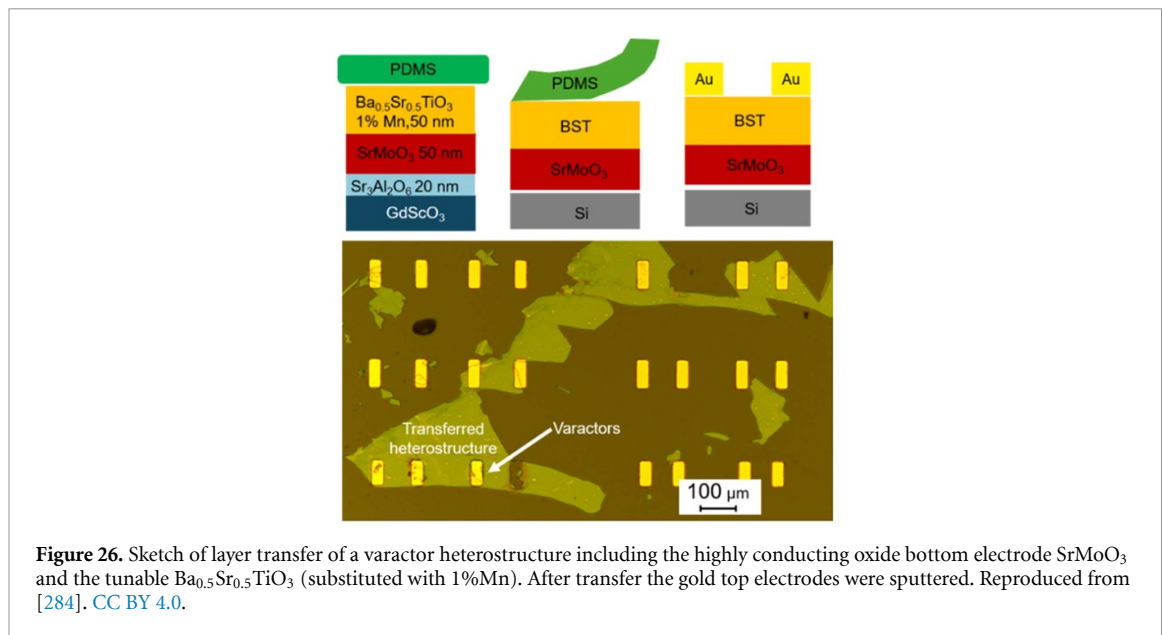


Figure 26. Sketch of layer transfer of a varactor heterostructure including the highly conducting oxide bottom electrode SrMoO_3 and the tunable $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ (substituted with 1%Mn). After transfer the gold top electrodes were sputtered. Reproduced from [284]. CC BY 4.0.

important degree of freedom is the possible rotation of layers during transfer, that would lead e.g. to vertically stacked twist grain boundaries. One famous example stems from vdW bonded graphene layers which even becomes superconducting if stacked under the ‘magic’ angle of 1.1° [44]. Further research focuses on the study of specific transfer defects such as cracks and wiggling. In materials science the view on defects is always twofold, following the questions: (i) Which defects occur and how do they alter material properties? (ii) How can such defects be used for novel devices? From the perspective of devices, the resulting challenge is how the transferred layers perform as functional part of the device. In case of tunable components, the obvious question is how the tunability is affected when a tunable dielectric such as $(\text{Ba}, \text{Sr})\text{TiO}_3$ is transferred. At the interfaces to the host structure, non-tunable dead layers may occur and also leakage currents can considerably increase. Figure 26 shows that the successful transfer of a complete device, in this case a varactor (but without top electrode), is possible and, indeed, the device properties remained almost unchanged (tunability, Q factor, leakage current) [284]. In this case, the bottom electrode and ferroelectric material were transferred jointly, the interface to the bottom electrode was thus protected. One of the key future requirements is the reproducible treatment of membranes before and after the insertion into a new host structure to reduce material defects occurring during the whole process. In case of oxide dielectrics, the membranes may suffer e.g. from oxygen vacancies, cracks, and surface adsorbates. Therefore, high-temperature annealing and plasma cleaning need to be included in the process chain. Another important question is the role of strain relaxation. The deviating (considerably lower) permittivity of thin films as compared to bulk of the same stoichiometry in $(\text{Ba}, \text{Sr})\text{TiO}_3$ often is attributed to strain and has to be compensated by varying the Ba to Sr ratio. An individually transferred layer of BST is relaxed except residual strain, which might contribute to improved devices.

In the long-term perspective, upscaling of transfer processes to the wafer level is a key issue that might decide about the large-scale applicability of oxide membranes besides niche products. The current state-of-the art achieves tens of micrometer-sized crack-free pieces of material and millimeter scale pieces with cracks. Micrometer-sized pieces are large enough for most electronic devices, but a technological process cannot rely on picking suitable flakes one by one. Another challenge in membrane transfer is the eventually required alignment of the membranes. Similar as in the case of 2D vdW materials, over time more and more sophisticated manipulators will be developed and transferred to start-up companies paving the way for controlled transfer and alignment procedures of multiple membranes.

Advances in science and technology to meet challenges

The key technological advancement required is a reproducible layer transfer process into devices. The technology development can build on recent advances in layer transfer of 2D vdW-bonded materials [285, 286]. However, it is important to note that there are key differences between vdW-bonding and oxide bonding, the latter being inherently three-dimensional. vdW layers can be easily peeled off by using scotch tape, while oxides require more complex chemical processes to break the strong covalent and ionic bonds. With the aid of sacrificial layers, oxides can be peeled off with controlled thickness

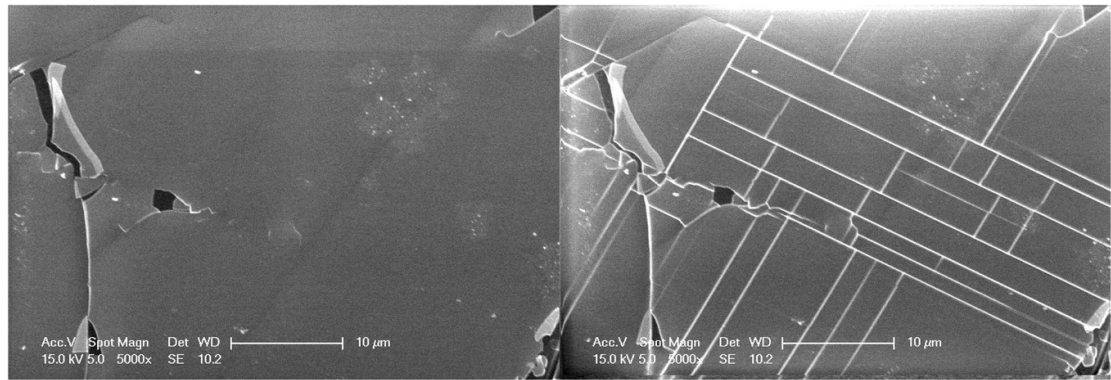


Figure 27. Left: $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ membrane on PDMS. Right: Surface cracks along specific directions appear in the same membrane after modest heating. Pre-defined cracking positions could be a step towards multiple membrane transfer on the wafer scale (sample and image by Josenhans, Prasanna, Kunkel, Cardoletti and Alff).

as required for the specific device. Therefore, vdW materials have advantages on the level of few unit cells, while (perovskite) oxides can be used for layers with thicknesses up to several 100 nm. While micromanipulators will be the first choice to drive fundamental research, automated processes need to be developed in the future that allow to transfer a large number of defined, defect-free membranes in one step. Depending on the individual requirements of the devices, cleaning processes might be essential for highest performance. Here, plasma cleaning or laser annealing have the potential to be incorporated in an automated work flow. Note that oxides have the inherent advantage that they are already oxidized at the surface (in contrast to vdW materials where surface oxidation is a critical issue). This fact could ease the requirements for additional cleaning processes. The most challenging aspect will be the upscaling of device yield. To our opinion, it is unlikely that crack-free lift off processes are achievable on the wafer scale (but we might be proven wrong). A route to go could be to use controlled cracking which occurs along preferential crystal directions and could be triggered by pre-structured substrates, potentially allowing to apply micro-stamping transfer techniques [273] with crack-free subunits (see figure 27). Note that massive parallel transfer using elastomer stamps is already a standard technology for semiconductor foundries and most likely will be adaptable to membrane technology. A combination of ion slicing (smart cut technique) and wafer bonding already lead to wafer-scale layer transfer of thin film lithium niobate onto silicon, yet a major enabler for the photonics industry [271]. The same technique can, in principle, be applied to other types of oxide membranes as well.

Concluding remarks

This section has been written having varactors and optical components in mind where perovskite-based components have already achieved technological maturity as thick films. In both cases, thin film oxide membranes offer the possibility to incorporate optimized materials in a device stack, partly in stacking sequences that are not at all available in conventional technology. In both cases, $(\text{Ba,Sr})\text{TiO}_3$ has proven to play a pivotal role as functional material [278, 305]. Oxide membrane technology has the potential to combine this perovskite with any other material platform such as CMOS technology, flexible substrates, and photonic integrated circuits. While BST might indeed become a key driver for membrane application-oriented research in the field, other (oxide) dielectrics/ferroelectrics could be easily introduced in an existing membrane technology. Beyond that, other available functionalities in perovskites such as magnetic and superconducting properties might also become available for complex circuits encompassing currently separated technologies.

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6.5. Freestanding oxides in integrated semiconductor circuits

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Status

Freestanding oxide thin films are emerging as a transformative class of materials in information technology, building on a rich history of oxide electronics research. While crystalline complex ferroelectric oxides have been integrated into semiconductor circuits in memory architectures at the front-end-of-line, high processing temperature incompatible with complementary metal-oxide-semiconductor (CMOS) devices, incompatibilities regarding substrate lattice-matching, thermal expansion, formation of unwanted interfacial layers and limited availability of suitable buffer layer materials are ongoing limitations of back-end-of-the-line (BEOL) integration [287, 288]. The utilization of FOMs grants the freedom to select the most optimal functional material without being constrained by the underlying substrate or devices. A major technological driver for this field is the pursuit of crystalline high-k perovskite oxides such as SrTiO₃ (STO) and BaTiO₃ (BTO) integrated as gate dielectrics in electronics [28, 289].

Freestanding oxides have been successfully integrated not only on conventional silicon [7, 290], but also on flexible platforms [27, 291] and with 2D materials [28], demonstrating the potential for new device form factors and improvements in device performance and versatility.

Where amorphous films, imperfect interfaces or growth-related damages limit transistor performance, single crystalline oxides offer outstanding performance in terms of equivalent oxide thickness (EOT). The introduced vdW-gap together with the ultra-high dielectric constant limit gate leakage, while superior interface quality suppresses interlayer formation and related trap formation. Resulting EOT below 2 nm and below 1 nm have been achieved for STO gated MoS₂ FETs with suitable subthreshold swing (SS) [28, 292].

Besides perovskites, single-crystalline HfO₂-based thin films have been integrated as high-k gate dielectrics in MoS₂ FETs, exhibiting additional ferroelectricity, figure 28(a). This integration achieved a subthreshold swing below 70 mV dec⁻¹, figures 28(b) and (c) with an EOT of 4.5 nm, attributed to the negative capacitance effect [293]. Although polycrystalline HfO₂, which is fabricated by post-annealing an amorphous ALD film with a low thermal budget, is CMOS-compatible and well-established, single-crystalline HfO₂-based thin films offer several advantages. Superior scaling potential, with demonstrated ferroelectricity down to 1 nm thickness and wake-up free switching are reported for crystalline HfO₂ with rhombohedral structure [294, 295]. The integration of freestanding membranes further mitigates the issue of direct damage to the 2D channels caused by ALD process of amorphous or polycrystalline HfO₂ and has already been successfully demonstrated with delaminated films down to 1 nm thickness [295, 296].

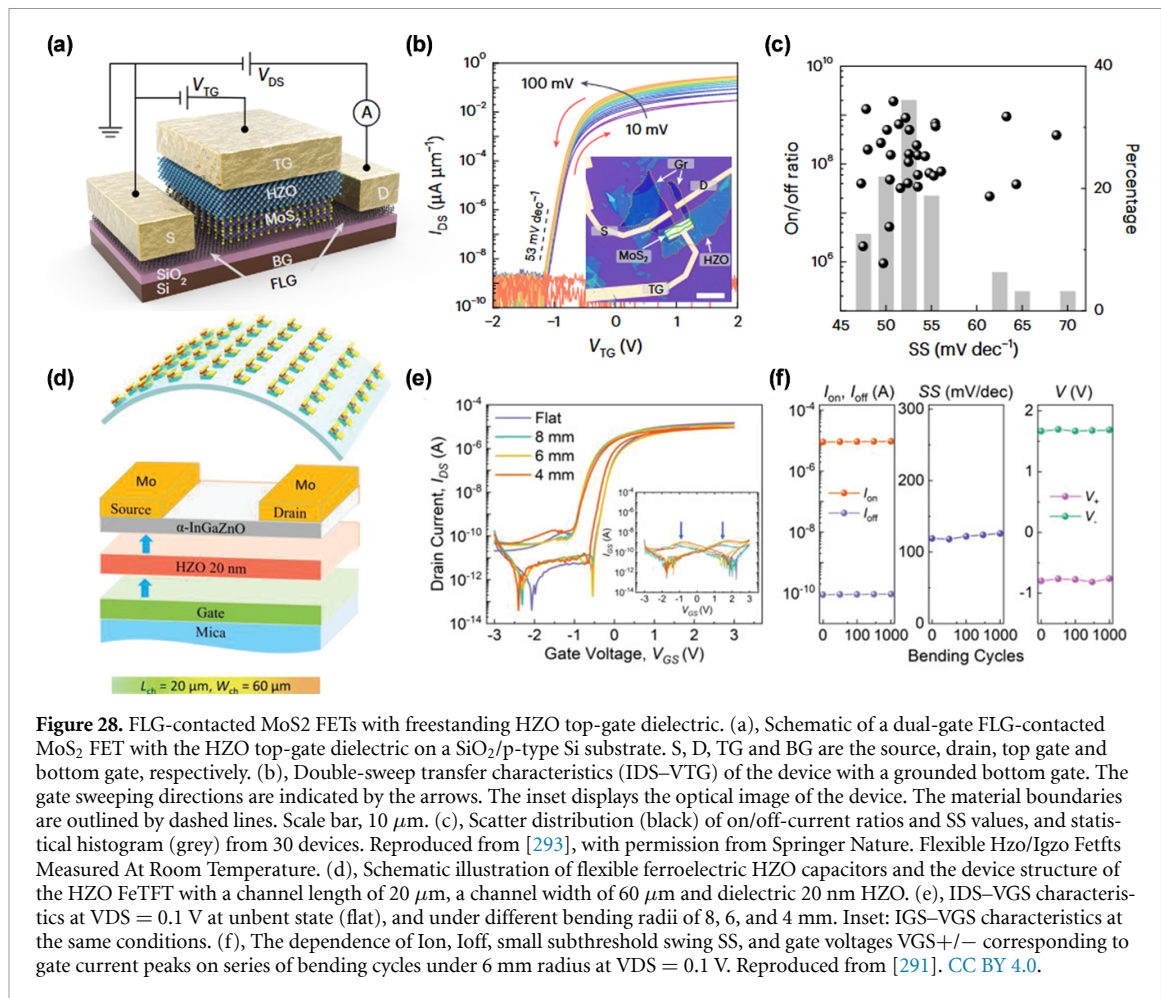
The de-clamping of ferroelectric thin films from their host substrate was found to enhance the domain wall mobility of BTO [297], or lead to coercive fields approaching single-crystal limits being among the lowest measured in BTO thin films [219]. Consequently, the integration of freestanding membranes can provide a route to ultra-low-voltage operation and faster switching dynamics in ferroelectric-based devices.

Another example of a metal oxide in which the crystalline phase plays a pivotal role in performance is the wide band-gap semiconductor β -Ga₂O₃, which is one of the most promising materials for power electronics. However, β -Ga₂O₃ has a relatively low thermal conductivity, which hinders the heat dissipation of power devices. Free-standing β -Ga₂O₃ offers the possibility of transferring high-quality crystalline structures to substrates with high thermal conductivity. Membranes of β -Ga₂O₃ have been successfully grown on single-crystal graphene by remote epitaxy and subsequently exfoliated [298].

Amorphous IGZO has been widely adopted as channel material, due to its high mobility. Nevertheless, recent work has demonstrated that crystalline IGZO or intermediate, nanocrystalline IGZO can provide advantages in carrier transport, thermal robustness, and device reliability [299, 300]. This bares great opportunity for freestanding membranes as crystallinity, or the degree of structural disorder can be tuned independently of underlying structures.

The second aspect that makes free-standing oxides particularly interesting is their mechanical flexibility, which makes them well-suited for integration into flexible electronics, such as wearables, and unlocks another path of new functionalities.

An early demonstration of HZO as flexible ferroelectric thin-film transistor demonstrated a promising SS of approx. 100 mV dec⁻¹, figures 28(d)–(f). The great mechanical stability is assigned to the



heteroepitaxy, where the weak vdW interaction between freestanding film and new substrate suffers little from the induced misfit strain and substrate clamping [291].

Overall, the field of fully integrated freestanding membranes is only at an early stage, rapid development and a wide expansion regarding possible material combinations, new properties and functionalities can be expected.

Current and future challenges

Due to the possibility of co-integrating complex crystal structures and materials in the form of oxide membranes with CMOS circuits, they offer a promising route toward the development of non-von Neumann computing systems, enabling novel functionalities and device concepts beyond traditional logic [299, 301].

Despite significant advances, the integration of freestanding oxides into very large scale integrated (VLSI) circuits or in power electronics remains constrained by several fundamental and engineering challenges. The fabrication of high-quality, wafer-scale freestanding membranes is particularly problematic.

The issue of sacrificial layers of the wet-etching approach represents another major bottleneck. Existing methods focus heavily on perovskite-based structures, with only a limited range of suitable sacrificial layers available. This severely narrows the material choices for freestanding membranes.

For the alternative remote epitaxy route via a 2D vdW material, e.g. graphene, the growth window for crystalline oxides is significantly reduced to avoid damaging the material [160].

Even when delamination is successful, transferring freestanding membranes to foreign substrates introduces additional challenges, leaving behind organic residues and inducing macroscopic defects, which disrupt subsequent growth and compromises device functionality, affecting long term stability and device-to-device variability. Continuous progress towards atomically precise interfaces is being made, approaching the short-term challenge of near defect-free transfer [302]. Long-term goal is the large-scale transfer and bonding of freestanding films without introducing macroscopic damage. This has not been demonstrated yet. Current approaches are usually focused on near-term opportunities, such as early

demonstrations and proofs of concept, limited to the stacking of individual thin film flakes, rendering them unsuitable for technology-relevant device manufacturing.

Further, there are indications that the removal of the substrate can induce significant structural and functional instabilities in the freestanding oxide itself.

At the interface of STO films the released energy of bond-breaking was found to cause a degradation of the crystalline order upon delamination of the ultra-thin film [126]. Similar mechanisms might be expected for other systems, too, when approaching ultra-scaled thicknesses.

Similarly, ferroelectric and domain-wall behavior have been found adversely affected, with reports showing orders of magnitude slower domain wall motion in BiFeO₃ (BFO), due to introduced inhomogeneities resulting from bond-breaking when the clamping effect of the host substrate is removed [303].

Collectively, these challenges, ranging from growth and transfer related material limitations to potential intrinsic structural instabilities, highlight that freestanding complex oxides still have to overcome major hurdles before a successful integration into VLSI circuits will be achieved.

Advances in science and technology to meet challenges

Overcoming the barriers to scalable and reliable integration of freestanding complex oxides into VLSI technology demands coordinated scientific and technological progress. A critical need is the development of residue-free, dry transfer techniques to reduce contamination, wrinkles, and damage during membrane handling. Here, the transfer of membranes including supportive structures like electrodes, thereby keeping critical interfaces unexposed, is a first step towards optimized stability and increased yield. The advantage of freestanding films to form sharp interfaces with the new host substrate needs to be achievable on large scale and at BEOL compatible temperatures to become relevant for information technology.

Alongside this, efforts must aim to move beyond flake-based devices toward scalable, reproducible fabrication of large-area freestanding membranes. To make such approaches industrially relevant, more automated fabrication, delamination and transfer methods compatible with existing semiconductor platforms will be essential. Complex processing schemes will need to be developed to address the specific requirements of free-standing oxides. Most critically, in order to meet scaling trends in VLSI, these schemes must also cover FinFET and gate-all-around FET geometries.

From a materials perspective, strain engineering and flexoelectric control can be key to mitigate or tune structural phase transitions in freestanding films with the goal of developing novel device concepts.

With respect to the co-integration of free-standing oxides with vdW materials in logic or memory devices, further research into the thermal and long-term stability of the vdW interfaces is needed to gain control over trap states and to ensure device reliability over extended operating time and cycles. Here, advanced characterization methods, such as operando TEM and strain mapping, can play a crucial role in real-time monitoring of structural integrity and defect formation during and after delamination and transfer.

Expanding the portfolio of usable materials for the freestanding membranes as well as sacrificial layers, beyond well-studied perovskites, is also vital, especially for diversifying functionalities.

Concluding remarks

Freestanding complex oxides hold transformative potential for next-generation computing architectures, offering excellent interface quality beyond the reach of traditional, substrate-bound systems.

Early demonstrations of integrated freestanding oxides already show promising performance. Further advances in scalable fabrication, defect management and membrane handling are key to achieving large-area integration with competitive transistor characteristics. This will bring these materials closer to commercial viability for use in flexible electronics and neuromorphic, low-power and ultra-scaled computing applications. Beyond replication of substrate-bound functionalities, the field should exploit newly discovered, distinct properties of freestanding membranes, such as altered ferroelectric and domain-wall behaviors, which can unlock entirely novel device concepts. Achieving these goals will be enabled by coordinated progress in materials engineering, growth techniques, and system-level integration.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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