

LaAlO₃/SrTiO₃ Heterointerface: 20 Years and Beyond

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This year marks the 20th anniversary of the discovery of LaAlO₃/SrTiO₃ (LAO/STO) oxide heterointerfaces. Since their discovery, transition metal oxide (TMO) interfaces have emerged as a fascinating and fast-growing area of research, offering a variety of unique and exotic physical properties which has provided a strong impetus for the rapid advances and actualization of oxide electronics. This review revisits the fundamental mechanisms accounting for the two-dimensional (2D) conducting interfaces, and how new models proposed to better account for the unique interfacial effects. Recent breakthroughs in the theoretical and experimental domains of oxide interfaces are also discussed including the detection and investigation of 2D quasiparticle. Moving beyond the well-known LAO/STO interface, this review delves into other systems where unconventional interfacial superconductivity, interfacial magnetism, and spin polarization are dealt with in greater detail. In terms of device applications, this review proceeds with a treatment on the recent developments in domains including field effect transistors and freestanding heterostructure membranes. By emphasizing the opportunities and challenges of integrating oxide interfaces with existing technologies, the review will end off with an outlook projecting the progress and the trajectory of this research domain in the years to come.

properties. The coupling between spin, charge, orbital, and lattice interactions in TMO has been found to play a fundamental role in a multitude of novel physical phenomena in condensed matter physics. These phenomena include metal-insulator transitions,^[1] interfacial superconductivity,^[2] 2D ferromagnetic and ferroelectric (FE) effects,^[3] spin-orbit orders and the list goes on.

This year marks 20th anniversary since the groundbreaking discovery of the interfacial 2D electron gas (2DEG) at the LAO/STO interface by Ohtomo et al.^[1] This quintessential interfacial system, a widely studied STO-based heterostructure, has served as a key platform for the study of strongly-correlated charge transport in low-dimensional systems and in the development of new oxide-based functional materials and integrating it with both established technological platforms and next-generation functional electronic devices. The progress made in the epitaxial growth techniques has progressively enabled the

synthesis of high-quality oxide heterointerfaces which in turn facilitated the exploration and understanding of strongly-correlated oxide heterointerfaces beyond the LAO/STO systems. Various other oxide interfaces have demonstrated remarkable properties

1. Introduction

Oxide interfaces have emerged as a fascinating and rapidly evolving field of research, offering a diverse range of unique and exotic

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and hold immense potential for novel device applications. These interfaces offer opportunities for developing magnetic storage devices, optoelectronic devices, spintronic devices, and other cutting-edge electronic devices.

A number of reviews have appropriately documented and summarized the progress made concerning the underlying mechanism governing the transport and other novel physical phenomena of the interfacial 2DEG systems and the efforts made in functionalizing these properties in diverse device applications.^[4] At the fundamental level, Gariglio and co-workers have provided a comprehensive review concerning the formation of interfacial 2DEG where special emphasis is made on the superconductive states and carrier confinement properties.^[4a] In addition, Sulpizio et al. have provided an in-depth summary of the important contributions made by nanoprobe in the understanding and the control of the oxide interfaces at the nanostructural level.^[4b] Works by Pai et al. and Christensen et al. have also delved into the diverse physical structures and quantum phenomena in STO-based heterostructures and discussed how these properties are regulated by external stimuli.^[4c,e] Huang et al. further provided a comprehensive review on the interfacial engineering strategies which allows reader to grasp the crucial role these strategies play in the development of next-generation electronic devices.^[4d] A recent review by Kornblum focuses on detailing recent developments in field-effect devices based on conducting oxide interfaces.^[4f] Ever since the publications of these reviews, significant advances have been made and the scope of oxide interfaces in the aspects of applications continue to expand. Apart from the propositions of new theoretical models and updates to existing ones to better account for the onset of interfacial 2DEG, new experimental evidence to substantiate the respective theoretical models, breakthroughs in the actualization and device applications of oxide interfaces, continue to be reported on a regular basis. Hence, this review would facilitate a deeper understanding of the physical mechanisms underlying the interfacial phenomena and foster exploration of the next generation of oxide electronic devices, which transcends the traditional LAO/STO system.

2. Mechanisms Governing the Interface Physics: Conventional Models and Updates

Since the first report of the 2DEG present at the LAO/STO interface, the origin and the underlying mechanisms governing these unique interfacial phenomena remains the focus of ongoing research and debate. Multiple complementary factors that are not mutually exclusive including charge-transfer, lattice distortions, electronic reconstructions, and interfacial coupling, have been proposed to account for this unique quantum effect absent from their individual parent components.

As one of the earliest and the most well-known models, the polar catastrophe model involves the electronic reconstruction caused by the polar discontinuity at the interface.^[1] As the number of LAO unit cell (u.c.) increases, there is a corresponding linear increase in the surface potential, which quickly leads to a non-physical surface potential (Figure 1a). Nevertheless, the *polar catastrophe* can be avoided with the transfer of $0.5 e^-$ u.c.⁻¹ from

the LAO layer surface to the LAO/STO interface, thereby resulting in the formation of the interfacial 2DEG (Figure 1b).^[5] This model has been successful in accounting for the metal-insulator transition and the emergence of interfacial conductivity at a critical thickness (L_c) of 4 u.c.^[8]

With the inevitable presence of oxygen vacancies in the STO layer during the synthesis process, they play a direct role dictating the interfacial charge concentration and mobility as evidenced in Figure 1c where the resistivity of samples synthesized under different oxygen pressures are displayed.^[6,9] The presence of oxygen vacancies further affects the structural and electronic properties of the interface, leading to variations in conductivity and superconductivity.^[10]

The cation mixing model (non-stoichiometric model) is another proposed mechanism where the interfacial intermixing/diffusion of cations (La and Sr ions for the LAO/STO system) results in the formation and charge dynamics of the interfacial 2DEG (Figure 1d).^[11] It is also noteworthy that the effects of charge transfer and electronic reconstruction play an indirect part in contributing to this cation mixing effect.^[12]

2.1. Limitations to Mechanisms Accounting for Conducting Interface

While these mechanisms have been successful to a certain extent in accounting for the interfacial 2DEG effects, there exist severe shortcomings which must be duly addressed. Particularly in the case of the polar catastrophe model, even though it can explain the formation of the 2DEG at the interface via the polar discontinuity between the LAO and STO layers,^[1,12b,13] it does not fully account for other emerging interfacial phenomena such as superconductivity and magnetism. With its primary focus on the electronic charge redistribution,^[14] this model tends to overlook the essential role that oxygen vacancies play in generating the interfacial 2DEG. Besides, there is still a significant mismatch between the magnitude of the theoretically predicted charge transfer with experimental reports. Specifically, while calculations based on the polar catastrophe model predicted a charge transfer of $0.5 e^-$ u.c.⁻¹ to fully compensate the polar potential in the LAO layers,^[5] the transfer of electrons from surface oxygen vacancies,^[15] can also achieve this which in turn results in an insulating LAO surface and a conducting LAO/STO interface. Even though high-energy optical measurements have confirmed the $0.5e^-$ u.c.⁻¹ charge transfer, transport measurements only account for $\approx 0.05e^-$ u.c.⁻¹ at the conducting LAO/STO interfaces.^[2,3,8,16] this is a measly 10% of the expected charge transfer contributes to the interfacial conductivity. One of the key shortcomings of the polar catastrophe model is that it predicts an incremental rise of 2DEG concentration with increasing LAO film thickness (n_{LAO}), which is not the case in actual experimental measurements. Instead, experimental study has revealed a sudden emergence of interfacial 2DEG at a L_c of 4 u.c. followed by a saturation of the interfacial charge concentration. It is essential to highlight that while the polar catastrophe model can account for the L_c of 4 u.c., it falls short in explaining the subsequent saturation of interfacial 2DEG concentration.^[17]

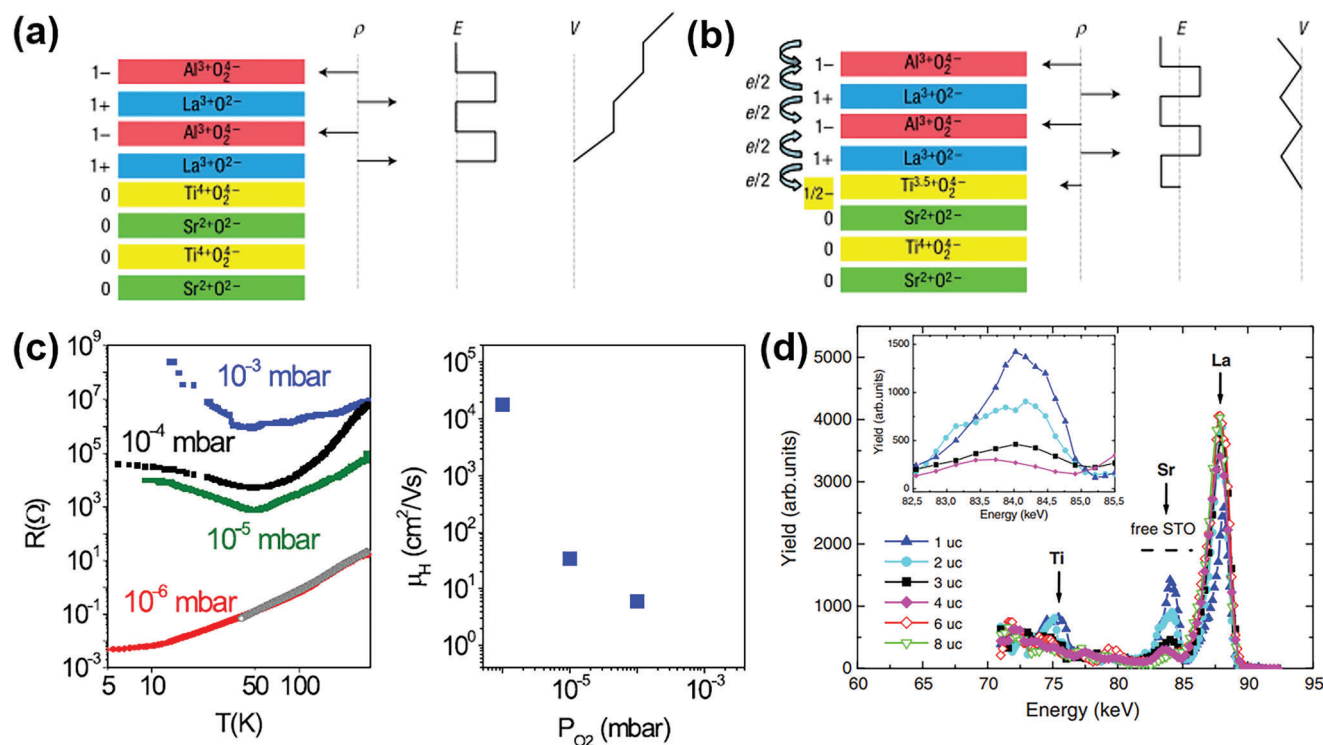


Figure 1. a) Neutrally charged (001) plane in the STO substrate while alternating net charges, σ , existing in the LAO(001) plane. The presence of the $\text{AlO}_2/\text{LaO}/\text{TiO}_2$ interface plane creates a non-negative electric field, resulting in the divergence of the potential, V , with increasing thickness of LAO. b) Polarization divergence is circumvented through electronic reconstruction, a transfer of $0.5e$ to the LAO/STO interface from the top LAO layer. c) Left: Temperature-dependent resistivity of samples grown under different oxygen pressures. Right: Relationship between interface mobility at 4 K and deposition pressure. d) Medium-energy ion spectroscopy observing the relationship between the interfacial ion mixing as a function of LAO thickness. The inset shows a local magnified view of the Sr peak at LAO thicknesses below 4 u.c. a,b) are reproduced with permission.^[5] Copyright 2006, Nature Publishing Group. c) is reproduced with permission.^[6] Copyright 2007, American Physical Society. d) is reproduced with permission.^[11b] Copyright 2009, American Physical Society.

As for the oxygen vacancy (V_{O}) model, it not only provides an explanation for the presence of the 2DEG by considering the role of oxygen vacancies in trapping additional electrons, but is also able to account for the 2D interfacial conductivity and some aspects of the electronic behavior. Nevertheless, studies have suggested that V_{O} concentration in the STO substrate is grossly insufficient to account for the observed 2DEG charge density and has to be compensated by vacancies from the LAO overlayer.^[18] In addition, the V_{O} model alone may not be sufficient to fully account for why the L_{c} is required for the interfacial conductivity to take place.^[12a] The coalescence of isolated interfacial conductive patches formed by the inhomogeneous distributions of oxygen vacancies could also account for the presence of a critical thickness for interfacial conductivity to take effect.^[19]

Meanwhile, the cation intermixing model have attracted considerable attention where the cation antisite defects between the LAO and STO can undergo site exchange and their relative proportions,^[11a,b] which is able, in principle, nullify the polar field and while promoting the interface 2DEG conductivity via electron doping.^[20] Nevertheless, deliberate attempts to dope the LAO/STO interface with atoms such as Cd and Mn have failed to generate any obvious metallic behavior.^[21] Instead, with the alloyed LAO/STO reported to remain insulating,^[22] this

raises further questions on the validity of the cationic mixing mechanism.

2.2. Mechanism of Surface Oxygen Vacancy Defects in Polar Field-Assisted Formation

Among the relatively conventional physical mechanisms mentioned above, each has certain shortcomings that cannot fully explain all experimentally observed physical phenomena. The interfacial conductivity at the LAO/STO interface and the mechanism governing the insulator-metal transition remain unexplained. Recently, a new model has been proposed to explain the origin of interfacial 2D conductance.^[15b,23] This newly proposed model still attributes the polar discontinuity as the driving force for the generation of interface physical phenomena, but the charge transfer mechanism is not Zener breakdown. Rather, the formation of oxygen vacancies at a L_{c} of 4 u.c. occurs at the LAO surface layer instead of at the interface. For each neutral V_{O} , two electrons are generated and transferred to the interface, causing an interfacial physical phenomenon.^[15b]

Although the polar catastrophe model and oxygen vacancies have been the focus of several theoretical studies, the relationship between defect generation and polarity has not been fully

demonstrated theoretically. Numerous studies have investigated the formation of interfacial defects assisted by polar field-assisted formation from different theoretical perspectives.^[10,15b,23,24] In addition to the oxygen vacancies in the interface, early theoretical calculations have shown that another possible source of electron doping could be the oxygen vacancies in the LAO surface layer,^[10a,24] which has a non-negligible effect on the interface. The formation of oxygen vacancies in the LAO surface layer results in the formation of gap states, which leads to the transfer of surface electrons to the STO conduction band at the interface to form the interfacial 2DEG, which also cancels out the interfacial discontinuity induced by the built-in field. This mechanism accounts for why core orbit shifts as expected in the polar discontinuity model was not observed.^[25] This physical mechanism was further confirmed both theoretically and experimentally.^[23,26] Lemal et al. revisited the polar field-driven mechanism at the origin of the 2DEG using first-principles calculations.^[23] They discovered that LAO surface oxygen vacancies were typically more stable at lower LAO layer thicknesses than at the onset of Zener breakdown. Therefore, Zener breakdown is unlikely to occur first for the standard growth conditions of LAO/STO heterostructures. Instead, the polar field-driven surface V_O modelling mechanism may be more suitable to explain the physical origin of the interface in polar LAO/STO heterostructures.^[23]

Recently, Song et al. further confirmed that the surface oxygen vacancies in the LAO layer are the most probable source of interfacial physics through a comprehensive scanning transmission electron microscopy (STEM) analysis combined with density functional theory (DFT) calculations.^[27] In addition, Yang et al. revealed the coexistence of surface oxygen vacancies and interfacial conductive states in LAO/STO by a combination of grazing angle resonance soft X-ray scattering and first-principles calculations.^[15c] It is shown that the polar field-assisted formation of surface V_O defects models plays a crucial role in determining the electronic properties of the oxide interface. The V_O defect mechanism has received more and more positive feedback as a formation mechanism of the interface physics and is highly compatible with most of the experimental observations, which is worthy of further in-depth study.

3. Theoretical and Experimental Breakthroughs in Multiple Facets

3.1. Computational Studies in the Modelling of Oxide Heterointerfaces

The development of theoretical models and computational modelling continue to play a crucial role in understanding the complex behavior of oxide interfaces. DFT based first-principles calculations,^[28] and Monte Carlo^[29] simulations are employed to investigate the electronic,^[29] magnetic, and transport properties of oxide interfacial systems that transcends the LAO/STO system. In the recent years, these theoretical approaches have made significant progress in providing invaluable insights into the underlying mechanisms and guide experimental investigations.

While the polarization catastrophe model and V_O have been at the focal point of multiple theoretical studies, the relationship between defect generation and polarity has not been theoretically

well-established. Multiple studies have emerged on the polarization field-assisted formation of interfacial electron sources from different theoretical perspectives.^[10a,c,15a,23,26,30] Beginning with the distinguishing behavior between the insulating *p*-type and conductive *n*-type LAO/STO interfaces,^[24] further progress have been made on the subject relating between with first-principles study at the atomic scale revealing a unified mechanism leading to interfacial conductivity and magnetism (**Figure 2**).^[15b,31] This breakthrough provides the very fundamental understanding that instead of electronic reconstruction, polarity field induced via defect formation holds the key to the onset of interfacial conductivity and magnetism.

3.2. A Different Perspective: Interfacial Structural Discontinuity

While spotlight is generally on the aspect of polar discontinuity, it is surprising that structural discontinuity at the LAO/STO interface is generally overlooked as the cause of the interfacial 2DEG. While the STO substrate presents itself in the undistorted cubic phase, the LAO layer comes in the form of antiferrodistortive (AFD) tilt of the AlO_6 octahedron at room temperature.^[32] As the AFD rotations in the LAO layer cannot coexist with FE polar distortion in most perovskite oxide systems, they are likely to be suppressed below the L_c of 4 u.c. On this regard, it is predicted that a built-in field will be generated to strongly polarize the LAO lattice, thereby resulting in a depolarized field.^[18a,33] It is then verified by Gazquez et al. in a study combining STEM and first-principles calculations that the strong competition between octahedral tilts and polar displacements results in the emergence of simultaneous reconstructions in the LAO/STO system (**Figure 3a**).^[34] There is a crossover from a bulk-like LAO structure with AFD rotations to a strongly polarized state without any AFD tilts at ≈ 3 u.c. (**Figure 3b,c**). This unveils the onset of a structural transition (at a L_c of ≈ 3 u.c.) that takes place apart from the electronic reconstruction typically reported. Interestingly, it is further highlighted that the results provide conclusive evidence in support of the “polar-catastrophe” model with the collective changes in structural distortion and electronic reconstruction at the LAO/STO interface.

By combining STEM analyses and DFT calculations, another study by Song et al.,^[27] showed that polar field-assisted formation of oxygen vacancies at the LAO/STO surface is a pivotal player in the formation of the interfacial 2DEG. This formation of oxygen vacancies is observed to occur concurrently with a localized structural transition (structural symmetry breaking) in the LAO layer that induces the AFD rotation while contributing electrons at the LAO/STO interface.^[27] While the FE distortion is favored below 4 u.c., surface V_O are formed simultaneously with a structural transition to the AFD octahedral rotation in the LAO layer above the L_c of 4 u.c. Their study revealed that the inhibition of AFD octahedral rotation in the LAO layer favors FE distortion below the L_c , whereas surface oxygen vacancies are formed above the L_c , leading to the development of FE distortion. On the same note, Min et al. further highlighted the interplay between interfacial lattice strain and polar field that results in the emergence of the AFD and FE structures in the LAO/STO system.^[28b] Specifically, the distribution of V_O distribution is intimately related to both these structural modes. Nevertheless, unlike the work by Song

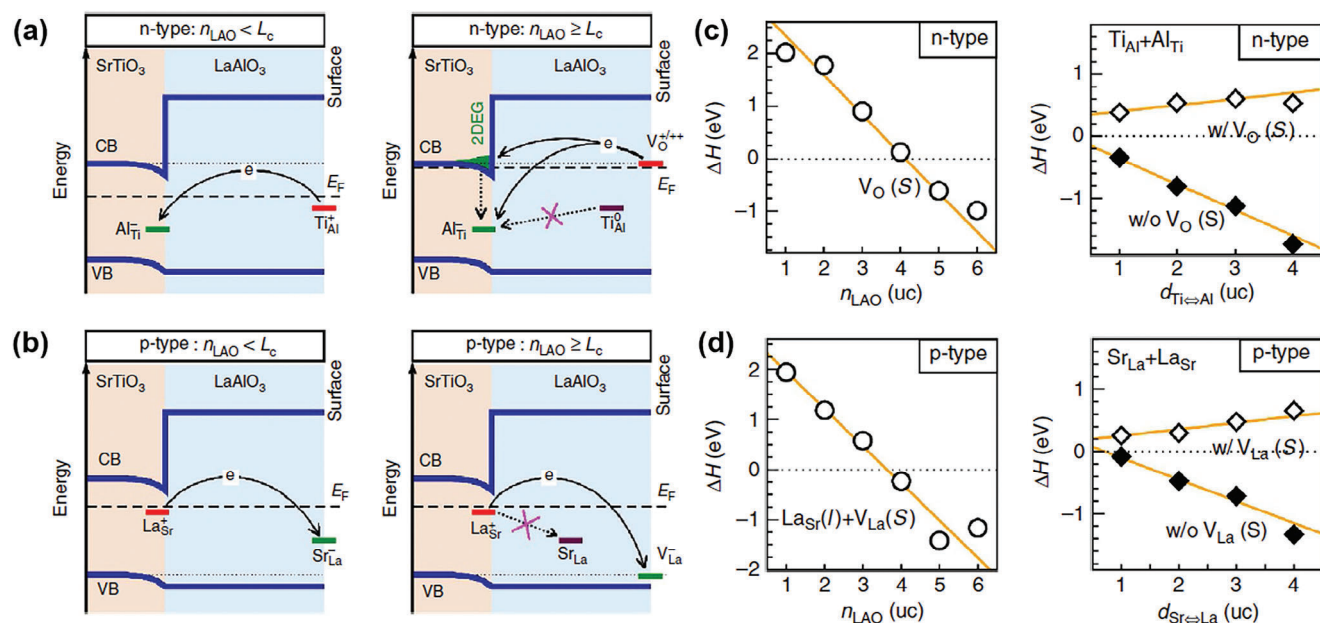


Figure 2. a) Left: All electrons transferred from $\text{Ti}_{\text{Al}}(\text{S})$ are captured by $\text{Al}_{\text{Ti}}(\text{I})$, preventing the formation of a 2DEG in n -type LAO/STO interfaces with $n_{\text{LAO}} < L_c$; Right: In n -type LAO/STO interfaces with $n_{\text{LAO}} \geq L_c$, surface V_O defects transfer 0.5 electrons to the interface, partially captured by $\text{Al}_{\text{Ti}}(\text{I})$, leading to the emergence of an interface 2DEG. b) Left and Right: In p -type interfaces with $n_{\text{LAO}} < L_c$ and $n_{\text{LAO}} \geq L_c$, all electrons transferred from $\text{La}_{\text{Sr}}(\text{I})$ are captured by $\text{Sr}_{\text{La}}(\text{S})$ and $\text{V}_{\text{La}}(\text{S})$ respectively, resulting in the absence of free carriers due to defects. c) Left: Variation of ΔH for n -type interface surface V_O defects under oxygen-rich conditions with n_{LAO} ; Right: Influence of $[\text{Ti}_{\text{Al}} + \text{Al}_{\text{Ti}}]$ defect pairs produced by n -type interface Ti-Al exchange, with and without surface V_O , on ΔH . d) Left: Relationship between ΔH of p -type interface $[\text{La}_{\text{Sr}}(\text{I}) + \text{V}_{\text{La}}(\text{S})]$ defect complexes and n_{LAO} ; Right: ΔH of $[\text{Sr}_{\text{La}} + \text{La}_{\text{Sr}}]$ defect pairs generated by ideal p -type interface La-Sr exchange, with and without $\text{V}_{\text{La}}(\text{S})$. Reproduced with permission.^[15b] Copyright 2014, Macmillan Publishers Limited.

et al., they reported the coexistence of the AFD and FE phases below 4 u.c. but disappear simultaneously at 4 u.c. and above. Both electron energy loss spectroscopy (EELS) and DFT calculations provide direct evidence for the formation of oxygen vacancies on the LAO (111) surface, which serves as the source of the interfacial 2DEG.

A comprehensive theoretical model depicting the coherent interplay between the concepts of electron reconstruction, lattice distortion, and surface oxygen vacancies is thereafter proposed by Zhou et al.^[15a] Specifically, the DFT study shows that lattice distortion and charge redistribution between the LaO and AlO_2 sublayers play a dominant role in the insulating state, while the conductivity and discontinuous transition at the LAO/STO interface are caused by the spontaneous appearance of $1/4 V_\text{O}$ and $0.5e$ charge transfer to the interface at each AlO_2 sublayer on the LAO surface. With interfaces synthesized under different growth conditions, this theoretical model is able to explain the varying properties appropriately.^[15a] Recent grazing-angle resonant soft X-ray scattering measurements by Yang et al. on the conducting LAO/STO interface has revealed the existence of surface oxygen vacancies.^[15c]

In attempt to draw a clear relation between theoretical models and experimental conditions governing the synthesis conditions, a recent DFT study by Li et al. has further reported the inevitable presence of highly concentrated oxygen vacancies on the surface of polar-nonpolar LAO/STO.^[35] Under chemical equilibrium conditions with parameters regulating the carrier density, LAO thickness, oxygen pressure and synthesis temperature, it has been shown that heterostructures with a LAO thickness of

above 3 u.c. will inevitably possess high V_O even under high oxygen pressure condition. The density of oxygen vacancies and carriers depends mainly on the thickness of the LAO and has little relationship with oxygen pressure and temperature, which also indicates the absence of intrinsic doping.

3.3. The Roles and Effects of Interfacial Electrolyte Field and Interfacial Charge Localization

When considering the role that oxygen vacancies play in the onset of 2DEG at oxide heterostructure interfaces, electrolyte gating is considered a viable means to investigate this phenomenon particularly due to its capability to tune the system's interfacial carrier density and to modulate any emerging novel interfacial phenomena. Besides, this gating process is an effective overlap between practical device applications and the quest to unravel the mechanism underlying the effects of oxygen migration and electrostatic charging at oxide heterointerfaces.^[36] The use of ionic liquids in electronic double layer transistors for the application of electric field has significantly enhanced the effectiveness of gating dielectrics and led to reports of novel phase transition processes in a diverse range of materials.^[37] As a quantum leap towards oxide-based electronics, carrier mobility in oxide heterostructures such as LAO/STO could be significantly enhanced via oxygen electromigration during ionic liquid-gating process while a chemically inert layer is inserted to protect the oxide surfaces.^[38]

By investigating the effects of electrolyte gating on the oxygen-deficient LAO/STO interfaces, Zeng et al. provided new insights

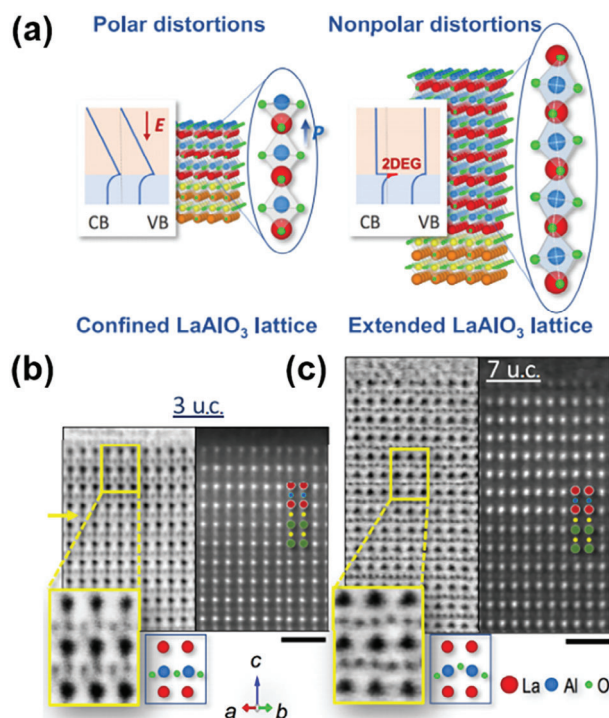


Figure 3. a) Schematic depicting the structure in the LAO layer below (left panel) and above (right panel) the L_c of 4 u.c. b,c) STEM images of 3 and 7 u.c. LAO/STO interfaces, respectively. While there are slight polar distortions at 3 u.c. thickness, AFD modes are observed at 7 u.c. thickness. Reproduced with permission.^[34] Copyright 2017, American Physical Society.

to the gating mechanism for buried oxides and the effects of oxygen electromigration along with its influence on the band structure at the LAO/STO interface (see Figure 4a,b for the device pattern and sample schematic, respectively).^[39] The electrolyte gating process results in the selective and irreversible filling of V_O because of oxygen electromigration at the amorphous LAO/STO interface. Not only does this process enhance the electron mobility and quantum oscillation of the conductance, but the filling of the V_O also leads to a significant transformation in the interfacial band structure. By further varying the crystallinity of the LAO layer, the V_O filling process via electro-gating can also be regulated.

In general, the interfacial 2DEG density can be modulated via the effects of localization and delocalization, of which, the effects of external stimuli in the form of electric field effect,^[41] control of LAO stoichiometry,^[42] oxygen partial pressure, temperature regulation,^[8,39] and laser irradiation play an critical role.^[43] While the effects of external stimuli may be extensive, it is necessary to account for the mode in which 2DEG is localized/delocalized and to explain the role that interfacial orbital hybridization plays in the regulation of the 2DEG concentration. Hence, as a follow-up study to the one by Zeng et al., Tang et al. employed X-ray Absorption Spectroscopy (XAS) to investigate the temperature-dependent properties and the effects of ionic liquid-gating on the interfacial 2DEG.^[40] Specifically, a significant reduction in 2DEG with decreasing temperature is attributed to the localiza-

tion of charges at the interfacial $O2p$ - $Ti3d(e_g)$ and $O2p$ - $Sr4d$ states that arise due to the interfacial orbital hybridization between the LAO and STO layer (Figure 4c). As observed in the temperature-dependent O K-edge spectra in Figure 4d, the dip in specific spectral regions with decreasing temperature indicates the filling of unoccupied interfacial 2D electronic states by the interfacial electrons in the interfacial hybridization states at the respective spectral region. While temperature control is a reversible method in regulating the charge localization processes at the respective interfacial states, this study serves as a confirmation on the work by Zeng et al, where ionic liquid-gating process can effectively and irreversibly modulate the interfacial carrier density of oxide heterostructures.^[36,39,44]

4. Interfacial Quasiparticle Dynamics

As a highly complex system with multiple interplay between charge, lattice, spin and orbital dynamics, quantum quasiparticles have inevitably emerged from the coupling between these degrees of freedom at oxide heterostructures and interfaces.

In the interfaces with partially filled d -orbitals, long-range electron-electron and electron-phonon correlations tend to play an influential role in the interfacial charge dynamics. Apart from the aforementioned onset of in-plane conductivity, superconductivity, and ferromagnetism, quasiparticle effects such as plasmon excitations, polaron dynamics and even density waves have been reported.

Plasmons are collective excitations of charge density, which is governed by a global long-range interaction between the electrons and they represent an elementary excitation for the Fermi liquid.^[45] After the theoretical prediction of plasmon features at the LAO/STO interface by Park and Millis,^[14a] Ruot-salainen et al. then employed non-resonant inelastic x-ray scattering and detected plasmon excitation on top of other forms of excitations including interband and semicore in the LAO/STO heterostructures.^[46]

In a relatively more complex multi-layer system, Faridi and Asgari provided a theoretical examination of plasmon excitations in the graphene-LAO/STO system which comprises an interface at between the graphene and the oxide heterostructure and also that at the LAO/STO interface.^[47] Unlike the typical LAO/STO interface with one optical and two acoustic plasmon dispersion modes, the compound graphene-LAO/STO system has an additional acoustic mode. There is a critical interlayer distance above which this acoustic plasmon mode could emerge.

Beyond LAO/STO, it has been noted that interfacial 2DEGs are favorable platforms for surface plasmon polaritons (SPP).^[48] Furthermore, with the onset of SPP and increase in electromagnetic field, properties of the 2DEG and the constituent materials could be significantly altered.^[49] Apart from studies of plasmons in transition conducting oxides such as indium-tin-oxide (ITO) and ITO-coated $LiNbO_3$ (LNO/ITO),^[50] the combination of a conducting oxide and one with good FE property with high polarity also serves as a good interface with formation of SPP. Hence, FE oxides would include $Sr_{1-x}Ba_xNb_2O_6$ and its close relatives $Ba_{2-x}Sr_xK_{1-y}Na_yNb_5O_{15}$,^[51] $LiTaO_3$,^[52] $BaTiO_3$,^[53] $KNbO_3$,^[54] and lanthanum-modified lead zirconate titanate,^[55] along with doped ZnO materials^[56] serve as good interfacial plasmonic materials.

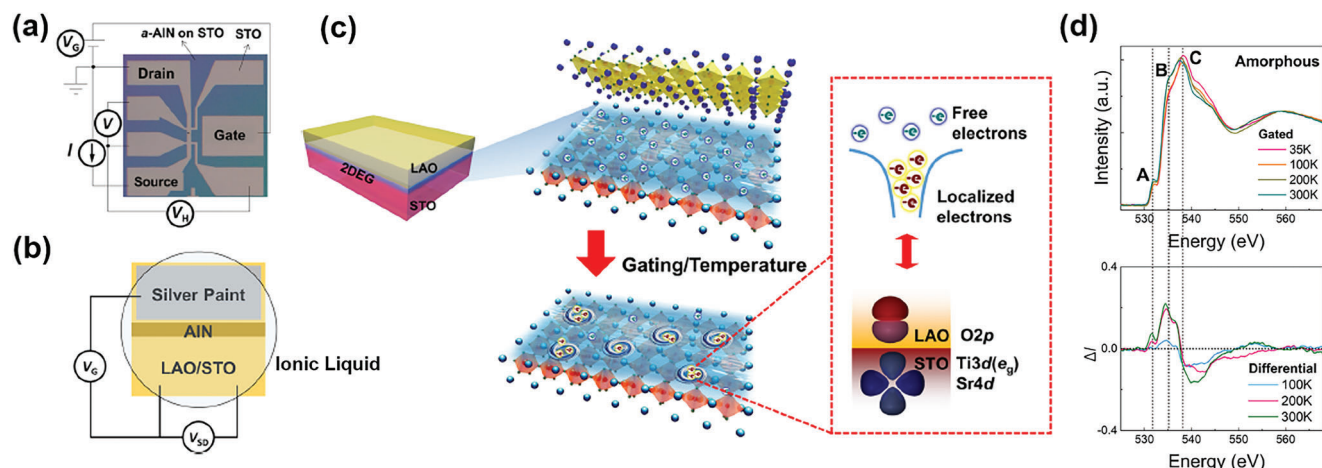


Figure 4. a) Optical micrograph, and b) schematic diagram of large-area samples of the LAO/STO device measurement circuit. c) Schematic depicting the 2D charges at the LAO/STO interface that can be reversibly regulated by temperature modulation or irreversibly controlled by ionic liquid-gating processes. Inset: Temperature reduction or ionic liquid-gating leads to the localization of 2D charges at the respective interfacial hybridized states. d) Temperature-dependent XAS analyses of pristine state amorphous 4.0 nm-LAO/STO. Reproduced with permission.^[40] Copyright 2022, AIP Publishing.

Polarons are quasiparticles formed as a result of coupling between excess electrons or holes with phonon vibrations. Depending on the scale of charge-lattice interaction and the spatial extent of polarization, the polarons formed are generally classified into small and large polarons. With their ease of formation, polarons can be detected in a range of systems including metal oxides,^[57] manganites,^[58] cuprates and 2D materials.^[59] Besides, polarons are known to exert significant effects on the systems' charge transport,^[60] surface reactivity and even on the systems' multiferroic properties.^[61]

The use of soft-X-ray angle-resolved photoelectron spectroscopy (ARPES) by Cancellieri et al. allows one to probe the charge carrier dynamics at the buried LAO/STO interface to identify the presence of interfacial large polarons (Figure 5a). By analysing the experimental spectral function,^[62] the formation of interfacial polaronic metal state is found to involve coupling with two active phonons – the hard longitudinal optical, LO₃ phonon and the soft transverse optical, TO₁ phonon (Figure 5b). Furthermore, the coupling between each active phonon components has a direct impact on the interfacial transport properties at different temperature range. At a relatively low temperature regime of $T > \approx 100$ K, while charge coupling with the LO₃-mode has a greater impact on the 2DEG mobility, coupling with the coupling with the TO₁ mode increasingly strengthens with rising temperature. Nevertheless, at sufficiently high density at low temperature, LO phonon scattering tends to be completely screened and the charge dynamics and mobility becomes dominated by electron scattering.^[63] A subsequent study by Geondzhian et al. compared bulk STO and LAO/STO interface and showed that the large polaron dynamics play a domineering role in their charge dynamics at low temperature.^[64] By studying the Ti L₃ edge resonant inelastic x-ray scattering (RIXS), the large polarons are manifested via intense *d-d* phonon excitations (Figure 5c). Besides, with increasing conductivity of the STO or LAO/STO system, the electron coupling with the LO₃ mode is found to be weakened correspondingly.

As highlighted earlier of a gross mismatch between theoretically predicted interfacial charge concentration of $0.5e \text{ u.c.}^{-1}$ transfer to the LAO/STO interface^[5,15a,b] and that of a measly $0.05 e \text{ u.c.}^{-1}$ by transport measurements.^[2,3,8,16] This is a clear indication that a significant proportion of the excess interfacial 2DEG has been localized with a tiny fraction left contributing to the actual interfacial transport properties. The formation of the more localized small polarons proves to be a key reason to account for this gross mismatch. It was first predicted by Kong et al. that 50% of the excess interfacial electrons are localized near the Ti-lattice sites to form small polarons, where they contribute very little to the interfacial conductivity.^[66] They further reported that while the high-localized polarons are spin-polarized, which however do not make any considerable contribution to the interfacial long-range magnetic ordering due to very weak coupling between neighboring polaron sites.

The experimental observation of the interfacial small polarons was subsequently reported in separate experimental studies. By employing high-resolution spectroscopic ellipsometry, a highly sensitive and non-destructive optical technique, Tang et al. reported the observation of small polarons at the LAO/STO interface (Figure 5d).^[65] By combining the experimental investigation with first-principles calculations, this work not only confirms that the polarons are 2D in nature (Figure 5e,f), but also shows that the hard longitudinal optical phonon mode, LO₃, is determined to play a pivotal role in the formation of this 2D small polaron in addition to its role in the formation of the interfacial large polarons.^[62] In attempt to draw a link between insulator-to-metal transition and the polaron dynamics of the LAO/STO conducting interface, Liu et al. employed sum-frequency phonon spectroscopy, an interface-specific and sensitive nonlinear optical technique, where an electronic reconstruction alongside strong polaronic responses are detected.^[67] Above the L_c of 3 u.c. LAO, the interfacial phonon mode at ≈ 101 meV highly-sensitive to localized STO lattice structure interacts strongly with the 2DEG –

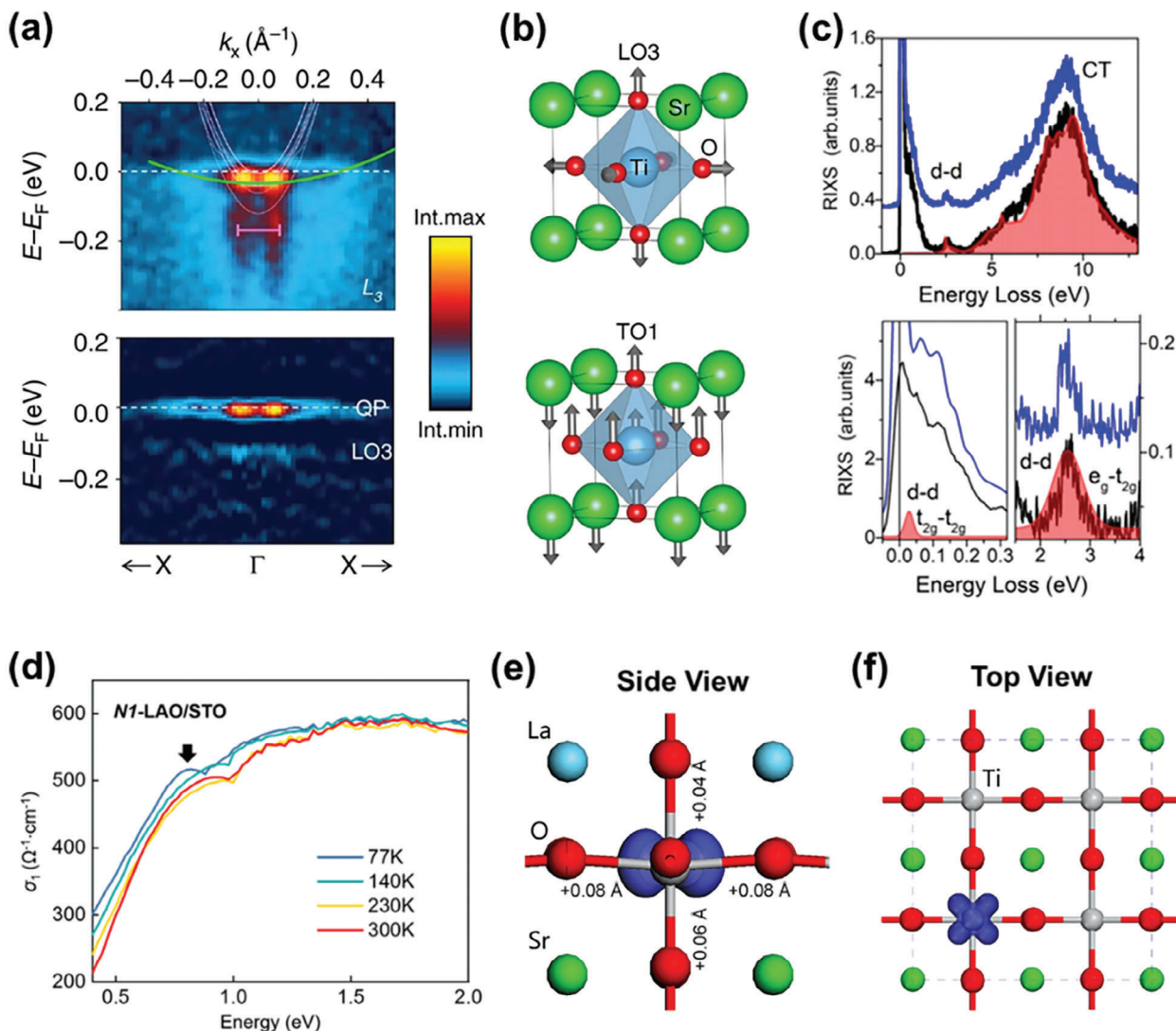


Figure 5. Experimental observations of both large and small polarons at the LAO/STO interface. a) High-resolution ARPES images along the TX line at the Ti L_3 -edge of the LAO/STO interface. Second derivative, $-d^2I/dE^2 > 0$ plot (bottom) clearly showing the quasi-particle peak attributed to the interfacial large polarons. b) Atomic displacements associated with the LO3 (top) and TO1 (bottom) phonon modes which make major contributions to the formation of the interfacial large polarons. c) Top panel: RIXS spectra of STO (black) and bilayer LAO/STO (blue) displaying the presence of $dd+$ excitation along with the charge transfer peak. Bottom Panel: Expanded view of the (left) low- and (right) mid-energy regions of the RIXS spectra where an additional intra- t_{2g} dd peak at ≈ 30 meV can be stimulated by atomic multiplet calculations. d) Optical conductivity, σ_1 , of LAO/STO elucidated from spectroscopic ellipsometry measurements where a near infrared feature (see arrow) has been identified as the interfacial small polarons. e) side, and f) top view of the LAO/STO interfacial structure overlaid with charge distribution of the small polaron states. Bond distortions provide further evidence of the interfacial small polarons. a,b) are reproduced under terms of the CC-BY license.^[62] Copyright 2016, The Authors, published by Springer Nature. c) is reproduced with permission.^[64] Copyright 2020, American Physical Society. d–f) are reproduced with permission.^[65] Copyright 2023, AIP Publishing.

a clear signature of small polaron at the LAO/STO conductive interface.

The detection of small polarons is important in two aspects. First, it accounts for a significant proportion ($\approx 50\%$) of the interfacial 2DEG interacting with the Ti-lattice to form the small polaron state, thereby explaining partially the mismatch between theoretical charge transfer and experimentally-derived transport measurements. In addition, by considering the many-body interactions beyond the LAO/STO interface, the presence of small polaron dynamics holds important implications on how quasiparticle dynamics mediates insulator-to-metal phase transition pro-

cesses and superconductivity in complex heterointerfaces including perovskite oxides and magic-angle twisted bilayer graphene where lattice distortion invariably breaks the periodic lattice symmetry.^[68]

5. Beyond LAO/STO

5.1. Unconventional Interfacial Superconductivity

Beyond the traditional LAO/STO interface, the quest to unlock and create novel states at oxide heterostructures have continue

to gain new grounds especially considering the rapid progress and advances in the synthesis, modelling and simulation of the systems at the atomic scale. Superconductivity that arises due to inversion symmetry breaking and strong electron-electron and electron-lattice interactions at the heterointerfaces is a particular case in point.^[69]

The observation of heterostructure superconductivity at the LAO/STO interface at $T_c \approx 250$ mK marks the beginning of this extensive investigation and discussion.^[2] The coexistence of interfacial ferromagnetism^[70] and the ability to gate-tune this superconductive phase^[41c,71] provides critical evidences that such interfacial superconductive phases are inherently unconventional and non-trivial.^[72]

The discovery of 2D superconductivity at the KTaO₃(KTO) interfaces with either an EuO or LAO top layer has marked a significant breakthrough beyond the conventional LAO/STO interface.^[75] This is of significant interest because of the contribution of the Ta5*d* electrons taking part in the interfacial orbital hybridization effects.^[76] Not only has a T_c of up to 2.2 K been attained – an order of magnitude higher than the ≈ 250 mK reported in the LAO/STO interface, but its superconductive property is also anisotropic in nature where it displays a strong dependence on the KTO substrate orientation. For instance, while 2DEG has been demonstrated at the LAO/KTO(001)^[77] and LaTiO₃/KTO(001)^[78] interfaces, no superconductivity has been reported with the 001-orientation KTO. It is further reported by Liu et al. in the same work that there is a spontaneous occurrence of an in-plane transport anisotropy before the superconductive phase sets in for EuO/KTO(111). This phenomenon may mark an emergence of a “stripe”-like charge order in this interfacial system. It is followed by a separate demonstration by Chen et al. that the LAO/KTO(111) interface can be tuned primarily from the superconductive into insulating phases by applying a gate voltage, V_G , across the KTO substrate instead of regulating the interfacial 2DEG concentration. This yielded a dome-shaped T_c - V_G phase diagram and with charge gating posing strong effect on mobility instead of on the charge concentration, this can be associated with the spatial profile of the interfacial charges and the effective disorder of the system.^[79]

In addition to the KTO(111) system, Chen et al. then reported the onset of superconductivity at the 2D LAO/KTO(110) interface albeit at a relatively lower $T_c \approx 0.9$ K with the superconducting layer thickness and the coherence length estimated at ≈ 8 and ≈ 30 nm, respectively (Figure 6a,b).^[73] Based on temperature-dependent sheet resistance, R_{sheet} , measurements under in-plane and out-of-plane magnetic fields, Hua et al. demonstrating the onset of 2D superconducting behavior of $T_c \approx 1.06$ K at the EuO/KTO(110) interface (Figure 6c–e).^[74]

While the discovery of superconductivity at KTO-based heterointerfaces is significant, it is particularly noteworthy that the superconducting properties in KTO systems are distinct from that of STO-based structures despite the common properties of these two substrates.^[80] It has earlier been predicted by Kozii et al. that the interplay between the strong spin-orbit coupling (SOC) effects and electron-electron correlations results in the onset of interfacial unconventional superconductivity alongside a mixture of spin-singlet and spin-triplet components.^[81] Specifically in the case of KTO-based superconducting interfaces, the Ta5*d*-orbitals exert a strong influence on the SOC properties on KTO-based

heterointerfaces on top of the interfacial electron-electron and electron-lattice interactions.^[82]

The presence of significantly larger SOC effects than that of STO has already been highlighted even in KTO(001)-based heterostructures.^[83] For instance, the observation of a considerable variation of Rashba SOC by Zhang et al. and a much larger maximum Rashba spin-splitting energy, Δ_{SO} , of ≈ 30 meV than that of STO-based heterostructures has been reported^[83b,84] while the SOC strength at the 2D EuO/KTO(110) interface can be varied with changes in band filling and the $\Delta_{\text{SO}} \approx 20$ meV reported.^[74] In addition, by depositing Al metal on KTO(001) single crystals, Arche et al. have reported the formation of an AlO_x/KTO interface where an interfacial Rashba parameter of $\alpha_R \approx -70$ meV Å⁻¹ can be obtained by bilinear magnetoresistance experiments, which is two to three times higher in absolute value than the value of the STO interface 2DEG.^[85] Recently, by investigating how Rashba SOC varies with LAO thickness at the LAO/KTO(111) interface, Liu et al. reported that interfaces with thicker LAO overlayers simultaneously exhibit a higher 2D interfacial carrier density but with a lower carrier mobility, thereby suggesting a tuning effect on the spatial confinement of the 2DEG.^[86] This in turn suggests a strengthened 2DEG spatial confinement favors a large Rashba SOC with a maximum of $\Delta_{\text{SO}} \approx 48$ meV.

Even though the studies presented above suggest the strong influence of SOC in controlling and regulating the unconventional superconductive properties at the KTO heterointerfaces, a clear causal relationship is not fully established between SOC and superconductivity. In the first place, no consensus has been arrived at concerning the primary superconducting mechanism underlying the KTO heterostructures. For instance, the “stripe”-like charge order at the EuO/KTO(111) interface could possibly be a superconducting phase,^[75] where its anisotropic transport properties could be a manifestation of the rotational symmetry breaking of superconductive phase in ferromagnetic EuO.^[87] In a study involving the YAlO₃/KTO(111) interface, Zhang et al. reported the observation of spontaneous rotational symmetry breaking with $T_c \approx 1.86$ K.^[88] While both the magnetoresistance and superconducting in-plane field display prominent two-fold symmetric oscillation within the superconducting state, such anisotropic behaviors disappear in the normal state. These are clear indications that such anisotropic property with in-plane rotational symmetry breaking is a fundamental property of superconducting YAlO₃/KTO(111) heterointerface, where it could be further categorized as a mixed-parity superconductor with a combination of both *s*- and *p*-wave pairing components. Apart from the above heterostructures, similar 2D superconductivity has also been observed at the interfaces of KTO with other epitaxial oxide heterostructures including AlO_x,^[89] TiO_x,^[90] LaMnO₃,^[91] LaSrMnO₃,^[92] and Hf_{0.5}Zr_{0.5}O₂.^[93]

5.2. Interfacial Magnetism and Spin Polarization

Emergent interfacial properties arise with the radical reconstruction of the interfacial band structures at oxide interfaces. Inherently non-magnetic insulators as separate entities, the emergence of ferromagnetism,^[3a] Rashba SOC,^[94] and the coexistence of the magnetic states with superconductivity^[70a,95] could

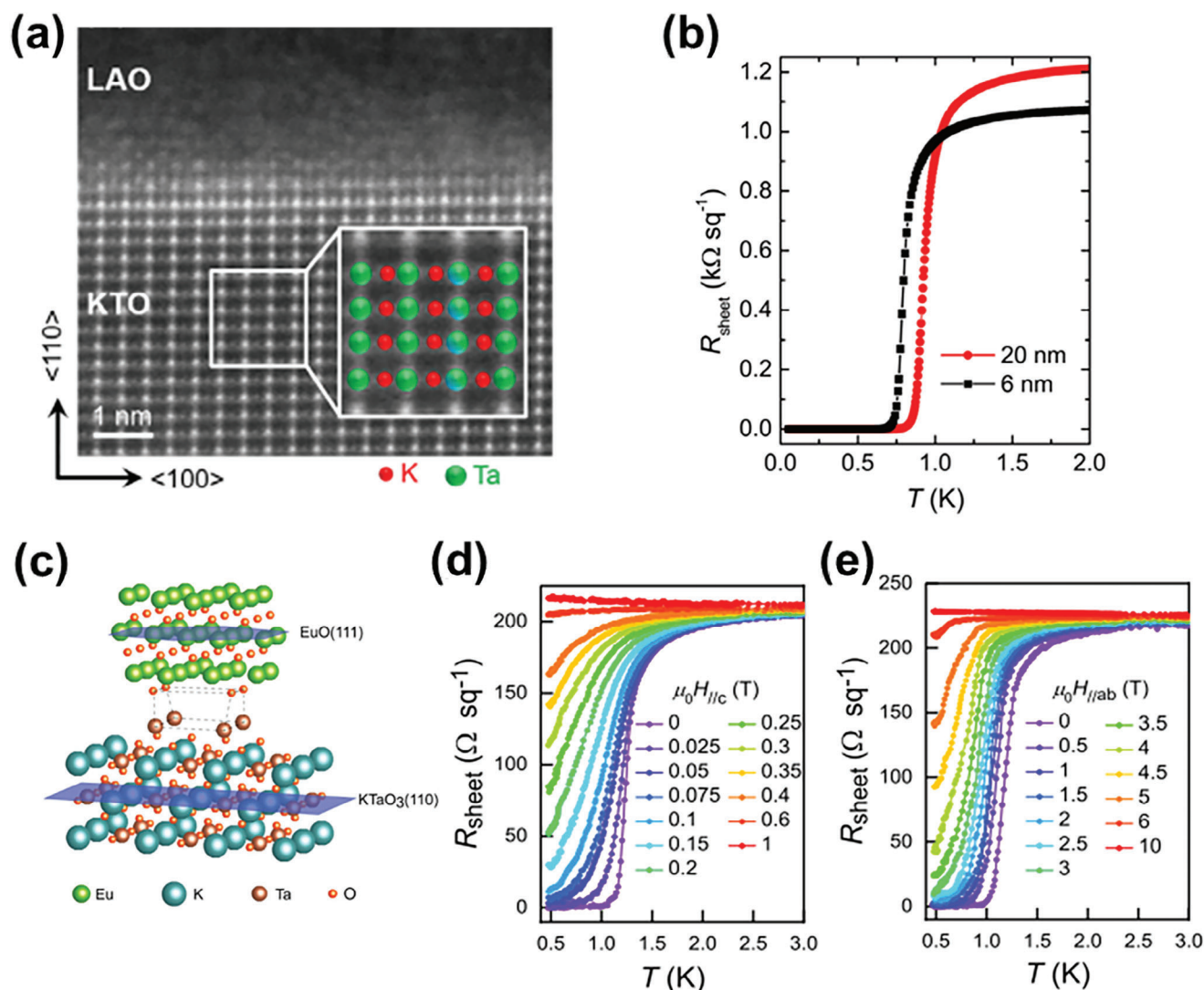


Figure 6. a) STEM image of 20-nm LAO/KTO(110) heterostructure interface. Inset: enlarged image depicting the atomic configuration of the KTO substrate layer. b) Temperature dependence of sheet resistance (R_{sheet}) for 6 and 20 nm LAO/KTO(110) interfaces at low temperatures, revealing distinct superconducting transitions with transition temperatures of 0.8 and 0.93 K, respectively. c) Schematic of epitaxial EuO(111) on KTO(110) (EuO/KTO(110)). d, e) Temperature-dependent R_{sheet} for EuO/KTO(110) interfaces under in-plane and out-of-plane magnetic fields, demonstrating 2D superconducting behavior. a, b) are reproduced with permission.^[73] Copyright 2021, American Physical Society. c–e) are reproduced with permission.^[74] Copyright 2022, Springer Nature.

possibly be the remarkable and completely unexpected properties that arise from the LAO/STO conducting interface. This can come in the form of charge-induced polarization where the polarity mismatch between the constituent layers results in a substantial lattice deformation at the interface.^[33b] As a consequence of crystal-field effect, the partially occupied $\text{Ti } t_{2g}$ -orbitals undergo a major band renormalization.^[96] Alternatively, the strong correlations between charge, spin-orbital degrees of freedom, modulates the charge density at the oxide interfaces and lead to such spin-orbital polarization phenomena. Moreover, this can enhance the ferromagnetic spin polarization at the oxide interface.^[97]

The spin-polarized interfacial 2DEG can be exploited in spintronic applications such as magnetic memory and spin-logic devices. As briefly described previously, Rashba SOC is gener-

ated by inverse symmetry breaking and is surprisingly functional in the control of electronic states at oxide interfaces on demand.^[98] The Rashba SOC is easily tunable and can be maximized in the topological region that avoids d_{xy} and d_{xz}/d_{yz} sub-band intersections.^[99] This has been demonstrated both theoretically and experimentally.^[84a,100] Today, in-depth research into the underlying Rashba physics is at the heart of spintronics, where SOC serves as the key parameter that controls non-equilibrium properties. In this case, information can be processed by manipulating the interplay between charge and spin degrees of freedom. For instance, Rashba SOC at the oxide heterostructure can achieve an efficient inter-conversion between spin and charge currents. Specifically, while charge current can be converted to spin current by direct Edelstein effect, the opposite process

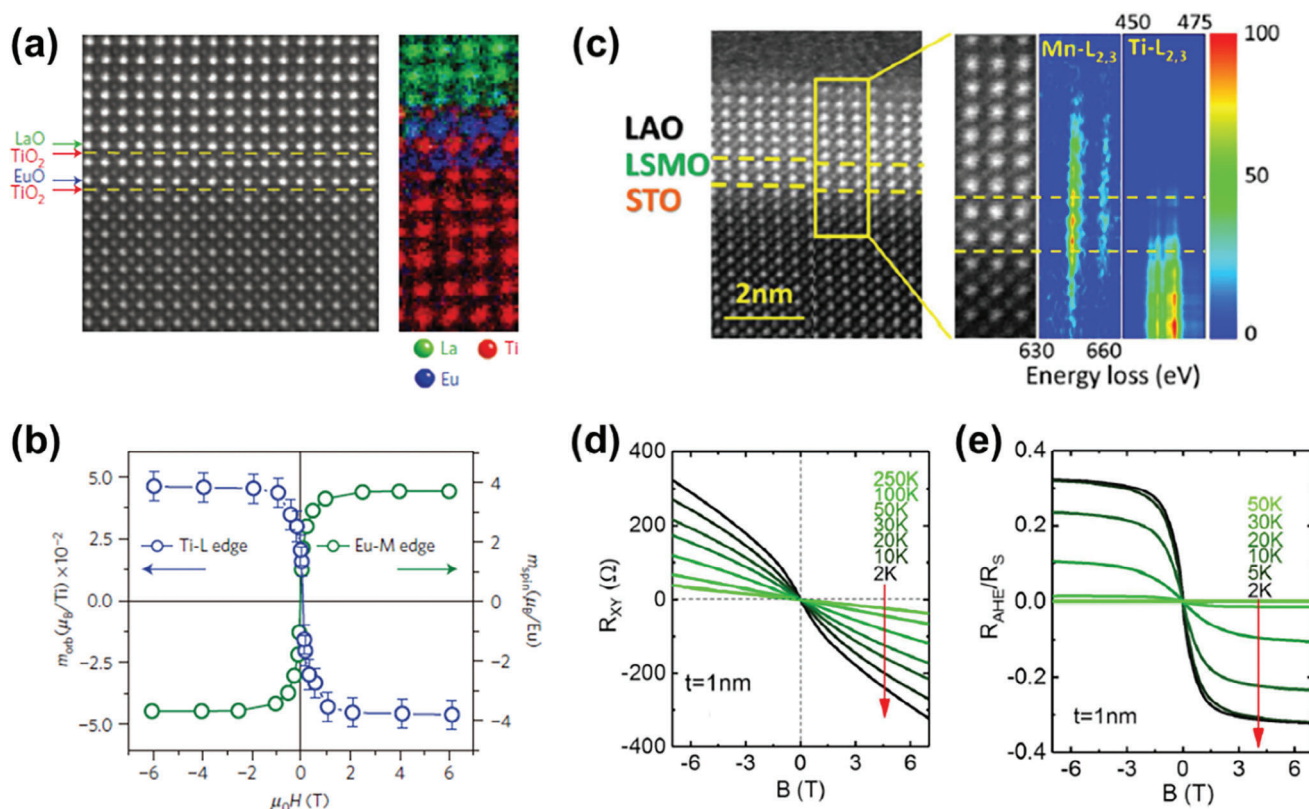


Figure 7. a) STEM image of the LAO/ETO/STO heterostructure detailing the specific locations of the atomic layers (left) and the EELS elemental mapping (right). b) Magnetic field dependence of the Eu spin moment (m_{spin}) and Ti orbital moment (m_{orb}), as derived via XMCD measurements of the heterostructure. c) STEM image of the LAO/LSMO/STO heterostructure along with the EELS mapping of the Mn-L_{2,3} and Ti-L_{2,3} edges. d) Magnetic field dependent Hall resistance (R_{xy}), and e) Anomalous Hall effect of the LAO/LSMO/STO interface in the temperature range between 2–250 K. a, b) are reproduced with permission.^[107] Copyright 2016, Macmillan Publishers Limited. c–e) are reproduced with permission.^[108] Copyright 2017, American Physical Society.

takes place via inverse Edelstein effect.^[101] This process of spin-charge conversion is particularly attractive in spintronic applications since it does not require the use of ferromagnets where spin precession can be manually operated in the absence of stray fields induced by ferromagnets. In addition, the magnetization of adjacent ferromagnetic layers in magnetic random-access memory devices can also be electrically controlled by the spin torque parameter.

Ferromagnetic metals are typically applied in conventional spintronics to achieve spin-charge conversion where it is usually induced by the exchange interaction between carriers and local spins. In the context of oxide interfaces, subsequent studies have revealed that this conversion can also be realized through Rashba effect taking place at the interface. Caviglia et al. were the first to observe this phenomenon at the LAO/STO interface.^[102] In which case, they uncovered a significant interfacial Rashba spin-orbit coupling (SOC) that is induced by the breaking of inversion symmetry in the interface space. This effect can be further modulated by applying an external electric field as evidenced by Lesne et al., who then utilized interface-driven spin-orbit coupling through the Rashba effect, thereby achieving unprecedented efficiency in spin-charge conversion.^[103] Apart from the exchange interactions between local magnetic moments and 2DEG which facilitates spin polarization, the role of Ti d_{xy} -orbitals

belonging to the degenerated anisotropic t_{2g} bands could also induce the magnetic properties.

To further enhance the intrinsic magnetism at the LAO/STO interface, multiple techniques have been proposed. Among the reported techniques, the most straightforward one is by means of doping the STO^[104] or the LAO.^[105] While for the LAO/STO(001) interface, X-ray magnetic circular dichroism (XMCD) and DFT calculations have suggested the existence of ferromagnetic ordering, in which the samples prepared at varied oxygen partial show different magnetic ordering strength and Curie temperatures.^[31] Another technique involves a considerably more complicated procedure via the insertion of a buffer ferromagnetic film between the LAO and STO layers.^[106] Recent notable studies would include the insertion of the ferromagnetic EuTiO_3 (ETO) to create a complex LAO/ETO/STO heterostructure (STEM image of the heterostructure in Figure 7a).^[107] Based on an XMCD characterization of the complex magnetic heterostructure, the magnetic contributions of the Eu spin moment, m_{spin} , and the Ti orbital moment, m_{orb} , could be elucidated (Figure 7b). At temperatures below ferromagnetic transition ($T_{\text{FM}} < 6\text{--}8\text{ K}$), results indicate that the Eu^{2+} ferromagnetic order is accompanied by a strong XMCD response even at a very low magnetic field of $\mu_0 H = 0.05\text{ T}$. Such a strong exchange interaction between the Ti $3d$ and electron-rich Eu $4f$ orbitals greatly facilitate the

spin-polarized 2DEG. In a separate study by Zhang et al., a magnetic 2DEG has also been produced at the LAO/STO interface buffered with a 1 nm-thick $\text{La}_{7/8}\text{Sr}_{1/8}\text{MnO}_3$ (LSMO) (Figure 7c). Magneto-transport measurements show the presence of nonlinear Hall effect (Figure 7d) and anomalous Hall effect (Figure 7e) in the LAO/LSMO/STO heterostructure, which conclusively indicated the presence of a tunable highly spin-polarized and highly conductive 2DEG.^[108]

Besides the above discussion of inducing magnetic behavior at the LAO/STO 2DEG interface via doping and intermediate-layer insertion, new strategies have been proposed. For example, current techniques have improved via the means of charge transfer and the utilization of epitaxial strain engineering where thin-films are grown on substrates with varying degrees of crystal parameters and symmetries.^[109] While LaTiO_3 (LTO) and EuTiO_3 (ETO) are antiferromagnetic insulating oxides, their polar/non-polar structures are expected to induce an interfacial 2DEG when stacked together due to the onset of polar discontinuity.^[110] Moreover, an exotic interfacial magnetic state is likely to be expected given ETO's non-trivial topological structure.^[111] Shin et al. reported a ferromagnetic LTO/ETO 2DEG interface at temperatures up to 5.5 K even though the respective entities are inherently antiferromagnetic.^[112] Apart from regulating the carrier concentration of the interfacial 2DEG, the interfacial magneto-transport can also be tuned by varying the thickness of the LTO overlayer. Thereby indicating that the anomalous Hall effect at the interface is directly intervened by the carrier concentration.

Given the limitations of the $\text{Ti}3d$ orbitals in STO-based heterostructures, even with the presence of spin injection or externally induced spin polarization processes, the interfacial 2DEG in STO-based interfaces remains the one with the highest spin-charge conversion efficiency.^[99a,103,109c] However, due to the greater influence of the $\text{Ta}5d$ atomic orbitals in KTO, it is inevitable to take into consideration the magnetic properties of KTO-based heterostructures. This is because the spin-orbit coupling is significantly stronger at this interface than in its STO-based counterparts.^[83a] On top of the study of superconductivity in KTO-based interfaces discussed in the previous section, heterostructures such as LTO/KTO^[78] and LAO/KTO^[77] have been studied with reports of interfacial 2DEG. By considering the greater susceptibility of $\text{Ta}5d$ electrons to couple with magnetic neighbors as compared to the $\text{Ti}3d$ -orbitals, Zhang et al. carried out the epitaxial growth of an EuO layer to induce magnetic proximity effect and generated spin-polarized 2DEG at the KTO interface (Figure 8a).^[109b] This provides a multi-functional platform that comprises both a highly conducting 2DEG and a spin-polarized interface (magnetization at 5 K displayed in Figure 8b) with a strongly hysteretic magnetoresistance up to 25 K (Figure 8c) as well as a well-defined anomalous Hall effect up to a significantly higher temperature of 70 K (Figure 8d). This is in stark contrast with the 0.4 and 10 K of the respective effects recorded at the LAO/STO interface.^[3a,95,107,113] This proximity effect can be attributed to the EuO layer that induces the ferromagnetic state in the TaO_2 layer, thereby triggering strong magnetic correlations between the EuO film and the interfacial 2DEG.

Beyond the conventional oxide-based heterointerfaces, which are generally isostructural, investigation of magnetic signatures has been made in other systems. Nevertheless, it is important to note that given the diverse classes of oxide interfaces and het-

erostructures, each possess their own unique characteristics and that the underlying mechanism that leads to the onset of interfacial 2DEG may differ accordingly. In certain instances, the properties of the interfacial conducting layer may also vary according to the crystallinity of the constituent oxide layers involved. A case in point would be the spinel/perovskite heterostructure in the form of $\gamma\text{-Al}_2\text{O}_3/\text{STO}$ (GAO/STO),^[114] in which case, a long-range magnetic order which can be coupled with both the 2DEG, and lattice degrees of freedom has been observed by Christensen et al. at the GAO/STO heterointerface.^[114b] The magnetic property is also tunable with the application of external mechanical forces. At a general level, spinel/perovskite heterostructures in the form of $\text{MAl}_2\text{O}_4/\text{STO}$ (where $\text{M} = \text{Ni}, \text{Co}, \text{and Fe}$) all show the coexistence of ferromagnetism and anomalous 2DEG transport properties. Note that all these three spinel oxides are ferromagnetic up to room temperature. In a comprehensive study involving all three $\text{MAl}_2\text{O}_4/\text{STO}$ species, Zhang et al. reported that high-mobility 2DEG and ferromagnetism are found at the interfaces of $\text{NiAl}_2\text{O}_4/\text{STO}$ and $\text{CoAl}_2\text{O}_4/\text{STO}$ while $\text{FeAl}_2\text{O}_4/\text{STO}$ is insulating.^[114d] In addition, the interfaces of the former two interfaces display anomalous Hall effect below 30 K which highlights the likelihood that there is magnetic proximity effect induced by the top spinel layer on the heterointerface.

6. Device Applications: LAO/STO and Beyond

The complex interplay between spin, charge, orbital, and lattice degrees of freedom results in the formation of new states of matter due to their complementary and competitive relationships. This has not only been of fundamental interests but has also attracted concerted efforts to capitalize on these unique properties in applications related to electronic devices, spintronics, catalysis, energy storage, and photovoltaics. While the quest for these practical applications has been two decades ago, the momentum still remains in full steam and it is gaining strength with significant improvements and maturity in the synthesis techniques.^[115] Meanwhile, it is important to address and alleviate severe limitations related to devices based on oxide interfaces such as leakage current,^[116] device stability,^[4e,117] interface quality.^[115c,118] The challenge of scaling up the fabrication and synthesis of high-quality interfaces is another concern that must be appropriately addressed.^[119] Recent advances and progresses in the synthesis and manipulation of complex oxide interfaces have also enabled the integration of such systems with other existing device structures and external stimuli.^[4e,120]

Apart from providing new latitude to reveal new fundamental behaviors of such complex oxide heterostructures,^[121] the ability to better manipulate and control these properties unleashes new opportunities and functionalities in developing low-power electronics, quantum computing elements, efficient catalytic systems, and high-performance energy storage devices on their own or by integrating it with other materials. Moreover, the exploration of other oxide interfaces beyond LAO/STO has broadened the scope of research, offering opportunities to design and engineer materials with tailored functionalities. This section discusses the recent progress in the applications and impacts of oxide heterostructures and the advances in the design and manipulation of the system for tailored functionalities and applications.

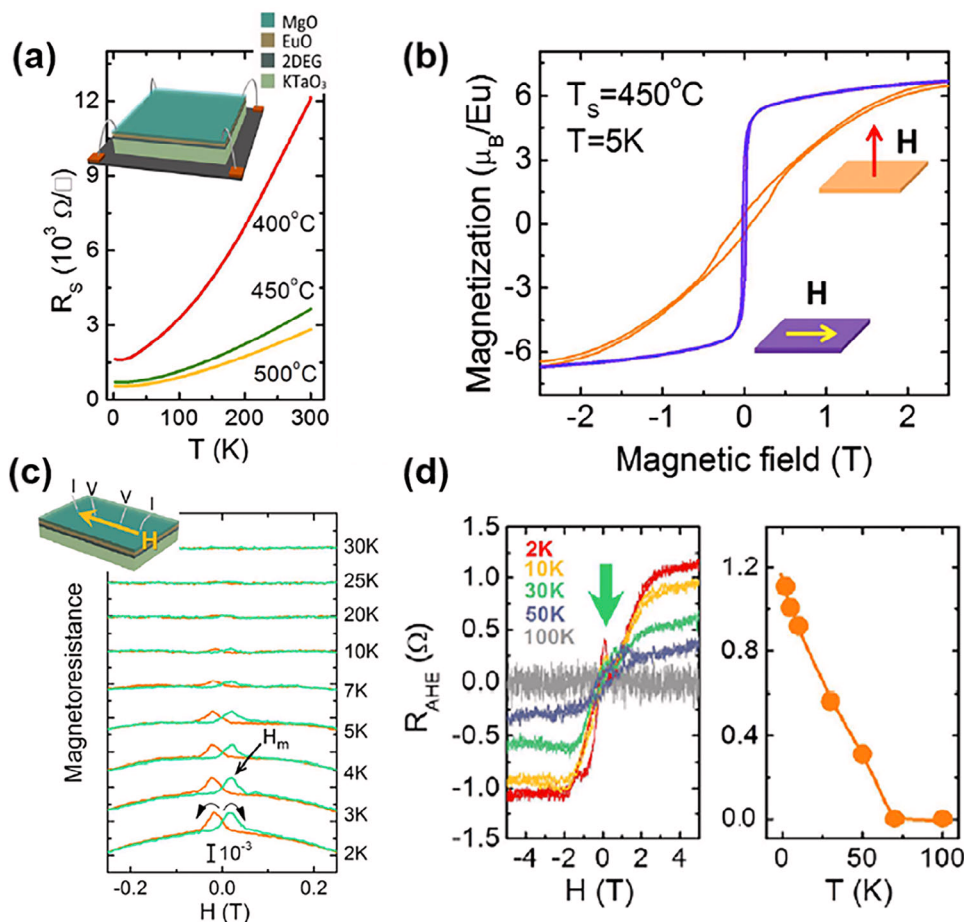


Figure 8. a) EuO/KTO interfaces prepared at different growth temperatures exhibit high conductivity, as illustrated in the schematic diagram of thin-film resistance measurements. b) Magnetization intensity dependence of the EuO/KTO interface measured under in-plane and out-of-plane magnetic fields at 5 K. c) The 2DEG at the EuO/KTO interface shows a pronounced magnetic hysteresis under an in-plane magnetic field and persists up to 25 K. d) Left and right graphs show the anomalous Hall resistance of the EuO/KTO interface as a function of magnetic field and temperature, respectively, demonstrating clear anomalous Hall effect. Reproduced with permission.^[109b] Copyright 2018, American Physical Society.

6.1. Field Effect Transistors and Beyond

Functionalization of LAO/STO-based systems generally capitalizes on the interfacial 2DEG in the fabrication of conventional field transistors^[41b,122] and quantum transport-based single-electron transistor.^[123] Charge transport properties of these devices are regulated by the electric field control of the interfacial carrier concentration in the form of field-effect transistors,^[41b,122] via side gating^[124] or through small LAO/STO islands in controlling the gate potential.^[123]

LAO/STO-based complex *n*-type metal-oxide-semiconductor integrated circuit that combines field effect transistors and resistors has been actualized by Jany et al.^[125] Based on a top-gate configuration, this device is capable of room-temperature operation with a gate voltage in the order ≈ 1 V. By adopting the side-gate configuration proposed by Stornaiuolo et al.^[124a] and exploiting the interfacial 2DEG as the active channel and gate electrode while using the STO substrate as the gate dielectric.^[116a,124b] Masarotti et al. has recently demonstrated the structural design of LAO/STO nanoscale field effect devices with the schematics and atomic force microscope (AFM) image displayed in **Figure 9a,b**,

respectively.^[126] In examining their performance in the cryogenic regime, they demonstrated that LAO/STO-based field effect transistors (FETs) are ideal candidates for superconducting electronic systems in the next-generation quantum devices.^[127] The LAO/STO field-effect devices in side-gate structures can achieve high efficiency. By applying a gate voltage smaller than 1 V, there is a significant drop in resistance by more than four orders of magnitude alongside a voltage gain of 50 (Figure 9c–e). In the dilution temperature range below 30 mK, R-T measurements further reveal that a gate voltage of only 200 mV is required to achieve complete superconductor-insulator conversion at temperatures (Figure 9f,g).

With reliable device performances and low threshold voltage, device structures in the form of scattered LAO/STO islands can also be utilized as contacts for charge injection into the STO layers. Müller et al. have demonstrated a lateral heterostructure comprising a narrow STO channel between two LAO/STO contacts conducts at bias voltages significantly below 100 mV.^[128] It is further demonstrated in the study that the tunnelling current can be easily controlled small gate-source voltages applied between a side gate and the channel. This study involving a

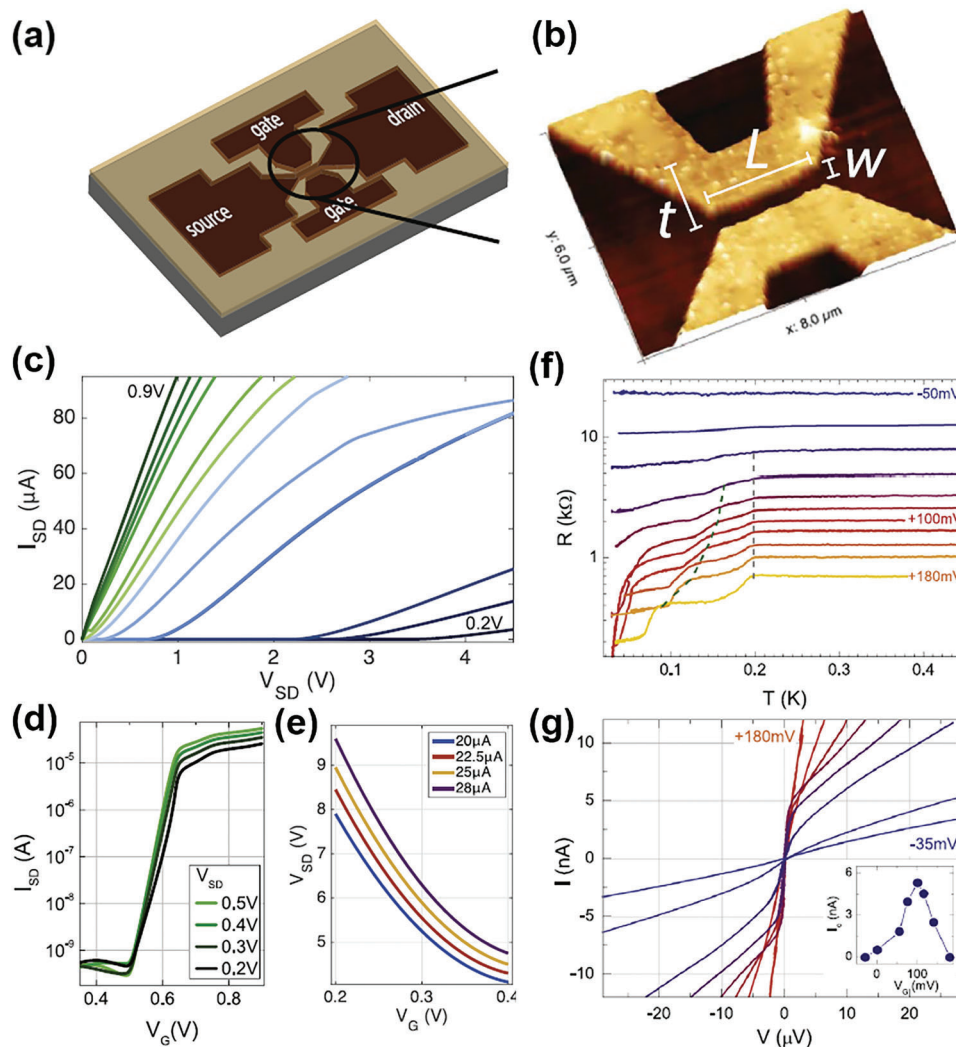


Figure 9. a) Schematic, and b) AFM image of the lateral gated field-effect device at the LAO/STO interface. c) Output characteristics of the lateral gated LAO/STO interface device, exhibiting unsaturated and upward trends at larger source-drain voltage, V_{SD} . d) Transfer characteristics of the lateral gated device at different source-drain voltages. e) Variation of output voltage V_{SD} with gate voltage, V_G , at different source-drain current, I_{SD} , values, showing significant voltage gain response. f) Gate voltage-induced transition of the interface nanochannel from a superconducting to an insulating state. g) I - V curve of the device at $T = 30$ mK, with an inset depicting the maximum critical current of 5 nA achieved at $V_G = 100$ mV. Reproduced with permission.^[126] Copyright 2020, IOP Publishing Ltd.

steep-subthreshold-slope device is fabricated entirely of oxide materials and is fabricated in a single-step, industry-compatible etching process.

Meanwhile, with the realization of other high charge density interfacial 2DEG systems based on rare earth oxide perovskites such as GdTiO_3 and LaTiO_3 on STO substrate,^[130] FET structures based on these heterostructures beyond the LAO/STO system have also been hugely promising. A particular case in point would be the high charge concentration 2DEG realized at the $\text{SmTiO}_3/\text{STO}$ (SmTO/STO) interface^[131] which display SmTO thickness-dependent tunable charge density and novel metal-insulator transition properties at the Mott–Ioffe–Rege limit.^[132] A recent study by Chandrasekar et al. addressed challenges concerning material synthesis, device design, sample fabrication,^[132b,133] and the presence of parasitic resistances (contact and access), which undermine device performances

by demonstrating high current density FETs with good pinch-off behavior using resistive SmTO/STO interfaces based on an I -shaped constriction transistor constriction as shown in **Figure 10a,b**.^[129] With such a device geometry and at a $2 \mu\text{m}$ constriction width, the saturation current density is elevated by alleviating parasitic series resistances and improving the control of electric field (Figure 10c). Their fabricated FET achieved a maximum saturation current density of 350 mA/mm and a transconductance of 200 mS/mm with $+1 \text{ V}$ gate voltage for a scaled device of gate length of $2.4 \mu\text{m}$ and a channel width of $2.2 \mu\text{m}$ at room temperature (Figure 10c). Notably, this is the highest reported $I_{D,\text{sat}}$ and g_m of any STO-based channel transistor reported in other studies (Figure 10d).^[132b,133,134]

Apart from the conventional STO-based interfaces, reports of 2DEG in another class of polar/nonpolar perovskite oxide interface in the form of $\text{LaInO}_3/\text{BaSnO}_3$ (LIO/BSO)^[135] and

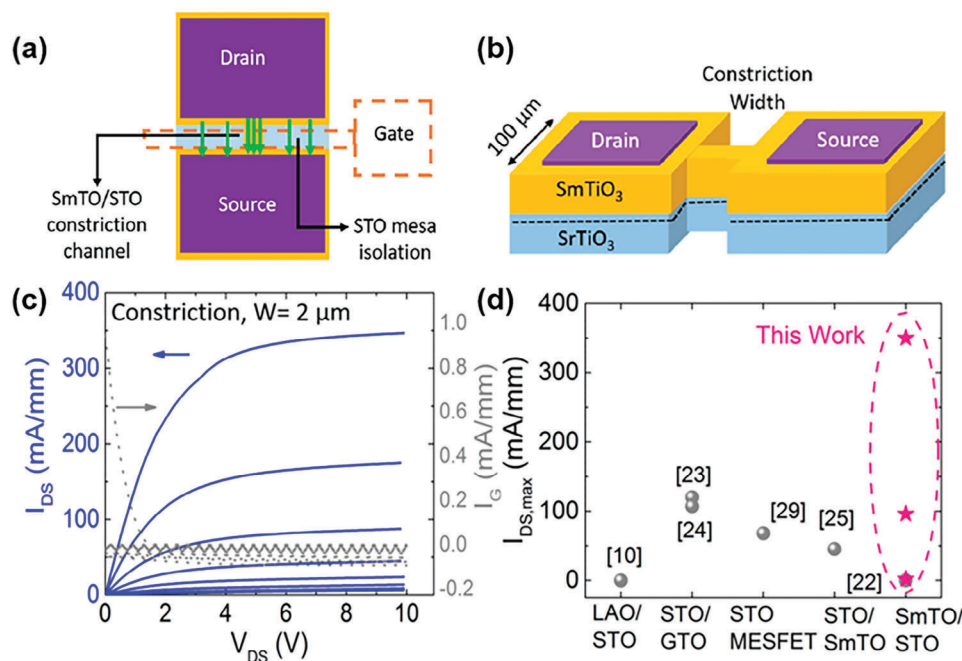


Figure 10. a,b) Schematic diagrams displaying the geometry of an I-shape constriction transistor based on a narrow SmTO/STO interfacial channel. c) Classical transistor output characteristics of the narrowed device with $W = 2 \mu\text{m}$ at room temperature, with gate current shown on the right axis. The narrowed device exhibits higher current density and cut-off behavior compared to planar transistors. d) Comparative plot of maximum current density reported in a series of STO-based field-effect transistors. Reproduced with permission.^[129] Copyright 2020, American Chemical Society.

$\text{LaScO}_3/\text{BaSnO}_3$ (LSO/BSO)^[136] have added further diversity to oxides-based FET systems. In addition to its high-temperature and oxygen stability, BSO can be doped readily using n-type dopants^[137] and it possesses high carrier concentration ($n_{3D} \approx 10^{20} \text{ cm}^{-3}$) and electron mobility ($\mu \approx 320 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$).^[138] As a heterostructure system, LIO/BSO shows an enhancement in room-temperature conductance by $\approx 10^4$,^[135a,b] where its high carrier mobility ($\mu_{FE} \approx 60 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and large on/off current ratio ($I_{on}/I_{off} \approx 10^9$). Meanwhile, with LSO being a high- κ dielectric material,^[139] LSO/BSO-based FET also displayed high field-effect mobility of $\mu_{FE} \approx 100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature.^[136]

Unlike conventional oxide heterostructures, it has been shown that 2DEG at the LIO/BSO and LSO/BSO interfaces do not arise due to oxygen vacancies or La interdiffusion.^[135a,d] Instead, the onset of 2DEG has been attributed to the so-called “interface polarization” model there exists an induction of 2DEG only near the interface with the BSO layer.^[136] Specifically, a polar perovskite with lattice constant similar to that of the BSO layer must be deposited coherently with the BSO layer in order to ensure a consistent and coherent in-plane epitaxial strain. Such an interfacial consistency induces an inversion symmetry breaking near the oxide interface. Thereby allowing the possession of polarization by the interfacial polar perovskite.^[139b,c] Conversely, such 2DEG interface will form for a perovskite with a large lattice mismatch even with its polar features. This is because of the inability for the in-plane lattice to be pinned down due to a large lattice mismatch, thereby resulting in a structural relaxation via the formation of interfacial dislocations. Such interfacial dislocations disrupt and reduce the interfacial polarization from the polar interface.^[140]

6.2. Freestanding Heterostructure Membranes – New Opportunities for Flextronics

When the functionalization of heterostructure materials is considered from a different perspective, the versatility in creating and manipulating the thin-film layer play a critical role in the actualization of the emergent physical properties found in perovskite heterostructures or for that matter, other forms of 2D heterostructures with novel functionalities.^[141] However, conventional applications and device fabrication techniques are generally restricted to the epitaxial growth of thin-film materials on single crystal substrates. The inherent brittleness of single crystal substrates limits the incorporation of thin-film materials into flexible devices. The extraction of these thin-film layers from the growth substrate is also challenging, thereby further restricting the capabilities of the heterostructures. Even in the case of integrating heterostructure systems with other thin-film materials, processes such as lithography can be severely limited^[120c,142] due to factors such as the requirement of very similar lattice parameters and crystal structures.^[143]

Lu et al. have proposed a general method for fabricating freestanding perovskite films to overcome the limitations of the need for a hard single-crystal substrate.^[144] The approach involves the epitaxial growth of water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$ on the perovskite substrate, followed by in situ growth of thin films and heterostructures (Figure 11a). The $\text{Sr}_3\text{Al}_2\text{O}_6$ layer can be etched in water to obtain a freestanding millimeter-scale single crystal film that can be transferred to any substrate and integrated with a semiconductor or layered compound heterostructures. Building upon this technique, Eom et al. have also reported the successful

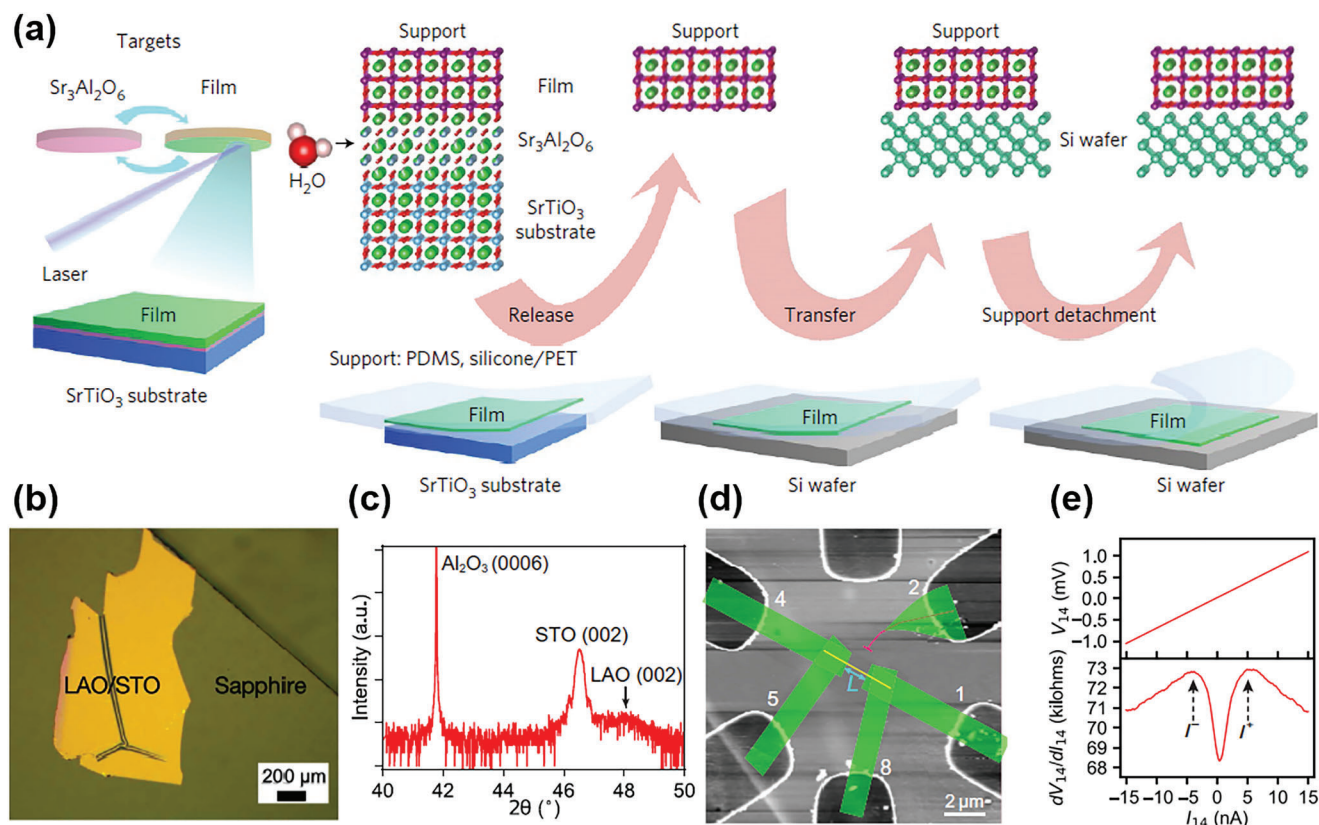


Figure 11. a) Schematic representation depicting the preparation procedure of perovskite oxide thin films. b) Optical microscopy image of LAO(10 u.c.)/STO (200 nm) suspended film that has been transferred onto a sapphire substrate. c) X-ray diffraction characterization shows that the LAO/STO crystallinity has been preserved. d) AFM image showing the formation of a nanoscale conductive channel at the LAO/STO film interface using c-AFM, with green regions indicating the extension of the gold electrode and the yellow line representing the main channel of length 1.2 μm . e) Top: Bi-terminal I - V curve obtained at 0 T and 50 mK; Bottom: Corresponding differential resistance curve (dV/dI curve) demonstrating clear superconducting features. a) is reproduced with permission.^[144] Copyright 2016, Springer Nature. b-e) are reproduced with permission.^[120a] Copyright 2021, American Association for the Advancement of Science.

preparation of a single-crystal freestanding membrane of LAO/STO heterostructure which can be directly integrated with other materials via van der Waals stacking – in this case on sapphire substrate (Figure 11b).^[120a] During this fabrication process, the key properties of the LAO/STO heterostructure remain intact and its crystalline phase is preserved while preserving its crystalline phase (Figure 11c) even after the formation of the membrane. This allows the creation of reversible patterns of nanoscale conducting regions through AFM lithography (Figure 11d) where its superconductive property has been preserved as confirmed via two-terminal I - V curve at 50 mK (Figure 11e).

As the techniques to extract freestanding thin-film materials develop, the functionalization and extraction techniques are no longer restricted merely to the LAO/STO heterostructures. Instead, with high-temperature superconductive materials such as $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) already well-established in a diverse range of practical device applications,^[145] the technique to extract oxide thin-film materials can be further extended to other classes of materials. The recent report by Jia et al. presented a novel approach for transferring water-sensitive YBCO films onto flexible substrates without the use of any buffer layer.^[146] Analyses revealed that the formation of a YBCO passivated layer protects the inner section of the freestanding YBCO during the etching pro-

cess, thereby effectively preserving the structural and superconductive properties of the highly water-sensitive YBCO layer.

Beyond the utilization of sacrificial layers, other techniques have been reported to achieve large-area freestanding thin-film membranes. A strain engineering approach by Sambri et al. has successfully demonstrated the self-formation of freestanding epitaxial LAO/STO membranes under a low-fluence and high-fluence regime (Figure 12a).^[147] By extending this self-formation technique, Dahm et al. further demonstrated that the positions and dimensions of these self-form LAO/STO membranes could be effectively regulated on different substrates while preserving their interfacial conductivity even after the membrane release process and being transferred to a silicon platform (Figure 12b).^[148] This is a particularly noteworthy breakthrough because of the potential to integrate interfacial oxide materials with existing semiconductor electronic manufacturing processes.

Even at cryogenic temperature, the conducting features of the freestanding LAO/STO heterostructure remain intact. Hence, in further attempt to leverage on the strain engineering method to produce freestanding LAO/STO, the heterostructure is then integrated with existing semiconducting technology by Erlandsen et al (Figure 12c-e).^[115d] This is executed via a

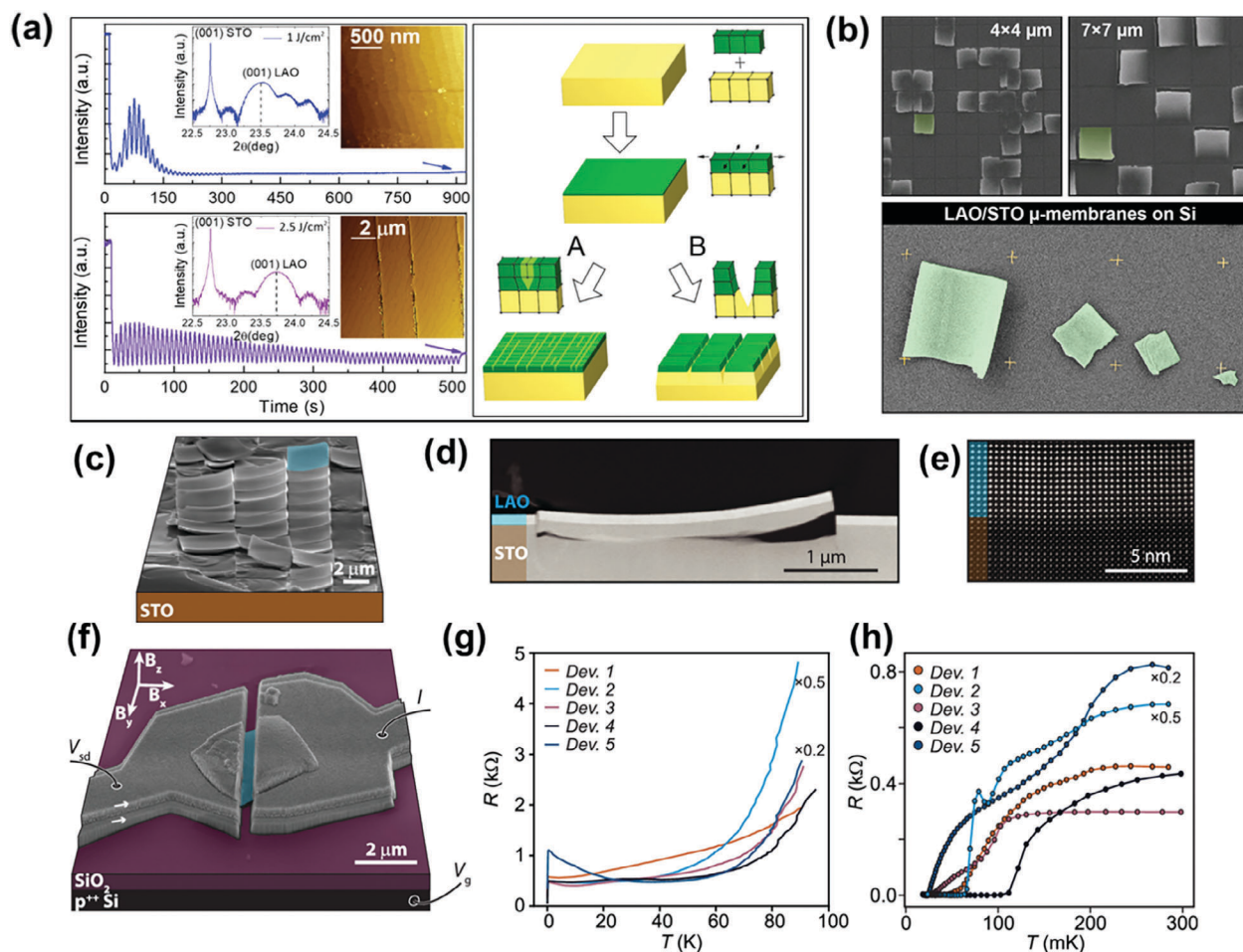


Figure 12. a) Comparing low-flux (left) and high-flux (right) growth modes in the self-formation technique of LAO/STO films. b) Scanning electron microscope (SEM) images of LAO/STO patterned structures with different dimensions. The transferred LAO/STO films can be arranged on a pre-patterned silicon substrate for further processing procedures. c) Tilted view SEM image of strain-engineered LAO/STO films after growth. d, e) Cross sectional images of the growth substrate obtained by low-magnification STEM and high-resolution image of the epitaxial LAO/STO interface, respectively. f) SEM image of a LAO/STO device fabricated on a p^{++} Si/SiO₂ substrate. g) Two-terminal resistance as a function of temperature for five devices exhibiting metallic behavior. h) Magnified view of the low-temperature region showing superconducting transitions as indicated in g). a) is reproduced with permission.^[147] Copyright 2020, Wiley-VCH. b) is reproduced with permission.^[148] Copyright 2021, American Chemical Society. c–h) are reproduced with permission.^[115d] Copyright 2022, American Chemical Society.

silicon-based backgate to regulate the critical current in the system with an image of the LAO/STO membrane device that is fabricated on a p^{++} Si/SiO₂ substrate displayed in (Figure 12f). The temperature-dependent two-point resistance measurement have shown that the metallic behavior of the integrated device remains (Figure 12g). Meanwhile, there are also signs of superconductivity at the mK-scale (Figure 12h). This is once again a particularly notable breakthrough due to the ability to integrate individual heterostructure membranes into existing silicon-based substrates at a significant quantity, which will be discussed in greater detail in the subsequent section.

6.3. Optoelectronic Devices

The so-called Persistent Photoconductivity (PPC) effect takes place as the conductivity of oxide heterointerfaces 2DEG in-

creases when illuminated at room temperature and that this elevated conductivity remains unchanged over an extended period even after the photon illumination is stopped. Based on this unique optoelectronic property, several oxide photoelectric devices can be developed as photodetectors, optical memory, phototransistor, photodiode, holographic memory.^[149] The LAO/STO interface is a case in point where it can be transformed from an insulating state to a metallic state under light stimulation and it remains metallic even after the photon stimulation is removed due to the continuous PPC effect. The on/off ratio of PPC to initial dark conductance is up to considerable 10^5 .^[149a] The metal state can also be converted back to an insulating state by applying a gate voltage.^[149a] The reversible properties of resistance switches based on the LAO/STO interface render it promising as a non-volatile memory system. Meanwhile, beyond the conventional LAO/STO interface, significant photoconductivity and persistent photocurrent properties have also been reported in other

oxide heterointerfaces,^[150] which could be used for optical switching or storage device applications.

The introduction of a buffer layer at the interface can effectively change the crystal and electronic structure to manipulate the properties of the interfacial 2DEG at the oxide heterointerface, particularly the photoconductivity and carrier mobility properties. The insertion of different buffer layers into the LAO/STO interface brings varying effects to the range, intensity and recovery time of the interfacial optical response.^[151] These works have enabled the study of how interfacial doping affects the properties of the LAO/STO interface and provides new understanding in the ability to regulate the properties of the oxide heterostructure. In addition, they provide the means of designing and exploring potential low-dimensional oxide materials for future optoelectronic devices.

In addition to the charge dynamics within the 2DEG interface, electrons can also undergo tunneling through adjacent conductive layers where they can be further modulated by light stimulation. Jeon et al. have achieved strong photo-response of the 2DEG in the Pt/LAO/STO heterostructure by adopting a vertical tunnel structure.^[152] The 2DEG tunneling current through the ultrathin LAO layer is significantly enhanced under ultraviolet irradiation. This strong and reversible photo response is attributed to the thermionic emission of photoexcited hot electrons from V_O defects of STO. They have further demonstrated that this reversible optical response is highly reproducible.^[153] Tunneling devices based on 2DEG in ultrathin oxide heterostructure therefore provide a new strategy for developing practical photoelectric applications, such as optically switchable tunneling transistors and wavelength modulation responsive multistage memory devices.

6.4. Resistive Random-Access Memory

The growing attention towards memory devices based on emerging electronic states at oxide interfaces has been notable. A case in point is the utilization of the interface 2DEG in resistive random-access memory (RRAM).^[154] Characterized by a simple metal-insulator-metal structure, RRAM devices consist of a resistive switching layer sandwiched between two electrodes. This design enables the storage of information by utilizing different resistance states. Beyond information storage, RRAM devices have been promising towards the next phase of novel computational paradigms due to the promise of faster processing speeds at lower energy consumption.^[155]

By incorporating 2DEG as a substitute for metal electrodes, oxide interface 2DEG has been employed in the fabrication of diverse types of RRAM. This approach enhances the design flexibility, improves performance, and introduces other intriguing features. Wu et al. pioneered the use of oxide 2DEG in RRAM devices.^[154c] Diverging from conventional resistive devices that employ metal layers as electrodes, they utilized a conductive layer proximal to the oxide interface as the bottom electrode. Voltage can be applied to the Pt top electrode to achieve resistive switching across the Pt/LAO/STO heterostructure. Furthermore, they substituted the top Pt electrode with a transparent ITO electrode, showcasing an optically transparent RRAM device.^[154e] Recently, Jeon et al achieved more reliable and gradual resistive switch-

ing devices by utilizing electrostatic potential to constrain the ultrathin LAO/STO heterostructure,^[156] suppressing the formation of localized conductive filaments, and introducing the collective control of oxygen vacancies in Pt/LAO/STO heterostructures. Additionally, RRAM devices utilizing 2DEG as electrodes have been reported in oxide interfaces such as $Ta_2O_{5-y}/Ta_2O_{5-x}/STO$,^[154d] $Pt/Al_2O_3/STO$,^[154a] $Cu/Ti/Al_2O_3/TiO_2$,^[154b] among others.

However, until now, all reported RRAM devices based on 2DEG have been singular demonstrations. Integrating RRAM devices based on 2DEG into crossbar arrays or 3D vertical structures poses significant challenges. On one hand, the relatively high sheet resistance of many 2DEG makes their utilization as bottom electrodes challenging. On the other hand, materials microfabrication requires the design of robust manufacturing techniques to produce the small features necessary for high-density RRAM arrays. In addition, the critical challenge impeding the development of RRAM devices is the proper understanding of the device switching mechanism, which has long been debated, and more rigorous analyses will be required in the future to better understand the switching mechanism of RRAM devices.^[154f]

6.5. Sensor

Changes in the conductivity of the oxide heterointerface can generally be used to sense the external stimulus applied to the sample especially under the effect of surface adsorbents.^[142,157] Thus designing gas detectors is conceivable in theory. By changing the top membrane material, different responses to different gas types can be achieved, leading to selectivity of detection. Gas sensors made of LAO/STO heterostructure surface modulated by Pd nanoparticles (NPs) were found to be highly sensitive under different ambient gases (H_2 , N_2 , H_2/N_2 , and O_2).^[158] Pd NPs, as catalysts, enhance charge coupling between the surface and the interface through direct charge exchange or changes in electron affinity, and hence improve gas sensitivity and selectivity.

The oxide heterostructure also has good sensitivity to organic compounds and pH. Meng et al. fabricated high-performance sensors for organic compounds using nanogold-modified single-crystal *p*-type $LaRhO_3/SrTiO_3$ heterostructure.^[159] They have achieved high response, fast response/recovery constant, and low operating temperature. Dong et al. fabricated a prototype pH sensor device based on the LAO/STO heterointerface 2DEG, which supported output current as a linear function of pH and exhibited high response over a wide range of pH values from 4 to 9.^[160] Recently, they also compared the performance of LAO/STO heterostructure sensors under different sensing thicknesses and preparation processes.^[161] It was found that surface damage is the primary cause of the decline in device current and sensing performance. This work provides insights into the development of oxide heterostructure sensors and offers experimental experience for obtaining highly sensitive sensor devices.

6.6. The Way Forward—Integration with Existing Technologies

Beyond FET and electronic devices, the novel properties underlying complex oxide interfaces accord them with a significantly wider myriad of applications in domains related to

ferroelectricity,^[162] ferromagnetism,^[15b,163] piezoelectricity,^[164] dielectric responses,^[165] magnetoelectricity,^[166] spin-structural coupling,^[28c,167] optoelectronics,^[168] and the list goes on.

As one considers the progress made in the synthesis, methodology and theoretical understanding of complex oxide heterostructures, it is perhaps advantageous to capitalize on the diverse functionalities of oxide heterostructures by integrating them with existing silicon-based technological platforms. In fact, epitaxial integration between TMO films and Si substrate predated the discovery of the conducting LAO/STO interface.^[169] Nevertheless, the ability to integrate multifunctional perovskite oxide interfaces with existing silicon technologies and fabrication techniques is promising and could possibly be the way forward in advancing the engineering integrated oxide-based electronics and photonics with the state-of-the-art.^[170]

While one considers the bright prospects of the future of computing, progress in this domain is particularly pertinent when one considers the immense challenges. With the exponential growth in the utilization of domains related to Deep Learning, Internet of Things, Cloud-based Computing, it is necessary to note that modern computing systems are consuming too much energy. The current rate of development especially with the rise of complex artificial intelligence applications will no longer be sustainable in the long run. Meanwhile, data centers are currently using a whopping 200-terawatt hours of energy each year in light of the preoccupation with the speed, accuracy and efficiency for computational operations and it is forecast to increase by an order of magnitude by 2030.^[171]

With the promise of energy-efficiency, adaptivity, parallel functionalization and fault-tolerance, neuromorphic computing serves as a promising candidate to power the future of computing.^[172] Metal-oxide based materials have been regarded as possible media in the fabrication of neuromorphic devices due to their promise of chemical stability and mechanical-flexibility.^[173] Furthermore, significant improvements have been made to the charge transport and mobility of the system through the integration of metal oxides in heterojunction systems.^[174] These improvements have allowed for the fabrication and integration of silicon and oxide materials beyond mere basic and planar geometries in more advanced and sophisticated applications. Beyond mere epitaxial Si/oxide/Si superlattices,^[175] the integration of perovskite oxides and Si has also been realized. BTO-based integrated photonic devices have been met with notable success.^[168a,176] Most notably, Ortmann et al. employed molecular beam epitaxy (MBE) and demonstrated the fabrication of 3D integrated Si(001)/TMO/Si(001)/TMO heterostructures.^[177] This work is particularly noteworthy due to their ability to alleviate the problem of spontaneous silicon oxidation and amorphization during the epitaxial deposition process. This fabrication process is realized by the inclusion of a wide-band gap oxide in the form of a LAO layer serving as an oxygen scavenging barrier.^[178] Meanwhile, the combination of thickness-dependent electronic structures and the ferromagnetic insulating character accord LaMnO₃/STO superlattices as another class of heterostructure considered to be integrated in a silicon-based device platform.^[163c,179] This has served as a basis by Chen et al. to deploy various techniques including MBE, pulsed laser deposition (PLD), and the introduction of multiple-layer buffer to integrate LaMnO₃/STO(001) into Si-based systems.^[180] By at-

tempting different integration techniques, the system comprising STO/Ca₂Nb₃O₁₀ is shown to possess the highest sample crystallinity due to the small lattice mismatch between the layers while the MBE-grown Si-integrated system produces a system with significantly suppressed saturation magnetization due to the presence of a large thermal strain. Even though it is still at the state of infancy, the successful integrated perovskite oxide/Si superlattice structures are ideal platforms particularly with their compatibility with existing Si-based wafer processing technology for the fabrication of highly-compact neuromorphic components and other electronic devices.^[168b,181]

7. Summary and Outlook

It has been 20 years since the pioneering discovery of 2DEG at the LAO/STO interface and this domain has grown extensively. Significant efforts have been made over the past two decades to understand the underlying physical mechanism governing interfacial 2DEG and a broad range of exciting and exotic phenomena, and they have been met with varying degrees of progress, and at the same time, success. This review focuses on the summary of the progress in the fundamental aspects of oxide heterostructures beyond the traditional LAO/STO interface and some of the progresses are not covered. This discipline has been burgeoning both in understanding the underlying mechanisms and means in which they could possibly be implemented for advance device applications. In addition, the number of oxide perovskite interfaces and the types of exotic physical properties are still growing.

Having discuss both the successes and limitations of existing theoretical models to account for the onset of 2DEG and other arising phenomena at the perovskite oxide interfaces, it is important to note that the model to describe the underlying mechanism adequately and sufficiently is yet to be established due to the complexity of such heterostructures. Nevertheless, these traditional theoretical models are mutually complementary via the combination of multiple factors including charge reconstruction, lattice distortion, and surface V_O to more consistently account for the physical phenomena observed at the oxide interface under different experimental conditions. Further concerted efforts on both the experimental and theoretical fronts are required to refine and better account for the origins of the interfacial 2DEG and other unexpected physical features.

Besides the realm of oxide heterostructures, the interest in integrating perovskite oxides with 2D layer materials is proven to be immense both in the aspects of heterostructure physics and potential practical device applications. With the multiferroic properties present in perovskite oxides and the essential optoelectronic features of 2D materials, both classes of seemingly unrelated materials can potentially play a highly complementary role via proximity effects or interfacial hybridization.^[182] In addition, the effects of interfacial hybridization without the issues of lattice mismatch allows for a wider variety of 2D material/perovskite oxide heterostructures to be studied and explored depending on the optoelectronic and magnetic effects required in the specific studies.^[183] As one considers the formation of such heterostructure systems, it could also be easily realized via straightforward and well-established processes such as mechanical exfoliation, wet/dry chemical transfer and even the direct synthesis of the 2D material on the perovskite substrate itself.^[184] While this

discipline is not exactly an uncharted research frontier, the fundamental studies and the actualization of such heterointerfaces in practical applications are generally restricted to the domain of interfacial FE effects.^[185] Therefore, such heterostructures can still be extensively explored and characterized moving forward^[168b] even before uncovering new possibilities in high-performance functionalities and applications.

Beyond the basic treatment of the scientific progress and the potential actualization that complex oxide heterostructures accords, it is also crucial to forecast the potential scientific and technological trends in the development and possible applications of this burgeoning discipline over the next few years and possibly decades. As a precursor to the novel technological applications to meet pressing global challenges, it is important to consider how the fabrication and implementation of oxide heterostructures could be actuated on a larger scale and how it could possibly be integrated effectively with existing technologies. Recently, Cohen-Azarzar et al. successfully realized an $\text{Al}_2\text{O}_3/\text{STO}$ conductive interface by employing a scalable and industrially compatible atomic layer deposition technique with NH_3 plasma pretreatment, propelling oxide electronics closer to mass production and practical applications.^[186] As highlighted in the previous section, the way to advance the knowledge of this technology and to apply it in practical scenarios may be further by integrating it with existing silicon technologies. Even though the integration of perovskite heterostructures with existing silicon technologies is still at the cradle phase, the breakthroughs to alleviate issues of amorphization and oxidation on the silicon surfaces are noteworthy breakthroughs in the fabrication of high-quality hybrid structural devices ideal for modern computational structures and systems. At the same time, with the integration of crystalline functional oxides with silicon being a longstanding and maturing technology developed over the past decades,^[169b,187] one could capitalize and adopt certain essential aspects of this know-how in the development of oxide heterostructure-based computational architecture.

One should note that a significant degree of technological development is still required to attain the current level of manufacturing standards found in conventional 3D silicon-based devices where their defect densities are measured in the scale of parts-per-billion. Conversely, 2DEG heterostructure systems synthesized primarily by the means of PLD and MBE still lag far behind with the defect concentrations and structural phase yield at a high percentage scale. While synthetic and elucidation techniques to derive high-quality oxide heterostructures at a large scale and oxide heterostructure-based device fabrication techniques have made considerable progresses over the past decades, a significant time and technological leap is still required for them to go beyond the level and quality currently restricted to the research laboratory. This is because of multiple manufacturability issues such as scalability and reproducible production of oxide-based heterostructure.

At this juncture, numerous publications have been made for the prototypical oxide heterostructure-based devices. The outcome has been promising with reports of high-performance and efficiency in a diverse range of applications including FET systems, radio frequency and thin-film transistors, neuromorphic components, light emitting diode emitters, solar cells, mechanical resonators, waveguides systems, and ultrasensitive sensors. These positive outcomes are compelling reasons that oxide

heterostructure-based systems will gradually grow in importance over the next few decades as ease of production and device performance improve with technological advances and integration with existing silicon-based technologies. With the attractive mechanical, optical, and electronic properties of oxide heterostructures, they will potentially become a ubiquitous component to complement conventional silicon-based devices that pervades virtually every aspect of future technologies ranging from communication infrastructures, smart/flexible wearables, to healthcare devices, where they could potentially be more energy efficient, ecological, versatile, robust, and perhaps, low-cost.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

interfacial superconductivity, oxide electronic device, perovskite oxide interface, quasiparticle dynamics, 2D electron gas

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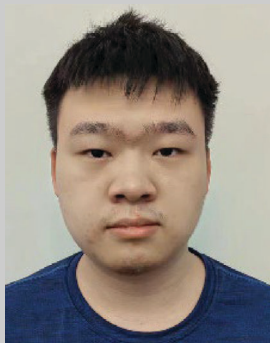
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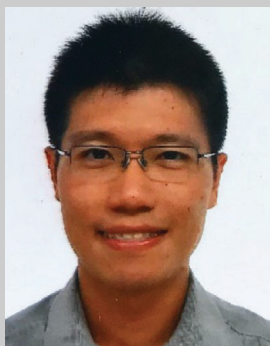
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