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Interface-Engineering and Applications in Oxide Heterostructures

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Complex oxide interfaces have mesmerized the scientific community in the last decade due to the tunable novel multifunctionalities, which originate from the strong interaction among charge, spin, orbital and structural degrees of freedom. Artificial interfacial modifications, which include defects, formal polarization, structural symmetry breaking and interlayer interaction have led to novel properties in various complex oxide heterostructures. These emergent phenomena not only serve as a platform for investigating strong electronic correlations in low-dimensional systems, but also of great potential for next-generation electronic devices with high functional density. This article reviews some recently developed strategies in engineering functional oxide interfaces and their emergent applications.

1. Introduction

Thanks to the advances in modern thin-film growth techniques such as pulsed laser deposition (PLD) and molecular beam epitaxy (MBE),^[1–5] atomic-level control of heterointerfaces has become feasible.^[6,7] Some subtle atomic structures that are not energetically-favored in three-dimensional bulk can be fabricated at two-dimensional interfaces, leading to the discovery of novel properties that are not observed or even expected in bulk materials.^[8] When compared to the interface between conventional semiconductors, the complex oxide interface with correlated electrons exhibits much richer interfacial phenomena, allowing diverse tunabilities

in a single oxide-interface-based device.^[9-12] In order to utilize the oxide interfaces in useful devices, significant effort has been made to understand the interfacial properties.^[13-16]

2. Strategies for Controlling Functional Oxide Interfaces

In this section, we concentrate our discussion on four different interfacial modification strategies which relate to: 1) defect engineering, 2) formal polarization, 3) structural symmetry breaking, and 4) interlayer interactions. Owing to the strong coupling among degrees of freedom such as lattice, charge, orbital and spin in correlated complex oxides,^[17-19] these strategies can effectively tune the interfacial functionalities, paving feasible routes for achieving all-oxide-based devices.

2.1. Defect Engineering

Defects are conventionally hard to control and traditionally play negative roles in material properties. They can induce for example local structural discontinuities and potential perturbations leading to degradation in the mechanical and electrical properties of the materials. However, owing to the development of modern experimental techniques, defects have become more controllable and designable – defects now not only induce disorders and randomness but can also provide alternate routes to new unexpected properties if properly deployed.^[20,21] Defect engineering is now becoming one of the frontiers in materials science.

Compared to conventional group IV and III-V semiconductors, oxide interfaces are apparently more suitable for defect engineering. *First*, the formation energy of defects is typically lower in complex oxides. The impurity and nonstoichiometry are highly charged due to the ionic nature of oxides, and to compensate these charges, defects are usually formed for charge neutrality. In fact, it is difficult to reduce the defect density in ‘dirty’ oxides to the level that has been achieved in conventional semiconductors. This is also one of the main reasons why functional oxides still lack real applications even though they have been shown

to exhibit rich and interesting properties. Nevertheless, this can be exploited further by artificially engineering the defects since oxides can maintain a larger amount of defects and thus their tunability is significantly high. *Second*, due to the flexibility of oxygen frameworks and multiple cation valencies, various species of defects can be stabilized in oxides for different purposes. Common examples can be found in the SrTiO₃ (STO)-based heterostructures.^[22] The introduction of oxygen vacancy can turn the bandgap insulator STO into a metallic oxide, which at low temperatures exhibits the dome-shaped superconducting phase as a function of carrier densities.^[23-25] Further, oxygen vacancies associated with formation of Ti³⁺ ions can lead to a ferromagnetic interaction in diamagnetic STO, which is an example of inducing magnetic interactions in a nonmagnetic material.^[26-28] Moreover, swapping the cation positions between Sr²⁺ and Ti⁴⁺ to form anti-site cationic defects of Sr_{Ti} or Ti_{Sr}, the paraelectric STO can become ferroelectric in a two-dimensional sheet even at room temperature.^[29] *Third*, due to the discontinuity of lattice and chemical environment, the interface is an open environment for defects with a lower formation energy. Also, when compared to bulk counterparts, the interfaces are expected to be more sensitive to defects, leading to more effective outcomes in defect-modulated interfacial functionalities. *Fourth*, it is convenient to induce targeted defects to the layer or interface *in-situ* or *ex-situ*. For the heterostructure prepared by the pulsed laser deposition, the *in-situ* preparation parameters such as growth temperature, ambient pressure, laser energy (frequency) and growth rate are all adjustable parameters in designing specific defect densities and species. Even after the *in-situ* fabrication, the interfacial defects are still tunable when a proper *ex-situ* thermal process is introduced. The convenience in inducing controllable defects in oxide heterostructures is a gateway for further exploration of the potentials of defect engineering in oxide electronic device applications.

2.2. Formal Polarization

The chemical bonds in complex oxides are usually dominated by ionic bonds, where electrons transfer from metal atoms to oxygen accompanied by the formation of cations and anions. Given the electrostatic effects, the separated positive and negatively charged ions can build up a polarization (i.e., formal polarization)^[30,31] giving rise to an electric potential within a neutral unit lattice. This can be clearly seen from the ionic structure of the perovskite ABO_3 oxides as sketched in Fig. 2. Looking at the STO lattices along [001] axis as sketched in Fig. 2(b), the alternating stack of neutral TiO_2^0/SrO^0 sub-layers results in a zero electric potential ($V = 0$), so STO is viewed as a *nonpolar* oxide along the [001] axis. By contrast, in the case of $LaAlO_3$ (LAO), the stacking sub-layers of AlO_2^-/LaO^+ become charged as shown in Fig. 2(c). Accordingly, a nonzero electric potential ($V \neq 0$) is established and thus LAO is a *polar* oxide along [001].

It should be noted that the *formal polarization* is different from the macroscopic *effective polarization* that is widely discussed in dielectric and ferroelectric materials.^[30] For examples, $BaTiO_3$ is a well-known ferroelectric oxide with a nonzero *effective macroscopic polarization*. However, following the idea of *formal polarization*, the $BaTiO_3$ (001) exhibits the *nonpolar* sub-layers of BaO^0/TiO_2^0 . The *effective polarization* in $BaTiO_3$ originates from the off-center displacement of Ti with respect to the oxygen octahedron, which is different from the *formal polarization* discussed here.

More importantly, the formal polarization in complex oxides is tunable. First, by viewing from different crystal orientations, the value of formal polarization varies. For instance, the (001) STO is nonpolar in Fig. 2(b), but the (111) STO is polar due to the stacking sub-layers of Ti^{+4}/SrO_3^{-4} as sketched in Figure 2(e). For LAO, the number of layer charges is also changed when varying the crystal orientation, as compared in Fig. 2(c) and (f). Second, the chemical doping is another way that can change the formal polarization. When doping the nonpolar STO with polar LAO, the $(Sr_{1-x}La_x)(Ti_{1-x}Al_x)O_3$ becomes polar even along its [001] axis and the value of formal polarization relies on the doping level x .^[31] A similar case can be

found in $(\text{La,Sr})(\text{Al,Ta})\text{O}_3$.^[32] Therefore, the crystal orientation and chemical composition are two key parameters for modulating the formal polarization.

Strikingly, the interface between nonpolar and polar oxides could lead to a divergent potential, namely, ‘polar catastrophe’.^[31,33-35] As shown in Fig. 3(a) and (b), growing polar LAO layers on top of the nonpolar (001) STO substrate, the electric potential V will diverge with the LAO layer thickness. Since this diverging V is not favored by nature some modifications need to be introduced to avoid such a diverging V . Therefore, the discontinuity of formal polarization is also an important driving force to modulate the interfacial properties in oxide heterostructures. However, this ‘polar catastrophe’ idea may not work in polar oxides in bulk form or at open surfaces, because a large amount of defects and surface adsorbates may compensate the formal-polarization-induced electrical potential. This explains why the LAO/STO interface on the STO_3 substrate can be conducting, while the insulating nature is always maintained at the similar interface prepared by growing STO layer on LAO substrate.^[36]

2.3. Structural Symmetry Breaking

One major difference between a thin layer and bulk is the crystal structure. Many early studies^[8,37-43] focused on a lattice-mismatch-induced strain engineering. When a layer is coherently grown on a substrate with a smaller lattice constant, the introduced compressive strain reduces the in-plane lattice constant of the layer with respect to its bulk counterpart, and vice versa. Such strain engineering can stabilize the crystal structure that is unstable in bulk,^[8,44-46] leading to large modulation of ordering temperatures and electronic states. Specifically, the manganite layer will exhibit a different orbital preference and thus phase separation under different types of strain – the A-type antiferromagnetic ordering is energetically favored under a tensile strain, while the C-type antiferromagnetic ordering in a compressively strain layer.^[37,47] On the other hand, for the STO layer, the biaxial strain can

establish ferroelectricity,^[48,49] modify the bandgap,^[50-52] manipulate the orbital occupancy and bandwidth,^[43] thereby controlling interfacial electronic transport behaviors.^[53] The strong coupling between lattice and orbital (spin) allows for effective strain engineering on modulating interfacial conductivity and magnetism.

We have shown above some examples that conventional strain engineering based on biaxial strain is able to tune the performance of thin-film devices due to the strong coupling between lattice and other degrees of freedom. However, the conventional strain engineering lacks the interface-resolved capability for controlling each individual interface in one single multilayer device. Recently, the strategy based on structural symmetry breaking when crossing the heterointerface has attracted significant interest.^[44,54-60] In ABO_3 perovskite oxides, the diverse crystal symmetries in perovskite oxides, including cubic, orthorhombic and rhombohedra structures, are closely related to the connectivity between octahedra,^[44,45,61,62] where the oxygen octahedral rotation pattern determines the bond angle of $B-O-B$ and thus the hybridization between the metal and oxygen.^[57,60] More importantly, in perovskite heterostructures, each ABO_3 monolayer may exhibit different oxygen octahedral rotation patterns, providing a feasible way to realize the layer-resolved engineering.^[57,60,63-66] In manganite-based heterostructures, when changing the oxygen octahedral rotation in the targeted manganite layer by selecting a suitable substrate and/or adjacent layer, the layer-resolved evolution of oxygen octahedral rotation and Mn-O-Mn bond angle is observed, affecting the switching of in-plane magnetic easy axis,^[56,60] Curie temperature,^[57,60,66] perpendicular magnetic anisotropy^[58] and interfacial magnetic coupling.^[67] Therefore, following this strategy based on oxygen octahedral connectivity, the substrate and spacer layer in perovskite multilayers are able to provide the additional engineering parameter by controlling oxygen octahedral rotation in each functional sub-layer, integrating multiple switchable states into single heterostructure for novel information storage devices.

2.4. Interlayer Interaction

The heterostructures may not just contain one single interface or one layer. In a multilayer or superlattice structure, interlayer interactions arising from two coupled functional layers that may be adjacent or separated by a spacer layer become important. One example is the interface between a superconductor and a ferromagnet.^[68] When the magnetic layer such as (La,Ca)MnO₃ is placed close to the superconducting layer such as YBa₂Cu₃O₇, the charge transfer and proximity effect occur at the interface, leading to the novel orbital order and spin states at the interface.^[69,70] Another example is the ferromagnetic tunnel junctions,^[71,72] where two ferromagnetic conducting oxide layers are separated by a thin insulating layer, the tunneling current can be switched ‘ON’ and ‘OFF’ depending on the relative spin orientation between two magnetic layer. If the spacer is not just a common insulator but a ferroelectric layer, the tunneling current will be controlled not only by magnetic field with different spin configurations, but also the applied electric bias that modifies the tunneling barrier by switching the polarization of the ferroelectric spacer.^[13,73] In this case, four states, or even more non-volatile states,^[74] can be achieved at room temperature,^[75] which is important for designing novel high-density information storage devices.

Beside the electric properties, the interlayer coupling can be also applied to tune the magnetic properties. The famous Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction,^[76] which appears when a thin conducting layer is sandwiched between two ferromagnetic layers, is a good example. By changing the thickness of the conducting spacer, the coupling strength between two ferromagnetic layers exhibits oscillating behavior even accompanied by a sign change. When the spacer layer is insulating, the interlayer interaction may depend on the spin-polarized tunneling that decays rapidly with increasing the spacer layer thickness.^[77,78] The complex interlayer coupling paves another path for controlling the interfacial properties and sheds some light on understanding electronic correlations in functional oxide heterostructures.

3. Emergent Phenomena at Functional Oxide Interfaces

In this section, we review some emergent phenomena that are important for interfacial functionalities, including interfacial electrical properties, magnetism and ferroelectricity. We link these interfacial properties to the interface-engineering strategies discussed in Section 2.

3.1. Modulations of Interfacial Electrical Properties

3.1.1. Insulator-Metal Transition at Oxide Interfaces

The emergence of conducting interface between two insulating oxides, such as LAO and STO, is one milestone for exploring novel functionality at oxide interfaces. Since the first report on the quasi-two-dimensional electron gas (q-2DEG) at the (001)-orientated LAO/STO interface,^[33] the STO-based interfaces have attracted immense attention due to their application potentials in novel electronic devices^[79-81] and rich physics^[82-90] arising from the complex electronic correlations. Although significant effort has been put in order to understand the LAO/STO interface, the origin of interfacial electrons is still under debate. One famous model is based on the discontinuity of formal polarization between (001)-orientated LAO and SrTiO₃.^[31,33-35] As sketched in Fig. 3(a) and (c), when grown on the (001) polar LaAlO₃ layer on the (001) nonpolar TiO₂-terminated STO substrate, the diverging electric potential V can be compensated by electron transfer from LaAlO₃ to the interfacial SrTiO₃ layer with the formation of the n -type interface. On the other hand, if the STO substrate is terminated by the SrO sub-layer as shown in Fig. 3(b) and (d), the formal polarization becomes upward and thus positive charges need to be created in the interfacial STO layer to form the p -type interface.

For the n -type interface, the build-up potential V must overcome the energy gap between the LAO valence band and STO conduction band. Accordingly, the critical thickness t_c for triggering the charge transfer at the polar/nonpolar heterointerface can be estimated by $t_c = \epsilon_0 \epsilon_P \Delta E / eP$, where ϵ_P is the dielectric constant of the polar material, ΔE is the energy gap

separating the valance band of the polar layer and the conduction band of the nonpolar material, e is the elementary charge and $\mathbf{P}$ is the electric polarization of polar layers.^[31] Taking $\epsilon_P = 24$, ΔE as STO bandgap of 3.2 eV, and $\mathbf{P} = 0.526 \text{ C m}^{-2}$ for the LAO/STO (001) interface, t_C is calculated to be 4 unit cells in perfect agreement with the experimental value.^[91] On the other hand, if $\mathbf{P}$ is tunable in the polar layer, the value of t_C can be tuned. When replacing the polar LAO with the STO-doped LAO, i.e., $(\text{La}_{1-x}\text{Sr}_x)(\text{Al}_{1-x}\text{Ti}_x)\text{O}_3$, $\mathbf{P}$ in polar layer depends on the doping level x . The experimental data reveal that in the $(\text{La}_{1-x}\text{Sr}_x)(\text{Al}_{1-x}\text{Ti}_x)\text{O}_3/\text{STO}$ system the doping-level-dependent t_C follows the model of polar discontinuity, suggesting the crucial role of formal polarization in controlling the interfacial electrical properties.^[31] Moreover, recent experiments suggested that other factors, including but not limited to oxygen vacancies,^[92,93] chemical stoichiometry^[94,95] and ionic point defects,^[96] can also influence the STO-based interface. So, more factors are also needed to be considered when estimating t_C .^[97-99]

For the p -type interface, the positive charges for compensating the upward formal polarization in LAO are believed to be generated by atomic reconstructions associated with oxygen vacancies.^[34] Those positive charges are usually less mobile or even localized at the p -type interface. However, when the p -type interface is fabricated by growing epitaxial SrTiO_3 layers on the AlO_2 -terminated LAO layers with a subtly designed band structure, the heterostructure can exhibit hole-doped conducting interface, or two-dimensional hole gas (2DHG) with ionic point defects being eliminated.^[100]

Although the interfacial conductivity at the (001) interface can be explained in terms of discontinuity of formal polarization as discussed above, the situation at the (110)- and (111)-orientated interfaces are more complicated. Figure 4 illustrates the case of the (110) LAO/STO interface. Based on the conventional structural model consisting of parallel layers, as shown in Fig. 4(a,b), there is no discontinuity of formal polarization at the (110)-orientated LAO/STO interface. Hereby, the (110) interface should maintain the insulating nature.

However, the calculation of density functional theory reveals that the buckled TiO_2/LaO interfacial structure, as shown in Fig. 4(c), is energetically favored at the (110) LAO/STO O_3 interface, where the discontinuity of formal polarization is still present. Such polar discontinuity arising from the buckled TiO_2/LaO causes the insulator-metal transition driven by the LAO thickness and the anisotropic superconductivity and magnetism at the (110) interface.^[101,102]

Beside the formal polarization, the defects are widely used in controlling interfacial conductivity. Examples are the conducting interfaces between amorphous oxide layer and single crystal STO substrate, so called amorphous STO-based oxide heterostructures. It must be noted that the amorphous interface lacks the ordered crystal structure and thus exhibits no formal polarization. So the key role here is an interfacial redox reaction. Regardless of the interface polarity, when depositing the oxide layer with a lower oxygen affinity on the STO substrate, such as LaCrO_3 ,^[103] LaMnO_3 ,^[104] and $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$,^[92] there is no redox reaction and the interface maintains the insulating nature. On the other hand, if the deposited oxide has a higher oxygen affinity, such as LAO ,^[93] Al_2O_3 ,^[92] yttria-stabilized zirconia (YSZ)^[92] and CaHfO_3 ,^[105] the deposited layer can absorb the oxygen from the STO surficial layer and create oxygen vacancies at the interface. The density of oxygen vacancies relies on the deposition pressure and layer thickness, resulting in the observation of insulator-metal transition driven by the depositing pressure and layer thickness.^[92,93] An advantage of the amorphous conducting STO-based interface is that it can be easily fabricated at room temperature, providing more flexibility for device designs.

Because both the oxygen vacancy and polar discontinuity can contribute to the interfacial conductivity as sketched in Fig. 5(a), there were intensive debates on the mechanism for the appearance of the conducting LAO/STO interface in early studies.^[93] The systematic studies on comparing the oxygen-deficient and well-oxidized LAO/STO interfaces reveal the differences between the oxygen-vacancy-induced and polar-discontinuity-dominated

interfacial conductivity.^[93] The first difference is the thermal stability of the interfacial conductivity, as shown in Fig. 5(b). If the oxygen vacancies play key roles in inducing the interfacial conductivity (like in amorphous LAO/STO interfaces), the conducting interface becomes insulating after high-temperature annealing in oxygen. By contrast, the polar-discontinuity-dominated interfacial carriers can be maintained after annealing. The second difference is the behavior of carrier density as a function of temperature, as shown in the inset of Fig. 5(c). Because the energy level of oxygen vacancies is below the conduction band minimum in STO, the mobile electrons can easily be trapped at lower temperature leading to a ‘carrier freeze-out’ phenomenon.^[106] However, for the polar-discontinuity-dominated conducting interface, the mobile electrons are less temperature dependent. For the as-grown single crystal STO-based interface where the oxygen vacancy and polar discontinuity provide mobile electrons, the carrier density will be higher and occupy multiple subbands, resulting in the transport behavior such as nonlinear Hall effect featured by multiple conducting channels.^[107]

3.1.2. Interfacial ‘Missing’ Charges

Although the polar-discontinuity model explains the appearance of t_C at well-oxidized interfaces, there is a large discrepancy on charge density. From the model based on polar discontinuity, half electron per unit cell area ($3.3 \times 10^{14} \text{ cm}^{-2}$) needs to be transferred to interfacial STO to completely compensate the polar discontinuity.^[34] However, the carrier density at the conventional LAO/STO interface is only 10% of the expected value ($\sim 10^{13} \text{ cm}^{-2}$) based on Hall measurement,^[83,86,93,108] and it becomes even lower ($\sim 10^{12} \text{ cm}^{-2}$) when estimated from the Shubnikov-de Haas (SdH) magnetoresistance oscillations.^[107,109-115] In order to find these ‘missing’ charges, other experimental methods, associated with the transport measurement, need to be applied to study the interfacial charges.

In order to directly measure both localized and delocalized charges near the Fermi level, one way is by using a combination of spectroscopic ellipsometry and ultraviolet – vacuum ultraviolet (UV-VUV) reflectivity,^[116] which can measure the optical conductivity (σ_1) of each individual layer in a multilayer system in a wide energy range of 0.5-35 eV. Owing to the optical f -sum rule,^[117] this approach can quantitatively measure the overall charge transfers, both localized and delocalized, between the LAO film and the interface layer, enabling one to unravel the actual mechanism behind the thickness-dependent interface conductivity of LAO/STO.^[118]

After thorough analyses,^[118,119] Figure 6(a,b) show the resulting σ_1 spectra of the LAO film and the interface layer for different thicknesses of LAO film, along with the optical transitions responsible for each peak.^[120-124] From the σ_1 spectra, the effective number of electrons participated in each optical transition, N , can be quantitatively obtained using the partial f -sum rule,

$$\frac{N}{V} = \frac{4m}{\hbar e^2} \int_{E_1}^{E_2} \sigma_1(E) dE \quad (1)$$

where e is the elementary charge, m is the electron mass and V is the unit volume. The E_1 and E_2 indicate the energy boundaries of a transition in the σ_1 spectra. The relative N of each layer relative to bulk values, n_{eff} , obtained using this method is shown in Fig. 6(c, d). By analysing the changes in n_{eff} , the various charge transfer and redistribution mechanisms that happen in each layer to counteract the potential divergence for both insulating and conducting LAO/STO interfaces can be observed, as follows.

Below 4 uc of LAO, the potential divergence is counteracted by the intra-layer charge redistribution mechanism between the sub-layers of LaAO without involving charge transfer into the interface, keeping the interface insulating. This charge redistribution is consistent with the ionic relaxation mechanism^[125] observed using surface X-ray diffraction^[126,127] and second harmonic generation.^[128] Above 4 uc of LAO, the potential divergence is instead

countered by the inter-layer $0.5e^-$ charge transfer from LAO film into the interface, as predicted by the polarization catastrophe model. This charge transfer is further stabilized by the presence of oxygen vacancies in the LAO film, to compensate the $0.5e^-$ hole left by such transfer.^[129-132] According to theoretical calculations,^[132] the interplay between these mechanisms is governed by their respective formation energy: in thinner films the ionic reconstruction is more energetically favorable while in thicker films the interfacial charge transfer is cheaper instead.

Besides LAO/STO, this technique (using spectroscopic ellipsometry and high-energy reflectivity to study the optical conductivity of materials) has also been utilized in other systems. Examples include studying metal-insulator transition in manganites,^[116] excitonic effects in grapheme,^[133] vacancy effects in vacancy-doped STO,^[134] mid-gap states in ambipolar cuprates,^[135] anomalous plasmonic excitations in $\text{Sr}_{1-x}\text{NbO}_{3+\delta}$,^[134] and interactions between localized and delocalized charges in $(\text{La}_{0.3}\text{Sr}_{0.7})(\text{Al}_{0.65}\text{Ta}_{0.35})\text{O}_3/\text{STO}$ interfaces,^[136] among others.

3.1.3. Carrier Mobility Modulations

The correlated electrons at the oxide interface trigger unconventional functionalities that are absent at the conventional interface. However, one great drawback is the low carrier mobility. For the conventional STO-based interface, the low-temperature mobility is only around $1,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$,^[86,93] which limits further studies on quantum transports at strongly correlation two-dimensional oxide interface and application potentials in novel all-oxide-based devices. A lot of effort has been placed in improving the carrier mobility at the STO-based interface and significant leaps forward in electron mobility were reported. A mobility of $30,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ was achieved in a δ -doped STO layer with a strong carrier confinement.^[137] By changing the growth conditions for suppressing the interfacial defects, the optimized LAO/STO exhibits the mobility around $6,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ with clear SdH oscillations observed

at low temperatures.^[109] The interfacial conductivity is also sensitive to the surface conditions. The interfacial mobility can reach around 10,000-20,000 cm²V⁻¹s⁻¹ when proper surface modification is applied by atomic force microscopy, thermal treatment and/or solvent.^[138] By capping a cuprate layer to removing the oxygen vacancies produced at the LAO/STO interface, the interfacial electron mobility is enhanced to 50,000 cm²V⁻¹s⁻¹,^[139] enabling the investigation of the subtle subband structure with a small energy splitting.^[113] Using the γ -Al₂O₃ layer to produce oxygen vacancies at the STO surface, the mobility can reach up to 140,000 cm²V⁻¹s⁻¹, but it is not stable after being annealed in oxygen.^[140] Also, the introduction of (La,Sr)MnO₃ layer as a buffer layer boost the mobility to 73,000 cm²V⁻¹s⁻¹,^[141] revealing the existence of multiple conducting channels at the interface.^[115] Another example for high-mobility interface is the (La,Sr)(Al,Ta)O₃/SrTiO₃ interface.^[32,107,136,142,143] When compared to the conventional LAO/STO interface, the (La,Sr)(Al,Ta)O₃/SrTiO₃ interface exhibit three major advantages. The first one is the small lattice mismatch of 1%. The second one is that the discontinuity of formal polarization is smaller than the conventional LAO/STO, resulting in a low density of ionic point defects.^[144,145] The third one is that there is negligible structural symmetry mismatch at the (La,Sr)(Al,Ta)O₃/STO interface. At room temperature, both the (La,Sr)(Al,Ta)O₃ and SrTiO₃ bulk possess a cubic structure, while at low temperature they both turn to a tetragonal structure.^[146,147] This means little effects from structural symmetry mismatch to reducing Ti-O-Ti bond angle for a low hopping integral. Hence, the (La,Sr)(Al,Ta)O₃/STO interface exhibit a robust high mobility of 35,000 cm²V⁻¹s⁻¹^[32] and better thermal stability^[142] for interfacial conductivity.

The effect of Ti-O-Ti bond angle on carrier mobility can also be inferred by comparing three two-dimensional electronic systems with different TiO₂-terminated surfaces – amorphous LAO grown on the TiO₂-terminated STO surface, anatase TiO₂ surface and rutile TiO₂ surface.^[148]

The sheet resistance as shown in Fig. 7(a) demonstrates delocalized metallic behavior down to 2 K in STO and anatase. However, in anatase there is an upturn below 100 K, which tends to saturate at very low temperature. Rutile surface exhibits strong localization below 25 K. The carrier density plotted in Fig. 7(b) as a function of temperature shows similar behavior for all the three systems at the temperature range of 300 to 100 K. In Rutile carrier density rapidly freezes out below 20 K, and it remains almost constant at low temperature for the other two oxides. The room temperature carrier density for STO is ten times higher than the low temperature value. The carrier density in anatase is more complex, reflecting the two-band nature of the transport. Figure 7(c) shows that the mobility increases by three-orders-of-magnitude with decreasing temperature for STO with saturation below 10 K than that of room temperature, whereas for anatase, the mobility follows inverse trend of carrier density as a function of temperature. Rutile interface shows three orders of-magnitude increase in mobility with decreasing temperature, and it then begins to drop below 20 K as the carriers freeze out.

These observed three distinct electrical transport properties have been explained based on orbital corrugation. Conductance in conventional cubic s-band metals such as copper, zinc is isotropic due to its symmetric Fermi surface. Unlike metals the transport of charge carriers in metal oxides occurs through overlap of cation and anion orbitals, which are directional. The motion of charge carrier in this system can be quite anisotropic. So the bond angle can greatly affect overlap of the orbitals, which could be a critical factor in determining the charge transport. To demonstrate this idea, authors have compared three different crystallographic TiO_2 surfaces where two dimensional electron gas was created by covering amorphous LAO. These three different surfaces allow to correlate the differences in the transport properties with the natural orbital corrugation. In the TiO_2 system, the Ti $3d t_{2g}$ orbitals overlap with the oxygen $2p$ orbitals. As in superexchange, the Ti-Ti transfer integral is highest for a bond angle of 180° . So higher the O-Ti-O angle means lower the effect in the charge transfer.

The two-dimensional character of the conduction in the occupied states in a thin layer at the titanate interface is expected to be sensitive to the in-plane Ti–Ti transfer integrals and bond angles. The bond angle is 166° in tetragonal STO at low temperatures, 155° in anatase, and 81° in rutile as shown in figure 8. The bond angles are linked to the lattice deformations that give rise to polarons in anatase.^[149] It is proposed that large polarons are formed in anatase^[149,150] and small polarons^[151] in rutile. However there is no small polaron formation in STO according to a prediction^[152]. This could be the reason why there is strong localization in rutile but only a weak increase in resistance at low temperature in anatase and none in strontium titanate.

3.2. Unconventional Magnetic Interaction

3.2.1. Interfacial Magnetic Interaction between Nonmagnetic Oxides

Both the LAO and STO bulks are nonmagnetic, but the interface between them can be magnetic. The discovery of magnetic interactions at the nonmagnetic oxide interface LAO/STO, characterized by the Kondo effect as shown in Figure 9(a), was first reported by Brinkman *et al.* in 2007.^[83] Later, the electronic phase separation (EPS), denoted by the coexisting ferromagnetism and superconductivity, was reported by Ariando *et al.* by superconducting quantum interference device (SQUID) measurement in 2011.^[86] The samples that were prepared at high oxygen pressures (above 10^{-2} mbar) show clear ferromagnetic hysteresis in magnetic-field-dependent magnetization ($\mathbf{M-H}$) measurements and superconducting diamagnetic signal in temperature-dependent magnetization ($\mathbf{M-T}$) curves, as shown in Fig. 9(b). This observation of EPS highlights the important role of the selective occupancy of interfacial Ti subbands.^[153-155] Henceforth, various methods have been applied to characterize the existence of interfacial ferromagnetism at the LAO/STO interfaces, including torque magnetometry,^[90] scanning SQUID microscopy,^[88] X-ray magnetic circular

dichroism (XMCD) and X-ray absorption spectroscopy (XAS),^[156,157] and magnetic force microscopy (MFM).^[158] Although most of experimental results are consistent, some of them are contradicting. For examples, large variations on magnetic moment density were reported.^[90,159] The ferromagnetic features are able to be observed at room temperature,^[86,158] or only detectable at low temperatures.^[87] The data from magnetotransport and SQUID showed that the magnetic interaction is enhanced by increasing an oxygen pressure during the sample growth, or reducing oxygen vacancy.^[83,86] However, the study on X-ray absorption spectroscopy (XAS) clearly demonstrated that formation of the oxygen vacancy is crucial for ferromagnetism at the interface.^[156,157]

In order to understand those inconsistent phenomena, complex magnetic interactions that involves different mechanisms need to be considered. One idea is to classify the magnetic LAO/STO interface into two categories.^[160] In the first category, the interfacial magnetic interaction is propagated by the coupling between local moments and itinerant electrons. The itinerant electrons can be provided by the two-dimensional electron gas – given the antiferromagnetic RKKY exchange between the mobile electrons and local moment,^[161] the interface becomes ferromagnetic when the mobile carrier density is lower than the local moment density. This explains why the ferromagnetic behaviors are observed in the relatively insulating samples.^[83,86,158] The local magnetic moment may be arising from oxygen with unpaired electrons and/or localized states from Ti.^[28,162,163] The second category is labeled as ‘metamagnetic’ phase, characterized by the anisotropic magnetoresistance (AMR)^[85,164,165] and Anomalous Hall effect.^[166] In order to observe this metamagnetic phase, the temperature must be low (~35 K) but above superconducting temperature (electron pairing but not superconducting), and the carrier density should be close to the Lifshitz transition point. If we assume that when carrier density is below Lifshitz transition point, there is a strong attractive interaction for the $3d_{xy}$ electrons, corresponding to the electron pairing and low-temperature superconductivity. When applying the magnetic field to overcome the pairing energy, or

electric field to crossing the Lifshitz point, $3d_{xz/yz}$ orbitals will be occupied. Apparently, $3d_{xz/yz}$ band is highly anisotropic, leading to the anisotropic magnetoresistance.^[85,164,165] And when electrons transfer from pairing $3d_{xy}$ state to impairing $3d_{xz/yz}$ state, the spin will not be zero and lead to the anomalous Hall effect.^[166] This model can explain the low-temperature correlated transport behavior, but why the $3d_{xy}$ electrons have strong attracting interactions is still unclear.

Now we focus on the spin-orbital coupling at the LAO/STO interface. Because of the polar nature of LAO and the built-in electric field parallel to the surface normal, the resulted inversion symmetry breaking leads to strong Rashba spin-orbit (SO) field which can be tuned by an external electric field.^[84,85] The simultaneous existence of ferromagnetism and tunable Rashba SO coupling (SOC) in LAO/STO allows the use of magnetotransport studies to probe the SO fields at the LAO/STO interface. The AMR occurs when there is a change in the angle between the applied current and the magnetization of a magnet. It was experimentally shown that AMR can be used as a powerful tool to study both magnetic ordering as well as the Rashba field in LAO/STO.^[167] Figure 10(a) shows a schematic for AMR measurements, in which a current ($I = I_{ac} + I_{dc}$) and an in-plane external magnetic field are applied to the LAO/STO device. Figure 10(b) shows the AMR curves, obtained by changing the angle between the applied current and magnetic field, with $I_{dc} = \pm 250 \mu A$. Both the AMR curves in Fig. 10(b) show a substantial deviation from the conventional $\cos^2\theta$ behavior. Moreover, the asymmetries at $\theta = 0^\circ$ and $\theta = 180^\circ$ are opposite for the opposite polarities of I_{dc} , suggesting a presence of a strong dc-current induced Rashba field (H_R) of 1.76×10^3 T (at a normalized current density of 10^{12} A/m²) which is substantially larger than that observed in the conventional metallic systems.

A comprehensive study^[168] of MR measurements in LAO/STO revealed the existence of a new type of angular dependent MR that is similar to the spin Hall MR^[169-171] observed in ferromagnet (FM)/heavy metal (HM) bilayers. In the case of FM/HM bilayers, it is not

straightforward to differentiate and quantify the bulk vs. interface contributions to the spin Hall MR. On the other hand, due to the interfacial nature of a LAO/STO, the observed phenomenon can be called the interfacial Rashba MR. Figure 10(c) shows the measured normalized MR (R_{xx}) in LAO/STO for the three rotations of magnetic fields (α , β , and γ). The applied dc-current for MR measurements is along the x -direction for all the three cases. Unlike in the α and γ rotations, there is no change in the angle between the applied current and magnetic field in the β rotation. Hence, the origins of observed change in R_{xx} in the β rotation is not from AMR but from the interfacial Rashba MR in LAO/STO. It is noted that the interfacial Rashba MR in LAO/STO ($\sim 7\%$) is much larger than the reported spin Hall MR of FM/HM structures ($0.01 - 1\%$).^[169-171]

Spin pumping techniques have been recently utilized to achieve spin to charge conversion using the Rashba SOC in the LAO/STO.^[172] In Ref. ^[172], the authors measured the spin pumping induced dc-voltage and subsequently estimated the value of inverse Edelstein length (λ_{IEE} , the figure of merit for 3D-spin to 2D-charge conversion) in the NiFe/LAO/STO for various gate voltages as shown in Fig. 10(d). The authors could tune the λ_{IEE} over one order of magnitude by changing the gate voltage. Furthermore, the maximum value of λ_{IEE} in LAO/STO (~ 6 nm) is much larger than the previously reported values in various other material systems.^[173,174]

The above studies establish the presence of strong Rashba SOC using magnetoresistance or spin pumping techniques. One practical application of this strong Rashba SOC in the LAO/STO in spintronics is to convert a charge current into spin current that can be used to control the magnetization of an adjacent FM using spin-orbit torques (SOTs).^[175,176] Very recently, Ref.^[177] demonstrated a giant room temperature charge-to-spin conversion efficiency (θ_P) in the STO/LAO/CoFeB structure using the spin torque ferromagnetic resonance (ST-FMR) technique. Figure 10(e) illustrates the generated spin current (σ) in the LAO/STO and the resultant SOTs (τ_P and $\tau_{\perp}$) on CoFeB. Figure 10(f) shows the extracted θ_P (square symbols)

for various temperatures. θ_P of the LAO/STO is 6.3 at room temperature, which is almost 2 orders of magnitude larger than the spin Hall angles in conventional heavy metals.^[175] Further analysis of the temperature dependent θ_P , which decreases at low temperatures, concluded that the spin transmission through the LAO layer at a high temperature range was due to the inelastic tunneling via localized states in the LAO band gap.

The LAO/STO interface with its strong current induced SOC is a promising system for spintronic research and applications. Highly efficient spin current generation using LAO/STO can find application in SOT based devices such as magnetic memories and oscillators, and may greatly invigorate research in oxide heterostructure based spin devices.

3.2.2. Emergent Ferromagnetic Interaction in Antiferromagnetic Thin Layers

The LaAlO₃/SrTiO₃ interface has set a good example where the formal polarization controls the interfacial conductivity, now we focus on the LaMnO₃/SrTiO₃ with interfacial magnetism. The t_{2g} and e_g orbitals occur when the d orbit of transition metal atom interplays with the crystal field created by the adjacent atoms. More intriguingly, facilitated and stabilized by the atomically-abrupt high quality oxide interface, novel electronic and magnetic state can be induced when the t_{2g} and e_g orbitals (see Fig. 11(a)) interact with external charges, which could be either electron (denoted as e^- in Fig. 11(a)) or hole (denoted as h^+ in Fig. 11(a)). The polar catastrophe has been well demonstrated at the atomically-abrupt insulator-to-metal at oxide interface between non-polar and polar oxides such as STO and LAO (Fig. 11(b)). More intriguingly, the transferred charges interact with d -orbitals like t_{2g} and e_g , consequently providing a possibility of a phase transition from non-magnetic to ferromagnetic states. The detailed electronic reconstruction is governed by the band structure at the interface, as schematically shown by case I in Fig. 11(c) and II in Fig. 11(d). For the former case, such as LAO/STO, when LAO thickness reaches a critical 4 unit cells, the build-up potential is large enough to drive the electron from the valance band of LAO to the conduction band of STO

(Fig. 11(c)). For the latter case, $\text{LaMnO}_3/\text{SrTiO}_3$ (LMO/STO) shows a different reconstruction performance at the interface.^[178] The valance band of LMO is smaller than that of STO while the LAO is just the opposite. Therefore, when the thickness of LMO goes up, electron will be transferred to the conduction band of LMO rather than to STO. Following the inspiration, real space distribution of magnetic flux on the other type of interface, *i.e.*, LMO/STO, was imaged.^[178,179] At this LMO/STO interface, critical thickness modulation^[180] and interfacial enhancement of magnetism^[181] were also investigated.

Beyond magnetic phase modulation induced by the polar catastrophe, antiferromagnetism could produce ferromagnetism in alternative ways, as demonstrated in Fig. 11(g,h,i,j). Electrostatic gating (Fig. 11(g)), in particular ionic liquid gating^[182], enables injection of unprecedented amount of charges. The carrier density could be as large as 10 times of conventional gating and may induce ferromagnetism. Ferroelectricity gating (Fig. 10(h)) is a non-volatile gating, which is highly useful for future energy efficient memory applications, including multiferroic tunnel junction.^[183,184] Multiferroicity simultaneous shows coupled ferroelectricity and ferromagnetism, which already enhanced both theoretical study and device design.^[183,185] Optical doping^[186] provides the possibility of manipulating the subtle electronic states and may interact with the spin at the 2DEG.

3.2.3. Magnetic Interlayer Interaction across an Insulator Spacer

For controlling the interfacial magnetism in a multilayer heterostructure, the interlayer interaction is important, such as the famous RKKY interaction when two ferromagnetic layer is separated by a conducting spacer.^[76] However, when the spacer is an insulator, the situation becomes different. The interlayer interaction can be governed by the spin-polarized tunneling.^[77,78] Recent study has demonstrated a complex phase diagram of interlayer interaction in the $(\text{La,Ca})\text{MnO}_3/\text{Ca}(\text{Ru,Ti})\text{O}_3$ multilayers, where the $(\text{La,Ca})\text{MnO}_3$ layer is the ferromagnetic conductor separated by nonmagnetic insulator $\text{Ca}(\text{Ru,Ti})\text{O}_3$.^[187] By changing

the temperature and magnetic field, the interlayer interaction between (La,Ca)MnO₃ layers can be antiferromagnetic with antiparallel spin alignments of adjacent (La,Ca)MnO₃ layers, ferromagnetic with parallel alignment for all the layers, or even ‘intermediate’ with antiparallel spin alignment for interior layers but parallel for the outer layers. This complicated and tunable interlayer magnetic interaction can be applied to fabricate synthetic antiferromagnets with small interlayer exchange coupling, suitable for providing layer-resolved magnetic switching for novel spintronic applications and studying the spin-flop transition and the domain wall dynamics.

If the interlayer interaction depends only on the tunneling current, the long-range magnetic coupling should not be established when crossing a relative thicker insulator oxide spacer. It is true for the case with the spacer being nonpolar insulator, but it fails when the spacer is polar oxides like LAO or NdGaO₃.^[188] Figure 12 demonstrates typical magnetic-field-dependent magnetization (M - H) hysteresis loops of 10 nm La_{0.67}Sr_{0.33}MnO₃ (LSMO) on LAO/STO at 10 K. The labels FC, M_s , H_c , n and H_{shift} denote to the field cooling process, spontaneous magnetization, coercivity, LAO thickness and the horizontal field-cooled shift of the hysteresis loop. Serving as reference results, there is no change observed in the magnetic parameters mentioned above, when $n \leq 4$, demonstrating that the polar catastrophe for the creation of the 2DEG is important to the modification of magnetism of the LSMO.

Without LAO, owing to a non-collinear ferromagnetic structure, a reduced magnetization ($2.2 \mu_B/\text{Mn}$) was observed in the 10-nm layer of LSMO.^[189] Nevertheless, the sign of the M - H loop shift oscillates with peaks at $n = \sim 7$ and 16, and minimum at $n = \sim 12$. Additional experiments were conducted to obtain a deeper understanding of the oscillation in magnetic interaction. In particular, control experiments with nonpolar ferromagnetic metal oxide SrRuO₃ (SRO), which replaces the upper LSMO layer in the previous heterostructure. As shown in Fig. 12(d), no loop shift was observed in this case. In addition, the top electrode was also replaced by a cobaltite La_{0.67}Sr_{0.33}CoO₃ (LSCO) electrode, similar to manganite. As

shown in Fig. 12(d), an opposite sign of the loop shift is observed in this case, as the sign of the spin–orbit coupling for cobalt is opposite to that of manganese. In order to understand those unexpected oscillating behavior, the magnetic-field-dependent excitations in a polar oxide couple with the transmission of magnetic information across oxide layer of LAO needs to be considered.^[190] Then, it further couples to electrons hopping at the Fermi level of LSMO. Both the hopping and the transmission of magnetic information are possibly influenced by standing waves in the LSMO.^[191]

3.3. Ferroelectricity at Low Dimensional Interfaces

Another example where a number of interfaces and/or interlayer coupling play important roles is in the so-called multiferroic tunnel junction (MFTJ), a device combining a ferroelectric barrier and ferromagnetic electrodes. Taking advantage of ferroelectric-based functionality may be the key for reducing the device feature size and thus increasing the information storage density in memory devices. One way is to stabilize the ferroelectricity in a low dimensional system. However, one major challenge is the depolarization field.^[192] Recently, it has been demonstrated that by taking advantage of polar nanoregions (PNR), which are usually induced by chemical defects or structural disorders, the ferroelectricity can be enhanced when reducing the film thickness or dimension.^[193] Although such PNR-induced ferroelectricity only shows a small amount of polarization, it opens new method to establish the low-dimensional ferroelectricity in heterostructures.

Although at the polarization or ferroelectricity is greatly suppressed at the low-dimensional system, the ferroelectric tunneling junction (FTJ) does need a very strong polarization and is a suitable platform for taking use of low-dimensional ferroelectricity. From the basic tunneling model, an FTJ is a functionalized tunneling device which the tunneling probability is modified by the ferroelectric polarization states of the ferroelectric tunnel barrier. Tunnel electroresistance (TER) is defined by $TER = [R(\leftarrow) - R(\rightarrow)] / R(\rightarrow) * 100\%$,

where $R(\leftarrow)$ and $R(\rightarrow)$ are the junction resistance value when the polarization point to left and right (see Figs. 13(a,b)), respectively. A typical R-V hysteresis loop of a BTO based FTJ is shown in Fig. 13(c), we could directly correlate the switching voltage with the polarization switching events, shown in Fig. 13(d), where ferroelectricity is poled and switched via a local probe in a piezoresponse force microscopy (PFM) set up. The principle of FTJs is analogous to magnetic tunnel junctions (MTJ), except the tunnel probabilities is different by the relative magnetization orientation (parallel or antiparallel) of two electrodes. Comparing to MTJs, the TER ratio is easily go up to 100, 00%, about two order magnitude larger than the tunnel magnetoresistance (TMR) ratio. The explicit reason is magnitude of polarization modulation on the tunneling barrier height are generally about 0.2 eV and above, due to the surface bound charge effect, resulting a large change in the tunneling probability and hence TER ratio. In a MTJ, the major origin of TMR lies in the asymmetry in the density of states of majority and minority spins of two magnetic electrodes, where the direct modification of tunneling profile is not very large. Fortunately, FTJs and MTJs are not exclusive but complementary each other as they address different parts in a tunnel junction. An example of MFTJ is Fe/BTO/LSMO heterostructure. In MFTJ, if there is a coupling between the ferromagnetic electrode and ferroelectric barrier, like Fe and BTO reported by Garcia,^[194] polarization controlled TMR could be realized. In another case, if the interaction is weak, a simple four state non-volatile memory could be realized, like the LSMO/BTO/LSMO devices reported earlier.^[195] The earlier is good for the voltage controlled magnetic devices while later is promising to realize future multifunctional devices responsive to both electric and magnetic stimuli.

One interesting question often asked is the compatibility between the ferroelectricity and tunneling. From previous experimental observations, ferroelectric properties generally degrade with reducing film thickness, but tunneling is only possible when the tunnel barrier is sufficiently thin, usually less than 3 nm. It has been however recently demonstrated a TER ratio about 400 % at room temperature in an FTJ consisting of Pt/BaTiO₃/Nb:SrTiO₃ with

BTO thickness of only 2 uc,^[196] see the different On and Off state I-V curves in Fig. 13(e). The nearly temperature independent tunneling properties, plus the exponential relationship between tunnel resistance area product value and the barrier thickness, conclusively proves that the measured properties are dominated by a direct tunneling process. Beyond 2 uc BTO ferroelectric performance in a device, it was discovered that 2 uc BiFeO₃ based FTJ functions normally at room temperature (TER ~ 2700 %) with the piezoelectric force microscope tip as a top electrode.^[197] The I-V curves measured from conducting AFM methods demonstrates the down scaling potential to sub 100 nm scale.

Beyond the classic behavior of the two level of resistance states in FTJs, multiple states have been realized utilizing both ferroelectric switching and oxygen vacancy migration.^[198] A three-state non-volatile memory R-V loop is shown in Fig. 13(f). This is enabled by comparison between the tunnel junctions between Pt/BTO/NSTO and Pt/STO/NSTO. From the comparison, it was found out oxygen vacancy migration is activated at different threshold electric field while ferroelectric polarization is switched at the coercive field. The difference in the switching field made it possible to address the states separately, from which a three-state non-volatile memory device using a simple FTJ geometry was developed.

A critical issue to be solved is the integration of FTJs with Si and large area applications^[199]. The devices can be fabricated for example on a STO buffered Si wafer, where STO is grown on Si by molecular beam epitaxy or pulsed laser deposition method. Even if there is a thin layer (~ 4 nm) of amorphous layer between STO and Si substrates, the STO layer is still expected to be of high quality crystalline. The epitaxial layer should include bottom electrode (*e.g.*, La_{2/3}Sr_{1/3}MnO₃), ferroelectric barrier layer (*e.g.*, BaTiO₃) and top electrode (*e.g.*, Pt). It has been reported that when the BTO film is about 2 nm (~5 uc), a large on-off ratio (~ 158,000%) at room temperature can be achieved. Furthermore, the FTJ shows about 10⁵ cycles without no much fatigue. It is a surprise that in BTO and BFO based FTJs, even the thickness is as thin as 2 uc, good resistance voltage loop is observed at room

temperature, in both device and local probe measurement geometries. The three-state nonvolatile memory device further opens the way to multi-state high density devices and last but not least, FTJ devices on Si is a demonstration of the feasibility to expand the FTJs from university labs to industrial large-scale applications.

4. Conclusion

The seminal observation of 2DEG at the oxide interface in 2004 kicked off an intense research effort that has rapidly led to advances both in the understanding of the underlying physical mechanisms and in the creation and tuning of novel properties at the interfaces, potential for future applications. Enhanced control at the materials science of this oxide interface, for example with respect to the capability of designing defects and structural symmetry breaking at the interface, a variety of new material combinations and interfaces can also now be prepared at unprecedented level of sophistication. Several schemes for the controlled creation of the interface have been demonstrated, including defect engineering, tuning formal polarization, structural symmetry breaking and interlayer coupling. 2DEG has been a routine observation. Low temperature superconductivity and large spin orbit coupling have been clearly observed, with tunable critical temperature and spin orbit field. The mechanism behind the electronic separation and the coexistence of superconductivity and magnetism are beginning to be understood. Some pending issues however still need to be addressed. For example while it is now possible to reproduce and stabilize the electronic properties of the interface, the relatively few measurements of Curie temperature and magnetization at the interface yield scattered results. A better understanding of the role of defects and of the influence of inhomogeneities in the material is also needed to achieve the better control of magnetism and other properties. With increasing use of surface sensitive measurement techniques and improved control of the oxide stoichiometry and defect content, it should become clearer what new physics is involved. It is to be expected that further discoveries will

arise from such interfaces, of which higher temperature superconductivity would be a most tantalizing prospect. The recent advances in the field make us optimistic for the medium-term realization of some of the devices that have been already proposed and to be integrated on silicon. Besides those described, many other device concepts can be anticipated: for example multi-state memories and components for bioinspired computing. The coexistence of different properties would make it possible to design new device paradigm in which switching devices with extremely low energy might be accessible. Because of their versatility and stability, the fact that they can be fabricated using relatively conventional thin film growth techniques, and the possibility to controllably create field effect devices and functional nanostructures, complex oxides and their interfaces present serious candidate materials systems for future nano-electronic device technologies.

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Figure 1. The interface-engineering strategies and some relevant phenomena. The Defect engineering can induce the novel ferromagnetism and/or ferroelectricity at the interface between nonmagnetic and/or non-ferroelectric oxides. The structural symmetry breaking at heterointerfaces is applied for manipulating the orbital reconstruction and bond angle for tuning the electronic behavior. The uncompensated formal polarization in polar oxide layers can trigger the charge transfer accompanied with electronic reconstructions at the oxide polar/nonpolar interfaces. The interlayer interactions play key roles in tuning the performance of multiferroic tunneling junctions and long-range magnetic coupling.

Figure 2. Formal polarization in perovskite ABO_3 lattices. (a) The sub-layers of AO/BO_2 with surface normal along $[001]$. (b) Nonpolar SrTiO_3 (001). (c) Polar LaAlO_3 (001). (d) The sub-layers of AO_3/B with surface normal along $[111]$. (e) Polar SrTiO_3 (111). (f) Polar LaAlO_3 (111).

Figure 3. Polar catastrophe and compensation. The discontinuity of formal polarization and divergent potentials at the (001) LaAlO₃/SrTiO₃ interface on the (a) TiO₂-terminated and (b) SrO-terminated SrTiO₃. In order to avoid the polar catastrophe, the negative charges are needed to transfer to the interfacial TiO₂ sub-layer to form the *n*-type LaO/TiO₂ interface in (c), while positive charge to SrO sub-layer for the *p*-type AlO₂/SrO interface in (d). Reproduced with permission.^[34] 2006, Nature Publishing Group.

Figure 4. (110) LaAlO₃/SrTiO₃ interface. (a) No polar discontinuity at the (110)-orientated LaAlO₃/SrTiO₃ interface, when the parallel interface is considered in (b). (c) The buckled interface which is energetically favored and with polar discontinuity of TiO₂⁰/LaO⁺. Reproduced with permission.^[101] 2013, Nature Publishing Group.

Figure 5. Mechanisms for the conducting LaAlO₃/SrTiO₃ interface. (a) Sketch of two mechanisms, polar discontinuity and oxygen vacancy, for inducing mobile electrons at the LaAlO₃/SrTiO₃ interface. (b) Room-temperature sheet resistance of 20-nm amorphous LaAlO₃/SrTiO₃ heterostructures prepared at different oxygen pressures before and after oxygen annealing in 1 bar of oxygen gas flow at 600 °C for 1 h. (c) Temperature-dependent sheet resistance (inset, temperature-dependent carrier density) of a 10-unit-cell crystalline LaAlO₃/SrTiO₃ heterostructure prepared at 10⁻³ Torr and 750 °C before and after oxygen annealing in 1 bar of oxygen gas flow at 600 °C for 1 h. Reproduced with permission.^[93] 2013, American physical Society.

Figure 6. Optical conductivity spectra and charge transfer mechanisms of LaAlO₃/SrTiO₃. (a) Optical conductivity spectra of LaAlO₃ films and (b) LaAlO₃/SrTiO₃ interfaces at different LaAlO₃ film thickness obtained using combination of spectroscopic ellipsometry and UV-VUV reflectivity, along with the respective optical transitions responsible for each peak. (c) Charge transfer and redistribution mechanisms in LaAlO₃ films and (d) LaAlO₃/SrTiO₃ interfaces at different LaAlO₃ film thicknesses obtained, using the f -sum rule, from their respective optical conductivity spectra. Reproduced with permission.^[118] 2014, Nature Publishing Group.

Figure 7. Transport properties at three interfaces with different Ti-O-Ti bond configurations. (a) Sheet resistance, (b) carrier density, and (c) mobility of the mobile electrons created by an amorphous LaAlO₃ overlayer on the 001 surfaces of strontium titanate (red), anatase (green), and rutile (blue). Inset of (a) shows the measurement geometry. Reproduced with permission.^[148] 2015, American Chemical Society.

Figure 8. Comparison of the crystal structures of strontium titanate, anatase, and rutile. The 001 axis is vertical, and the surface in each case is a plane with composition TiO_2 . The Sr, Ti, and O ions are colored in green, silver, and red, respectively. Reproduced with permission.^[148] 2015, American Chemical Society.

Figure 9. Magnetic interaction and electronic phase separation at the LaAlO₃/SrTiO₃ interface. (a) Temperature dependence of the sheet resistance for *n*-type LaAlO₃/SrTiO₃ conducting interfaces prepared under different partial oxygen pressure. (b) The 1 kOe field-cooling (FC) and zero-field-cooling (ZFC) in-plane magnetization as a function of temperature and measured by a 0.1 kOe magnetic field applied while warming the sample from 2 to 300 K (solid black lines) for the ten unit cells of LaAlO₃/SrTiO₃ samples prepared at an oxygen partial pressure (P_{O_2}) of 1×10^{-2} mbar. (c) LaAlO₃/SrTiO₃ scanning SQUID image mapping the ferromagnetic order. Inset: Scale image of the SQUID pick-up loop used to sense magnetic flux. Reproduced with permission.^[83,86,88] 2007, 2011, 2011, Nature Publishing Group.

Figure 10: Spin-orbital coupling at the LaAlO₃/SrTiO₃ interface. (a) Measurement schematic for longitudinal resistance for a LAO/STO device. (b) Angular AMR data measured from LAO (5 unit cells)/STO at 2 K for $I_{dc} = \pm 250 \mu A$ with fits. (c) Angular dependence of normalized MR (R_{xx}) from LAO (12 unit cells)/STO as a function of the rotating angle (α , β , and γ) with an applied field of $B = 9$ T. (d) Gate dependence of the inverse Edelstein length, λ_{IEE} measured from the NiFe (20 nm)/LAO (2 unit cells)/STO at 7 K. (e) Illustration of spin current (σ) generation and SOTs (τ_P and $\tau_{\perp}$) in the STO/LAO (8 unit cells)/Co₁₅Fe₆₀B₂₅ (7 nm) structure during ST-FMR measurements. (f) SOT efficiency, θ_p (squares) extracted from the STO/LAO/Co₁₅Fe₆₀B₂₅ structure as a function of temperature with the fit. Reproduced with permission.^[167,168,172,177] 2014 (AIP), 2017 (APS), 2016 (NPG), 2017 (ACS).

Figure 11: Magnetism induced by formal polarization at the interface between non-polar and polar perovskite oxides. (a) Symbols of orbit and charge used in this figure. (b) Schematics of atomic arrangement of oxide interfaces between polar and non-polar perovskite oxides. (c,d) Two representative cases of interfaces where transferred charge due to polar catastrophe interacts with orbitals at different sides of interfaces. (e,f) Real space imaging of magnetic flux distribution of oxide interfaces of (e) $\text{LaAlO}_3/\text{SrTiO}_3$ and (f) $\text{LaMnO}_3/\text{SrTiO}_3$ measured by scanning SQUID microscopy. (g,h,i,j) Alternative charge-orbital interactions based on (f) electrostatic gating, (h) ferroelectric gating, (i) multiferroics, and (j) optical doping. Reproduced with permission. ^U Copyright Year, Publisher.

Figure 12. Long range exchange coupling in oxide heterostructures. (a) Hysteresis loops and a reverse case (only for 8 uc) with different LAO thickness after 7 T field cooling (FC) at 10 K. (b - d) The horizontal shift of the M - H loop (H_{shift}), coercivity (H_c) and saturation magnetization (M_s) as a function of LAO thickness. The solid lines in b - d are all fits to an exponentially damped sinusoidal oscillation. (d) The H_{shift} for a 10 nm LSMO bottom layer with different top layers. No shift observed for nonpolar metal (SRO). The LSCO produced a shift opposite to that of LSMO, due to the opposite signs of spin-orbit coupling in LSCO. Reproduced with permission.^[188] 2015, Nature Publishing Group.

Figure 13: Ferroelectric Tunnel Junction (FTJ). (a, b) Schematic images of the tunnel barrier profile of the Pt/BTO/NSTO ferroelectric tunnel junctions at ON and OFF state, respectively. (c) A typical resistance voltage (R-V) hysteresis loop of a Pt/BTO/NSTO FTJ. (d) The phase and amplitude voltage hysteresis loops, measured by PFM, of ferroelectric BTO layer. The switching voltages are almost the same as the switching voltages in the corresponding FTJs shown in (c). (e) I-V curves of the ON and OFF states of a 2 uc BTO FTJ. (f) Dual R-V hysteresis loop shows the 3-state resistance memory device in an FTJ with BTO containing oxygen vacancies. Reproduced with permission.