Diluted Oxide Interfaces with Tunable Ground States

The metallic interface between two oxide insulators, such as LaAlO3/SrTiO3 (LAO/STO), provides new opportunities for electronics and spintronics. However, due to the presence of multiple orbital populations, tailoring the interfacial properties such as the ground state and metal-insulator transitions remains challenging. Here, we report an unforeseen tunability of the phase diagram of LAO/STO by alloying LAO with a ferromagnetic LaMnO3 insulator without forming lattice disorder and at the same time without changing the polarity of the system. By increasing the Mn-doping level, \( x \), of LaAl\(_{1-x}\)Mn\(_x\)O\(_3\)/STO (0 ≤ \( x \) ≤ 1), the interface undergoes a Lifshitz transition at \( x = 0.225 \) across a critical carrier density of \( n_c = 2.8 \times 10^{13} \) cm\(^{-2}\), where a peak TSC = 255 mK of superconducting transition temperature is observed. Moreover, the LaAl\(_{1-x}\)Mn\(_x\)O\(_3\) turns ferromagnetic at \( x \geq 0.25 \). Remarkably, at \( x = 0.3 \), where the metallic interface is populated by only \( d_{xy} \) electrons and just before it becomes insulating, we achieve reproducibly a same device with both signatures of superconductivity and clear anomalous Hall effect (7.8×10\(^{12}\) cm\(^{-2}\) < \( n_s \) ≤ 1.1×10\(^{13}\) cm\(^{-2}\)). This provides a unique and effective way to tailor oxide interfaces for designing on-demand electronic and spintronic devices.
Nanoscale patterning of electronic devices at the amorphous LaAlO3/SrTiO3 oxide interface using an electron sensitive polymer mask

A simple approach is presented for designing complex oxide mesoscopic electronic devices based on the conducting interfaces of room temperature grown LaAlO3/SrTiO3 heterostructures. The technique is based entirely on methods known from conventional semiconductor processing technology, and we demonstrate a lateral resolution of similar to 100 nm. We study the low temperature transport properties of nanoscale wires and demonstrate the feasibility of the technique for defining in-plane gates allowing local control of the electrostatic environment in mesoscopic devices. (C) 2018 Author(s).

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Tuning the Ground State of Oxide Interfaces by an Electron Sink

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Tuning the Two-Dimensional Electron Gas at Oxide Interfaces with Ti-O Configurations: Evidence from X-ray Photoelectron Spectroscopy

Chemical redox reaction can lead to a two-dimensional electron gas (2DEG) at the interface between a TiO2-terminated SrTiO3 (STO) substrate and an amorphous LaAlO3 (a-LAO) capping layer. When replacing the STO substrate with rutile and anatase TiO2 substrates, considerable differences in interfacial conduction are observed. Based on X-ray photoelectron spectroscopy (XPS) and transport measurements, we conclude that the interfacial conduction comes from redox reactions, and that the differences among the materials systems result mainly from variations in the activation energies for the diffusion of oxygen vacancies at substrate surfaces.

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Controlling the Carrier Density of SrTiO$_3$-Based Heterostructures with Annealing

The conducting interface between the insulating oxides LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) displays numerous physical phenomena that can be tuned by varying the carrier density, which is generally achieved by electrostatic gating or...
adjustment of growth parameters. Here, it is reported how annealing in oxygen at low temperatures ($T < 300 \, ^\circ\text{C}$) can be used as a simple route to control the carrier density by several orders of magnitude. The pathway to control the carrier density relies on donor oxidation and is thus applicable to material systems where oxygen vacancies are the dominant source of conductivity. Using STO capped with epitaxial $\gamma$-Al$_2$O$_3$ (GAO) or amorphous LAO (a-LAO), the pathways for changing the carrier density in the two STO-based cases are identified where oxygen blocking (GAO) and oxygen permeable (a-LAO) films create interface conductivity from oxygen vacancies located in STO near the interface. For a-LAO/STO, the rate limiting step ($E_a = 0.25 \, \text{eV}$) for oxidizing oxygen vacancies is the transportation of oxygen from the atmosphere through the a-LAO film, whereas GAO/STO is limited by oxygen migration inside STO ($E_a = 0.5 \, \text{eV}$). Finally, it is showed how the control of the carrier density enables writing of conducting nanostructures in $\gamma$-Al$_2$O$_3$/STO by conducting atomic force microscopy.

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**Giant tunability of the two-dimensional electron gas at the interface of $\gamma$-Al$_2$O$_3$/SrTiO$_3$**

Two-dimensional electron gases (2DEGs) formed at the interface between two oxide insulators provide a rich platform for the next generation of electronic devices. However, their high carrier density makes it rather challenging to control the interface properties under a low electric field through a dielectric solid insulator, i.e. in the configuration of conventional field-effect transistors. To surpass this long-standing limit, we used ionic liquids as the dielectric layer for electrostatic gating of oxide interfaces in an electric double layer transistor (EDLT) configuration. Herein, we reported giant tunability of the physical properties of 2DEGs at the perovskite/spinel interface of $\gamma$-Al$_2$O$_3$/SrTiO$_3$ (GAO/STO). By modulating the carrier density thus the band filling with ionic-liquid gating, the system experiences a Lifshitz transition at a critical carrier density of $3.0 \times 10^{13} \, \text{cm}^{-2}$, where a remarkably strong enhancement of Rashba spin-orbit interaction and an emergence of Kondo effect at low temperatures are observed. Moreover, as the carrier concentration depletes with decreasing gating voltage, the electron mobility is enhanced by more than 6 times in magnitude, leading to the observation of clear quantum oscillations. The great tunability of GAO/STO interface by EDLT gating not only shows promise for design of oxide devices with on-demand properties, but also sheds new light on the electronic structure of 2DEG at the non-isostructural spinel/perovskite interface.

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Suppressed carrier density for the patterned high mobility two-dimensional electron gas at γ-Al₂O₃/SrTiO₃ heterointerfaces

The two-dimensional electron gas (2DEG) at the non-isostuctural interface between spinel γ-Al₂O₃ and perovskite SrTiO₃ is featured by a record electron mobility among complex oxide interfaces in addition to a high carrier density up to the order of 10¹⁵ cm⁻². Herein, we report on the patterning of 2DEG at the γ-Al₂O₃/SrTiO₃ interface grown at 650 °C by pulsed laser deposition using a hard mask of LaMnO₃. The patterned 2DEG exhibits a critical thickness of 2 unit cells γ-Al₂O₃ for the occurrence of interface conductivity, similar to the unpatterned sample. However, its maximum carrier density is found to be approximately 3×10¹³ cm⁻², much lower than that of the unpatterned sample (~10¹⁵ cm⁻²). Remarkably, a high electron mobility of approximately 3,600 cm²V⁻¹s⁻¹ was obtained at low temperatures for the patterned 2DEG at a carrier density of ~7×10¹² cm⁻², which exhibits clear Shubnikov-de Hass quantum oscillations. The patterned high-mobility 2DEG at the γ-Al₂O₃/SrTiO₃ interface paves the way for the design and application of spinel/perovskite interfaces for high-mobility all-oxide electronic devices.
Transport and excitations in a negative-U quantum dot at the LaAlO$_3$/SrTiO$_3$ interface

In a solid-state host, attractive electron–electron interactions can lead to the formation of local electron pairs which play an important role in the understanding of prominent phenomena such as high $T_c$ superconductivity and the pseudogap phase. Recently, evidence of a paired ground state without superconductivity was demonstrated at the level of single electrons in quantum dots at the interface of LaAlO$_3$ and SrTiO$_3$. Here, we present a detailed study of the excitation spectrum and transport processes of a gate-defined LaAlO$_3$/SrTiO$_3$ quantum dot exhibiting pairing at low temperatures. For weak tunneling, the spectrum agrees with calculations based on the Anderson model with a negative effective charging energy $U$, and exhibits an energy gap corresponding to the Zeeman energy of the magnetic pair-breaking field. In contrast, for
strong coupling, low-bias conductance is enhanced with a characteristic dependence on temperature, magnetic field and chemical potential consistent with the charge Kondo effect.
Tuning the ground state of polar LaAlO$_3$/SrTiO$_3$ interface by an electron sink

Most of the intriguing properties of two-dimensional electron gases (2DEGs) at the LaAlO$_3$/SrTiO$_3$ (LAO/STO) interface are sensitive to the electrons located in 3d-orbit of Ti. However, tuning the electronic structure of the system remains challenging due to the intrinsic high carrier density. Herein, instead of using LaMnO$_3$ (LMO) as buffer layers [1], we show that Mn doping in LaAlO$_3$ (LAMO) creates an electron sink that alters the ground state of 2DEG by suppressing the carrier density at the interface, without changing the polarity of the system. By precise control of the Mn-doping level, we found that 2DEGs in our system experience a change from two-band to one-band transport with decreasing carrier density, which is accompanied by a Lifshitz transition at a critical carrier density of $2.76 \times 10^{13}$ cm$^{-2}$ at 2K. Significantly, the peak value (255.7mK) of superconducting transition temperature is observed at Lifshitz point. In addition, our experiments realize the coexistence of ferromagnetism (FM) and superconductivity (SC) by Mn doping.

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Two-Dimensional Electron Gases at Modulation-doped Oxide Interfaces

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Electric field control of the γ-Al₂O₃/SrTiO₃ interface conductivity at room temperature

Controlling interfaces using electric fields is at the heart of modern electronics. The discovery of the conducting interface between the two insulating oxides LaAlO₃ (LAO) and SrTiO₃ (STO) has led to a number of interesting electric field-dependent phenomena. Recently, it was shown that replacing LAO with a spinel γ-Al₂O₃ (GAO) allows a good pseudo-epitaxial film growth and high electron mobility at low temperatures. Here, we show that the GAO/STO interface resistance, similar to LAO/STO, can be tuned by orders of magnitude at room temperature using the electric field of a backgate. The resistance change is non-volatile, bipolar, and can be tuned continuously rather than being a simple on/off switch. Exposure to light significantly changes the capabilities to tune the interface resistance. High- and low-resistive states are obtained by annihilation and creation, respectively, of free n-type carriers, and we speculate that electromigration of oxygen vacancies is the origin of the tunability.
New Insights into the Creation of High-Mobility Two-Dimensional Electron Gas at Oxide Interfaces: Control of Interfacial Redox Reactions by an Electron Sink

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**Abstract**

This study presents new insights into the creation of high-mobility two-dimensional electron gas (2DEG) at oxide interfaces. Specifically, it focuses on controlling interfacial redox reactions through the use of an electron sink. The research is conducted by a team from the Department of Energy Conversion and Storage, Electrofunctional materials, University of British Columbia, Canadian Light Source, and Chinese Academy of Sciences.

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Magnetization reversals, Electric fields, Vacancies, Electrical resistivity, Heterojunctions

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Quantization of Hall Resistance at the Metallic Interface between an Oxide Insulator and SrTiO$_3$

The two-dimensional metal forming at the interface between an oxide insulator and SrTiO$_3$ provides new opportunities for oxide electronics. However, the quantum Hall effect, one of the most fascinating effects of electrons confined in two dimensions, remains underexplored at these complex oxide heterointerfaces. Here, we report the experimental observation of quantized Hall resistance in a SrTiO$_3$ heterointerface based on the modulation-doped amorphous-LaAlO$_3$/SrTiO$_3$ heterostructure, which exhibits both high electron mobility exceeding 10,000 cm$^2$/V s and low carrier density on the order of ~10$^{12}$ cm$^{-2}$. Along with unambiguous Shubnikov-de Haas oscillations, the spacing of the quantized Hall resistance suggests that the interface is comprised of a single quantum well with ten parallel conducting two-dimensional subbands. This provides new insight into the electronic structure of conducting oxide interfaces and represents an important step towards designing and understanding advanced oxide devices.

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Patterning of high mobility electron gases at complex oxide interfaces

Oxide interfaces provide an opportunity for electronics. However, patterning of electron gases at complex oxide interfaces is challenging. In particular, patterning of complex oxides while preserving a high electron mobility remains underexplored and inhibits the study of quantum mechanical effects where extended electron mean free paths are paramount. This letter presents an effective patterning strategy of both the amorphous-LaAlO3/SrTiO3 (a-LAO/STO) and modulation-doped amorphous-LaAlO3/La7/8Sr1/8MnO3/SrTiO3 (a-LAO/LSM/STO) oxide interfaces. Our patterning is based on selective wet etching of amorphous LSM (a-LSM) thin films, which acts as a hard mask during subsequent depositions. Strikingly, the patterned modulation-doped interface shows electron mobilities up to $\sim 8700$ cm$^2$/V s at 2 K, which is among the highest reported values for patterned conducting complex oxide interfaces that usually are $\sim 1000$ cm$^2$/V s at 2K. © 2015 AIP Publishing LLC.

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