Emergent Phenomena in two-dimensional electron gases at oxide interfaces

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Outline

- Complex oxide heterostructures
- Oxide thin film growth
- Two-dimensional electron gases at complex oxide interfaces
- Electron correlations in extreme density quantum wells of SrTiO$_3$
- Interface-induced magnetism
- Summary
**Introduction**

Quantum-confined structures in III-V semiconductors have lead to a wealth of phenomena and new technologies.

**III-V Semiconductor 2D Electron Gases**

- **The Four Main Families of Devices Originating from Ultrathin, Well-Controlled Semiconductor Homo- and Heterostructures**

  - **Two-Dimensional Systems**
    - SDHT-TEGFET-HEMT-MODFET
    - NPI
    - Quantum Wells
    - Quantum Hall Devices
  
  - **Charge Transfer Systems**
    - SDHT-TEGFET-HEMT-MODFET
    - NPI
    - Real Space Transfer Devices
  
  - **Bandgap Engineered Structures**
    - Double-Heterostructure Lasers
    - Graded-Gap APD
    - Heterostructure Bipolar Transistors (graded base or not)
    - Separate Absorption-Multiplication APD
    - Staircase Solid State Photomultiplier

  *Note that the same structures can belong to several of the families and that, using the term bandgap engineering in its most general description of engineered structures with desired properties obtained by a tailoring of the band structure, all of the structures can be considered “bandgap-engineered.”*

**Complex Oxide 2D Electron Gases**

- GdTiO₃
- SrTiO₃
- 3 nm

Fundamentally different:
- Electrons in relatively narrow d-bands
- Occupy a significant fraction of the d-band
- Electron correlations, exchange...dominate transport
- Rich physical phenomena (magnetism, superconductivity, ...)
- Underlying physics poorly understood
- Exploration in its infancy
- High-quality films are needed

Quantum-confined structures in III-V semiconductors have lead to a wealth of phenomena and new technologies.
Why Complex Oxide Heterostructures?

- Two-dimensionality and quantum confinement
- Enhance strong electron correlations
- Proximity effects
- Unique charge and magnetically ordered states

External control of strongly correlated phenomena through electrostatic doping
- Modulation doping
- Electric field effect
High electron densities are key for strong electron correlation physics.

Short range Coulomb interactions require significant probability for two electrons to occupy the same site.

Opposite of the usual correlation regime investigated in conventional semiconductor 2DEGs.
Complex Oxide Materials: Perovskite Titanates

Mott Insulators
- \( RTiO_3 \) where \( R = \text{La} \ldots \text{Y} \)

Band Insulators
- \( \text{SrTiO}_3, \text{BaTiO}_3, \ldots \)

- Transition metal-oxygen octahedra: tilts and rotations
- Determine physical properties
- Modified in thin films through strain, proximity, ...
Complex oxide heterostructures

Oxide thin film growth

Two-dimensional electron gases at complex oxide interfaces

Electron correlations in extreme density quantum wells of SrTiO$_3$

Interface-induced magnetism

Summary
Most complex oxides films are deposited by PLD or sputtering

Main issues:
- Impurities (H, C, transition metals, ...)
- Energetic deposition creating defects
- Poor stoichiometry control during deposition

Oxide Molecular Beam Epitaxy

- High purity
- Low energetic deposition
- Stoichiometry control?

Without an MBE growth window, stoichiometry control requires precise flux control → only possible to 0.1 - 1 %

Corresponds to defect concentrations of $10^{20} - 10^{21}$ cm$^{-3}$
A wide MBE growth window is largely responsible for the ease and success of III-V MBE.

Below this line, solid As will not precipitate → excess As desorbs in the chamber

Above this line, As will condense on a Ga-rich GaAs surface

J. Tsao, Materials Fundamentals of Molecular Beam Epitaxy.
No practical growth window in oxide MBE using solid sources

- Requires precise flux control → only possible to 0.1 - 1%
- Corresponds to defect concentrations of $10^{20}-10^{21}$ cm$^{-3}$
Oxide Molecular Beam Epitaxy

Titanium tetra isopropoxide (TTIP)

Persistent RHEED oscillations indicate layer-by-layer growth mode [only been reported for a few systems: Si, Pt, AlAs]

Transition to step-flow growth

Streaky RHEED indicates atomically smooth film surface

Surface reconstructions

$2 \times$ along [110] (always); $4 \times$ along [110] (only for stoichiometric films after growth)

Further investigations are required to understand origin of persistent RHEED oscillations.

Oxide Molecular Beam Epitaxy

MBE growth window

TTIP desorption leads to a growth window at practical substrate temperatures and fluxes

Stoichiometry is self-regulating within the growth window

No need for precise flux control

Shift to higher TTIP/Sr flux ratios with increasing temperatures shows that desorption of TTIP is responsible for growth window


Oxide Molecular Beam Epitaxy

- 1:1 correspondence of La-concentration and free carrier concentration over several orders of magnitude
- Excellent control over carrier concentrations $\rightarrow$ concentration of unintentional defects below doping level
- Higher mobility than single crystals
- Fewer charged impurities, such as Al, Fe ..., in MBE films


2DEGs at Oxide Interfaces

Polar/Non-polar Interfaces: Extreme 2D Carrier Densities

- Mobile, compensating charge density
- Fixed charge at (001) interfaces between $RTiO_3$ and $SrTiO_3$
- Free charge carrier density at the interface of $1/2$ electron per interface unit cell, or $\sim 3 \times 10^{14}$ cm$^{-2}$
- Order of magnitude higher charge density than what is achievable in conventional semiconductors
- See also early work by Harrison on Ge/GaAs W. A. Harrison et al., Phys. Rev. B 18, 4402 (1978).
Well-oxidized LaAlO$_3$/SrTiO$_3$ interfaces show $\sim 1 \times 10^{13}$ cm$^{-2}$ carrier densities, an order of magnitude lower than predicted.

Significant debate in the literature as to the origin (or lack thereof) of the free charge carrier density at this interface.
Two-dimensional Electron Gases at Oxide Interfaces

- Lack of charge at the LaAlO$_3$/SrTiO$_3$ interface:
  - Origin of charge carriers: compensating charge for polar discontinuity or defect-related (residual vacancies, interdiffusion,...)?
  - If polar discontinuity is indeed compensated by a “mobile” charge, are the free carriers trapped or localized due strong electron localization?
- Carrier confinement is weak for low-density electron gases in SrTiO$_3$

**Low density electron gases**

- At low densities, charge is spread-out
- Energy separation of subbands is extremely small, multiple bands are filled

![Graph showing average carrier separation from surface versus carrier density.](image)

\[
\dot{w} \sim \left( \frac{\hbar^2 \varepsilon}{4 \pi m e^2 n_T} \right)^{1/3}
\]

\[
\frac{\hbar^2}{m \omega^2} \sim 10^{-4} \text{eV}
\]

\[
w \sim 50 \text{ unit cells}
\]

Guru Khalsa and A.H. MacDonald,
Two-dimensional Electron Gases at Oxide Interfaces

- New insights using alternative interfaces between two insulating oxides
- Charge densities close to theoretical prediction of $3 \times 10^{14}$ cm$^{-2}$
- Test of theoretical predictions
- Strongly confined two-dimensional electron gas
- Strong electron correlations in the 2DEG?
- Interactions between the magnetic insulator and the 2DEG?
- Correlation with superconductivity in the SrTiO$_3$?
- Close proximity of unique properties: two-dimensionality, superconductivity and magnetism
MBE of GdTiO$_3$/SrTiO$_3$ Interfaces

HAADF/STEM

SrTiO$_3$ after growth (4x indicates cation stoichiometry)

RHEED

GdTiO$_3$ on (001) LSAT after growth

Superlattices

One-unit-cell SrTiO$_3$ quantum well embedded in GdTiO$_3$

- GdTiO$_3$ is insulating (p-type)
- Remarkable drop in sheet resistance even for one unit cell of SrTiO$_3$
- Sheet resistance independent of SrTiO$_3$ thickness for all thickness greater than 20 nm
- Sheet carrier density (n-type) is independent of SrTiO$_3$ thickness
- Conduction in a space charge layer with constant thickness
- Sheet charge carrier density corresponds to the theoretical expected density of $\sim 3\times10^{14}$ cm$^{-2}$.

**GdTiO₃/SrTiO₃ Superlattices**

- Sheet carrier concentration scales with number of multilayer repeats (interfaces)
- Independent of SrTiO₃ or GdTiO₃ thickness
- Each interface contributes a constant sheet charge carrier density $\sim 3 \times 10^{14} \text{ cm}^{-2}$.


Note: each repeat $x$ contains two interfaces.
Theory of high-density 2DEGs in SrTiO$_3$

2DEGs in SrTiO$_3$ at carrier densities of 0.5 electron per interface unit cell

May be able to see $d_{yz}$ in SdH, but very large mass

Many, mostly $d_{xy}$-derived bands, close spacing, will be washed out in Shubnikov-de Haas experiments

Large energy separation between bottom $d_{xy}$ band and higher-lying subbands $\rightarrow$ 2D behavior

160 - 220 meV

Quantum oscillations from SrTiO$_3$/GdTiO$_3$ interfaces

Oscillations in tilted fields depend only on $B_\perp$, consistent with a two-dimensional system.

The product of the frequency ($a$) of the Fourier transform of the oscillations in $1/B$ and $\cos \theta$ is independent of $\theta$, as expected for a two-dimensional system.

Quantum oscillations from SrTiO$_3$/GdTiO$_3$ interfaces

At low $B$-fields, observe two periodicities:
Oscillations are due to a single, spin-split subband with an electron density of $f_a = n_a h/e$, where $n_a = 2.8 \times 10^{13}$ cm$^{-2}$
- Consistent with oscillations due to the bottom d$_{xy}$ subband
- Density roughly agrees with theoretical predictions
- Landau level and and Zeeman spin splitting likely comparable in SrTiO$_3$*
- Only one periodicity at high $B$-fields
- Spin splitting is $B$-field dependent
- Influence of Rashba effects?
- Theory lacking


Characteristics of the high-density 2DEG

- Theoretical calculations for high-density 2DEGs in SrTiO$_3$
- Basic picture:

![Energy diagram](image)

- Large energy separation between bottom $d_{xy}$ band and higher-lying subbands → 2D behavior
  - $160$ - $220$ meV
- Many, mostly $d_{xy}$-derived bands, close spacing, will be washed out in Shubnikov-de Haas experiments
- Maybe able to see $d_{yz}$ in SdH, but very large mass

- Shubnikov-de Haas oscillations consistent with basic picture
- Unlikely that the other subbands will be resolved in SdH
- Need alternative method to probe subbands

2DEGs in SrTiO$_3$ and carrier densities of 0.5 electron per interface unit cell
Characteristics of the high-density 2DEG

Experiment:
- Voltage bias is applied across the two 2DEGs (top and bottom)
- Resonant tunnelling occurs when subbands of the two 2DEGs align in energy and match conservation criteria
- In-plane (parallel to the barrier) momentum is conserved ($d_{xy} \leftrightarrow d_{xy}$)

\[ E - \frac{\hbar^2}{2m^*} \left( k_{||}^2 + k_z^2 \right) = E + \frac{\hbar^2}{2m^*} \left( k_{||}^2 + (k_z')^2 \right) \]
Characteristics of the high-density 2DEG

Resonant tunnelling features are seen in the $dI/dV$ and $d^2I/dV^2$ characteristics.

Features under a positive bias indicate tunneling from subbands in bottom 2DEG into the subbands of the top 2DEG, and vice-versa.

Subband spacing of the two 2DEGs can be derived from $d^2I/dV^2$.

Characteristics of the high-density 2DEG

Excellent qualitative agreement between experiments and theory
Subband spacing reflects the small asymmetry in carrier density between top and bottom 2DEG
Larger subband spacing in experiment may be due to compressive strain from the LSAT substrate or calculations underestimating the confinement (mass).

### Resonant Tunneling Experiment

<table>
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<tr>
<th>Subbands (top 2DEG)</th>
<th>Subband spacing (meV)</th>
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### Theory

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| (pure xy-pure xy)    | (mixed xy-
| mixed xy)            | (mixed xy -
| pure xy)            |
| Zhong et al. [3]     | 219       | 57.14     | 28.5      | 42.85     |
| (xy-xy)              | (xy-xy)   | (xy-xy)   | (xy-xy)   |
| Zhong et al. [3]     | ~214      | 64.28     | 64.28     | 100       |
| (xy-xy)              | (xy-xy)   | (xy-xy)   | (xy-xy)   |
| Popovic et al [4]    | 270.96    | 25.8      | 38.7      | 45.16     |
| (xy-xy)              | (xy-xy)   | (xy-xy)   | (xy-xy)   |
| Delugas et al [5]    | 160       | 80        | 64        | 56        |
| (xy-xy)              | (xy-xy)   | (xy-xy)   | (xy-xy)   |


Characteristics of the high-density 2DEG

- Excellent qualitative agreement between experiments and theory
- Subband spacing reflects the small asymmetry in carrier density between top and bottom 2DEG
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Alternative Routes to 2DEGs: Modulation Doping

Separates donors from mobile charge → high mobility
Controllable carrier density → low density electron gases
Enabled devices (HEMT) and scientific discoveries (fractional quantum Hall effect)
Requires suitable band alignments (dopants in a wide-band gap semiconductor) and controlled doping

2DEGs in SrTiO$_3$ by Modulation Doping

- SrZrO$_3$/SrTiO$_3$ have favorable band offsets for modulation doping
- Use Sr(Ti,Zr)O$_3$ as the analog of AlGaAs for smaller lattice mismatch and to facilitate doping
- Use La as the dopant in the Sr(Ti,Zr)O$_3$


Shubnikov-de Haas oscillations provide evidence for a confined 2DEG at the La:Sm(Ti,Zr)O$_3$/SrTiO$_3$ interface. Angle-dependence confirms 2D nature. Interpretation of SdH underway...


Also see: Phys. Rev. B 88, 115304.
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Electrons are located in a band insulator. For electron densities approaching 1 electron/unit cell, expect transition to a Mott insulating state. Two-dimensional, Artificial Mott Insulator. Depends critically on the degree of the confinement of the 2DEG.
Designing a two-dimensional electron gas with on-site Coulomb repulsion (Mott physics)?

- Very large 3D carrier densities in a SrTiO₃ quantum well
- “2 SrO” quantum well: only 1 electron/three Ti layers
- On-site Coulomb repulsion (electron correlations)?
Electron Correlation Effects in High-Density 2DEGs

\[ \rho(T) = \rho_0 + AT^2 \]

\[ A = \frac{4\pi^2k_B^2 am_b}{e^2\hbar^2 n} \left( \frac{\lambda_{tr}(0)^2}{v_F} \right) \]


- Quantum wells with thickness greater than 2 SrO layers are metallic
- Quadratic temperature dependence over a wide temperature range: Fermi liquid (electron-electron scattering)
- A coefficient should decrease with carrier density
Electron Correlation Effects in High-Density 2DEGs

Above a critical carrier density of \( \sim 7 \times 10^{21} \text{ cm}^{-3} \), A coefficient increases with carrier density.

For all titanates, including the thin SrTiO\(_3\) quantum wells, mass-enhancement in proximity to a Mott transition. A coefficient should diverge at the transition.*

Short-range, on-site Coulomb interactions (Mott-Hubbard physics) in a two-dimensional band insulator.

Carriers introduced by electrostatic doping.

\[ \rho(T) = \rho_0 + AT^2 \]

\[ A = \frac{4\pi^2 k_B^2 a m_b}{e^2 h^2 n} \left( \frac{\lambda_{tr}(0)^2}{v_F} \right) \]


Quantum wells thinner than 2 u.c. are **insulating**

Disorder/interface roughness scattering (Anderson) or Mott transition?

Anderson: variable range hopping (VRH)

At $T > 200$ K: thermally activated transport

Small activation energy typical for small polarons*

Variable range hopping

$$\rho(T) \sim \exp\left(\frac{T_0}{T}\right)^y$$

$y = 1/2$ for strong electron interaction and Coulomb gap (Efros-Shklovskii law)

Arrhenius

$$\rho(T) \sim \exp\left(\Delta E/kT\right)$$

Electron Correlation Effects in High-Density 2DEGs

$t_\perp \sim 0.68\text{eV}$

$t \sim 0.32\text{eV}$

Dimer Mott state


- Theory predicts a “Dimer Mott Insulator” state for 1 SrO layer
- Different orientation than experiments
- Structural distortions on the SrO layers are key
Nearly all properties of Mott insulators are coupled with structural distortions (oxygen octahedra tilts).

In $\text{RTiO}_3$, large tilt angles favor ferromagnetism.

What are the structure of extreme electron density SrTiO$_3$ quantum wells?

---

Correlation with Structure?

Gd shifts suppressed at SrTiO$_3$ interface

Gd positions form zigzag pattern in bulk
Gd displacements are correlated with oxygen octahedral tilts
Correlation with Structure?

Shown are the A-site displacements (deviation from 180° angle in cubic SrTiO$_3$).

- Sr atoms show displacements in the 2 SrO-thick, insulating quantum wells, consistent with octahedral tilts.
- Sr atoms are not displaced in the metallic quantum wells.
- The Gd displacements are reduced near the interface.
- Structural changes coincide with the transition to insulating phase.
- Similar to bulk RTiO$_3$.

Electron Correlation Effects in High-Density 2DEGs

- Oxide interfaces allow for narrow quantum wells bound by high-density, two-dimensional electron liquids
- Extremely large carrier densities without chemical doping
- Access fundamental Mott physics
- Evidence for onset of short range Coulomb interactions in a band insulator
  - Mass enhancement in metallic quantum wells
- A correlated insulator emerges at the highest densities (thinnest quantum wells)
- Insulating state is correlated with structural distortions (octahedral tilts), similar to bulk Mott insulators
- Pathway towards the Artificial Mott Insulator
Complex oxide heterostructures
Oxide thin film growth
Two-dimensional electron gases at complex oxide interfaces
Electron correlations in extreme density quantum wells of SrTiO$_3$
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Summary
Two-dimensional Electron Gases at Oxide Interfaces

- Strong electron correlations in the 2DEG?
- Interactions between the magnetic insulator and the 2DEG?
- Close proximity of unique properties: two-dimensionality, superconductivity and magnetism
Proximity Effects at Ferrimagnetic Insulator/2DEG Interfaces

- Very strong ferromagnetic hysteresis, provided that quantum wells are thinner than about 4 SrO layers
- Angle-dependent behavior consistent with anisotropic magneto-resistance (AMR)
- Spin-polarized, high-density 2DEG at an epitaxial oxide interface
- Negative AMR ($\Delta \rho_A = \rho_A - \rho_A < 0$)
  - Similar to III-V 2DEGs under compressive strain
  - Indicative of spin-orbit coupling in the 2DEG

\[ \rho_{xx} = \rho_\perp + \Delta \rho_A m_x^2 \]
\[ \rho_{xy} = \Delta \rho_A m_x m_y \]
\[ m_x = \cos \alpha \cos \beta \]
\[ m_y = \sin \alpha \cos \beta \]

Proximity Effects at Ferrimagnetic Insulator/2DEG Interfaces

- Ferromagnetism in SrTiO$_3$ quantum well is distinct from that of GdTiO$_3$
- Metallic conducting is GdTiO$_3$ not magnetic
- Onset of hysteresis in quantum well at about 5 K, vs. $T_c = 20$ K in GdTiO$_3$
- Coercive fields are different

4 nm GdTiO$_3$ magnetism $T_c \sim 20$ K

Proximity Effects at Ferrimagnetic Insulator/2DEG Interfaces

Replace GdTiO$_3$ with SmTiO$_3$

No hysteresis

Ferromagnetism in SrTiO$_3$ quantum well is a proximity effect

Summary: Proximity Effects in High-Density 2DEGs

- Proximity effects in correlated, high density 2DEGs at epitaxial interfaces
- Spin-polarized, high-density 2DEG
- Conditions favoring unusual states of matter are present in these quantum wells
  - Noncentrosymmetry
  - Strong electron correlations
  - Proximate magnetism
  - Spin-orbit coupling
- Can test more complex spin states and fluctuations, and other proximity effects, such as superconductivity
Thank you