Novel Electronic and Magnetic Phenomena in Nanoscale Quantum Materials

BRIEF DESCRIPTION OF INNOVATION

The focus of this program is the study of novel phenomena in nanostructured materials, such as ultra-thin films, one-dimensional nanowires, and molecular nanosystems created on stepped surfaces or crystalline substrates with the help of molecular beam epitaxy methods and/or atomic manipulation techniques. The aim is that of defining alternative pathways for the design of new materials and functional devices, which hold so much promise for technological advances in fields as diverse as electronics, computer science, telecommunications, and biomedicine. The electronic and magnetic properties of these nanostructured materials will be studied with some of the most powerful tools for experimental condensed matter physics currently available in AMPEL at UBC, such as Angle-Resolved Photoelectron Spectroscopy (APRES), Electron Energy Loss Spectroscopy (EELS), Scanning Tunnelling Microscopy (STM), and other photoelectron spectroscopy techniques accessible on dedicated beamlines at synchrotron facilities, such as the Canadian Light Source, the Stanford Synchrotron Radiation Laboratory, the Advanced Light Source, the Australian Synchrotron, and SPring-8 in Japan (this in particular for X-ray absorption and both elastic and inelastic resonant X-ray scattering).

The main innovative aspect of this research plan is that of combining the most powerful ideas developed in the study of the physics of strongly correlated electron systems (such as the 3d and also 4d transition-metal oxides), with the state-of-the-art capabilities of engineering materials and devices at a nanometer scale. This could lead to new concepts, materials, and technological advances, since by lowering the system dimensions (from three-dimensional bulk over two-dimensional thin films to one-dimensional nanostructures) the physical properties can change dramatically. In particular, this approach could provide us with the opportunity to realize novel collective phenomena in systems generally characterized by conventional physical properties.

We do believe that the direct experience which we have gained over the years in the study, by optical, microwave, and photoelectron spectroscopy, of the electronic correlations in low-dimensional spin systems, high-Tc superconductors, spin, charge, and orbital-ordered materials, will be crucial to the success of the present research plan.

STATEMENT OF THE PROBLEM AND SCIENTIFIC SIGNIFICANCE OF PROPOSED RESEARCH

Novel materials are at the hart of the scientific and technical progress in fields as diverse as electronics, telecommunications, computer science, and biomedicine, as dramatically exemplified by the development and widespread establishment of the semiconductor-based technology. In more recent years, the leading research effort in solid state physics has been directed towards the field of the so-called strongly correlated electron systems, in which the valence electrons self-organize into novel ground states substantially different from those of conventional metals and insulators. Strong electron correlations, in concert with electron-phonon interactions, can give rise to a large variety of spectacular phenomena, including colossal magneto-resistance and high-temperature superconductivity, as observed in the transition-metal oxides.

At this stage, we are once again witnessing the beginning of a new era in condensed matter physics, which has started with the so-called “nanoscience revolution”. Delving into the world of nanoscience - where the scale of structures and devices is between 100 and 1,000 times smaller than what we have today - creates major challenges for scientists and engineers in a broad range of fields. One of the critical issues facing the pioneers of nanoscience and nanotechnology is the fact that the basic physical properties of materials can change dramatically when the characteristic scales are reduced below a few nanometers, necessitating the development of new concepts and designs. One key to overcoming this challenge is understanding the electronic structure of complex systems and nanomaterials. It is in fact the low-energy electronic structure, and in particular the intimate interplay between fundamental characteristics of the electrons (such as spin, charge, and orbital degrees of freedom), that determines the physical properties and application potential of these novel materials, which hold so much promise for important technological advances.

On the fundamental side, the goal is to define alternative pathways for the design of new materials and functions and to understand the underlying microscopic mechanisms responsible for the observed physical properties. On the applied side, the challenge is to learn how to actively control those mechanisms and proceed to the fabrication of specifically engineered nanostructures and functional devices. In this regard, as described in more detail in the following pages, my attempt will be to realize novel magnetic and electronic systems by implementing at the nanometer scale, with atomic manipulation techniques, those mechanisms and concepts which have been observed and/or developed in the field of the correlated electron systems.

A crucial aspect of this research plan is the development of an ultra-high vacuum system for the in-situ fabrication of specifically designed complex systems and nanostructures, such as ultra-thin films, artificial superlattices, and low-dimensional self-assembled structures on lattice-matched substrates. These structures, in which constituents with different chemical and/or physical characteristic are combined to obtain new properties substantially different from those of conventionally synthesized materials, will be the basic building blocks of modern solid-state electronics and spintronics. In order to study their fundamental properties, it is mandatory to have a dedicated preparation chamber directly connected to the electron spectrometers, as the exposure to air would result in severe and permanent damage. Furthermore, given the level of accuracy nowadays achievable with epitaxial and atomic manipulation technique, the intrinsic low-dimensionality of the electronic structure, and the complexity of the system we would like to engineer and study, the photoelectron spectroscopy experiments could and, as a matter of fact, should be performed with better resolution and at lower temperature than what is available in state-of-the-art spectrometers. Therefore, an important part of this research project, which is presently the main effort in the Quantum Materials Lab in AMPEL, is the development of an innovative apparatus for very low temperature (2.2K) ARPES and EEELS experiments with energy resolution in the sub-meV regime combined with angular resolution better than 0.1 degrees. These breakthroughs would allow the study of the temperature-dependent electronic structure of novel complex systems and nanostructured materials at an unprecedented level of detail.

PLAN OF PROCEDURE

The main objective of this research program is the study of nanoscale phenomena in complex systems and nanostructured materials, such as novel magnetic properties and metal-insulator transition in oxide thin films, and electronic properties of molecular nanosystems. It is well known that complex materials show very interesting phenomena, the most spectacular being probably Mott-Hubbard insulating behaviour [1], unconventional and high-temperature superconductivity [1], and colossal magneto-resistance [2]. In addition, by lowering the system dimensions (from three-dimensional bulk over two-dimensional thin films to one-dimensional nanostructures) the physical properties can change dramatically, which could lead to new concepts, materials, and technological advances. On the fundamental side, the aim is to understand the underlying microscopic mechanisms responsible for the physical properties of these systems. On the more applied side, the goal is to learn how to actively control those mechanisms and to define alternative pathways for the design of new materials and functional devices. In this context, in addition to the more traditional work performed on high-quality single crystals, the growth of thin films on crystalline substrates and the fabrication of molecular nanostructures, with molecular beam epitaxy methods and/or atomic manipulation techniques using a scanning-tunnelling microscopy apparatus, will provide exciting and qualitatively new research opportunities. In fact, these methods will allow the production of systems that can not be obtained through the more conventional synthesis processes.

For the development of microscopic theoretical descriptions and, in turn, the exploitation of the full application potential of complex materials and nanostructures, a key aspect is understanding the fundamental electronic and magnetic properties. In this regard, because of the nanometer scale of these systems, ‘surface-sensitive’ techniques such as scanning-tunnelling microscopy (STM) and spectroscopy (STS), angle-resolved photoemission spectroscopy (ARPES), and electron energy loss spectroscopy (EELS) constitute ideal experimental probes. A novel apparatus for ARPES and EELS experiments to be performed at temperature as low as 2.2K, with energy resolution in the sub-meV regime combined with angular resolution better than

0.1 degrees, is currently under development in the Quantum Material Lab at UBC. The STM systems will soon be available in the newly formed group at UBC led by Dr J. Barth. These complimentary tools will be used for the comprehensive investigation of the elementary excitations. It will be possible to study both the energy and momentum dependence of the detected excitations, as well as the electron removal and addition spectra. It should also be emphasized that state-of-the-art STM and angle-resolved techniques such as ARPES will provide us with the opportunity of performing a *microscopic* study of the electronic structure in both *real* and *momentum space*, respectively.

1. (Transition-metal) Oxide nanostructures: novel magnets, nanowires, and metal-insulator transition

The epitaxial growth of ultra-thin films on crystalline substrates and the fabrication of nanostructures with atomic resolution will provide exciting, qualitatively new research opportunities. In fact, this growth method would allow the production of systems that can not be obtained through conventional chemical processes. For instance, we would like to investigate whether a new class of magnetic systems can be obtained by introducing dilute specific point defects in the crystal structure of conventional materials, which could play an important role in particular in the new field of *spintronics* [3]. For example, half-metallic ferromagnets could be obtained from simple nonmagnetic band insulators such as CaO [4], and half-metallic antiferromagnets from antiferromagnetic insulators such as NiO [5], which would all be ideal materials for innovative spin-dependent electronics. In either case, we will start by first attempting to introduce either Ca or Ni vacancies in polycrystalline furnace-grown materials, or alternatively oxygen-nitrogen substitution by annealing the samples in a source of atomic nitrogen. Eventually, when all the equipment will be in place, we will introduce similar dilute point defects with "atomic resolution" in epitaxial thin films. In addition, one-dimensional nanostructures will be also investigated with respect to the possibility of creating nanowires or magnetic chains that exhibit new interesting physical phenomena. These nanostructures could be created on stepped surfaces [6], as well as on substrates or films, with the help of molecular beam epitaxy methods and/or atomic manipulation techniques using an STM apparatus [7,8]. Particularly interesting is the prospect of studying kinks and terraces in system such as CaO and NiO; in fact, due to the reduced coordination one may observe the spontaneous self-organization of magnetic nanostructures. At last, it is worth emphasizing that the epitaxial growth of thin films on crystalline substrates will also be applied as a new approach to study the metal-insulator transition in correlated metal such as, for instance, $CaVO_3$ [9]. By opportunely choosing the film-substrate lattice mismatch, we can attempt to actively tune fundamental parameters such as chemical bond angles and bond lengths (and in turn the overlap integrals between the valence electrons), and this way drive the system through the metal-insulator transition.

2. Molecular nanosystems: organic macromolecules on highly polarizable surfaces

A topic, which is at present of extremely high interest from the point of view of both fundamental scientific research and technological applications, is the fabrication of molecular nanosystems consisting of olygomers and macromolecules on metallic and semiconducting surfaces. These systems hold much promise for important technological advances in fields as diverse as molecular electronics, molecular magnetism, and biotechnology. It has become clear that the physical properties of these molecular materials can change dramatically at the nanometer scale in proximity to the interface with polarizable surfaces, thus necessitating the development of new concepts and designs. For example, even the widely accepted description in terms of "band-bending" for the modification of the electronic structure at semiconductor-metal or semiconductor-semiconductor interfaces has recently been questioned on the basis of photoemission and inverse photoemission experiments on a monolayer of C_{60} on a Ag surface [10,11]; in particular, it was suggested that also the severe reduction of the C_{60} band gap due to screening effects by the metal should also be taken into account [10]. A spectacular example of the unexpected physical properties that can be realized in molecular nanosystems is the recent detection of a temperature-dependent gap on a C_{60} monolayer chemisorbed on a Ag(100) single crystal [11], which is observed at temperature as high as 230K. Although this temperature is approximately 100 K larger than any value of superconducting T_C ever reported [1] and to date there is still no conclusive interpretation of these results, this behavior is very reminiscent what is observed on the high-T_C superconductors. As emphasized by the above examples, one crucial key to overcoming the challenge associated with the fabrication and characterization of functional molecular devices is understanding the electronic structure of complex molecular nanosystems. This will in particular require state-of-the-art techniques such as STM and STS, which can be used for the manipulation and investigation of these materials with atomic resolution and provide direct information on both the electron addition and removal spectra, as well as ARPES and EELS, which can provide complementary insights on the momentum dependence of the elementary excitations. In the particular case of on C_{60} on Ag(100) in order to more directly address the nature of the observed excitation gap we will at first study the effect on the low energy electronic excitations of dilute magnetic and nonmagnetic impurities deposited on the C_{60} surface; for instance, if a suppression of the temperature-dependent gap was observed only in the presence of magnetic impurities (such as, e.g., Gd), this would strongly suggest a direct connection to superconductivity.

REFERENCES

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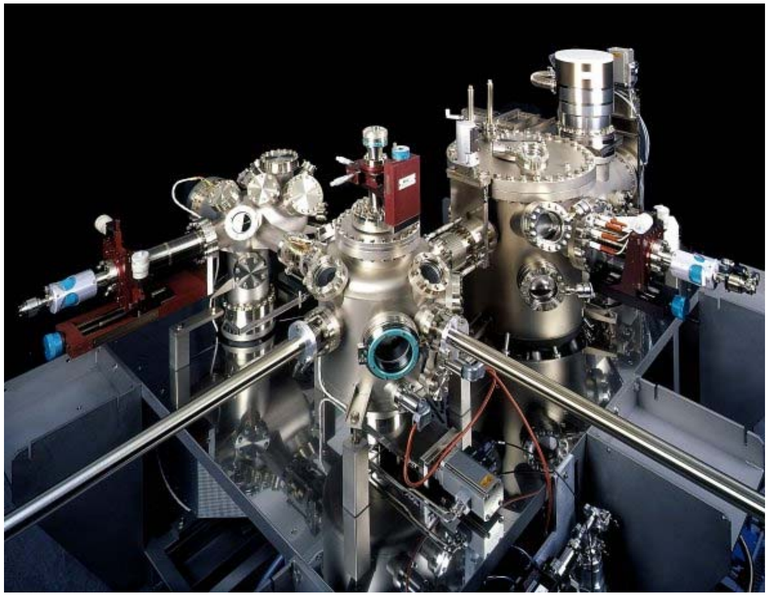
Transition-Metal Oxides films: Growth & Characterization

TMO Molecular Beam Epitaxy

- Evaporation cells, RHEED
- Gas flow system (O_2, N_2, H_2)
- Radical atom source (O, N, H)
- Ozone source

Devices fabrication

- Ag, Au, Ti, Cr evaporators
- In-situ masking system



Analysis tools (low-Temperature)

• LEED	(low-energy electron diffraction)
• XPS	(X-ray photoelectron spectr.)
• UPS	(UV photoelectron spectr.)
• EELS	(electron-energy loss spectr.)
• MOKE	(magneto-optical Kerr effect)
• STM	(scanning tunneling microsc.)

- Transport properties ($h\nu$,**B**)

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SPECS: 3,060,000 CND

(all inclusive)

ThermoVG: 3,500,000 CND

(STM not included)

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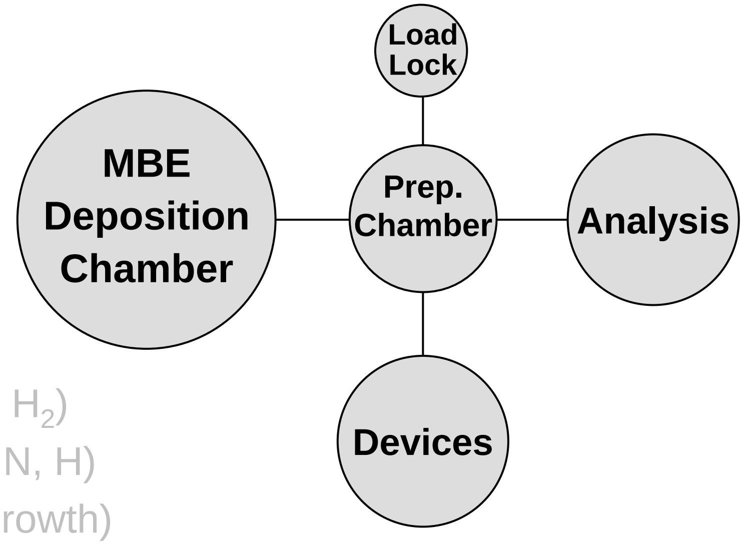
Transition-Metal Oxides films: Growth & Characterization

Equipment to be developed in AMPEL



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- Mass spectrometer
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- Cryogenic paneling

Sample Preparation Chamber

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- Outgassing & parking
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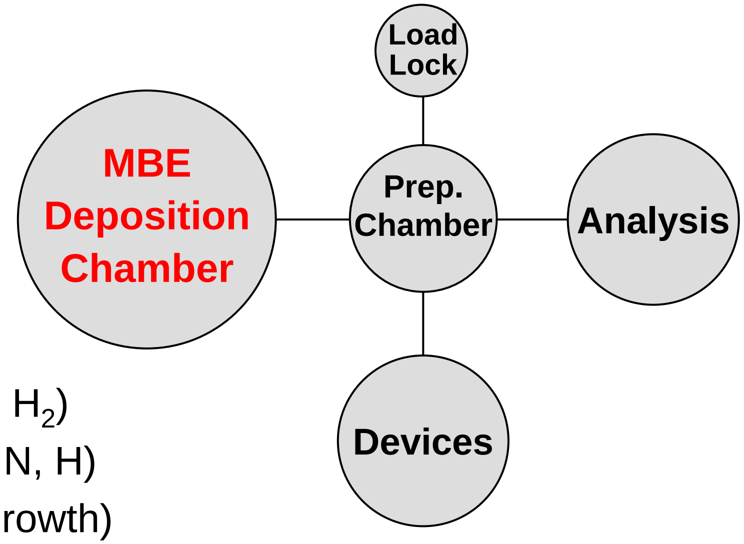

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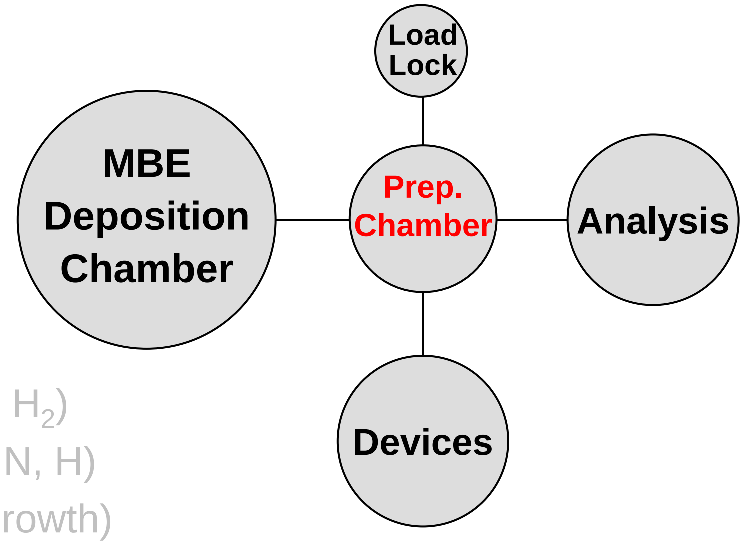

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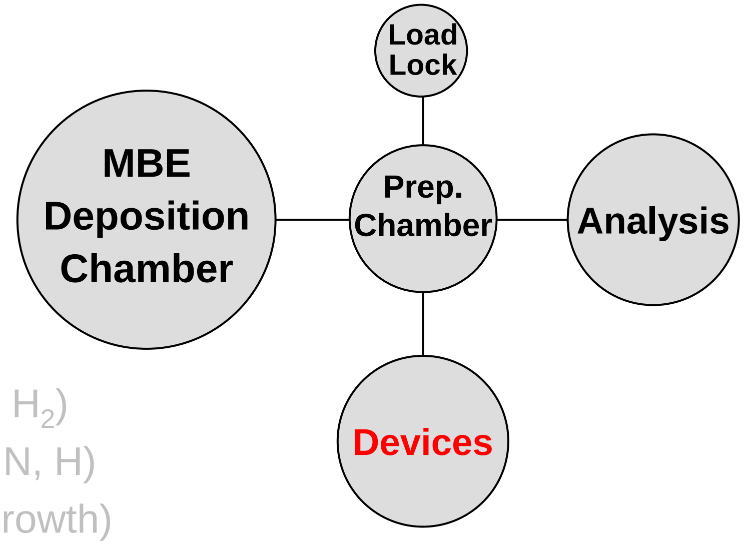

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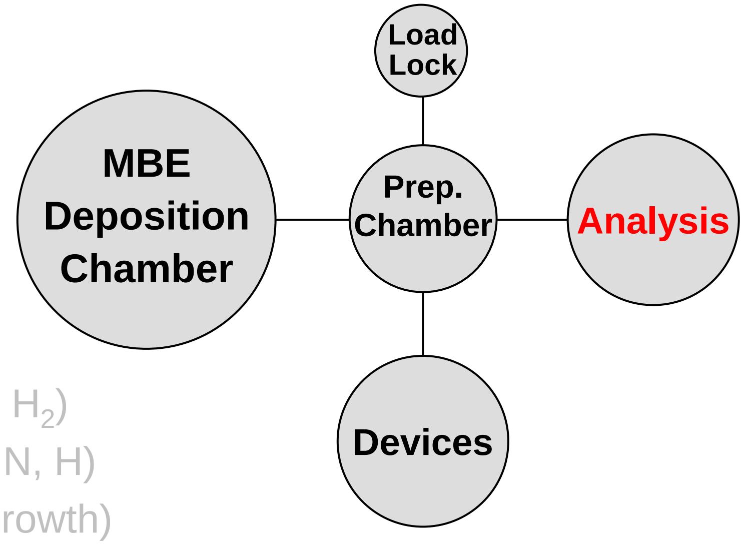

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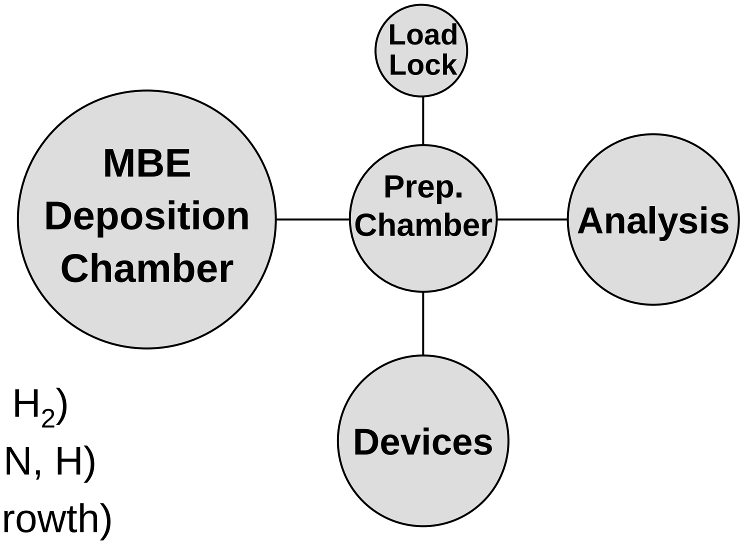

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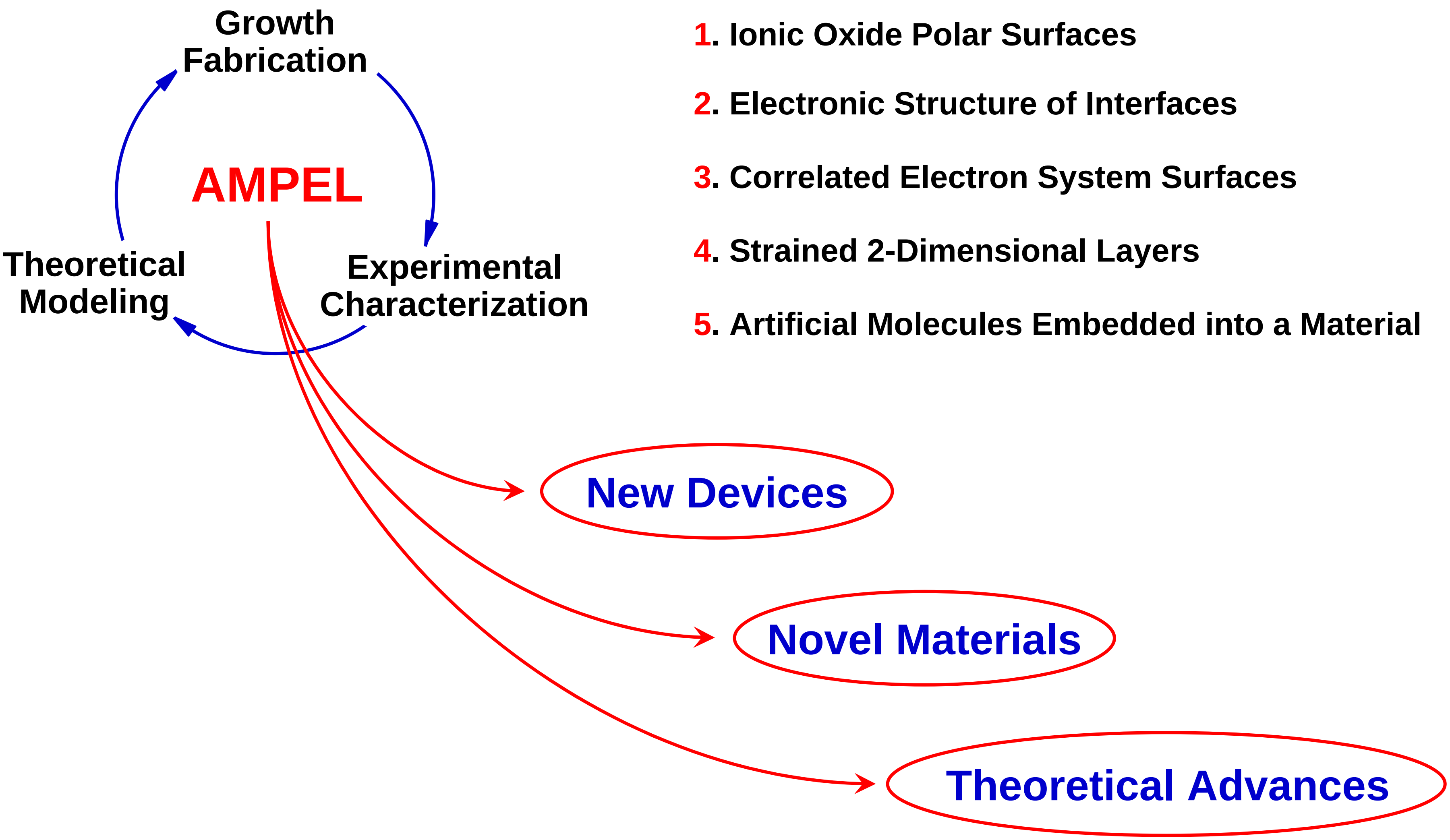
Novel Nanoscale Phenomena in TM Oxides

World-class Material Science program in Canada

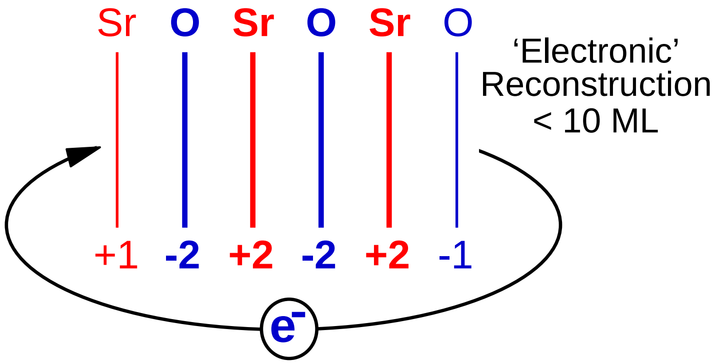


1. Ionic Oxide Polar Surfaces
2. Electronic Structure of Interfaces
3. Correlated Electron System Surfaces
4. Strained 2-Dimensional Layers
5. Artificial Molecules Embedded into a Material

Novel Nanoscale Phenomena in Transition-Metal Oxides

Ionic Oxide Polar Surfaces

Stabilization of polar surfaces by epitaxy



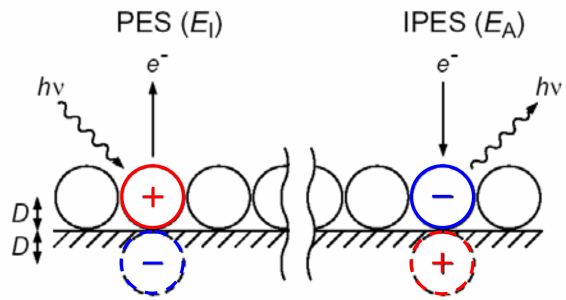
‘Electronic’ Reconstruction < 10 ML

Transparent insulator → ½ metallic FM

Applications: Spintronics; CMR

Electronic Structure of Interfaces

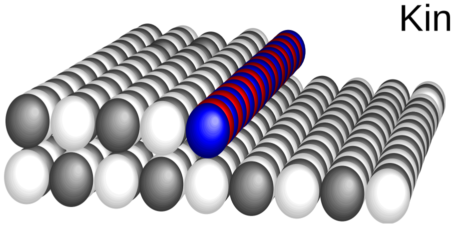
Metal-Insulator interface: gap suppression



Applications: Molecular Electronics; Fuel Cells; Thermal Barrier Coatings

Ionic Correlated Electron System Surfaces

Tuning the gap via the Madelung potential in ionic insulators



Kinks and steps stabilized by epitaxy

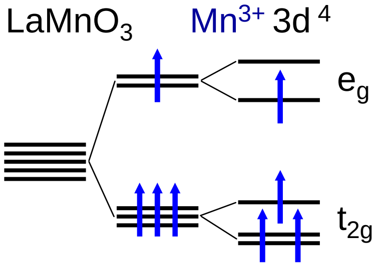
NiO (100) → 1D Metallic steps

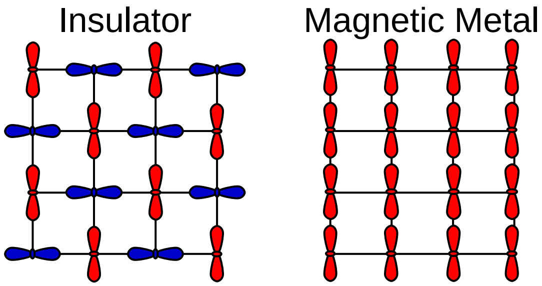
Superconducting Copper oxides

Applications: Novel SC; QuBits

Strained 2D Layers

Positive and negative pressure

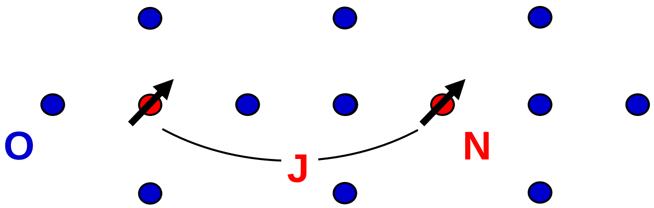




Applications: CMR; M-I Transition; Orbital Ordering

Artificial Molecules Embedded into a Material

Ca, Mg, Sr, Ni vacancies or O-N substitution in oxides

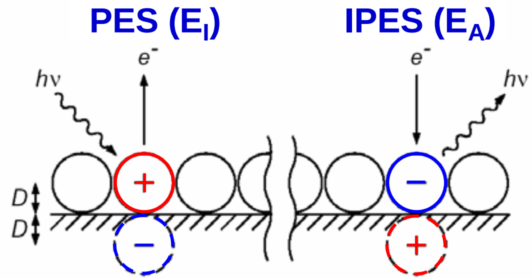


New class of magnetic materials by ‘‘low-T’’ MBE growth

Applications: Spintronics; Novel Magnets

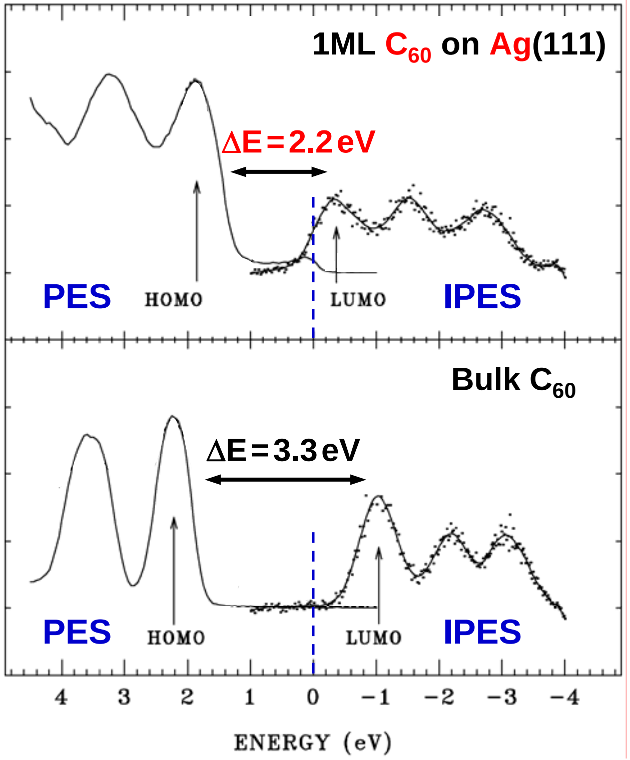
Electronic Structure of Interfaces

1 Monolayer on metallic substrate



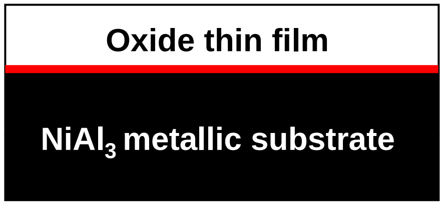
Conventional Semicond.: Band bending

Molecular Insulator: **Gap collapse**



Thermal Barrier Coatings

NiO or Al_2O_3 film on $NiAl_3$

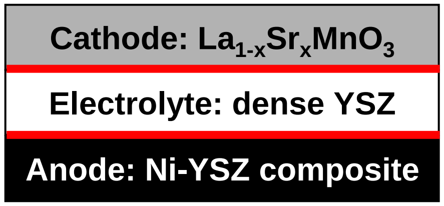


Oxide thin film

$NiAl_3$ metallic substrate

Fuel Cells

$La_{1-x}Sr_xMnO_3$ / YSZ



Cathode: $La_{1-x}Sr_xMnO_3$

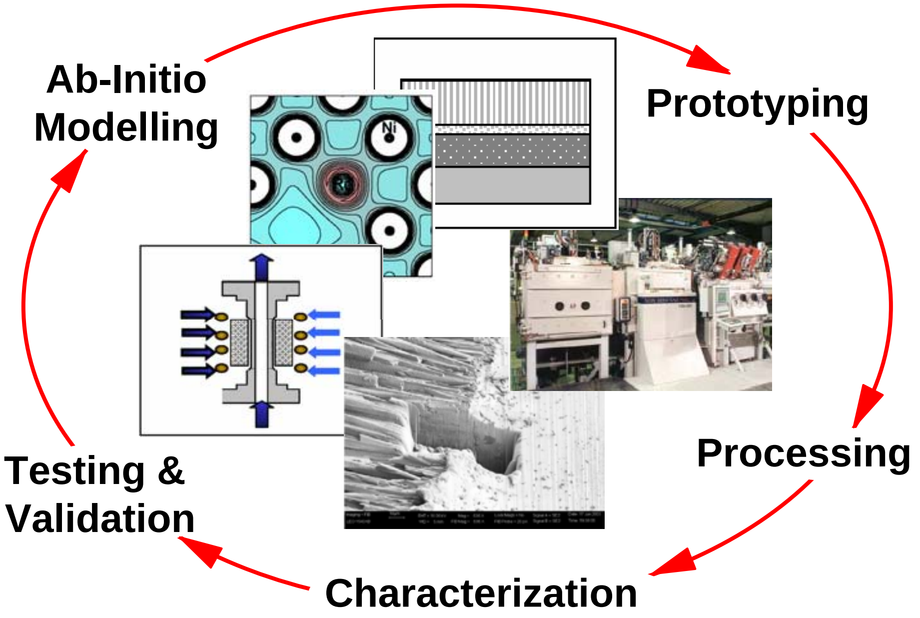
Electrolyte: dense YSZ

Anode: Ni-YSZ composite

What happens at interfaces?

Coatings → **Interface influences coating adhesion**

Fuel Cells → **Change in transport across interface**



Ab-Initio Modelling

Prototyping

Processing

Characterization

Testing & Validation