

Perspective article

Perspectives of cross-sectional scanning tunneling microscopy and spectroscopy for complex oxide physics

Aaron Wang, TeYu Chien*

Department of Physics and Astronomy, University of Wyoming, Laramie, WY 82071, USA

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ABSTRACT

Complex oxide heterostructure interfaces have shown novel physical phenomena which do not exist in bulk materials. These heterostructures can be used in the potential applications in the next generation devices and served as the playgrounds for the fundamental physics research. The direct measurements of the interfaces with excellent spatial resolution and physical property information is rather difficult to achieve with the existing tools. Recently developed cross-sectional scanning tunneling microscopy and spectroscopy (XSTM/S) for complex oxide interfaces have proven to be capable of providing local electronic density of states (LDOS) information at the interface with spatial resolution down to nanometer scale. In this perspective, we will briefly introduce the basic idea and some recent achievements in using XSTM/S to study complex oxide interfaces. We will also discuss the future of this technique and the field of the interfacial physics.

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1. Introduction

Complex oxide materials are known for its rich physical properties and achieving control over these physical properties has been the focus in recent years. For examples, efforts have been put to alter the magnetic properties in SrRuO₃ [1–3], and the electronic properties in SrTiO₃ [4–8]. Also, the complex oxide heterostructures are the other focus since the interfaces of dissimilar materials lead to new states that do not exist in the bulk counterparts [9–12]. These controllable physical properties and new two-dimension phases at interfaces have possible applications in the next generation electronic devices.

For researchers, two important aspects need to be achieved to fully investigate the complex oxide interfaces. First aspect is the synthesis method to produce high-quality samples. Since the oxides are involved in the fabrication process, unlike traditional semiconductor heterostructures, the control over the partial pressure of oxygen during synthesis is crucial. Nowadays, molecular beam epitaxy (MBE) with ozone as the oxygen source and pulsed laser deposition (PLD) are the two most powerful tools to synthesize desired quality complex oxides and complex oxide heterostructures. The other aspect is the characterization methods. The probing tools generally either have great spatial resolution but lack of capability

of probing physical properties; or capable of measuring physical properties but with poor spatial resolution. For example, high resolution transmission electron microscopy (HRTEM) [1,13–17] can have spatial resolution down to atomic scale, however, cannot provide much information on physical property at each location. On the other hand, X-ray magnetic circular dichroism (XMCD) [18], and X-ray linear dichroism (XLD) [19] are capable of revealing physical properties averaging over a macroscopic volume of the materials, *i.e.* poor spatial resolution.

To gain further insights on complex oxide interfaces, one would need a tool with excellent spatial resolution while capable of probing certain physical properties. Owing to its great spatial resolution and capability of measuring variety of physical properties, scanning probe microscopy (SPM) is then the ideal category of technique for this purpose [7,20,21]. While there are plenty of variety of SPM, here we will focus on the scanning tunneling microscopy and spectroscopy (STM/S) technique, which is a unique tool with spatial resolution down to atomic scale and capable of extracting electronic local density of states (LDOS) information.

In 1982, STM was invented by Gerd Binnig and Heinrich Rohrer and was awarded the Nobel Prize in 1986 [22]. Over decades, STM has been developed and more powerful versions have been developed and used to study materials in unprecedented ways. Among them, cross-sectional scanning tunneling microscopy and spectroscopy (XSTM/S) is an expansion technique of STM developed to study the interfaces of heterojunctions. In recent years, it has been applied to complex oxide interfaces to reveal the novel

* Corresponding author.

E-mail address: tchien@uwyo.edu (T.Y. Chien).

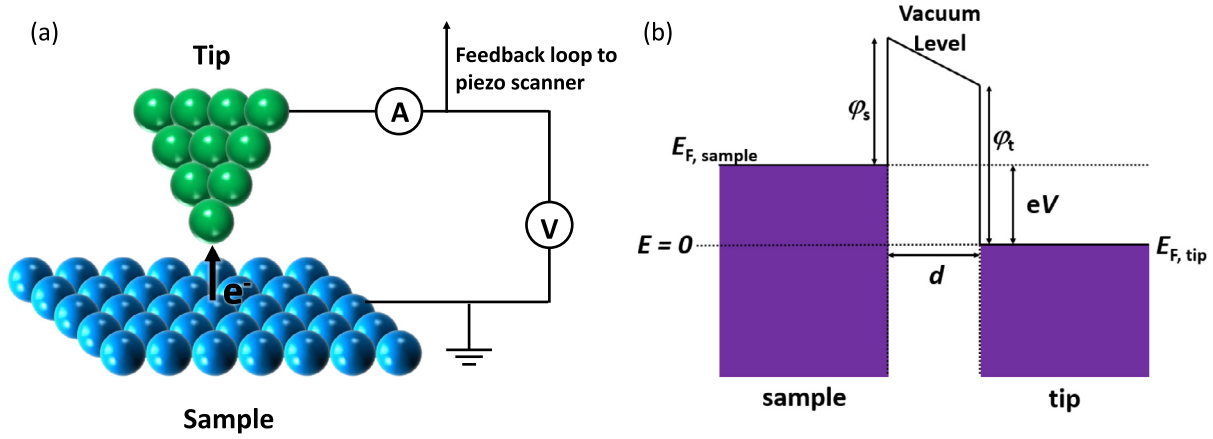


Fig. 1. (a) Schematics shows STM setup. (b) Electronic DOS of tip and sample upon electric bias application for STM measurements.

interfacial properties found at the interfaces. Specifically, XSTM/S is achieved by fracturing materials to create contamination-free, cross-section surfaces of the interface regions for STM measurements. In this perspective, we will be focusing on discussing the XSTM on studying the complex oxide materials, especially the complex oxide interfaces. In particular, we will discuss how the XSTM could provide novel insights on the interfacial physics and the future of this field.

2. Complex oxides and complex oxide interfaces

Complex oxide materials have drawn plenty of attention in recent decades due to their intriguing and versatile physical properties [23–25]. Unlike semiconducting materials, in which weakly interacting electrons dominating the physical properties, complex oxides exhibited a highly coupled environment where charge, spin, lattice and orbital are heavily mingled. The use of the semiconductor physics in the highly coupled environment in complex oxides is then questionable. The complex interacting nature of the complex oxide physics is also responsible for the various functionality not found in conventional semiconductors. For examples, ferromagnetism [16,17,26,27], antiferromagnetism [28–31], electrical conductivity [15,32], superconductivity [28,33,34], and multiferroics [35], have all been reported in various types of oxides with similar crystal structures and chemical formula. More intriguing, these functionalities have been proven to be easily altered with external stimuli, such as electric field [36], temperature [37], magnetic field [38], and strain [3]. Thus, these intriguing functionalities make the complex oxides not only possible for providing an ideal playground for studying the underlying many-body physics but also for the development of the next generation devices.

Similar to the semiconductor interfaces, the emerging properties by combining two or more complex oxides provide a vast playground on studying the bizarre and yet novel physics at the complex oxide interfaces, which may become the bases of novel devices in the future. For examples, two-dimensional electron gas (2DEG) at the interfaces of two band insulators, $\text{LaAlO}_3/\text{SrTiO}_3$, has drawn plenty of attention due to the fascinating physics and possible application in electronic devices [39]. Novel magnetic states could also be created at the complex oxide interfaces. In particular, the ferromagnetism emerged at the interfaces of paramagnetic metal CaRuO_3 and antiferromagnetic insulator CaMnO_3 [40] may provide a brand new view in understanding the magnetic interactions as well as possible novel magnetic devices. Proximity effects between oxide based superconductors and ferromagnets, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, is another interesting topic regarding the competing superconducting and ferromagnetic phases [41, 42].

3. Cross-sectional scanning tunneling microscopy and spectroscopy (XSTM)

Scanning tunneling microscopy and spectroscopy (STM/S) utilizes quantum tunneling effects across the tip–sample junction to perform topographical imaging and related measurements. Fig. 1(a) shows a typical STM setup. An atomically sharp tip is brought near the sample surfaces with tip–sample distance ~ 1 nm or less. While applying voltage across the tip–sample junction, electrons will tunnel through the vacuum energy barrier and form tunneling current. The tunneling current is used in a feedback loop to maintain constant current condition during scanning by adjusting tip height. The tip height is recorded as the topography information. Based on the quantum tunneling effect, the tunneling current as function of tip–sample bias and location of the tunneling, $I(V, \vec{r})$, across the tip–sample junction is expressed as:

$$I(V, \vec{r}) = \int_{-\infty}^{\infty} N_s(E - eV, \vec{r}) f_s(E - eV) |M|^2 N_t(E) \times [1 - f_t(E)] dE \quad (1)$$

where $N_s(E, \vec{r})$ is the sample electronic LDOS at location \vec{r} at energy E relative to the sample Fermi level; $N_t(E)$ is the tip electronic DOS at tip apex at energy E relative to the tip Fermi level; $f_{s/t}(E)$ is the Fermi Dirac distribution function for sample and tip, respectively; and $|M|^2$ is the tunneling matrix element. Fig. 1(b) shows the DOS of tip and sample as well as electric bias relationship under STM operational conditions. By applying WKB approximation, one would find the following relationship between the tunneling matrix and the tip–junction distance, d , as:

$$|M|^2 \propto e^{-\kappa d} \quad (2)$$

where κ is a constant related to electron energy and applied bias. The exponential dependence of the tip–sample distance of the tunneling matrix is the source of the excellent spatial resolution of STM.

Furthermore, for constant temperature measurements and with constant tunneling matrix assumption, Eq. (1) could be expressed as:

$$I(V, \vec{r}) \propto \int_{-\infty}^{\infty} N_s(E - eV, \vec{r}) N_t(E) dE \quad (3)$$

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