Characterization and Manipulation of Complex Oxide Architectures via Synchrotron Radiation

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Abstract

Complex oxides are gifted systems that have attracted significant attention over past decades. These materials provide a wide spectrum of intriguing physical properties, including superconducting, piezo/ferroelectric, magnetic, dielectric, ferromagnetism, colossal magnetoresistance, metal-insulator transition and so on…. Great efforts have been made to reveal the origin and coupling of those fascinating properties. Using modern epitaxy methods, controlling the interplays between charge, lattice, orbital and spin degrees of freedom in complex oxides allows us to design materials or architectures with new properties. Along this vein, fundamental understanding of the physical origin of the intriguing phenomena is crucial in order to provide advanced basis for materials design and the development of next-generation nanodevices. In this talk, we will present some of our previous achievements upon optical control and advanced growth of complex oxides, which have been greatly facilitated by synchrotron radiation-based techniques.

As the first part of this talk, we will present non-volatile and ultrafast optical modulation of multiferroics. With the application of short laser pulses, non-volatile switch between different phases and correlated ferroic orders have been achieved on nanosecond and femtosecond time frames. Moreover, the configuration of optically written ferroelectric domains can be further tuned by symmetry breaking of charge distribution, taking advantages of competing elastic and electrostatic energies. In the second part talk, using multiferroic $BiFeO_3$ as a model system, we will present an efficient approach to fabricate twisted lateral homostructures with various conjunction tunability, including crystalline orientation, epitaxial constrain and phase stability. ties. Our results evidence the excellent controllability and unbounded conjunction tunability of the lateral homostructures using the proposed method, allowing epitaxial films to be assembled at particular position in the plane, as if they were artificially “weaved”. In the end, we will talk about our perspective regarding the combination of novel oxide nanoarchitectures and synchrotron-based technologies. We hope that our contribution can not only provide a better understanding of complex material architectures, but also delineate an different scenario for epitaxial growth.

首次觀測到缺陷保護之稀磁性氧化物中的雜質能帶: 藉由極化硬 X 光光電子能譜儀實現

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摘要

稀磁半導體 (DMS) 同時具有半導體和鐵磁特性，在自旋電子應用中具有巨大潛力。 III-V DMS 中的鐵磁性由載流子介導機制控制已得到許多實驗證實，但 III-V DMS 中的鐵磁居里溫度遠低於室溫，很大地限制了其實際應用價值。另一方面，儘管有非常多研究報導了各種過渡金屬 (TM) 摻雜氧化物，即所謂的稀釋磁性氧化物 (DMO)，其居里溫度 (Tc) 可高於室溫，但 DMO 之鐵磁性的起源至今仍不清楚。事實上，即使已經數千篇相關論文發表，這個問題仍然是一個很大的爭論難題。

要進一步闡明 DMO 的鐵磁性成因的困難來自兩個方面: (1)如何保存 DMO 樣品中的缺陷。DMO 中的鐵磁性與結構缺陷(如鋅間隙或氧空位等)密切相關，然而結構缺陷在大氣環境條件下很容易回補，尤其是在樣品表面附近，因此 DMOs 的磁性通常是不穩定的。 (2)另一個關鍵因素是找到對分析工具。

在本工作中，我們設計了一種保留缺陷的方法，以驗證 DMO 中摻雜濃度、缺陷密度和鐵磁性的相關性。我們專注於 Co 摻雜的 ZnO，這是實驗研究最多的 DMO 材料。 我們使用氫化來控制 Co 摻雜 ZnO 中的缺陷數量並覆蓋 ZnO 保護層以保護 Co：ZnO 中的缺陷，以下稱為 PROT-Co：ZnO。 我們利用日本春八同步輻射加速器之硬 X 光光電能譜 (HArd X-ray PhotoEmission Spectroscopy, HAXPES) 技術，一種對塊體(bulk)和電子軌道敏感的分析工具，我們可以檢測費米能級附近的價帶的電子軌道態密度 (DOS)。我們發現雜質能帶存在於 PROT-Co:ZnO 樣品中，此雜質能帶隨 Co 含量和缺陷密度的減少而減少，而雜質能帶是通過 Co 和結構缺陷的交換相互作用而形成。

我們突破以往相關研究中樣品製備和測量技術的局限，本研究結果非常有助於重新審視和闡明 DMO 中室溫鐵磁性的機制。

Quantum Materials in the Spotlight of Momentum Microscopy: From Symmetry to Topology

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Abstract

Quantum materials are considered a key resource for the 21st century, promising a wealth of novel phenomena with respect to spin transport, superconductivity and magnetism. The macroscopic electronic properties of quantum materials are generally determined by specific quantum effects, which can be caused by broken symmetries, topology or chirality. The microscopic driving forces for quantum phenomena are the competition of various spin-dependent interactions, such as spin-orbit coupling and exchange interaction. These fundamentally spin-dependent interactions lead not only to the peculiar dispersion of the electronic states associated with Dirac and Weyl points, but also to complex spin textures in momentum space [1].

In order to understand the physical properties of quantum materials on a fundamental level, we need to explore these electronic states in detail and disentangle the role of the various interactions. Only recently, the comprehensive experimental access to the spin-resolved band structure at every point in the Brillouin zone became feasible by spin-resolved momentum microscopy [2]. This novel concept combines high resolution imaging of the spectral function in two-dimensional (k_x, k_y) planar sections through the valence electronic structure with an imaging spin filter [3].

With this comprehensive spin-resolved information of the electronic states we discuss the role of the individual interactions and symmetry-breaking mechanisms based on the prototypical Dirac and Weyl semimetals $NiTe_2$ and $MoTe_2$, differing in their crystal structure in terms of non-broken/broken inversion symmetry. Detailed Fermi surface maps of the spin-texture and circular dichroism of the topological states obtained by momentum microscopy provide evidence for a link between the spin-orbital texture and the intrinsic chirality of the wave functions that form a topological state [4].

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Electronic Structures of Topological materials

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Abstract

Understanding the fundamental physics in different phases of matter is one of the most important goals of condensed matter physics. Traditionally crystalline solid systems can be classified as insulators and metals according to energy band theory. In 2005, scientist discovered that the Bloch state wavefunction can be mapped to a nontrivial topological structure, leading to an entirely new material phase, called topological phase. The topological material is featured by a bulk energy gap originating from spin-orbit coupling and time-reversal symmetry protected gapless surface states, which is distinct from the conventional band insulator. Because of this property, Angle resolved photoemission spectroscopy (ARPES) has become the most powerful experimental tool for studying topological materials. In this talk, I will introduce our recent works on topological semimetal [1], magnetic topological insulator [2-4], and topological heterostructures [5-7] based on first-principles calculations and ARPES.

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Probing the electronic and chemical structures of 2D materials by soft X-ray spectroscopy techniques

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Due to the ultra-thin nature of monolayer two-dimensional (2D) material systems, it is difficult to study their intrinsic electronic and chemical properties by conventional spectroscopic means. To overcome such barrier, the surface sensitive synchrotron radiation (SR) based soft X-ray spectroscopy techniques are most suitable to study the 2D material systems. In this talk, I will present some examples to show the advantages of those techniques in studying 2D materials, to show the versatileness of soft X-ray SR techniques.

Resonant Tunneling and Negative Differential Resistance in 2D materials Transistors

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Abstract:

During the last decade, tremendous research efforts have been focused on two-dimensional (2D) materials due to their rich physics and great potentials for many applications. In this talk, I will share with the results relevant to resonant tunneling and negative differential resistance (NDR), which has been extensively studied for various electronics applications. In both stacking transition metal dichalcogenides (TMD) [1] and lateral growth of two kind of TMD material [2], resonant tunneling behavior is observed through discrete quantum state. Based on tunneling mechanism, high frequency operation (64GHz) is presented in graphene base hot electron transistors [3]. In another hand, according to the theoretical simulation, the NDR is predicted to be affected by defects in monolayer 2D materials. We have experimentally verified the theoretical prediction in monolayer MoS_2 and clay graphite with simple fabrication processes controlled the appropriate amount of defect [4-5]. We believe that the resonant tunneling and NDR effect in 2D transistors may pave a way to develop more electronic applications in the future.

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The modeling quantum many-body calculation to analyze Soft X-ray absorption spectroscopy

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Abstract

The soft X-ray absorption spectroscopy (XAS) at L-edge is a powerful tool to study in 3d transition metal-oxide the correlation between spin, charge, and orbital degrees of freedom. However, the atomic multiplet effects arising from the interaction between the core hole and the valance elections complicate the information delivered from XAS. It thus requires aid from the modeling calculations to look insight into the XAS. In this talk, I will introduce a new computational code developed in our group that can analyze the XAS. In this code, the full atomic-multiplet, local solid-state effects, and spin-orbital coupling are employed to solve the quantum many-body ground state wavefunction. And Green's function is employed to **propagate** the ground state to the final state to generate the XAS. By fitting with the experimental XAS, interesting quantum information like spin, charge, orbital, and the correlation between them can be obtained.

Probing 2D Materials with Photoelectron Momentum Microscopy at TPS 27A2

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Two-dimensional (2D) materials have been intensively studied recent years due to its unique properties and its potential applications, however, the lateral dimension of a single flake of 2D van der Waals materials is roughly in a size of a few to few tens of microns. This implies that the probing tools are required to be position-sensitive.

The photoelectron related image and nano-spectroscopy (PRINS) end-station at the Taiwan Photon Source (TPS) 27A2 is now commissioning a momentum microscope [1] that aims to work on photoelectron-related imaging and nano-scale spectroscopy through the combination of an imaging-type electron column integrated with a hemispherical electron energy analyzer and an imaging spin filter. The microscope is able to conduct full-field imaging by collecting photoelectrons in either real-space or momentum-space with spatial, energy, and spin resolution. The main microscope system was delivered at the TPS at the end of 2021, and its on-site testing has been initiated in the first half of 2022. Although the soft X-ray beamline will not be ready until early 2023, the off-line commissioning by He-I radiation and a Hg lamp has delivered preliminary commissioning results in real-space, momentum-space, and spin-resolved imaging.

In this report, I will briefly introduce the performance of this momentum microscope and its great potential on probing the electronic structures of 2D materials by taking advantage of its power of combining real- and momentum-space imaging. A couple of selected commissioning results performed on an Au(111) single crystal and single-layer/multilayer transition metal dichalcogenides (TMDs) will be demonstrated as examples to shed light on what/how NSRRC can help in 2D material research.

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