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Cryogenic Electron Microscopy on Strongly Correlated Quantum Materials

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Quantum materials refers to a class of materials with exotic properties that arise from the quantum mechanical nature of their constituent electrons, exhibiting, for example, high-temperature superconductivity, colossal magnetoresistivity, multiferroicity, and topological characteristics. Quantum materials often have incompletely filled d- or f-electron shells with narrow energy bands and the behavior of their electrons is strongly correlated. One distinct characteristic of the materials is that their electronic states are often spatially inhomogeneous, thus is well suited for study using spatially resolved electron beams with its great scattering power and sensitivity to atomic ionicity. Furthermore, most of these exotic properties only manifest at very low temperatures, posing a challenge to modern electron microscopy. It requires extraordinary instrument stabilities at cryogenic temperatures with critical spatial, temporal, and energy resolutions in both static and dynamic manner to probe these materials. On the other hand, the ability to directly visualize the atomic, electronic and spin structures and inhomogeneities of quantum materials and correlate them to their functionalities creates enormous opportunities. At the most elementary levels of condensed matter physics, understanding the competing orders of electron, spin, orbital, and lattice and their degrees of freedom, the impacts of defects and interfaces, and the site-specific quantum phenomena and phase transitions that give rise to the emergent behaviors allows us to discover and control novel materials for quantum information science and technologies [1].

In this presentation, several of our research examples are selected to highlight the use of Cryo-EM to study strongly correlated quantum materials. We focus on the critical roles of heterogeneity, interfaces, and disorder in crystal structure, electronic structure, and spin structure to understand the physical properties of the materials. We show how electron diffraction and diffuse scattering analysis at low temperatures empowers us to reveal the nature of structural disorder and phonons under thermal equilibrium and far-from equilibrium [1]; how atomically resolved imaging and electron energy-loss spectroscopy at 10K can be used to understand the interface-enhanced superconductivity (Fig.1); and how to use Lorentz phase microscopy to explore the intriguing transformations among various topological chiral spin states under applied magnetic field at various cryogenic temperatures (Fig.2). Finally, we review our recent unprecedented development of a closed-cycle cryocooler for Cryo-EM without the need of refilling liquid He. The system is designed to reach a temperature as low as 4k with temperature stability and control better than a few mK [2].

References:

[1] Zhu, Y., "Cryogenic Electron Microscopy on Strongly Correlated Quantum Materials", Invited review article, special issue on Cryogenic Electron Microscopy. Guest editors: Y. Cui and L. Kourkoutis, Accounts of Chemical Research, 54, 3518−3528 (2021).
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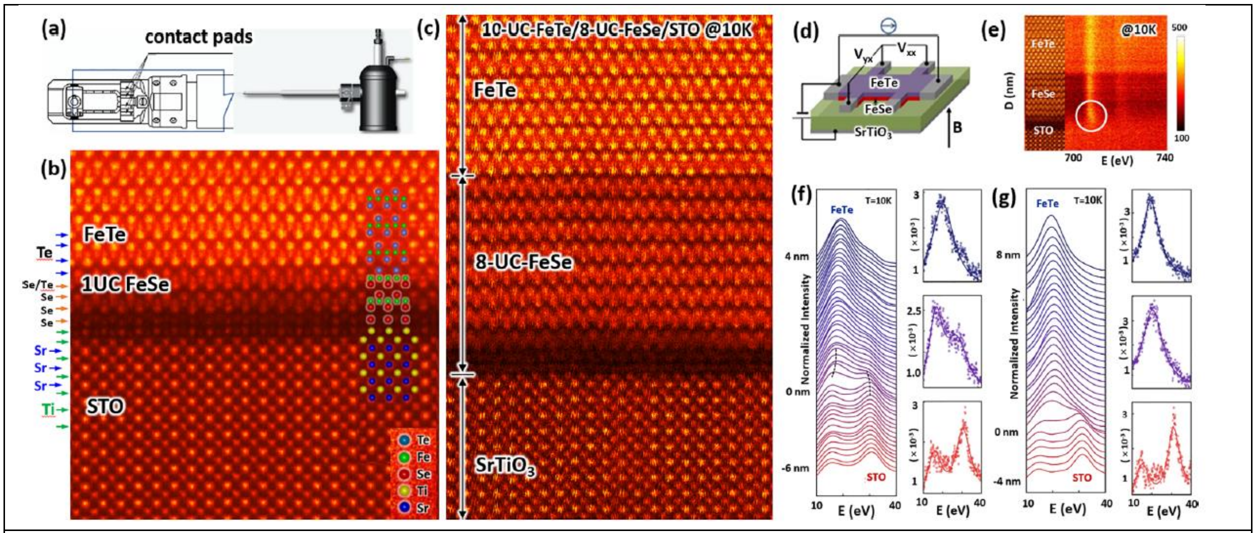

Figure 1. First atomically resolved imaging and EELS at 10 K in FeSe/$SrTiO_3$. (a) Custom-made liquid-He biasing-holder by Gatan-UK for Brookhaven in 2014. (b, c) STEM images showing the cross-section heterostructure of the 10-unit-cell (UC) capping layer FeTe on 1UC- (b) and 8UC- (c) FeSe superconductor grown on $SrTiO_3$. The interfacial structure with an additional Se layer is identified. (d) Schematic of the same sample as a gate tuned Hall-bar device for transport measurement to understand interface enhanced superconductivity. (e) Fe L3 EELS map of the film showing a blue-shift within 2UC (marked by the circle) from the FeSe/STO interface. (f, g) Valence EELS across the interfaces from FeTe/1UC-FeSe/STO (f) and FeTe/ 8UC-FeSe/STO (g). Black dashed lines in panel f indicate a red-shift in plasmon peaks near the interface (for details see [1]).

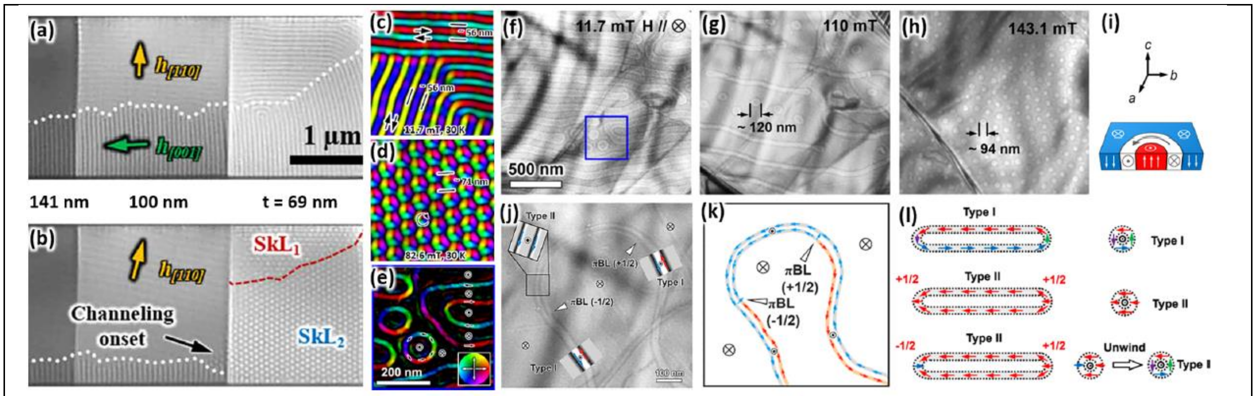

Figure 2. Field-temperature dependence of topological chiral spin textures. (a−d) Lorentz images of helical-to-skyrmion phase evolution at 25 K in Te-doped Cu_2OSeO_3 for sample thickness t = 69, 100, and 141 nm and applied magnetic field of 11.7 mT (a) and 62 mT (b), showing anisotropic scaling of two orthogonal helical phases and edge-induced skyrmion nucleation and channeling. (c, d) Induction maps of helical spin states (c) and hexagonally packed skyrmion lattice (d) reconstructed from t = 69 nm area in panels a and b, respectively. (e−l) Chiral spin stripe−skyrmionic bubble transition induced by magnetic field cooling (H//c-axis) (f−h) in van der Waals $Cr_2Ge_2Te_6$ at 20 K. (e) Induction map of stripes and skyrmion bubbles from the boxed area in panel f. (j−l) Bloch-line (BL) pairs and their unwinding under external fields showing type-I and type-II stripe domains bounded by a pair of π BLs and the stripe−bubble transformation process (for details see [1]).