
Hoffman Laboratory Research Overview

Research in the Hoffman laboratory is motivated by the conviction that technological solutions are driven by new materials, and that the materials frontier lies in the design of emergent properties at interfaces and small length scales. Our research combines atomic layer-by-layer growth and atomic resolution imaging of materials, to uncover new physics and applications inaccessible via bulk synthesis and probes. We have used scanning tunneling microscopy (STM) and magnetic force microscopy (MFM) to investigate fundamental mechanisms of electron pairing and technical challenges of vortex pinning in high- T_c superconductors. We have used conducting atomic force microscopy (CAFM) to control the metal-insulator transition in VO_2 at the nanoscale. We have used STM to map the band structure of the topological materials Sb and SmB_6 . We have incorporated a molecular beam epitaxy (MBE) system to create novel heterostructures and tailor their properties using atomic resolution feedback from *in situ* STM. Here we describe in more detail some of our technological capabilities, past research successes, and future directions.

Microscopy Techniques

The Hoffman lab has built 3 scanning probe microscopes, with custom design and software to enable new research directions. We highlight below a few new capabilities we have developed.

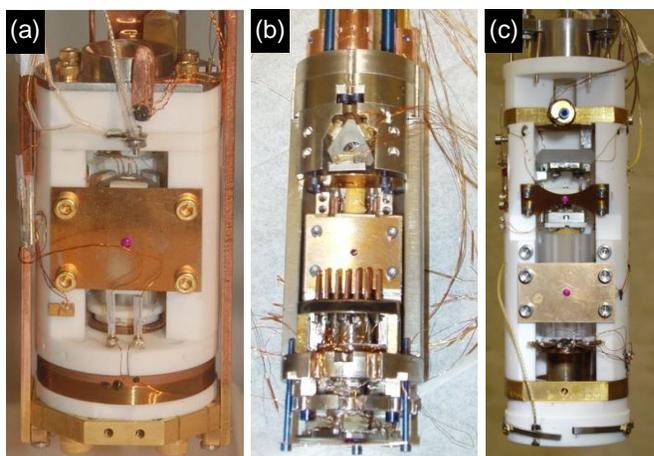


Fig. 1: Three home-built microscopes. **(a)** STM, optimized to study cleavable materials. Operates between 2-50K, in a 3-axis magnetic field (9T vertical, 3T in-plane). The same atomic field of view can be followed throughout the temperature and field range. **(b)** Versatile force microscope, for contact and non-contact imaging and nanoscale phase manipulation, with insulating, conducting, or magnetic tips. Operates in 5T field, and from 2-350K. **(c)** UHV STM, for the *in situ* study of MBE-grown films. MBE is currently equipped with Fe, Se, Si, Pb, Bi, Co, Cu, Ni, and Ag sources. STM operates in a 2-axis magnetic field (9T/3T), and from 2-300K.

Algorithm for picoscale imaging

To search for subtle signatures of intra-unit-cell symmetry-breaking electronic order in cuprate superconductors, we implemented a new algorithm for picoscale spatial resolution STM. Our algorithm implements Lawler's drift correction [1], then efficiently creates an ultra-high-resolution average unit cell or 'supercell' by overlaying, with picoscale precision, portions of an STM topography separated by hundreds of Angstroms, encompassing more than 10,000 unit cells [2]. The new algorithm allowed us to image a 5 picometer inversion-symmetry-breaking structural distortion throughout the normal and superconducting phases of bismuth-based cuprates. More broadly, this algorithm can serve to visualize picoscale intra-unit-cell electronic structure in any crystalline material.

Capacitive method for cantilever Q control

We introduced a simple and versatile method to control the quality factor Q of a conducting cantilever in an atomic force microscope (AFM) via capacitive coupling to the local environment [3]. Using this method, Q may be reversibly tuned to within 10% of any desired value over several orders of magnitude. Such control increases the AFM functionality by allowing greater control of parameters such as scan speed and force gradient sensitivity. We have filed for a provisional patent on this method.

Method for nanoscale thermometry

The diverse temperature-dependent electronic properties of thin film oxides position them at the forefront of materials research, but there are few methods for thermal imaging with the spatial resolution necessary to fully understand these properties. We introduced a new method for nanoscale thermal imaging of insulating thin films. By scanning a conducting atomic force microscope tip in contact with a VO_2 film, and sweeping the tip voltage at each location, we locally triggered and detected an abrupt insulator-metal transition. By fitting the Poole-Frenkel conduction regime immediately preceding the transition, we calculated the local temperature at transition [4]. This method for thermal imaging could be fruitfully applied to a wide variety of insulating films.

Spin-polarized imaging of cleavable materials

We optimized an *ex situ* etching procedure to sharpen bulk Cr tips for spin-polarized STM. We then evaluated *in situ* the spin-polarized tunneling characteristics of these Cr tips by imaging the layered antiferromagnetism of nanoscale terraces on $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ at 6.5 K [5]. We thus demonstrated a simple method for spin-polarized tip preparation and evaluation using cleaved crystal planes, circumventing the need for ultra-high vacuum preparation of clean surfaces or films.

Superconductivity

- Goal: raise T_c in cuprates by better understanding the pseudogap & its effect on the Fermi surface
- Goal: enable high-current applications by improving vortex pinning
- Moonshot: discover room temperature superconductivity based on interface effects

Superconductivity – the lossless flow of electrical current – promises transformative technologies including efficient energy usage, fast transportation, accessible communication bandwidth, and novel medical imaging – if only it could be harnessed within practical temperature and magnetic field regimes. Today’s highest- T_c superconductors, the cuprates, host a mysterious ‘pseudogap’ phase – manifest as a depression in the density of states near the Fermi level. The pseudogap is controversially believed to strengthen superconducting pairing but weaken its phase stiffness, or to compete with superconductivity altogether. We have used STM to image the effects of single atom defects [6], picoscale structural distortions [2], and charge density wave (CDW) order [7] on the pseudogap phase in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$. Because superconductivity arises from pairing of electrons on the Fermi surface, we are now focusing especially on the cuprate Fermi surface. We recently used field-dependent quasiparticle interference imaging to discover a quantum phase transition in Fermi surface topology, and to prove *d*-wave pairing on the recovered antinodal segments of the Fermi surface [8].

To gain additional insight into the complex phase diagram of cuprates, we investigated NbSe_2 , a model system which also hosts superconductivity and a well-known CDW phase. We discovered a new 1D (stripe) CDW smoothly interfacing with the familiar 2D (triangular) CDW on the surface of NbSe_2 [9]. We used this strain-tuned quantum phase transition to resolve two longstanding debates about the anomalous spectroscopic gap and the role of Fermi surface nesting in the CDW phase of NbSe_2 , and also to highlight the importance of local strain and quantum phase transitions in correlated superconducting systems such as cuprates.

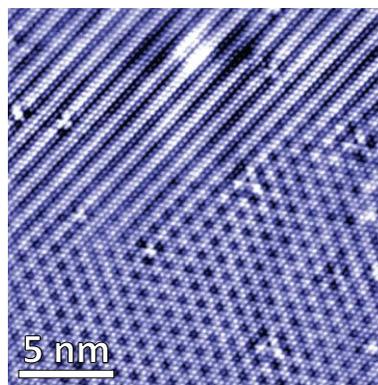


Fig. 2: Quantum phase transition from 2D to 1D CDW on NbSe_2 .

Meanwhile, the 2008 discovery of high- T_c superconductivity in Fe-based materials renewed the worldwide drive towards superconducting applications. Dissipative vortex motion and large electronic anisotropy have been limiting factors in cuprate-based technology. We used STM to provide the first atomic resolution images of impurities and vortices in an Fe-based superconductor, $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ (Fig. 3a) [10]. The lack of correlation between vortices and surface defects demonstrated strong native pinning, as well as low anisotropy, in striking contrast to cuprates [11]. Our follow-up study on $\text{K}_x\text{Sr}_{1-x}\text{Fe}_2\text{As}_2$ suggested that dopant size mismatch and consequent clustering may be responsible for the strong native pinning [12]. We performed a complementary study using an MFM tip to manipulate individual vortices in $\text{NdFeAsO}_{1-x}\text{F}_x$ (Figs. 3b-c), and quantify the intrinsic single-vortex pinning force of 8 picoNewtons [13]. This is an important step towards deliberate defect engineering to increase the critical current.

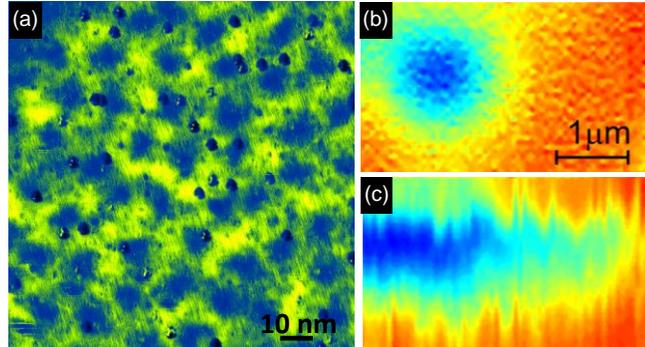


Fig. 3: (a) STM image shows non-correlation of vortices and surface defects in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. (b) MFM scan of a vortex in $\text{NdFeAsO}_{1-x}\text{F}_x$ at large tip-sample distance. (c) Vortex at small tip-sample distance, where $F_{\text{tip-vortex}} > F_{\text{pin}} = 8 \text{ pN}$.

Today, we are poised at a singularly exciting time in superconductivity research, with the recent discovery of T_c up to 110K in single-layer FeSe films on SrTiO_3 , compared to $T_c=8\text{K}$ in bulk FeSe [14-15]. This finding launches an effort to understand the mechanism of the extraordinary order-of-magnitude increase, and a race to discover higher- T_c interface superconductivity. Having just completed the assembly of a new molecular beam epitaxy (MBE) system with our existing low- T STM (Fig. 4), our group is perfectly positioned to be at the forefront of this endeavor, employing the following strategies. (1) We will use STM to characterize the local electronic effects of surface and near-surface oxygen interstitials

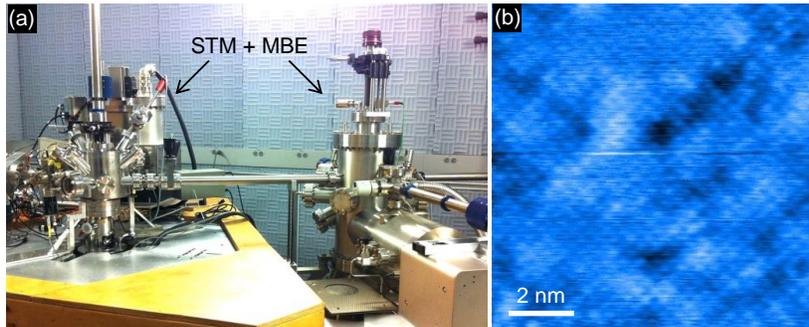


Fig. 4: (a) Photo of Hoffman lab MBE+STM system. (b) Single layer FeSe on SrTiO_3 , grown and imaged in the Hoffman lab.

and vacancies and picoscale strain in SrTiO_3 . (2) We will tune the FeSe carrier concentration by gating. (3) We will investigate epitaxial strain effects by growing FeSe on other substrates such as BaTiO_3 . (4) We will introduce Te dopants and other controlled atomic impurities into FeSe to probe the superconducting order parameter [16].

Our ultimate goal is to use the knowledge gained from FeSe to engineer higher- T_c materials and films. One target system is rare-earth-doped CaFe_2As_2 , which hosts a small-volume-fraction high- T_c phase of unknown origin. Our own STM images of $\text{Pr}_x\text{Ca}_{1-x}\text{Fe}_2\text{As}_2$ argue against rare earth dopant inhomogeneity [17], but other bulk measurements suggest indirectly that coordinated layers of As vacancies could double T_c [18]. This hypothesis could be well tested with controlled MBE growth and STM imaging. We will also return to cuprates, to investigate the T_c -boosting interface effects in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ [19]. We propose a novel hybrid MBE-organic growth path [20] for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ using $\text{Cu}(\text{C}_5\text{H}_7\text{O}_2)_2$ [21] which may circumvent the stringent requirement for perfect La/Sr:Cu flux ratio.

Vanadates

- Goal: understand the microscopic electronic mechanisms of phase transitions in VO₂
- Moonshot: find a stable parameter regime to separate electronic & structural transitions

VO₂ undergoes an insulator-to-metal transition as a function of temperature, strain, or electric field. Useful properties such as 80 fs switching time, resistivity ratio up to 10⁵, large change in optical reflectivity, and tunability near room temperature suggest wide-ranging applications including energy-saving window coatings [22] and low-dissipation electronics [23]. However, repeated cycling through the apparently concomitant structural transition degrades VO₂ crystals [24]. It is therefore crucial to understand the relationship between electronic and structural transitions of VO₂.

We demonstrated the first controlled nanoscale electronic switching of a VO₂ film, using a biased conducting AFM tip (Fig. 5a-b) [25]. Furthermore, our AFM can independently and simultaneously image both transitions of VO₂: the cantilever deflection measures the structural transition while the cantilever current measures the electronic transition. In polycrystalline VO₂ films, we found preliminary evidence that the electronic and structural transitions can be decoupled (Fig. 5c). It is known that an M1→M2 structural transition can precede the insulator-to-metal (M→R) transition [26], but to our surprise we have occasionally observed a structural transition *after* the insulator-to-metal transition. However, local variations in strain and grain orientation have hampered our efforts towards systematic understanding. We aim to use MBE-grown VO₂ films with controlled strain and orientation, to search for a controlled parameter regime to access the electronic transition without structural degradation of VO₂.

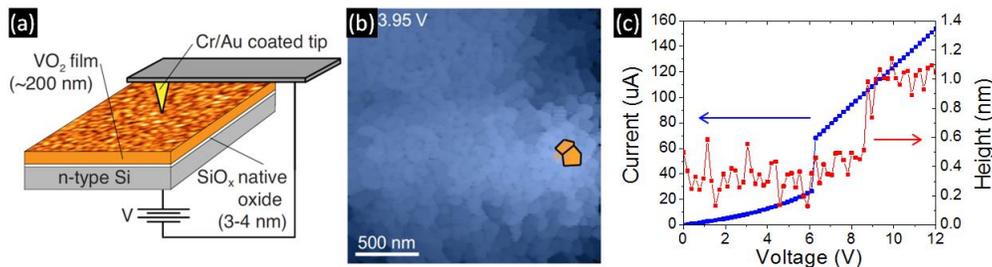


Fig. 5: (a) Schematic of the force microscope measurement geometry. (b) Conductance map showing the nucleation of a nanoscale metallic puddle upon local application of 3.95 V [25]. (c) Separation of electronic (blue) and structural (red) phase transitions in a single grain of VO₂, as the voltage is swept.

Topological Materials

- Goal: establish proof & understanding of the first topological Kondo insulator
- Goal: establish proof & understanding of the first topological p_x+ip_y superconductor
- Moonshot: manipulate a Majorana fermion with a magnetic force microscope

Topological materials present exciting opportunities for low-dissipation spintronics and fault-tolerant quantum computing. Major challenges include (1) mitigating defects which scatter spin-polarized surface states; (2) controlling bulk conduction which can swamp the surface states in transport devices; (3) realizing Majorana fermions for topological qubits.

On the first front, we have an ongoing effort to image and understand the effects of atomic defects in Bi- and Sb-based topological materials [27-28]. We found that surface states in the semimetal Sb are significantly longer-lived (as evidenced by the appearance of 27 sharp Landau levels in Fig. 6a) than in first-generation Bi₂X₃ topological “insulators”. Sb therefore serves as an optimal model system to understand protection against back-scattering, and to explore fundamental behaviors of magnetic and superconducting topological proximity effects. We fit the Landau levels [29] to provide the first measurement of the g -factor of topological surface states, $g=12$, which determines their immunity to

magnetic impurities and stray field. Furthermore, we aim to use spin-polarized scanning tunneling microscopy [5], to image the predicted RKKY spin coupling between magnetic impurities to determine the length scale over which these impurities impact the surface.

On the second front, we are investigating SmB_6 which was predicted to be a topological Kondo insulator (TKI) with clean insulating bulk [30], in comparison to first-generation Bi_2X_3 materials whose apparently unavoidable defects render their bulk conducting [31]. The Kondo hybridization gap Δ

would set the T scale for device operation, but bulk measurements of Δ have varied by over an order of magnitude in SmB_6 . We recently used STM to compare polar and non-polar cleaved surfaces and to quantify Δ locally in SmB_6 [32]. We are continuing to search for evidence of the topological surface state in SmB_6 and other proposed TIs including YbB_6 , BiTeCl , and BaBiO_3 .

On the third front, we will launch a new effort to create and control a Majorana fermion, by coupling a topological material to a superconductor using MBE. We will start by depositing Sb films on a variety of superconductors. We will use magnetic-field-dependent QPI imaging to determine the superconducting coherence factors [8] and distinguish between classes of superconductors. Once we have understood the signatures of topological superconductivity in this Sb model system, we will move to true topological *insulators*, and use STM to search for the quantized conductance expected to arise from Majorana bound states in vortex cores [33]. Finally, we aim to use MFM to manipulate these vortices (similar to Figs. 3b-c), and thereby braid their attached Majorana fermions. We look forward to additional theoretical interactions to envision a readout mechanism for this manipulation.

Atomic Scale 3D Printing

The long-term vision of the Hoffman laboratory is to establish “atomic 3D printing”, using a combination of MBE and scanning probe microscopy to create and explore novel quantum materials and heterostructures. Quantum materials exhibit diverse properties that depend not only on their internal chemical composition, but also on the details of their interfaces. Molecular beam epitaxy (MBE) has revolutionized the control of materials in the vertical (z) dimension, allowing the custom stacking of individual atomic layers to form interfaces not available in nature. We aim to take a key step forward by integrating scanning probe lithography (SPL) to assemble quantum heterostructures for the first time in all three dimensions, using sharp tips to manipulate individual atoms in the lateral (x,y) dimensions that MBE alone cannot control.

Our first concrete goal builds upon our successful growth of single-layer FeSe on SrTiO_3 (Fig. 4b), the remarkable system where superconducting $T_c > 100\text{K}$ has been recently reported, although the mechanism remains poorly understood. We aim to use STM to manipulate sparsely deposited adatoms as local “spoilers” of superconductivity in FeSe, bounding clean regions of arbitrary shape to explore the limits of high- T_c superconductivity at small sizes – a goal which has been elusive in cuprates due to their complex crystal structure and susceptibility to damage during nano-patterning [34]. Furthermore, we envision useful devices such as ultra-precise superconducting metamaterials for THz optics or electron lensing, or the world’s smallest SQUID. With true 3D printing, i.e. lateral manipulation in subsequent,

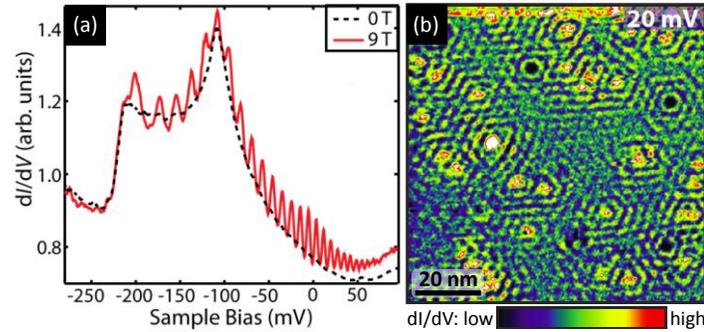


Fig. 6: First simultaneous observation of Landau levels (LLs) and quasiparticle interference (QPI) on any material. **(a)** STM spectra on Sb(111) showing 27 LLs, more than have been observed in any topological material to date. **(b)** QPI of surface states on Sb(111), showing scattering from a few defects. [28]

separated layers of FeSe, a second current loop could be added to the SQUID to make the world's smallest magnetic susceptometer.

To exploit "atomic 3D printing" more broadly, we aim to catalyze collaborations linking fundamental theory, materials synthesis, and our own imaging to master the materials ("ink"); computational modeling to suggest desired atomic configurations ("drawings"); our own MBE and SPL ("printing") and spectroscopic and transport characterization ("proofing"); with eventual engineering of novel applications such as ultra-fine wires for fast computing with charge or spin, ultra-dense memory, ultra-precise metamaterials for shaping electromagnetic fields, or ultra-sensitive magnetic detectors.

Outlook

The Hoffman laboratory combines a versatile set of tools and expertise, to grow new materials and to image and manipulate their nanoscale electronic and magnetic properties. The specific projects discussed above represent only a few examples in which MBE and SPM can lead to breakthroughs in the understanding and utility of materials. We are committed to an energetic and collaborative research program to continue to address forefront problems in fundamental science and high-impact technology.

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