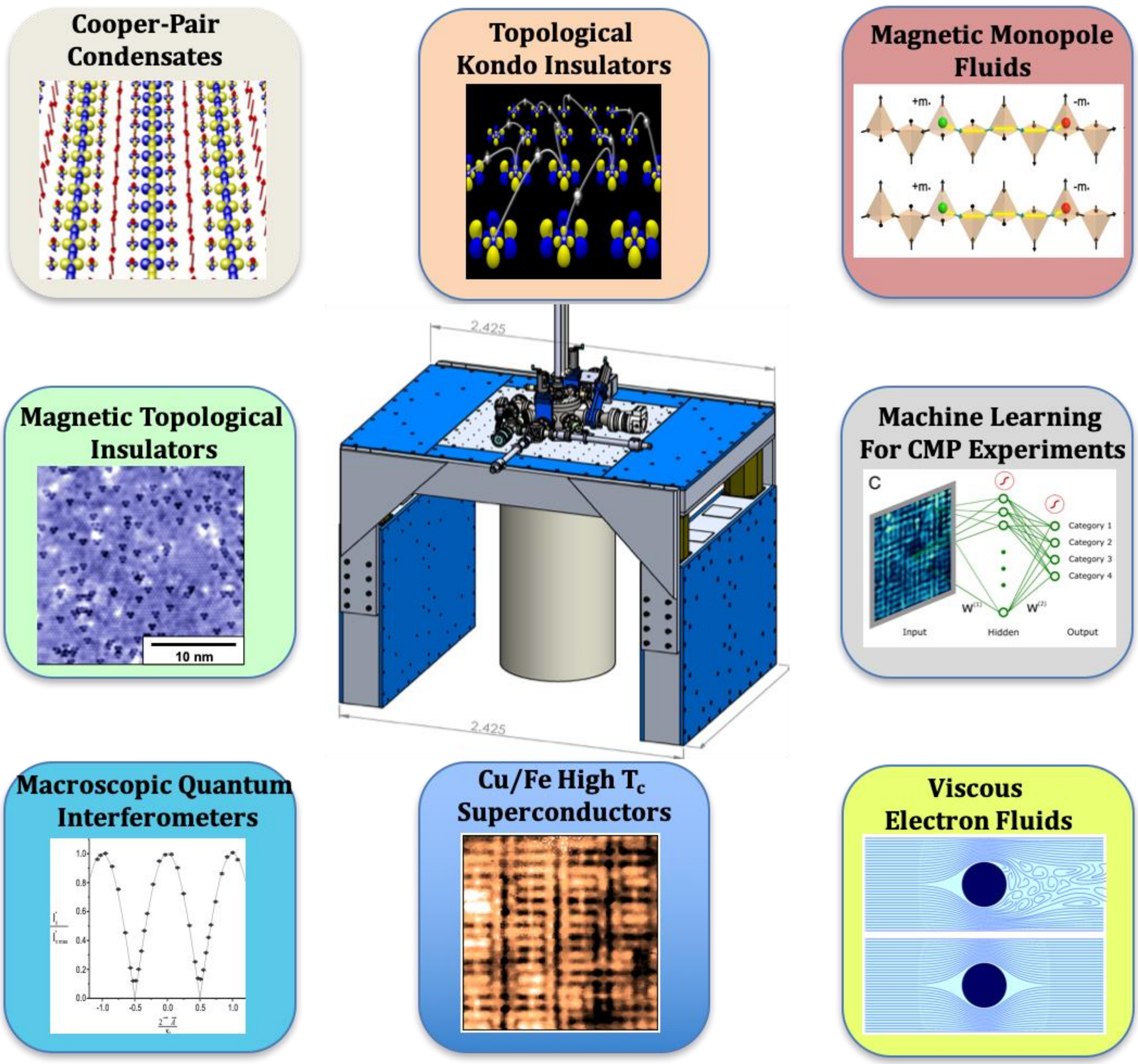
Visualizing Quantum Matter at the Atomic-Scale

Davis Group research concentrates upon the fundamental physics of electronic, magnetic and atomic quantum matter. A specialty is development of innovative instrumentation to allow direct visualization (or perception) of characteristic quantum many-body phenomena at atomic scale. Among the fields of active interest today are:



Davis Group plans to operate two suites of ultra-low vibration laboratories, one in Beecroft Building at Oxford University (UK) and the other in the Kane Building at University College Cork (IE). Ours is as single research group conducting scientifically harmonized studies with complementary scientific instruments at Oxford and Cork. The overall objective is to exploit the distinct capabilities and facilities at both laboratories to maximize scientific efficiency.

Our immediate research objectives (and associated collaborators) include:

Cooper-Pair Condensates (Prof. A.P. Mackenzie - MPI CPfS)

Research Status: We recently introduced nanometer resolution Scanned Josephson Tunneling Microscopy (SJTM), a technique allowing imaging of Cooper-pair tunneling from a superconducting STM tip to the Cooper-pair condensate of a superconductor. The SJTM operates at millikelvin temperatures and sequentially forms an array of 65,500 nanoscale Josephson junctions, whose Josephson critical current I_c is then measured to form the condensate image (***Nature*** 532, 343 (2016)). For the first time in superconductivity research, one can visualize the Cooper-pair condensate itself (Fig. 1A).

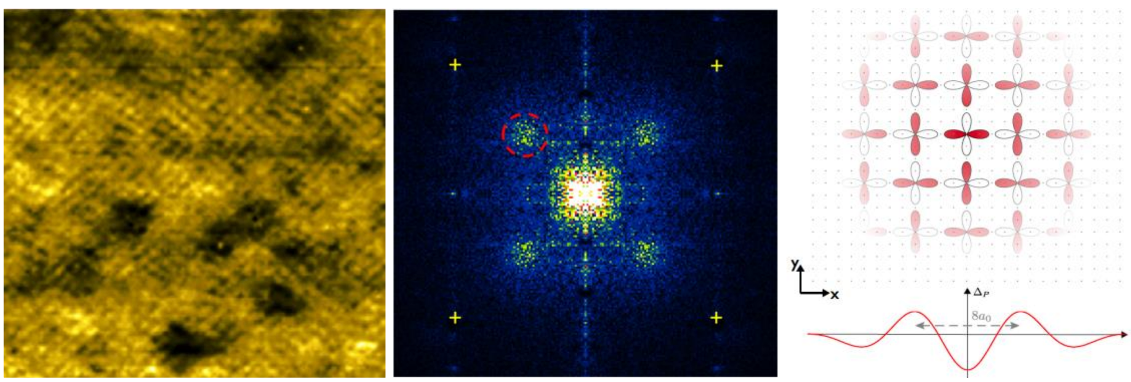


Fig. 1A Josephson critical current $I_c(r)$ images with atomic-resolution in 75nmX75nm FOV; B) Fourier transform of A shows the existence of 4a0-periodic modulations of Cooper-pair density due to the PDW state; C) Schematic representation of a biaxial *d*-symmetry PDW.

Research Plans: SJTM is a very promising new approach to research into all kinds of heterogeneous superconductivity. Projects of immediate research interest include:

a) The Cooper-pair density wave (PDW) state occurs when the density of Cooper-pairs modulates periodically in space at wavevector $\boldsymbol{Q_P}$. Only one instance has ever been detected (***Nature*** 532, 343 (2016)). Now we plan a search for new PDW states in several classes of materials. Transition metal dichalcogenides appear ideal, because they often host both superconductivity (SC) and charge density waves (CDW); Ginzburg-Landau theory predicts that a PDW state must be induced by the interactions between the SC and CDW states. Heavy-fermion superconductors e.g. $CeCoIn_5$ at high fields are also reported to host PDW states. Copper-based high temperature superconductors (CuHTS) materials, e.g. $YBa_2Cu_3O_7$ and La_2BaCuO_4, are widely predicted to host a strong-coupling PDW state.
b) In CuHTS, an exceptional new electronic phase appears at highest magnetic fields. It supports unexplained quantum oscillations and an unidentified density wave (DW) state. Although generally referred to as a CDW, theory indicates that this could actually be a PDW state. Because this field-induced DW state is accessible in the "halo" surrounding quantized

vortex cores (*Science 295*, 466 (2002)), we now plan to image this "halo" DW using SJTM to determine directly if it is a PDW.

Magnetic Monopole Fluids (Prof. S. Blundell - Oxford)

Status: Magnetic monopoles are hypothetical elementary particles exhibiting quantized magnetic charge $m_0 = \pm(\mathrm{h}/\mu_0 \mathrm{e})$ and quantized magnetic flux $\Phi_0 = \pm \mathrm{h}/\mathrm{e}$. A classic proposal for detecting such magnetic charges is to measure the quantized jump in magnetic flux Φ threading the loop of a superconducting quantum interference device (SQUID) when a monopole passes through it. Naturally, with the theoretical discovery that a fluid of emergent magnetic charges should exist in several lanthanide-pyrochlore magnetic insulators including $Dy_2Ti_2O_7$, this SQUID technique was proposed for their direct detection (Castelnovo *et al* ***Nature 451***, 42 (2008)). Experimentally, this has proven extremely challenging because of the high number density, and generation-recombination (GR) fluctuations, of the monopole plasma. Recently, however, theoretical advances by Prof. S. Blundell of Oxford University have allowed the spectral density of spin-noise $S_\Phi(\omega, T)$ due to GR fluctuations of $\pm m_*$ magnetic charge pairs to be determined.

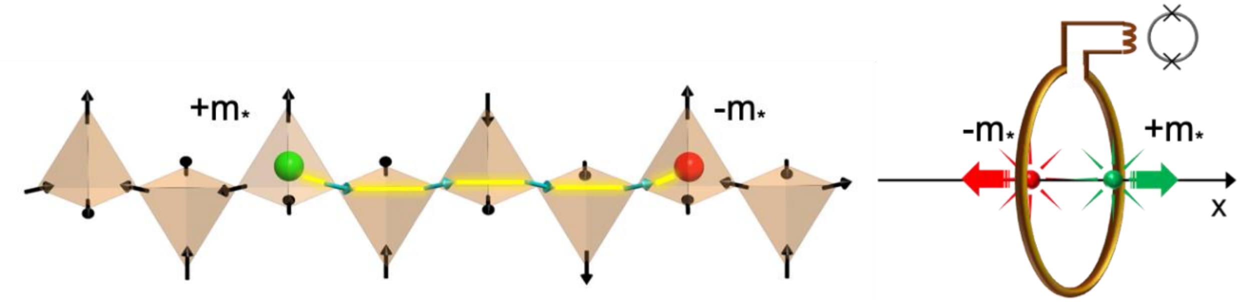


Fig. 2A The sequence of Dy spin flips in $Dy_2Ti_2O_7$ that generate two magnetic monopoles of opposite charge propagating through the material. B) When these monopoles are created within the input coil of a DC SQUID and depart to infinity in opposite directions, the flux through the SQUID jumps by $\Phi = m^* \mu_0$.

In 2018 we developed a high-sensitivity, SQUID based spin-noise spectrometer, and measured the frequency and temperature dependence of $S_\Phi(\omega, T)$ for $Dy_2Ti_2O_7$ samples. Virtually all the elements of $S_\Phi(\omega, T)$ predicted for a magnetic monopole fluid, including the existence of intense magnetization noise and its characteristic frequency and temperature dependence, are detected. This provides the first direct access to the microscopic physics a monopole fluid.

Research Plans: High precision measurement of the spin-noise spectrum is an innovative approach to magnetic quantum fluids. It opens a wide variety of new research avenues including the following projects of immediate interest:

a) $Ho_2Ti_2O_7$ is a pyrochlore magnetic insulator with many similar characteristics to $Dy_2Ti_2O_7$ and it is widely believing to also contain a fluid of emergent magnetic monopoles. We plan to use our spin-noise spectroscopy (SNS) technique to search for the flux noise $S_{\Phi}(\omega, T)$ signature of magnetic monopole fluid in $Ho_2Ti_2O_7$.

b) Based on our measurements, we estimate that the flux jump of individual magnetic monopoles in $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ should be detectable in sub-micron scale samples and at mK temperatures. We plan to develop a millikelvin SNS instrument for this purpose, and to search for individual magnetic monopoles in these and other compounds.

c) Eventually, out SNS approach will be generalized into a visualization technique in the form of a Scanned Spin-Noise Microscope (SSNM). We plan development of this new instrument as part of the suite of new quantum microscopes at our Beecroft Building laboratories.

Magnetic Topological Insulators (Dr. Genda Gu – BNL)

Status: Surface states of topological insulators (TIs) are expected to exhibit many valuable new electronic phenomena when a 'mass gap' is opened in their Dirac spectrum by ferromagnetism (FM). Such ferromagnetic topological insulators (FMTI) should exhibit phenomena including the *Quantum Anomalous Hall Effect* (QAHE), the *Jackiw-Rebbi Solitons* (JRS), and *Emergent Axionic Electrodynamics*. The QAHE has indeed been observed but, mysteriously, it is only detected at mK temperatures.

To explore the intriguing physics of FMTI, we recently developed the first visualization technique for the Dirac mass of FMTI surface states. We found that the Dirac mass $m(\boldsymbol{r})$ is extremely disordered and correlates with the local density of the magnetic dopant atoms generating FM state. This chaotic Dirac-mass landscape $m(\boldsymbol{r})$ poses far more questions on FMTI than it answers.

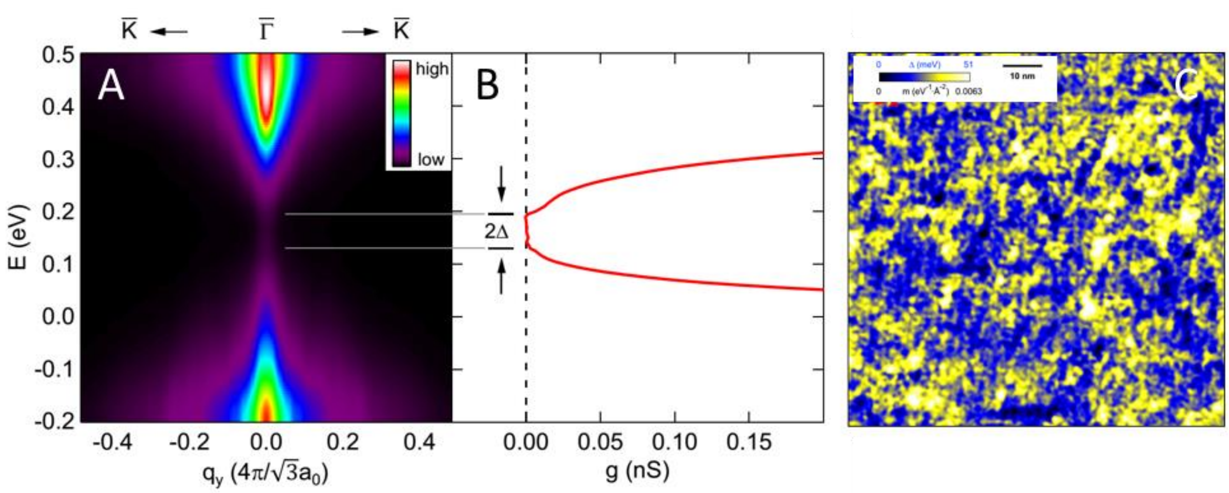


FIGURE 3A. Intensity of tunneling conductance *g(q,E)* into the Dirac spectrum of surface-states of $Cr_{0.08}(Bi_{0.1}Sb_{0.9})_{1.92}Te_3$; the ferromagnetism opens a gap Δ_{FM}~20meV around the Dirac point where the conductance reaches zero. B. Tunneling conductance at a single atomic location; again Δ_{FM}~20meV where the conductance reaches zero. C. Typical spatial map of $\Delta_{FM}(r)$ in $Cr_{0.08}(Bi_{0.1}Sb_{0.9})_{1.92}Te_3$

Research Plans:

a) In general, ferromagnets exhibit both FM domains and magnetic hysteresis, and FMTI are no different. But these phenomena should, in theory, have a profound influence on JRS and QAHE. We plan to measure the atomic-scale electronic structure throughout the hysteresis loops of $Cr(BiSb)Te_3$ and $Va(BiSb)Te_3$ and thus to visualize the evolution of FM domains and the network of JR states that should exist between regions of opposite magnetization.

b) The QAHE only stabilizes at temperatures T<<1K. This likely means that nanoscale disorder (Fig. 2C) somehow shorts out the chiral edge currents, allowing them to pass through the centre of the sample so that the conductance is not quantized. Precisely how this happens is unknown. We plan to image topological surface states of FMTI approaching QAHE with falling temperature, to visualize how the bulk currents are destroyed and the QAHE edge current stabilized.

c) The interplay of electric field ***E*** and magnetic field ***B*** at the surface of FMTI should be analogous to that predicted theoretically for axions. We plan to pursue proposals for how to observe this effect by generating axionic phenomena with an STM tip and observing the nanoscale ***B***-field response.

Topological Kondo Insulators

Status: In a crystal with a sub-lattice of localized *f*-electron states, the Kondo effect generates a heavy-fermion band structure. At high temperatures, a conventional (light) electronic band coexists with localized *f*-electron states on each magnetic atom. At lower temperatures, hybridization between this light band and the *f*-electron states results in opening a hybridization gap $\Delta_{HF}(k)$, and its splitting into two new very flat bands with greatly enhanced density-of-electronic-states $N(E)$ within just a few meV of E_F. We developed a dilution-refrigerator-based mK SISTM instrument for mapping simultaneously the ***r***-space and ***k***-space electronic structure of heavy-fermion systems at temperatures down to 20 mK. Demonstration of the feasibility of this approach for visualizing heavy-fermion formation, and measuring heavy-fermion band-structures, launched the field of STM studies of heavy fermions (***Nature 465***, 570 (2010)).

Research Plans: The capability to image heavy fermions (Fig 4) opens exciting new avenues for research into strongly entangled electronic quantum matter.

a) The theory of topological Kondo insulators (TKI) postulates a strongly anisotropic $\Delta_{HF}(k)$ that inverts the parity of bulk heavy-fermion states. The resulting prediction is for heavy-fermion topological surface states to appear at three points of the surface BZ. To explore these phenomena, we plan to apply high-resolution heavy-fermion visualization technique at millikelvin temperatures to measure the ***k***-space structure of $\Delta_{HF}(k)$ of the TKI SmB_6.

b) Such mK SISTM techniques also represent an exciting opportunity to achieve direct visualization of electronic quantum criticality. When quantum fluctuations become sufficiently strong, heavy-fermion systems often undergo a quantum phase transition to a new ground state. Indeed, understanding this type of quantum critical electronic matter is one of the key challenges of condensed matter physics. $YbRh_2Si_2$ is a heavy-fermion system with a QCP near B=0.66 Tesla (and no superconductivity). We plan to apply mK visualization techniques in magnetic field, to determine the heavy-fermion band structure, and to characterize the quasiparticles in the quantum critical regime surrounding the antiferromagnetic QCP of $YbRh_2Si_2$.

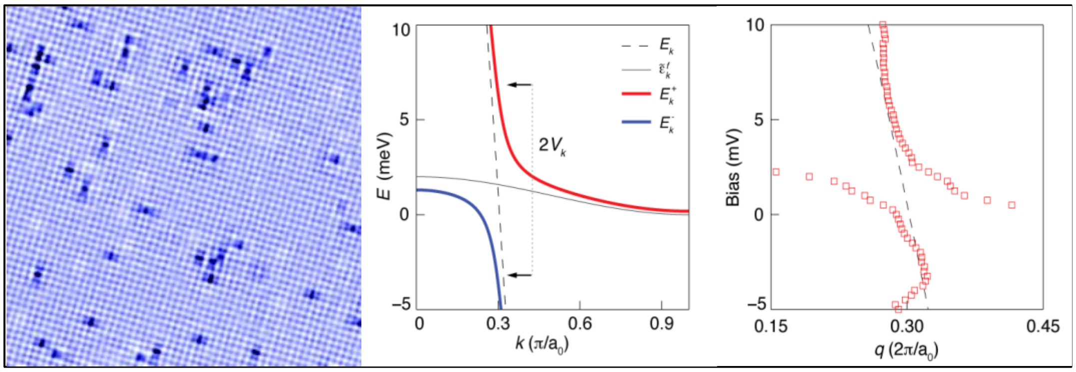


Fig.4A URu_2Si_2surface with Th substitution sites dark; B) the expected heavy-fermion band formation due to hybridization of the localized/magnetic Ur electrons with delocalized Ru electrons; C) our QPI measured heavy-fermion band formation in of URu_2Si_2.

Cu/Fe HT Superconductors (Dr. H. Eisaki – AIST & Prof. P. Canfield – AMES)

Status: Novel 'electronic liquid crystal' phases have long been predicted for correlated electronic materials, especially those where the intense correlations generate the highest temperature superconductivity. By using direct atomic-scale visualization we have discovered several of these phases including the smectic (DW) state in CuHTS (*Science 295*, 466 (2002); *Nature 430* , 1001 (2004) ; *Science 315*, 1380 (2007)); the nematic phase in CuHTS (*Nature 466*, 374 (2010); *Science 333*, 426 (2011)); the famous nematic phase of FeHTS (*Science 327*, 181 (2010); *Science 357*, 75 (2017)) the Cooper-Pair Density Wave (PDW) state in CuHTS (*Nature 532, 343* (2016)).

Research Plans: Having established the existence of these broken-symmetry electronic liquid crystal states, the challenge now is to understand their relationship to the HTS.

a) Recently the effects of quenched disorder on such a two-dimensional DW state have been discovered. While long range order of a unidirectional incommensurate DW cannot exist in the presence of quenched disorder, its short-range remnant survives up to a certain critical disorder strength but in the form of a $\boldsymbol{Q}$=0 broken rotational-symmetry state. This state was dubbed a *vestigial nematic* (VN). We plan to search for the VN state by determining if energy scale of nematic state is the same as that of the DW state throughout phase diagram.
b) Intense theoretical interest has emerged in whether a PDW state is actually the competing phase to superconductivity in CuHTS. Thus, we plan to test if the reported charge

modulation phenomenology is actually a secondary effect of a fundamental PDW state. We will image conventional density-of-states $N(r,E)$ of charge modulations, simultaneously with imaging of Josephson $I_C(r)$ to visualize the PDW. Comparison between the first ever such pairs of $N(r,E)$: $I_C(r)$ images will be highly revealing as to which state is fundamental.

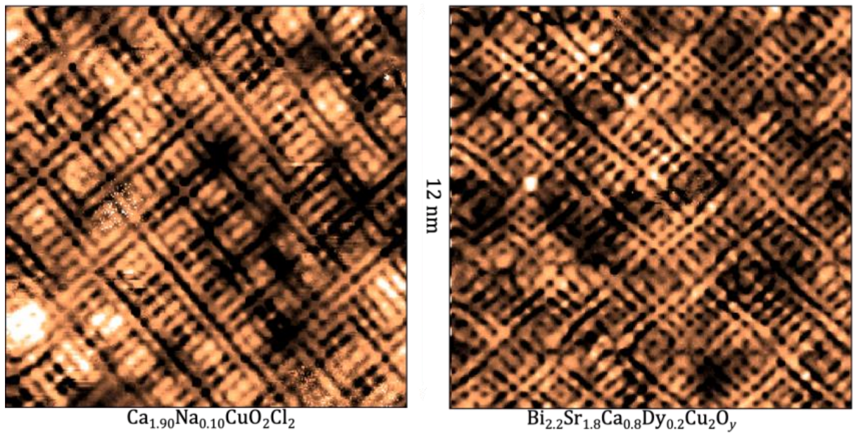


Fig. 5A,B The characteristic DW observed in virtually all CuHTS system has very short correlation lengths and appears to be lattice commensurate and 4a0 period.

Viscous Electron Fluids (Prof. A.P. Mackenzie - MPI CPfS)

Status: There is now widespread interest in whether some electron fluids exhibit viscosity. Key evidence for this phenomenon comes from studied of ultra-pure dellafossite crystals (A.P. Mackenzie *Rep. Prog. Phys.* **80** 032501 (2017)).

Fig. 6 All familiar viscous fluid flow in the geometry(L): viscosity (ν): density (ρ) regime that exceeds Reynolds number R=ρvL/ν~3000, generates turbulence. Here the viscous flow from left to right past an obstacle generates a vortex train.

Research Plans: A profound challenge for this field is to detect turbulence of an electronic fluid. No phenomena e.g. Fig. 6 have ever been observed for any electron fluid. Thus, exploratory studies to visualize viscous phenomena in an electron fluid are of great interest

(1) We plan to attempt visualize the impurity scattering interference in Co-dellafossite crystals whose Fermi surface si already very well understood. Subsequently, a large electric current generating (electron fluid flow) will be applied and its effects visualized directly at atomic scale (in a conventional electron gas no detectable effects would be expected) .
(2) If effects of electron fluid flow are observable, then the Reynolds number for an atomic scale perturbation will be used to predict the current density necessary to cause turbulence, for which we will then search.

Macroscopic Quantum Interferometers

Status: Superfluid Josephson junctions use nano-aperture arrays through which the fluid can flow quantum mechanically. We invented such junctions and then discovered superfluid Josephson oscillations (***Nature 388***, 449 (1997)), the current-phase relationship of a superfluid Josephson junction (***Science 278***, 1435 (1997)), π-states within the Josephson junction (***Nature 392***, 687 (1998)).

The key technical outcome was development of the first superfluid macroscopic quantum interferometer (DC-SQUID; ***Nature 412***, 55 (2001)). This device is completely insensitive to electromagnetism but couples directly to inertial accelerations and rotations.

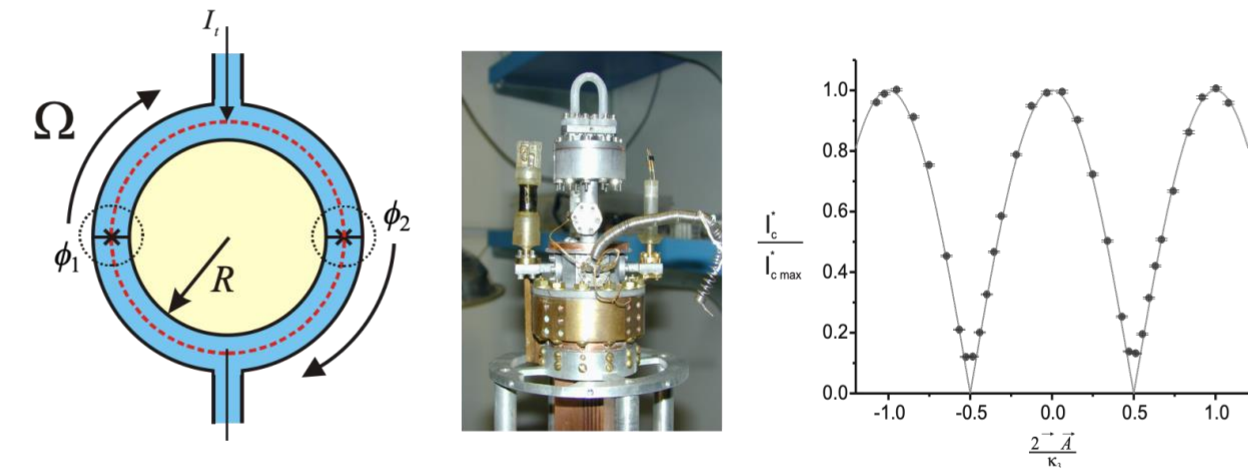


Fig. 7A Schematic of a superfluid macroscopic quantum interferometer, here sensing rotation Ω. B) Image of a functional superfluid macroscopic quantum interferometer with sensing area ~3cm², C) Measured interference patterns of superfluid macroscopic quantum interferometer (DC SQUID) with changing rotation.

Research Plans: Because the inertial mass of an 4He atom is ~ 10^9 that of an optical photon, quantum interference of atom wavefunctions is concomitantly (extraordinarily) more sensitive to inertial effects.

a) Development of large-scale (meter) version of a superfluid interferometer (***Nature 412***, 55 (2001)) as a prototype for modular atomic interferometry
b) Development of large-scale quadrupolar sensitivity superfluid ^4He interferometer in an ultra-low vibration environment, for low frequency studies of metric stain dynamics.

Quantum Microscope Development

For a decade after the invention of the scanning tunnelling microscope (STM), comprehensive mapping of the complete electronic structure of a material in *r*-space $N(r, E)$ or of its *k*-space electronic structure E(k) had not been contemplated. We introduced and demonstrated (*Rev. Sci. Instrum.* 70, 1450 (1999)) a design for a spectroscopic imaging scanning tunnelling microscope (SISTM) allowing electronic structure visualization as a powerful general tool for solid-state physics research. Building on that method, we have developed instruments to achieve the following quantum visualization techniques: *Spectroscopic Imaging STM* (*Nature 403,* 746 (2000)); *Quasiparticle Interference Imaging* (*Science 266,*455 (2002)); *Superconducting Order Parameter Determination* (*Science 357,* 75 (2017)); *Kondo Heavy-Fermion Visualization* (*Nature 465,* 570 (2010)); *SJTM Cooper-pair Condensate Visualization* (*Nature 532,* 343 (2016)); *Orbital Selective Quasiparticle Visualization* (*Science 357,* 75 (2017)).

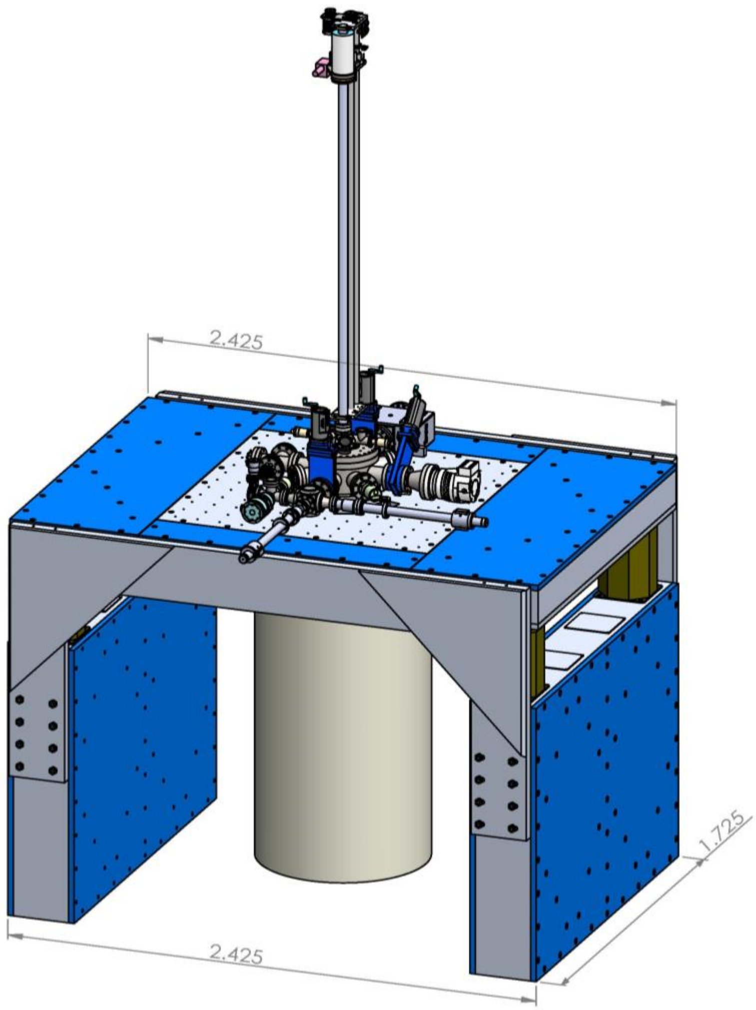


Fig. 9 Schematic of the externals of fourth the generation quantum microscopes designed specifically to operate in the ultra-low vibration (ULV) laboratories in the basement (20 m underground) of Beecroft Building – Oxford, where three instruments with distinct capabilities are planned. Three other quantum microscopes with distinct and complimentary capabilities will be housed in the Kane Building at UCC.

Research Plans: For the Davis Group research program, we will design, fabricate, install and operate fourth-generation quantum microscopes with advanced, multi-functional and modular capabilities. All the studies outlined above (except atom interferometry) will be carried out using the following unique set of instruments:

a) A mK SISTM quantum microscope, operating down to T=250mK and at magnetic fields up to 16T. Its planned uses include *Cooper-Pair Condensates & Topological Kondo Insulators*.
b) A mK SJTM quantum microscope, operating below T=50mK and at magnetic fields up to 12T. Its planned uses include *Magnetic Topological Insulators & Cu/Fe HT Superconductors.*
c) A mK SNS quantum microscope, operating down below T=50mK in zero field. Its planned uses include *Magnetic Monopole Fluids & Viscous Electron Fluids.*