Nature and origin of unconventional superconductivity in ultra-clean UTe_2

F. M. Grosche, A. Huxley, A. G. Eaton, S. S. Saxena, P. Wahl, A. Hermanns, G. G. Lonzarich

Case for support

Uranium-based unconventional superconductors are surprisingly abundant and continue to defy a comprehensive understanding. Foremost among these is the new superconductor UTe_2, which displays at least three distinct superconducting states and in which superconductivity can survive up to fields exceeding 60 T, indicating triplet pairing. With ultra-clean crystals of UTe_2 now available, this project combines wide-ranging experimental, computational and theoretical studies into the origins of superconductivity and the associated anomalous normal states in UTe_2 and related uranium-based quantum materials.

Vision: UTe_2 as a model unconventional superconductor

Superconductivity resurgent. Superconductivity research is ramping up globally, driven by (i) the recognition that superconductors facilitate large-volume applications for instance in fusion research, accelerators, MRI scanners, generators and motors, and power distribution, as well as device applications in computing and sensing; (ii) exciting breakthroughs in fundamental research across different material systems ranging from the cuprates and Fe-based high temperature superconductors to organics, twisted bilayer graphene and f-electron systems; (iii) materials breakthroughs, including the ability to induce near-room temperature superconductivity in supercompressed superhydrides [1, 2], the discovery of 80 K superconductivity in a pressurised, novel nickelate [3], and the discovery of multiple field resilient superconducting states in $CeRh_2As_2$ [4] and UTe_2 [5, 6].

In conventional superconductors, the pairing interaction is communicated by lattice vibrations. Fundamental and applied superconductivity research are increasingly examining unconventional superconductors, which instead harness the strong electronic interactions that are also responsible for magnetism and that are known in some cases to reach coupling strengths equivalent to several thousand Kelvin [7, 8]. Like rare minerals that occur in seams, these superconductors are thinly spread across the space of all accessible materials but richly concentrated within those families on which most current research is focused, which include, for example, various copper oxide, iron or cerium compounds.

Uranium-based superconductors make up a large fraction of the overall still limited number of unconventional superconductors (Table). This material family is highly diverse in terms of crystal and electronic structure. Studying and learning from these U-based superconductors can accelerate the wider search for unconventional superconductors with desirable properties, but the bewildering diversity and complexity of phenomena and materials challenges in this class of materials renders a detailed and comprehensive understanding at best time-consuming and difficult.

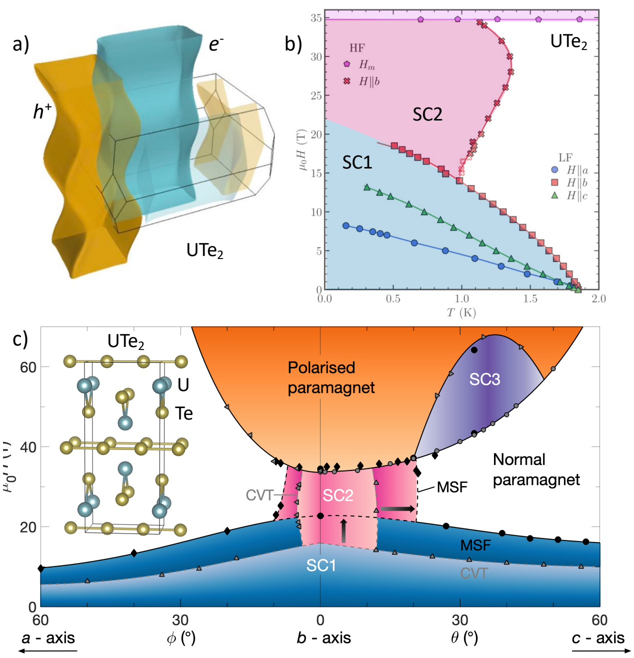

FIG. 1. Key properties of UTe_2. (a) Crystal structure and Fermi surface geometry deduced from quantum oscillation measurements, showing compensated electron and hole pockets of constant cross-section that undulate slightly along the *c*-axis. (b) Temperature dependence of the superconducting upper critical field B_{c2} along the *b*-axis, showing transition between two superconducting states, SC1 and SC2. (c) Angle dependence of the low temperature B_{c2}, showing the resilience of SC2 to applied fields reaching up to a metamagnetic transition at $\simeq$ 35 T and a third superconducting state SC3 that reaches to far higher field values, still. Our improved 'MSF' crystals reach higher critical fields in the SC1 state (dark blue) and superconductivity extends over a wider angle range in the SC2 state (pink) than previous 'CVT' generations of samples.

Examining the new superconductor UTe_2 can produce important insights for understanding unconventional superconductors more generally: (i) it holds at least three, probably more, distinct superconducting states, which can be selected by varying applied field, temperature and pressure, (ii) at least some of these are triplet pairing states, as demonstrated for instance by NMR measurements and by the unusual resilience of superconductivity to applied field of up to $\simeq$ 60 T in certain field directions, (iii) low temperature magnetic order identified at moderate pressure and strong magnetic fluctuations observed by neutron scattering at ambient pressure strongly suggest a central role for a magnetic pairing mechanism, a strong contender also in other unconventional superconductors such as the Ce- and Yb-based heavy fermion systems and Fe- or Cu-based high temperature superconductors.

Clean crystals for a clearer view. Until recently, work on UTe_2 was hampered by poor crystal quality. Long electronic mean free paths are required to establish anisotropic forms of order, such as unconventional superconductivity. Previous generations of samples exhibited a residual Sommerfeld ratio C/T in the low-T limit within the superconducting state, which caused much theoretical speculation that could quickly be dismissed once $C/T \rightarrow 0$ could be established in cleaner crystals more recently. Likewise, informative investigation methods such as quantum oscillation measurements require high quality samples and proved impossible for several years, causing ample speculation about the electronic structure near the Fermi energy. New growth methods pioneered by our project partners at Charles University, Prague, now produce pristine single crystals with superior quality and electronic mean free path as measured by their residual resistance ratio (RRR) of order 500, an order of magnitude improvement on previous best efforts. Using this new generation of ultra-clean crystals grown using the molten salt flux (MSF) technique, we were able to detect quantum oscillations with unprecedented clarity, enabling us to resolve the Fermi surface structure of UTe_2 [9].

Simple Fermi surface. Because disorder is always relevant in unconventional superconductors, many initial findings in UTe_2, need to be reexamined in these new crystals. We have already found that the superconducting critical fields are significantly enhanced, whereas the metamagnetic transition remains unchanged [10]. Moreover, our recent quantum oscillation and quasiparticle interference oscillation measurements [9, 11] have revealed a surprisingly simple Fermi surface geometry, which consists of just two compensated, cylindrical, slightly corrugated pockets (Fig. 1), populated by heavy quasiparticles. The accuracy and simplicity of this result is reminiscent of the case of Sr_2RuO_4 [12]. It presents a solid point of departure for modelling the magnetic or charge response functions in UTe_2 and ultimately for understanding in detail the nature and origin of its superconducting states.

UTe_2 now presents a clean model system, because of the pristine quality of newly available single crystals and the simple Fermi surface geometry. Our primary vision for this project is to investigate those key aspects of UTe_2 that – like the electronic structure – reveal underlying principles and can lead towards a working model for superconductivity and magnetism in this intriguing material. These involve, in particular, careful mapping out of the high pressure, field and temperature phase diagram, the determination of the magnetic order induced at high pressure, and the investigation of normal state properties and Fermi surface by multi-probe studies and quantum oscillation measurements at high pressure. As the project unfolds we will turn this methodology also to related U-based superconductors such as UGe_2.

Research questions

Our project will address the following key research questions of wider relevance in superconducting quantum materials:

a) Superconducting states: UTe_2 hosts at least three distinct superconducting states, which can be selected in applied field. NMR data [] and the magnitude of the upper critical field B_{c2} strongly suggest that at least some of these involve triplet Cooper pair states. This produces a rare opportunity to study the nature and origin of triplet superconductivity in a clean system: what are the spatial order parameter wavefunctions (a large number of candidates are allowed by symmetry, but experiments can rule in or rule out candidates). Why is triplet superconductivity favoured? What causes high critical fields, in particular if they far exceed the paramagnetic limit (UTe_2 SC2 and SC3 states) or if superconductivity becomes reentrant (UTe_2 SC3)? What is the origin of residual C/T in the low T limit in less pure samples of UTe_2, and can this be quantitatively attributed to impurity bound states? What is the nature of the vortex lattice in the mixed state, and does it change as we tune UTe_2 between the different superconducting states SC1-3? Are there further superconducting states accessible under pressure, as preliminary studies suggest [], and what is the role of spin-orbit coupling in determining the supercnoducting state?

Addressing these questions provides new insights for applications in other material families: what determines B_{c2}, and what material properties can help maximise it? How do we identify anisotropic order parameters experimentally, how can they be manipulated, how do we find materials that host them?

b) Correlated 'normal' state: superconductivity in UTe_2 and other U-based systems arises out of a strongly correlated normal state. What is the origin of the high electronic heat capacity and enhanced quasiparticle mass in UTe_2 and other U-based heavy fermion compounds, which typically exceeds density functional theory (DFT) values by at least an order of magnitude? Many U-based superconductors have more than one f-electrons on each U site. This introduces strong local correlations via Hund's coupling, which may produce significant baseline mass renormalisa-

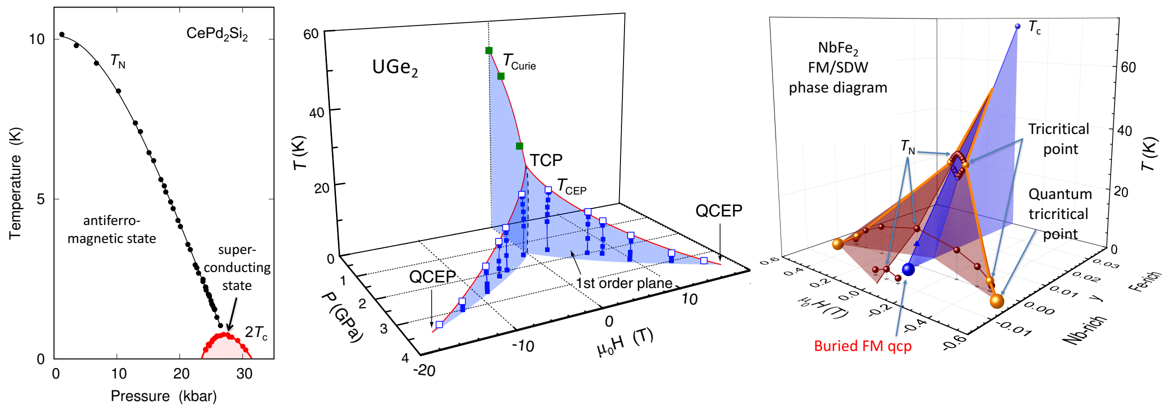

FIG. 2. **Template phase diagrams guiding materials exploration:** (a) High pressure phase diagram of $CePd_2Si_2$, showing a superconducting region attached to the threshold of antiferromagnetism [15]. Considering the added effect of magnetic field adds a third dimension: in UGe_2 (b), superconductivity appears within a ferromagnetic region, which itself branches into two metamagnetic sheets [13]. In PrPtAl or $NbFe_2$ [**?**] (c), by contrast, ferromagnetism is replaced by an antiferromagnetic or spin-density wave region. *** Use URhGe rather than UGe_2 for middle panel ***

tion, as demonstrated, for instance, in our recent QO study of YFe_2Ge_2 [14]. What is the f-electron count in UTe_2, does it change with applied pressure, and is the mass renormalisation uniform across the Fermi surface, supporting the Hund's metal scenario outlined above? Do normal state transport and thermodynamic properties universally follow Fermi liquid theory, or can we reach – under applied field or pressure – (quantum critical) regions in the phase diagram in which the electrical resistivity takes a sub-quadratic temperature dependence or the heat capacity Sommerfeld ratio C/T does not saturate to a constant at low T?

c) Magnetic, charge, and 'hidden' order: superconducting and normal state properties are controlled by the effective interaction between charge carriers, or quasiparticles. In contrast to the bare Coulomb interaction, the effective interaction can be dynamic, it can couple to spin, and it can be tuned by varying underlying material parameters. In many currently known unconventional superconductors, the interaction is predominantly magnetic [7], but different mechanisms are possible. These might involve density, valence, quadrupolar or orbital degrees of freedom, individually or in combination, which may be assessed by investigating ordered states nearby. In UTe_2, magnetic order is induced by moderate applied pressure $\simeq$ 17 kbar, but what is the precise nature of this magnetic order, and how is it affected by applied field and pressure? Because the pairing interaction can be mediated by magnetic fluctuations [7], where are the regions in the pressure-field-temperature phase diagram (see Fig. 2 for examples), where this interaction can become long-ranged? Moreover, charge density wave order (CDW) may be widespread in UTe_2 and other unconventional superconductors. What is the nature of CDW order, how does it affect superconductivity, and can we tune it by applied pressure or strain? In at least one prominent U-based superconductor, URu_2Si_2, a thermodynamic phase transition into a still unidentified 'hidden order' state continues to defy resolution. Is such hidden order more widespread across U-compounds, and what is its nature and origin? FM qcp -> 1st order or SDW or something else altogether (URu2Si2). Lessons from NbFe2, PrPtAl. Fermi surface instabilities Central e.g. to UGe2 story. Maybe happens more generally?! Local vs. band, orbitally selective Mott transitions?

Approach: multi-probe studies in clean crystals

The research programme capitalises on our recent breakthroughs in UTe_2 research [9–11] and our long track record of discovery research in heavy fermion superconductors such as $CePd_2Si_2$ and $CeIn_3$ [15], UGe_2 [16], $CeCu_2Si_2$ [**?**], and $CeSb_2$ [17].

The planned experiments (Table I) exploit our expertise and facilities in scanning tunneling spectroscopy (St. Andrews) and high precision transport, magnetic, thermodynamic and ultrasound measurements under extreme conditions (Cambridge and Edinburgh) of hydrostatic pressure (piston-cylinder and anvil cell devices, reaching up to >100 kbar), magnetic field (up to 20.4 T in the lab, with higher fields available at international facilities) and low temperature (down to <0.03 K). We will continue to refine and extend experimental methods, with particular emphasis on high pressure temperature modulation calorimetry, ultrasound and quantum oscillation measurements [17**?**].

The programme is structured into four work packages (WP). WP1 maps out the rich phase diagram of UTe_2 in field, temperature, pressure and strain. WP2 addresses the need to resolve and understand the low temperature ordered states, magnetic, superconducting or otherwise. Quantum oscillation experiments probing the electronic

Technique \ Environment	Field	Pressure	Strain	Who
Transport	✓	✓	✓	C, E
Heat capacity	✓	✓	(✓)	C
Magnetisation (SQUID)	✓	✓	✗	C
Magnetisation (torque)	✓	✗	✗	C
Ultrasound	✓	✓	✗	E
rf (TDO)	✓	✓	✓	C
Scanning Tunneling	✗	✗	✗	A
X-ray diffraction	✗	✓	✗	E
Neutron scattering	✗	✓	✗	C, E
Muon spin rotation	✓	✓	✗	C
Quantum oscillations	✓	✓	(✓)	C, E

TABLE I. **Techniques** to be used in this project. In the column 'Who', C=Cambridge, E=Edinburgh, A=St. Andrews. The columns show which sample environments (applied field, hydrostatic pressure or uniaxial strain) can be combined with each measurement technique.

Fermi surface play a central role in this effort, but will be supplemented by numerous complementary probes such as tunneling spectroscopy, transport studies, ARPES etc. Where possible experiments will extend to high pressure to investigate properties in those regions of the phase diagram that are of particular interest. The crucial WP3 concerns the growth and characterisation of clean crystals of our candidate materials as well as increasingly the exploration for new materials of interest, and WP4 covers the development of new instrumentation underpinning all of our studies.

Work package 1 (WP1): mapping the phase diagrams

The power of mapping out magnetic field, pressure and composition phase diagrams is illustrated in Fig. 2. The recent example of UTe_2 demonstrates that unexpected twists such as the ultra-high field superconductivity SC3, resilient up to 60 T for a narrow range of field orientations, could easily be missed without careful examination of a material's phase diagram over wide parameter ranges. Producing and refining the multidimensional phase diagram of an unconventional superconductor like UTe_2 provides the point of departure for more probing studies focusing on critical regions of the phase diagram and moreover by itself produces important clues for the nature of the underlying low temperature states. For instance, the surprising resilience of the superconducting states to applied field already points towards a triplet character of the superconducting order parameter, motivating follow-up by specialised probes. More generally, shape of the pressure, temperature, field phase diagram is of overarching importance, because it allows us to identify quantum critical points, where a pairing interaction could be expected to become relevant, and it would furthermore allow us to track the evolution of the enigmatic SC3 and SC2 superconducting pockets. We will use transport (electrical resistivity, Hall effect), thermodynamic (heat capacity), magnetic (magnetisation, muSR) and structural (XRD, ultrasound) techniques at applied pressure or strain and in applied fields to survey pressure/strain/field/temperature phase diagrams and thereby (i) delineate distinct magnetic, CDW or superconducting states and (ii) locate critical regions where response functions are expected to peak, e.g. near quantum phase transitions.
Objective 1: *Map out field, temperature, pressure and strain phase diagrams in high purity samples of UTe_2 to correlate superconducting and normal state properties with magnetic quantum phase transitions.* **Objective 2:** Widen these studies to other U-based superconductors, in particular UAu_2 and UGe_2.

WP2: resolving the low temperature states

Having identified regions of interest, the associated low temperature states – superconducting, magnetic, charge density wave – and also the properties of the underlying normal state need to be resolved.

Superconducting states: the nature of the superconducting order parameter can be inferred from a combination of bulk and surface probes. The former include measurements of the temperature dependence of heat capacity, thermal conductivity, nuclear magnetic resonance, or ultrasound attenuation. Of particular importance is the temperature dependence of penetration-depth, which can be determined using the tunnel-diode oscillator technique (Edinburgh, with Bristol) and via careful analysis of muon spin rotation studies. A key technique available in this project is scanning tunneling spectroscopy (STS) in the vicinity of surface defects, which has proven to be successful in effectively imaging the gap geometry []. At least for superconducting states accessible in low fields, we will apply this approach to the new generation of ultra-clean UTe_2 samples.

Further information can be inferred from the response of the superconducting states to an artificially reduced electronic mean free path in the bulk. Electron irradiation offers a highly controlled approach for varying defect concentration. By tracking the response of the three distinct superconducting states in UTe_2 to increasing defect concentration, we can detect changes in the superconducting order parameter. Analysis will incorporate the role of impurity bound states, which for a sign-changing gap produce distinct signatures in all low T properties, by numerical studies as in [**?**] and by varying the impurity level.

Magnetic order , which in UTe_2 is induced at high pressure > 17 kbar, will be investigated by high pressure neutron diffraction, muon spin rotation or X-ray magnetic circular dichroism, as well as by high pressure magnetisation measurements. to probe magnetic or superconducting ground state properties.

Fermiology: key input for any theoretical description derives from the observation of quantum oscillations in high magnetic fields, a precise signature of the electronic Fermi surface and carrier mass. Quantum oscillations have already been observed by us and others in UTe_2 at ambient pressure, but important questions are still unresolved. Moreover, it will be important to track the evolution of the Fermi surface and carrier mass as the magnetically ordered state is approached and crossed with pressure. We have pioneered rf tunnel-diode based techniques for tracking quantum oscillations in anvil pressure cells, which we will apply now to UTe_2 and, later, to other U-based superconductors. Ambient pressure quantum oscillation measurements in UTe_2 and other materials of interest will be referenced against information derived from STS studies (see also above). Ambient pressure and high pressure quantum oscillation surveys on the Cambridge 20.4 Tesla/dilution refrigerator cryomagnet will be augmented by measurements up to 30 Tesla at the new all-superconducting facility in Beijing, where we have recently carried out preliminary measurements, up to 37 Tesla at the HFML Nijmegen facility or higher fields, still, at NHMFL Tallahassee.

WP n Quantum oscillation studies

Understanding the electronic properties of the normal state out of which unconventional superconductivity emerges is crucial for subsequently interrogating the microscopic character of the superconducting state(s). In our recent (ambient pressure) high-field quantum oscillation (QO) measurements at NHMFL, Florida, USA we resolved the Fermi surface (FS) of UTe2 [cite Eaton et al]. Unlike several other uranium-based superconductors the UTe2 FS is remarkably simple, consisting of just two cylindrical sheets (see fig X). This simplicity proffers the tantalising possibility that an accurate microscopic description of UTe2 may soon be within our grasp.

A key outstanding question concerns the evolution of the FS under pressure - particularly as the putative QCP at $p_c \simeq 17\,\mathrm{kbar}$ is crossed. A magnetically ordered state at $p > p_c$ has been observed, which is proposed to be antiferromagnetic in nature but is yet to be conclusively determined (see muSR section). Mapping the FS through p_c will therefore provide vital clues pertaining to the role of magnetic and quantum critical fluctuations in driving the various exotic superconducting phases.

Tracking QOs under pressure is a challenging problem, but one for which we are ideally placed to tackle. Contactless RF resistivity measurements will be performed using our methodology recently deployed in [cite Semeniuk PNAS]. At ambient pressure we recently made the surprising observation of quantum interference oscillations (QIOs) in UTe2 [cite Weinberger et al], in addition to 'conventional' QOs from Landau quantisation. These provide information about the k-space gaps between neighbouring Fermi sheets along with the difference in the effective carrier masses; recently we have also observed QIOs in preliminary measurements under pressure. By combining detailed QO and QIO measurements as a function of pressure we will build a full map of both the Fermi surface geometry and the Fermi velocity distribution, yielding valuable insight as to the hybridisation of the Te p-bands with the U d- and f-bands. To our knowledge no such synergistic combinatory investigation – of both QOs and QIOs simultaneously – has previously been performed on any heavy fermion compounds. This novel approach may therefore be of particular interest in a range of other materials, with this study forming the groundwork for future QO+QIO investigations to directly resolve the FS geometry and fermi velocity distribution.

WP n+1 High magnetic field measurements

The phase diagram of UTe2 - as revealed by prior measurements on CVT specimens – is remarkably rich, with intertwined charge- and pair-density wave orderings coexisting with the groundstate (SC1) superconductivity. Under applied magnetic fields two further superconducting states emerge (SC2 & SC3) with SC3 spectacularly extending to 70 T [cite Ran Nat Phys & Helm et al], the highest ever recorded upper critical field for a re-entrant superconducting phase, or indeed for any heavy fermion superconducting state. In the new generation of crystals our preliminary measurements have already revealed that this rich phase landscape is markedly different, with the angular domain of SC2 showing an acute sensitivity to crystalline quality [cite Wu et al, see fig. X]. This naturally calls for a detailed mapping of the SC3 state in MSF samples, requiring measurements at pulsed magnetic field facilities for which we have recently been granted time.

Furthermore, under pressure four distinct superconducting states have been identified [cite Aoki review], with complex field-angle dependencies. Understanding how these phases evolve as a function of pressure and magnetic field tilt angle is challenging, as the axial length of typical pressure cells greatly restricts their ability to be rotated in the narrow bores of resistive magnets. Thus to date measurements have only been performed on separate samples with individual field orientations, with phase diagrams then stitched together, which greatly complicates the understanding of this remarkably complex phase landscape.

To address this, we have recently designed a miniaturised piston cylinder cell [see fig X] in collaboration with NHMFL, Florida, USA, specifically designed to fit into their recently constructed wide bore series-connected 36 T hybrid magnet. This will allow us to pioneer the combination of applied pressure and magnetic field tilt angle, enabling the full UTe2 phase diagram to be mapped on a single sample in-situ for the first time, thereby eliminating uncertainty as to how this plethora of exotic emergent phases evolves under pressure. This wide-bore magnet is also compatible with the Razorbill strain cell [see section X], which will allow us to pioneer the combination of high field and uniaxial pressure as well - which will yield important insights pertaining to the superconducting order parameter of the SC2 state [maybe cite & discuss Ramshaw/write about strain separately?]

Non Fermi liquid signatures will be examined using high-precision thermodynamic and transport measurements across pressure, magnetic field and temperature, in order to pin down the regions in the phase diagram where they extend to lowest temperature and correlate them with quantum critical phenomena arising from nearby ordered states. The role of disorder will be examined in samples of varying purity levels.

Objective 3: *Probe the superconducting states in UTe_2, UAu_2 and UGe_2 with complementary techniques in order to resolve the superconducting order parameter structure.*

Objective 4: *Develop a theoretical understanding of superconductivity and of anomalous normal state properties in all four material systems.*

Objective 5: *Resolve the Fermi surface and carrier mass and its evolution with pressure in UTe_2 by quantum oscillation surveys, extending later to related materials.*

Objective 6: *Survey non-Fermi liquid signatures using high precision temperature sweeps into the milli-Kelvin range, in fields up to* 20 T *and pressures up to* 100 kbar.

WP3: crystal growth and materials discovery

a. MSF b. Induction furnace c. CVT

Availability of depleted uranium for crystal growth

Crystal quality plays a central role in the discovery of new collective phenomena in quantum materials.

Selected materials

For these reasons, our project will initially investigate three material systems available as <u>high-quality single crystals</u>, in which discoveries and enabling breakthroughs have occurred as recently as last summer (Fig. 3):

a) UTe_2: Although synthesising single crystals of UTe2 has proved to be straightforward via chemical vapour transport progress in improving sample quality has required the systematic optimization of parameters (Cairns J. Phys.: Condens. Matter 32 (2020) 415602, Rosa et al Nature Commun Mater 3, 33 (2022)). As the ratio of Te/U in the deposition zone (controlled by the reagent composition and temperature) grows towards 2:1,

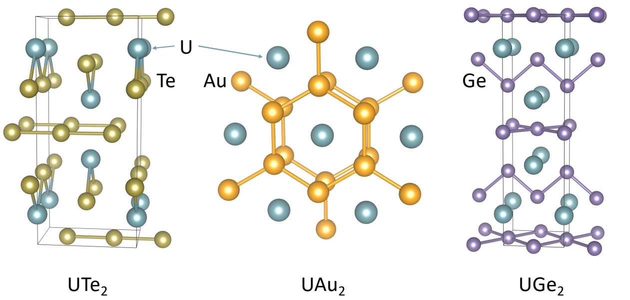

FIG. 3. Material families identified for the first stage of this project. As the programme unfolds, the investigation will widen first to other U-based superconductors (see table) and eventually to *d*-metal compounds with strong Hund's coupling and hence underlying similarity to the headline materials.

UAu_2	AFM, s/c under pressure, A. Huxley 21, 22 PNAS, possible multi-component order parameter, QO under pressure?
UBe_{13}	
UCoGe	FM
U_6Fe	Perhaps CDW at 110K? Whitley PhD. Looks like interesting linear-in-T rho(T) but little good transport data published. RRR 10
UGe_2	FM
UIr	
UPd_2Al_3	AFM, TN 14K, Tc 2K
UNi_2Al_3	AFM, TN 1.4 K, Tc 1 K
UPt_3	
URhGe	FM
URu_2Si_2	Hidden order
UTe_2	dHvA under pressure Strain in high field, to investigate multi-component SC1/SC2 Pioneer miniaturised pressure cells – map full p-B-T-angle phase diagram QIOs under pressure, combined with dHvA gives full 3D details of FS without needing to rotate

Tc of the resultant crystals increases, but then falls abruptly. The dramatic drop in Tc is attributed to U deficiency. As well as its role in controlling the composition in CVT a lower growth temperature also appears to lower the intrinsic concentration of U vacancies formed. Growth from molten salt (NaCl/KCl) flux occurs at lower temperature than achieved with CVT (with iodine as transport agent) and has resulted in higher Tc and RRR crystals. The molten salt grown crystals are however smaller (sub mm) and have a large scatter in quality within a single batch. The current crystals grown in Edinburgh by CVT have sharp heat capacity transitions above 2.0 K, but with varying non-SC fractions. While the thermodynamics of the processes involved in the growth process are straightforward to quantify, the kinetics is equally important and is not well characterised. Methodology.

Both CVT and molten salt growth will be carried out. We will characterise our crystals alongside those form our collaborators (CU-Prague) to ensure we have the best material available for subsequent study. Characterisation will by inhouse Laue, Energy dispersive X-ray (including FIB), PPMS (heat capacity and susceptibility to 300K - 350mK, 7 Tesla) and homebuilt heat capacity (to 4K - 20 mK 14 Tesla) as well as transport.

We aim to better optimise the synthesis process. We will do this by incorporating fibre optics into our CVT furnace to image the growth process in real time. This will give a better understanding of the kinetics. For the molten salt growth as well as following known recipes with a standard vertical furnace we will apply RF heating directly to the carbon crucible to better control the temperature gradient at the conical growth tip to maximize crystal yield and size.

The work programme will combine optimizing growth to produce the best samples possible and making

When flux growth or chemical vapour transport are not productive, we use cold-crucible induction melting, and we will explore Czochralski and Bridgman growth for single crystal production. We will continue to improve these techniques by using higher quality starting materials, by tuning the growth protocol and by optimising the annealing procedure.

We will widen our programme

1. UAu_2 and UGe_2 Text about UAu_2 and UGe_2

2. to other well-known U-based heavy fermion superconductors such as UPt_3 and URu_2Si_2. With recent advances in instrumentation a re-examination of the superconducting and magnetic states in the former is becoming timely. Little is known, moreover, regarding its evolution with pressure and strain. In the latter, which hosts an enigmatic hidden order state below about 17 K at ambient pressure, quantum oscillation measurements to higher magnetic fields than were possible in the past will reveal much-needed information about Fermi surface geometry and carrier mass.

3. to related U-based systems such as U_6Fe or UBe_{13}.

4. to other materials: we expect the expertise gained in the growth of UTe2 to also enhance capability to grow other U -Te. There are several interesting 2D van der Waals magnetic systems with higher Te content that could be synthesised. These may also give insight into UTe2 and appear as defects.

5. *d*–metal compounds that may mimic some of the properties of the U-based superconductors which form the central objective of this project.

Sample characterisation will involve powder and single-crystal x-ray diffraction as well as electron microprobe analysis, and the determination of magnetic, thermodynamic and transport properties using our dedicated SQUID magnetometer and PPMS (both with ^{3}He inserts). As part of WP2, more detailed investigation of the nature of disorder and impurities will be carried out in collaboration with ... using high resolution single crystal x-ray diffraction and electron microscopy. d. Characterisation: transport/thermodynamic/magnetic; XRD; TEM

Materials discovery: we will follow up fresh opportunities in targeted searches for altogether new unconventional superconductors. Pressure-assisted high throughput surveys play a central role in these searches, as in previous discoveries (e.g. Figs. 3b-d). Further acceleration is possible by more accurate selection of candidate materials, for which we will increasingly complement heuristic filters by numerical calculations with collaborators (see also WP2).

Objective 7: *Further improve the quality of UTe_2, UAu_2 and UTe_2 crystals by studying the origins of disorder in these material systems, and grow superior crystals for studies of superconducting and normal states. Grow related systems and substitution series to map out composition phase diagrams.*

Objective 8: *Explore new superconducting quantum materials in pressure-assisted high-throughput surveys guided by heuristic and – increasingly – computational filters (also WP 2).*

WP4: Instrumentation and techniques

We will develop novel instrumentation needed for many of the studies listed above, which largely results from combining a diverse range of probing experiments with tuning parameters such as pressure, strain or magnetic field.

Low-T magnetometry under pressure

Strain experiments in high magnetic fields

AC calorimetry into 100kbar range Note success in piston-cylinder cells [17].

Ultrasound will be used to measure sound velocity and sound attenuation. The sound velocity measures different elastic constants, selected by the polarisation and propagation axes. These are thermodynamic quantities, which like the heat capacity are sensitive to phase transitions, providing a reliable method for mapping out phase diagrams as a function of temperature, pressure and field. The attenuation (at low temperature) measures the electronic density of states but is also sensitive to defects in particular the coupling of latter to different order parameters. The different sources of attenuation can be distinguished by measuring at different sound frequencies. Ultrasound was one of the key experimental probes that validated the BCS theory as described in the original 1957 paper. The version we employ takes this to a new level harnessing advanced ultrafast electronics developed for modern telecommunications. Ultrasound is particularly sensitive for detecting CDW order as well as SC.

A planned extension of this work will be to look for acoustic quantum oscillations at high magnetic field [18–20]. Such oscillations are much less susceptible to harmonic mixing and will help confirm the interpretation of the quantum interference described in WP 2.

Since ultrasound is a directional probe it is ideally suited to determining the presence of order parameter nodes along different crystal directions to determine the symmetry of the order parameter.

Methodology: The pulse-echo measurement technique will be used since it works in both piston cylinder and anvil pressure cells (see below), allows measurements on the same sample at different frequencies and is more easily interpreted than resonance techniques.

In our realisation of this technique 1mm disk LiNBO3 transducers are excited with a short (sub microsecond) pulsees of sound at a harmonic overtone of the transducer f>100Mhz. The transducer both generates the sound in the sample and is then switched to listen to the echoes of sound which is successively reflects back-and-forth through the sample. The captured echoes allow very precise measurement of changes of velocity (with ppm resolution) and of the attenuation.

We have refined this set up to successfully measure U6Fe under pressure, covering both the CDW and superconducting states (figure). In this case the single crystals are 3mm long and the transducer is attached directly to a polished sample. For UTe2 the current best crystals have dimensions of less than 1mm. To measure sub mm crystals we use a sapphire buffer rod to temporarily separate the detection and generation of sound. We have tested this successfully with a sound frequency of 353 MHz on UTe2. For Indium we have also demonstrated that we can measure the change in attenuation due to superconductivity in samples as thin as 20 microns (figure inset); this demonstrates that the method can be used on small samples in a diamond or sapphire anvil cell.

We will study different quality crystals of UTe2. The work will also be expanded to look at other compounds synthesisised in WPxxx.

Miniature piston-cylinder and anvil cells for rotation studies

Plan of work, management, risks

The experimental, theoretical and computational expertise of numerous UK and international partners complements our strengths in materials growth, exploration and discovery as well as high pressure, high magnetic field measurements.

Plan of work: the attached chart outlines the project schedule, which is organised along the three work packages

Management: Selection of materials, contingency planning and new opportunities will be decided during weekly group meetings or, in case of urgency, at additional impromptu meetings. Collaborative work with multiple project partners can carry on in parallel and will be coordinated via long-distance communications. Visits to collaborating groups will be prepared by the investigators concerned and finalised in the weekly meetings.

Risks and rewards: we have carefully considered the risks and rewards of our ambitious proposal and conclude that they are adequately balanced. Risks are mitigated by (i) the spread of projects, which range from immediately achievable to extremely challenging, (ii) our combined experience over many years of research and the state-of-the-art capabilities of our facilities, (iii) the large and expanding pool of materials that can be investigated, (iv) the great diversity of quantum phenomena of theoretical and practical interest that are expected to arise beyond those discussed above.

National importance

Societal and economic impact: the superhydride discoveries show that the technological benefits of superconductivity are not fundamentally limited to low temperatures. New superconducting materials with superior properties, be it transition temperature, critical magnetic field, metallurgy or cost, can unlock transformative impact, often with particular relevance to sustainability or health: (i) powerful magnets already used in MRI scanners, fusion research (ITER and private enterprises Commonwealth Fusion Systems and Tokamak Energy), and accelerators (LHC), requiring thousands of tons of high critical field superconducting wire; (ii) lightweight generators already used in wind turbines and motors/generators now examined for use in airplanes; (iii) radio-frequency and microwave devices such as exceedingly sharp, low-noise filters for base stations of radio communications systems; (iv) ultra-fast, ultra-low-power electronics with applications in communications and computing, where traditional electronics is reaching its performance limits; (v) solid-state based quantum computing such as Google's “quantum supremacy" breakthrough.

The project will prepare the ground for a systematic exploration of new unconventional superconductors, and likely serendipitous discoveries carry the potential for entirely unanticipated new technologies. Further impact arises from the advanced training our graduate students and PDRAs receive in condensed matter physics and methodology. This work contributes to the UK effort in a key scientific area and feeds new materials and techniques as well as skilled problemsolvers and entrepreneurs into our emerging network of high technology instrument makers.

Academic beneficiaries: This project contributes to the strong UK research in quantum materials. It connects with work on cuprate and iron-based superconductors in Bristol and Oxford, uranium-based superconductors and high pressure research in Edinburgh, non-centrosymmetric superconductors, organic superconductors and topological materials in Warwick, ruthenates and other 2D materials in St. Andrews and Birmingham, and Yb-based superconductors at RHUL, with theory work at Bristol, Oxford, Kent, Loughborough, Birmingham, KCL, RHUL, UCL and Cambridge, and with numerous other quantum materials research initiatives throughout the UK. Motivated by the high scientific and economic impact of quantum materials research, leading industrial nations have invested heavily in this field, notably the USA, China, Japan and the other large European countries. To ensure that the UK can benefit from any breakthroughs and know-how arising, we must push forward with ambitious research programmes which leverage existing strengths. The project falls within the EPSRC research areas *Condensed matter: electronic structure* and *magnetism and magnetic materials* as well as *Superconductivity*, and within the EPSRC themes *Physical Sciences* and *Energy*. It is relevant to the Physics Grand Challenges *Emergence and physics far from equilibrium* and *Quantum physics for new quantum technologies.*

[1] A. P. Drozdov *et al.*, Nature **569**, 528 (2019).
[2] M. Somayazulu *et al.*, Phys. Rev. Lett. **122**, 027001 (2019).
[3] H. Sun *et al.*, Nature (2023).
[4] S. Khim *et al.*, Science **373**, 1012 (2021).
[5] D. Aoki *et al.*, J. Phys. Soc. Jpn. **88**, 043702 (2019).
[6] S. Ran *et al.*, Science **365**, 684 (2019).
[7] P. Monthoux *et al.*, Nature **450**, 1177 (2007).
[8] M. R. Norman, Science **332**, 196 (2011).
[9] A. G. Eaton *et al.*, "Quasi-2D Fermi surface in the anomalous superconductor UTe2," (2023).
[10] Z. Wu *et al.*, "Enhanced triplet superconductivity in next generation ultraclean UTe2," (2023).
[11] T. I. Weinberger *et al.*, "Quantum interference between quasi-2D Fermi surface sheets in UTe2," (2023).
[12] C. Bergemann *et al.*, Advances in Physics **52**, 639 (2003).
[13] H. Kotegawa *et al.*, J. Phys. Soc. Jpn. **80**, 083703 (2011).
[14] J. Baglo *et al.*, Phys. Rev. Lett. **129**, 046402 (2022).
[15] N. D. Mathur *et al.*, Nature **394**, 39 (1998).
[16] S. S. Saxena *et al.*, Nature **406**, 6 (2000).
[17] O. P. Squire *et al.*, Phys. Rev. Lett. **131**, 026001 (2023).
[18] D. Shoenberg, *Magnetic Oscillations in Metals*, 1st ed. (Cambridge University Press, Cambridge, 1984).
[19] M. Yoshizawa *et al.*, Physica B: Condensed Matter **281–282**, 740 (2000).
[20] A. Suslov *et al.*, in *AIP Conf. Proc.*, Vol. 850 (AIP, Orlando, Florida (USA), 2006) pp. 1173–1174