

Nature and origin of unconventional superconductivity in ultra-clean UTe₂

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

Case for support

Uranium-based unconventional superconductors are surprisingly abundant but remain incompletely understood. Foremost among these is the new superconductor UTe₂, 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 UTe₂ now available, this project combines wide-ranging experimental, computational and theoretical studies into the origins of superconductivity and the associated anomalous normal states in UTe₂ and related uranium-based quantum materials.

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 under extreme pressure in supercompressed superhydrides, the discovery of 80 K superconductivity in a pressurised, novel nickelate, and the discovery of field resilient superconducting states in CeRh₂As₂ and UTe₂.

Research and, increasingly, applications move away from conventional superconductors, which are facilitated by a phonon-mediated electronic interaction and turn towards *Unconventional superconductors*, which 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.

Only a few material families are so far known to exhibit superconductivity that is not mediated by lattice deformations, or phonons, alone [? ?]. 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. Among these material families based on, for instance, copper oxide, iron or cerium, *uranium-based* superconductors form one of the most diverse set, not only in terms of the number of materials but also in terms of the variety of phenomena. The wider search for unconventional superconductors with

The new superconductor UTe₂

Uranium-based superconductors make up a large fraction of the overall still limited number of unconventional superconductors (Table). Moreover, this material family is highly diverse in terms of crystal and electronic structure. Superconductivity in U compounds is often found near the threshold of ferromagnetism, but many U superconductors are antiferromagnetic or appear far from the threshold of magnetism.

.

Clean crystals for a clearer view. Long electronic mean free paths are required to establish anisotropic forms of order, such as unconventional superconductivity, and without high quality samples, such discoveries could easily be missed. After the initial discovery, informative investigation methods such as quantum oscillation measurements require high quality samples, and the availability of clean single crystals opens the door to external collaborations (work package 2, below). And while the added complexity introduced by disorder itself produces interesting effects, this hinders the initial understanding of already challenging phenomena.

Research questions

Studying the wide family of U-based unconventional superconductors will address the following key research questions about superconducting quantum materials:

a) Superconducting state: what is the symmetry of the superconducting order parameter? What causes high critical fields, in particular if they far exceed the paramagnetic limit? What is the origin of residual C/T in the low T limit in many samples of unconventional superconductors such as UTe₂, and can this be quantitatively attributed to impurity bound states? Can we explain not only why some materials superconduct but also why other, very similar materials do not? What factors determine the variation of T_c within the same material family?

b) Anomalous ‘normal’ state: can we understand quantitatively the high electronic heat capacity and enhanced quasiparticle mass in U-based heavy fermion compounds, which typically exceeds density functional theory (DFT) values by at least an order of magnitude? A breakdown of the standard model of condensed matter physics, Fermi liquid theory, can be signalled by a sub-quadratic temperature dependence of the electrical resistivity $\rho(T)$ at low temperature. Under-

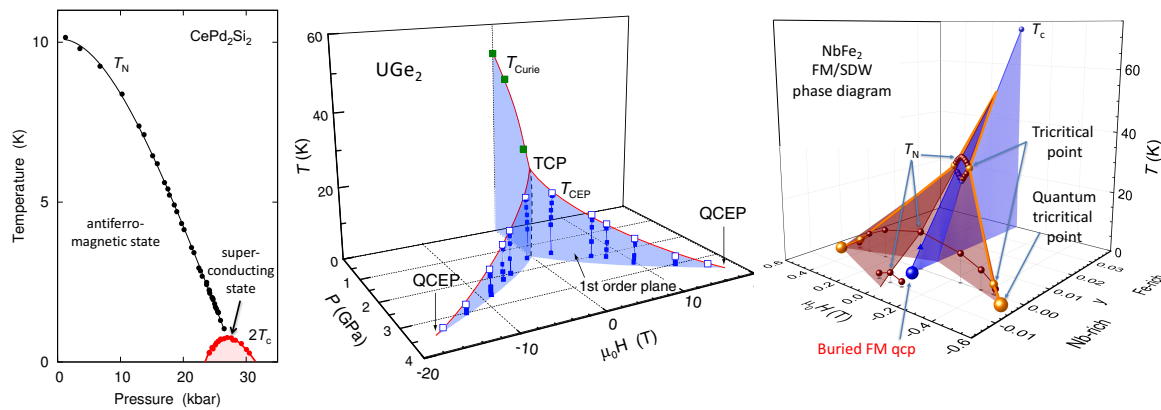


FIG. 1. Template phase diagrams guiding materials exploration: (a) High pressure phase diagram of CePd_2Si_2 , showing a superconducting region attached to the threshold of antiferromagnetism [?]. Considering the added effect of magnetic field adds a third dimension: in UGe_2 (b), superconductivity appears within a ferromagnetic region, which itself branches into two metamagnetic sheets [?]. In PrPtAl or NbFe_2 [?] (c), by contrast, ferromagnetism is replaced by an antiferromagnetic or spin-density wave region.

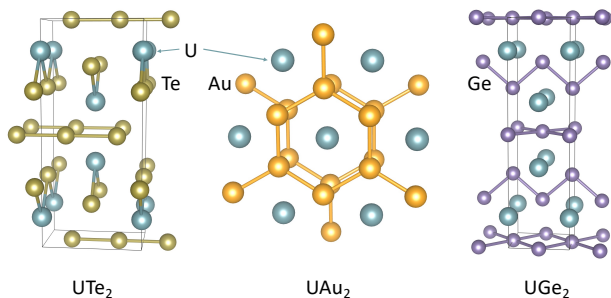


FIG. 2. Material families identified for the first stage of this project. As the programme unfolds, the investigation will widen first to other U-based superconductors (see table) and eventually to d -metal compounds with strong Hund's coupling and hence underlying similarity to the headline materials.

standing this wide-spread phenomenon (e.g. ??, but also other heavy fermion and transition metal compounds such as the cuprates), which often coincides with unconventional or strong-coupling superconductivity, is a fundamental challenge in condensed matter physics. Are signatures of Fermi liquid breakdown confined to the immediate vicinity of quantum critical points? Can they be understood quantitatively in terms of observable low-energy excitations, or soft modes – charge, orbital, nematic, magnetic or vibrational? How do they relate to Planckian dissipation (e.g. [? ?]) as a universal ceiling on scattering rate?

c) Nature and tunability of effective interaction: 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 [?], but different mechanisms are possible. These might involve density,

valence, quadrupolar or orbital degrees of freedom, individually or in combination. How does the form of the effective interaction connect to microscopic models such as the Hubbard model for correlated metals near Mott localisation, the Kondo lattice model for $4f$ -electron heavy fermion superconductors, or the Hund's metal in some of the Fe-based superconductors [?]? Can we understand and control the energy scales that enter these microscopic models, and can we exploit their tunability to vary superconducting and normal state properties?

More ideas:

- Most early hf superconductors were U-based. Maybe because it's actually widespread in U compounds?
- C/T is intermediate between heavy d-metal compounds (e.g. KFe_2As_2 , YFe_2Ge_2) and Ce-compounds
- Local vs. band, orbitally selective Mott transitions? What is the number of itinerant f electrons? Age-old problem of e.g. UPt_3 QO
- On-site correlations, Hund's metal How do we actually pick that up experimentally?
- Role of Spin-orbit coupling?
- FM qcp \rightarrow 1st order or SDW or something else altogether (URu_2Si_2). Lessons from NbFe_2 , PrPtAl
- What determines B_{c2} , and what can we learn from it? And are there ways to maximise B_{c2} ?
- Are there vortex state transitions? Maybe lessons to be learned for applications
- Multicomponent order parameters How do we identify them? How are they manipulated? How can we find new materials that host them?

- Fermi surface instabilities Central e.g. to UGe₂ story. Maybe happens more generally?!
- Nature of hidden order states, as in URu₂Si₂

Selected materials

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

a) UTe₂: using a new generation of ultra-clean crystals grown using the molten salt flux technique by project partners at Charles University, Prague, we were able to detect quantum oscillations with unprecedented clarity, enabling us to resolve the Fermi surface structure of UTe₂ [?]. These high quality crystals with residual resistivity ratio (RRR) of order 500 display a significantly enhanced T_c compared to previous generations of samples. Because disorder is always relevant in unconventional superconductors, many initial findings in UTe₂, starting with the observation of residual Sommerfeld ratio C/T within the superconducting state, need to be re-examined in these new crystals. We have already found that the superconducting critical fields are significantly enhanced, whereas the metamagnetic transition remains unchanged [?]. With at least three distinct superconducting states reported in UTe₂ in a complex field/pressure/temperature phase diagram, these new, cleaner crystals offer the opportunity to clarify many of the open scientific questions surrounding this interesting but complex material.

b) UAu₂: Text about UAu₂

c) UGe₂: Text about UGe₂

Programme and Methodology

The research programme capitalises on our recent breakthroughs in the material systems listed above. The programme is structured into four work packages (WP). WP1 addresses the need to resolve and understand the low temperature ordered states, magnetic, superconducting or otherwise. WP2 studies electronic, magnetic or vibrational excitations. Quantum oscillation experiments probing the electronic 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 a broad range of new instrumentation underpinning all of our studies.

UAu ₂	AFM, s/c under pressure, A. Huxley 21, 22 PNAS, poss
UBe ₁₃	
UCoGe	FM
U ₆ Fe	Perhaps CDW at 110K? Whitley PhD. Looks like interest
UGe ₂	FM
UIr	
UPd ₂ Al ₃	AFM, TN 14K, T _c 2K
UNi ₂ Al ₃	AFM, TN 1.4 K, T _c 1 K
UPt ₃	
URhGe	FM
URu ₂ Si ₂	Hidden order
UTe ₂	dHvA under pressure Strain in high field, to investigate

The planned experiments exploit our expertise and facilities in high precision transport, magnetic and thermodynamic measurements under extreme conditions of hydrostatic pressure (piston-cylinder and anvil cell devices, reaching up to >100 kbar), magnetic field (up to 20.4 T) and low temperature (down to <0.03 K in this project). We will continue to refine and extend experimental methods, with particular emphasis on high pressure temperature modulation calorimetry and quantum oscillation measurements [? ?].

Work package 1 (WP1): ordered states

Spectroscopic studies using neutron diffraction, X-ray diffraction, muon spin rotation or X-ray magnetic circular dichroism to probe magnetic or superconducting ground state properties. These largely facilities-based experiments will be complemented by laboratory-based studies such as scanning tunneling spectroscopy (St. Andrews) and penetration-depth measurements using the tunnel-diode oscillator technique (Edinburgh, with Bristol).

Superconducting states: 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.

Quantum phase transitions and phase diagrams: the power of mapping out magnetic field, pressure and composition phase diagrams is illustrated in ???. The recent example of UTe₂ demonstrates that unexpected twists such as the ultra-high field superconductivity, 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. We will use transport (electrical resistivity, Hall effect), thermodynamic (heat capacity), magnetic (magnetisation, μ SR) and structural (XRD, ultrasound) techniques at applied pressure and in applied fields to survey phase diagrams and clarify outstanding questions in the comparatively new superconductors UTe₂ and UAu₂.

Objective 1: Probe the superconducting states in UTe₂, UAu₂ and UGe₂ with complementary techniques in order to resolve the superconduct-

ing order parameter structure.

Objective 2: Map out pressure and field phase diagrams in next-generation high purity samples of UTe_2 , UAu_2 and UGe_2 to correlate superconducting and normal state properties with magnetic quantum phase transitions.

WP2: Excitations

Electronic excitations: 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. Ambient pressure and high pressure quantum oscillation surveys will be carried out on all four materials systems. Studies on the Cambridge 20.4 Tesla/dilution refrigerator cryomagnet will be augmented by measurements up to 37 Tesla at the HFML Nijmegen facility. Complementary information is provided by Scanning Tunneling Spectroscopy using the quasiparticle interference technique and by ARPES.

Magnetic excitations: neutron scattering studies will map out the magnetic fluctuation spectrum and thereby inform theories for the superconducting pairing mechanism and for normal state heat capacity [?] and transport properties. RIXS?

Theoretical and computational studies: Results arising in WP1 and 2 will feed into work by theorists in the UK and abroad, including project partner XXX listed above. The resulting insights will help refine the filters used to select new candidate materials in collaboration with XXX?.

Non Fermi liquid signatures will be examined using high-precision thermodynamic and transport measurements across pressure, magnetic field and temperature in all four material systems, 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. The absolute scale of the electrical resistivity will be compared to expectations from the hypothesis of Planckian Dissipation, which assumes that scattering rates are limited to a universal ceiling of $k_B T/\hbar$ in strongly correlated materials.

Objective 3: Resolve magnetic, electronic and vibrational excitations by neutron scattering, ARPES, ultra-high field quantum oscillation measurements and Raman spectroscopy.

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

Objective 5: 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.

Objective 6: Resolve the Fermi surface and carrier mass in UTe_2 , UGe_2 and UAu_2 by quantum oscillation surveys, extending later to high pressure and related materials.

WP3: crystal growth and materials discovery

a. MSF b. Induction furnace c. CVT Crystal quality plays a central role in the discovery of new collective phenomena in quantum materials.

Crystal growth: UTe_2 , UAu_2 and UGe_2 will be grown using our carefully optimised ... We will further improve our growth protocols ...

We will widen our programme

1. 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.

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

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

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.

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 3He 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. ??b-d). Further acceleration is possible by more accurate selection of candidate materials, for which we will increasingly com-

plement 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

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 [?].

Ultrasound under pressure

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 inter-

est 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 (ITER), 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 problem-solvers 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*.

[] H. Kotegawa *et al.*, J. Phys. Soc. Jpn. **80**, 083703 (2011)