

Uranium-based quantum materials

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

Part 1: Track record

About the applicants

The applicants have found (UGe_2 , URhGe , UAu_2)

Malte Grosche (FMG) is head of the Quantum Matter group at the Cavendish Laboratory, with a 25 year track record of quantum materials research, which includes all the projects mentioned above. Other recent projects feature quantum oscillation measurements in the pressure-metallised Mott insulator NiS_2 [1, 2] (Fig. 1), and the identification of quantum tricritical points in the band magnet NbFe_2 [3]. His publications have attracted over 5,500 citations. FMG will coordinate the overall management of this project, liaise with project partners, and oversee materials selection, data analysis and dissemination of results.

Andrew Huxley (AH) is ...

Alexander Eaton (AGE) is ...

Siddarth Saxena (SSS) is ...

Peter Wahl (PW) is ...

Gilbert Lonzarich (GGL) is an Emeritus Professor and Fellow of the Royal Society, who has made pioneering contributions in key areas of correlated electron physics. These include (i) quantum oscillation measurements in correlated electron systems, (ii) magnetic fluctuations in metals near the threshold of magnetism and their role in facilitating superconductivity, (iii) quantum phase transitions and quantum critical phenomena. He has been awarded the IOP Mott medal and prize, the HP Europhysics Prize, the IOP Max Born medal, the IOP Guthrie medal, the Royal Society Rumford medal and the Kamerlingh Onnes Prize. His publications, which include twelve in *Science* and *Nature*, have attracted more than 12,500 citations. Recent highlights include studies of the electronic structure of high- T_c superconductors [4, 5] and of quantum critical fluctuations in ferroelectric materials [6, 7]. GGL will lead on the interpretation of results and the computationally assisted search for new superconductors.

Other researchers

Jiasheng Chen (JC) is a postdoctoral researcher, who has been studying superconductivity in YFe_2Ge_2 since its discovery [8]. Having systematically eliminated the main causes of disorder, he produced the first high quality bulk superconducting samples [9] and established a horizontal flux growth method that produces ultra-pure single crystals of YFe_2Ge_2 [10], LuFe_2Ge_2 , and CeNi_2Ge_2 . His transport and heat capacity measurements suggested that superconductivity in YFe_2Ge_2 is unconventional [11], and he has already led preliminary neutron and μSR studies.

Relevant publications by the participating researchers

Discovery	[8, 9, 16–19]
High quality crystal growth	[1, 3, 10, 11]
Tuning by pressure or chemical substitution	[3, 9, 15, 19–21]
Quantum oscillation, transport or thermodynamic measurements	[1, 2, 4, 5, 22–29]
Technical developments	[1, 12–15]

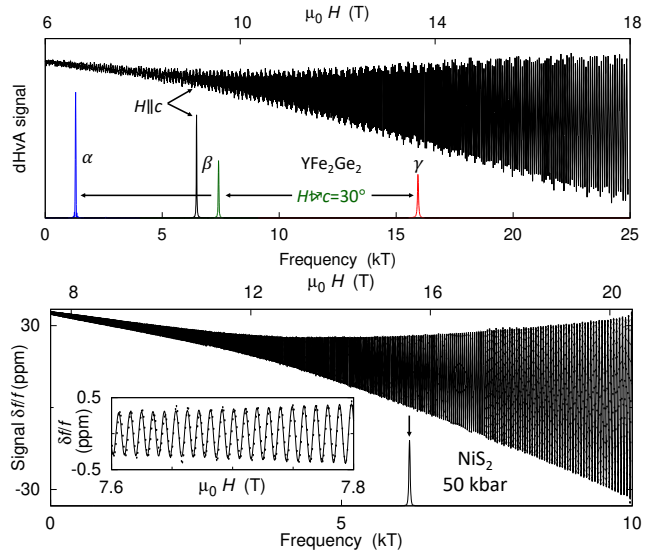


FIG. 1. Recent highlights: The table lists selected publications in relevant areas. The figures show quantum oscillations signals recorded in the unconventional superconductor YFe_2Ge_2 (upper panel) [29] and in the correlated metallic state on the threshold of Mott localisation in high pressure NiS_2 (lower panel) [1, 2], resolving key aspects of the electronic structure and demonstrating the high quality of in-house-grown crystals.

JC will be in charge of crystal growth and characterisation and will take a central role in measurements at large facilities.

Patricia Alireza (PLA) is a senior postdoctoral researcher with 20 years experience in high pressure techniques for low temperature measurements. PLA has pioneered transport and magnetic high pressure methods, which have been taken up widely by the community. These include the introduction of miniature coils into the sample space of anvil pressure cells for susceptibility, skin depth and NMR measurements [1, 2, 12, 13], and the construction of ultra-low-background miniature anvil cells for use in commercial SQUID magnetometers [14, 15], which enabled the detection of the Meissner effect in high pressure hydrogen sulfide by the Erements group. PLA oversees high pressure development and trains incoming graduate students.

Key project partners

Vladimir Sechovsky, Michal Valiska, Charles University Prague, will make available high quality crystals of UTe_2 and other uranium based quantum materials.

Antony Carrington, Sven Friedemann, University of Bristol, will carry out penetration depth measurements using the tunnel-diode oscillator technique at ambient and elevated pressure and pursue transport and Raman measurements to ultra-high pressures.

Devashibhai Adroja, Rutherford Appleton Laboratory, will lead on muon spin rotation and neutron scattering studies of superconducting and magnetic states as well as magnetic excitations.

Research environment and prior work

The programme benefits from substantial prior work (Fig. 1) and from sustained investment in modern research equipment, which includes a newly upgraded 20.4 T/dilution refrigerator high field facility, a 15 T/300 mK cryomagnet, and a 7 T/100 mK cryogen-free demagnetisation cryostat. Experiments demanding still higher magnetic fields will be taken to international facilities, where we have successfully bid for magnet time in the recent past (nine weeks since 2014). Two more weeks of magnet time at HFML Nijmegen have already been granted for work on this project. A 9 T PPMS and a 7 T SQUID magnetometer, both equipped with Helium-3 inserts, are available for sample characterisation and rapid turnover measurements. Crystal growth facilities include two arc furnaces and a mirror furnace as well as numerous box and tube furnaces for flux and vapour transport growth. High-quality crystals of key materials in recent studies, such as NiS_2 and YFe_2Ge_2 , were produced in our group (Fig. 1). Advanced electron-microscopy and x-ray characterisation equipment as well as focused ion beam facilities are available within the Cavendish, and we can access additional growth and characterisation facilities at the new Henry Royce Institute for Materials in Cambridge.

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Part 2: Case for support

New materials are the lifeblood of condensed matter physics. This project combines wide-ranging experimental, computational and theoretical studies into the origins of superconductivity and the associated anomalous normal states in uranium-based quantum materials.

Superconductivity resurgent

- Large volume applications
- Fundamentals
- Materials breakthroughs

One of the most exciting recent developments in condensed matter research has been the demonstration of superconductivity in superhydrides near room temperature but at very high pressure [? ? ?]. The compressed superhydrides demonstrate the power of engineering a phonon-mediated superconducting pairing mechanism towards optimal outcomes. Further gains are possible by widening the scope 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. We urgently need to find new unconventional superconductors: not only are they scientifically interesting – with every case studied, the guiding principles for finding new superconducting material families can be refined. An example of such a guiding principle is illustrated in Fig. 2, namely to home in on the threshold of magnetic order. There, at a so-called quantum phase transition, magnetic excitations reach to low energies. They mediate a long-ranged interaction which can stabilise superconductivity with an unconventional order parameter structure [?]. Such non-phononic pairing interactions are strongly tuneable. This causes superconducting domes which in some cases are surprisingly narrow, explaining why this type of superconductivity is often found not by random searches but by scanning phase diagrams systematically near the border of magnetism.

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 UTe_2 , 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. Understanding this wide-spread phenomenon (e.g. Fig. 3, 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?

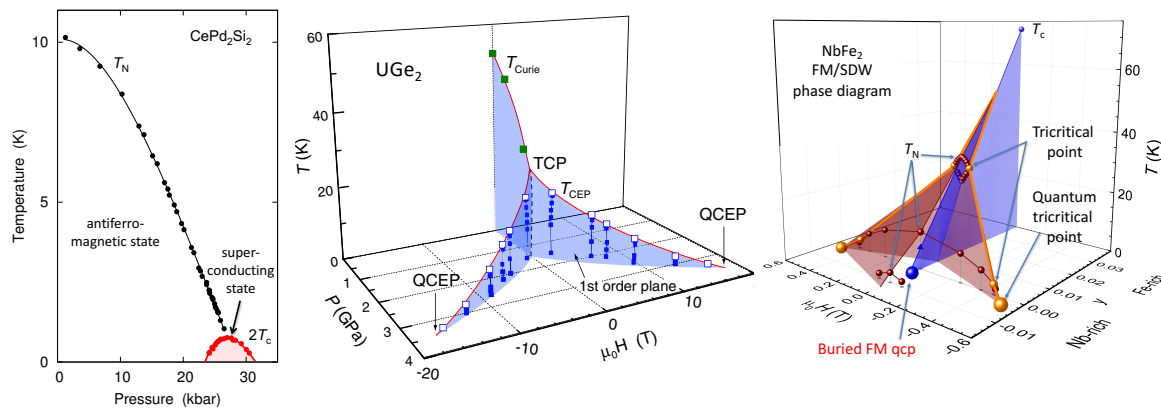


FIG. 2. Template phase diagrams guiding materials exploration: (a) High pressure phase diagram of CePd_2Si_2 , showing a superconducting region attached to the threshold of antiferromagnetism [16]. Considering the added effect of magnetic field adds a third dimension: in UGe_2 (b), superconductivity appears within a ferromagnetic region, which itself branches into two metamagnetic sheets [31]. In PrPtAl or NbFe_2 [3] (c), by contrast, ferromagnetism is replaced by an antiferromagnetic or spin-density wave region.

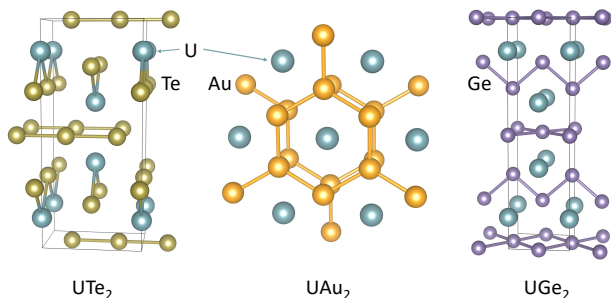


FIG. 3. Material families identified for this project, which combine an anomalous normal state with unconventional or strong-coupling superconductivity. The choice of materials is motivated by our recent discoveries of unconventional superconductivity in YFe_2Ge_2 , LuFe_2Ge_2 and high pressure CeSb_2 , the realisation of the importance of sliding modes in incommensurate host guest phases such as high pressure Bi [30] and Sb, progress in crystal growth, which has for the first time produced high quality crystals of the unconventional superconductor CeNi_2Ge_2 , and advances in high pressure techniques.

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 con-

trol 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_3QO
- 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 Bc_2 , and what can we learn from it? And are there ways to maximise Bc_2 ?
- 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 UGe_2 story. Maybe happens more generally?!
- Nature of hidden order states, as in URu_2Si_2

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 (Fig. 3):

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.

They 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 [1, 2].

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 Fig. 2. 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: *Explore the phase space surrounding YFe₂Ge₂, CeNi₂Ge₂ and CeSb₂ in high pressure and chemical substitution studies. Search for structural instabilities in quasiperiodic high-pressure phases of Bi, Sb and Ba.*

Objective 2: *Probe the superconducting states in all four material systems with complementary techniques in order to resolve the superconducting order parameter structure.*

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. In YFe_2Ge_2 , $\rho(T)$ takes a non-Fermi liquid form at low T , but the observed strong quantum oscillations are interpreted in terms of Fermi liquid quasiparticles [29]. This presents a paradox which invites closer examination using transport measurements and quantum oscillation experiments at low applied fields (see also below). 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 in all four material systems 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 YFe_2Ge_2 , LuFe_2Ge_2 and CeNi_2Ge_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 ...

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 induc-

tion 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. 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: Exploration and Instrumentation

Low-T magnetometry under pressure

Strain experiments

AC calorimetry into 100kbar range

Ultrasound under pressure

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 (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*.

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