Superconducting and normal states in quantum materials

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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.

Mike Sutherland (MLS) is an Affiliated Lecturer and Fellow of Corpus Christi College, having previously held a Royal Society University Research Fellowship. MLS has 20 years experience in precision transport and magnetic measurements at low temperatures and in high magnetic fields (e.g. [4]). Recent highlights include thermal transport studies in the Kondo insulator SmB_6 [5], in non-Fermi liquid materials [6, 7] and in superconductors [8, 9]. MLS will oversee measurements in our 20.4 T cryomagnet facility and coordinate in-house and collaborative thermal transport studies.

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 [10, 11] and of quantum critical fluctuations in ferroelectric materials [12, 13]. 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 [14]. Having systematically eliminated the main causes of disorder, he produced the first high quality bulk superconducting samples [15] and established a horizontal flux growth method that produces ultra-pure single crystals of YFe_2Ge_2 [16], $LuFe_2Ge_2$, and $CeNi_2Ge_2$. His transport and heat capacity measurements suggested that superconductivity in YFe_2Ge_2 is unconventional [17], and he has already led preliminary neutron and μSR studies. JC will be in charge of crystal growth and characterisation and will take a central role in measurements at large facilities.

Puthipong Worasaran (PW) has in late 2021 finished his PhD in our group, during which he demonstrated outstanding expertise in challenging transport and magnetic measurements in anvil-cell devices at hydrostatic pressures exceeding 100 kbar. He will be in charge of most of the high pressure measurements.

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, 18, 19], and the construction of ultra-low-background miniature anvil cells for use in commercial SQUID magnetometers [20, 21], which enabled the detection of the Meissner effect in high pressure hydrogen sulfide by the Eremets group. PLA oversees high pressure development and trains incoming graduate students.

Key project partners

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.

Andrey Chubukov, University of Minnesota, will contribute wide-ranging expertise in numerical and analytic studies of quantum materials and help with interpreting experimental results.

Research environment and prior work

The programme benefits from substantial prior work (Fig. 1) and from sustained investment in

Relevant publications by the participating researchers

Discovery	[14, 15, 22–25]
High quality crystal growth	[1, 3, 16, 17]
Tuning by pressure or chemical substitution	[3, 15, 21, 25–27]
Quantum oscillation, transport or thermodynamic measurements	[1, 2, 4–11, 28, 43]
Technical developments	[1, 18–21]

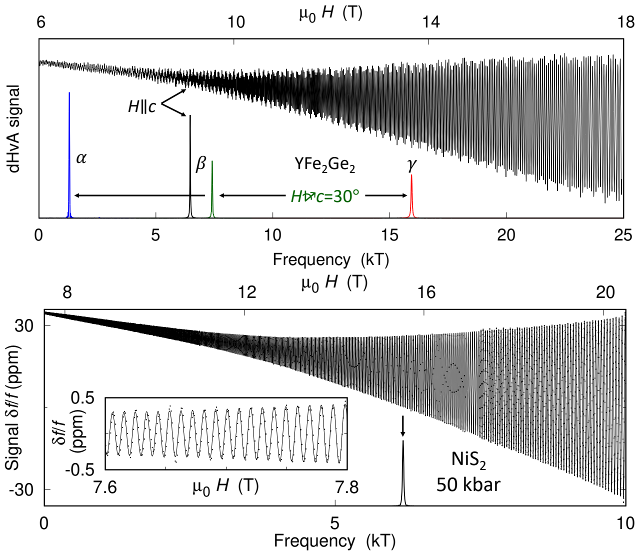

FIG. 1. **Recent highlights:** The table lists selected publications in relevant areas. The figures show quantum oscillations signals recorded in the unconventional superconductor YFe_2Ge_2 (upper panel) [43] and in the correlated metallic state on the threshold of Mott localisation in high pressure NiS_2 (lower panel) [1, 2], resolving key aspects of the electronic structure and demonstrating the high quality of in-house-grown crystals.

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 four novel material systems. Our findings will refine the criteria guiding the search for new superconductors, increasingly targeted for practical applications.

Prospecting for new superconductors

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 [31–33]. 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.

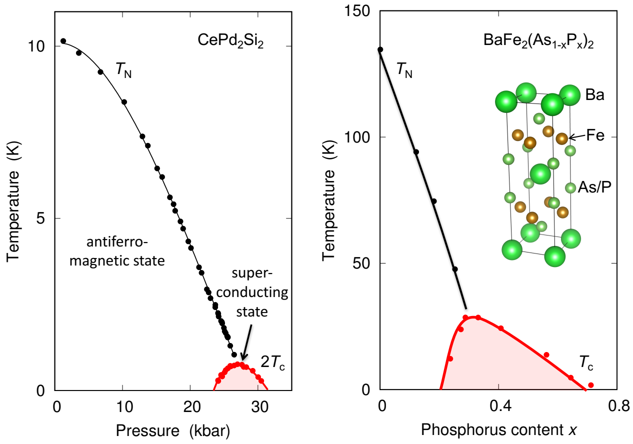

FIG. 2. **Guiding principle:** (left) High pressure phase diagram of CePd$_2$Si$_2$, showing a superconducting region attached to the threshold of antiferromagnetism [22]. In BaFe$_2$As$_2$ (right), superconductivity appears when antiferromagnetic order (and an associated nematic state, not shown) is suppressed under isoelectronic substitution of As by P, or by applied pressure (from [36]). Numerous Ce- and Fe-based superconductors were identified once the search focused on the threshold of magnetism.

Unconventional superconductivity is rare. Only a few material families are so far known to exhibit superconductivity that is not mediated by lattice deformations, or phonons, alone [34, 35]. 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 [34]. 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.

Real materials are complicated. Numerous additional factors – for instance competing interaction channels, disorder, structural transitions, the role of orbital and charge degrees of freedom – require attention. Every such complication also represents a tuning parameter which may be used to advantage: can a combination of vibrational and magnetic excitations be engineered to boost superconductivity, for example? This defines the project plan: to boost the success rate for finding new superconducting quantum materials, we refine existing guiding principles by studying new superconductors and, importantly, also by investigating why very similar reference materials are not superconducting despite prior expectations. The search becomes increasingly precise as more superconductors are uncovered and investigated, ultimately targeting functional materials with superior useful temperature or field range, metallurgical properties, or manufacturing cost for a wide spectrum of applications.

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.

Selected material families

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

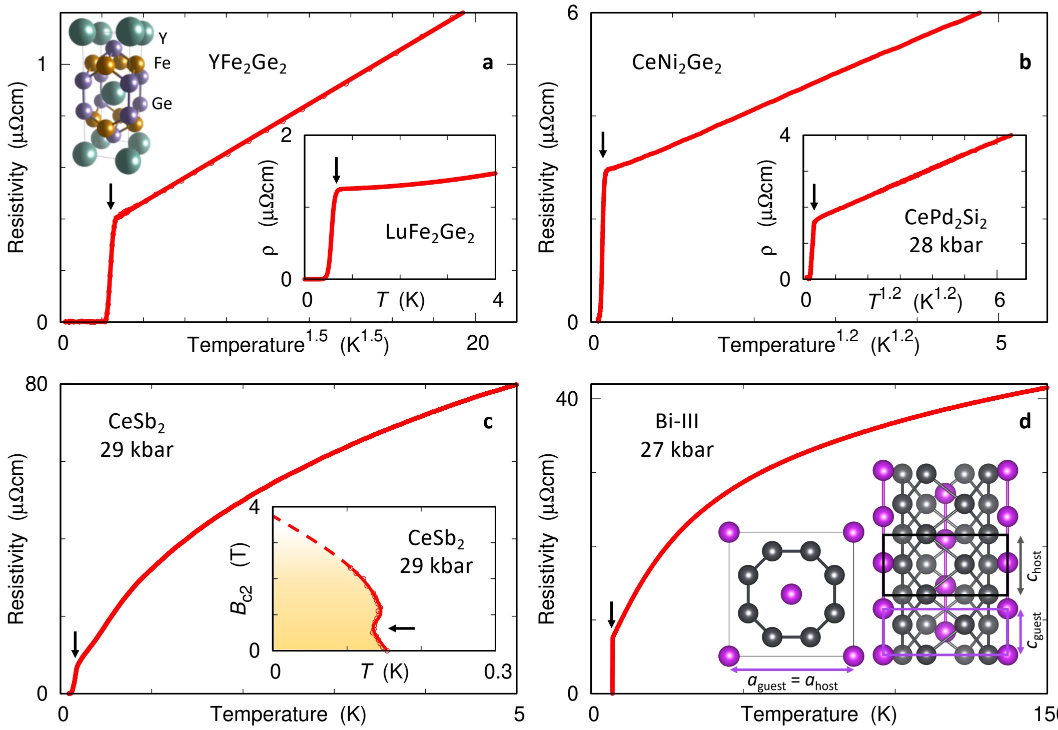

FIG. 3. **Material families** identified for this project, which combine an anomalous normal state with unconventional or strong-coupling superconductivity. The choice of materials is motivated by our recent discoveries of unconventional superconductivity in YFe_2Ge_2, $LuFe_2Ge_2$ and high pressure $CeSb_2$, 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.

a) Iron-based superconductors including the new system YFe_2Ge_2 and its relative $LuFe_2Ge_2$, which straddle an antiferromagnetic quantum phase transition and exhibit an unusually high heat capacity C at low temperature T ($C/T \simeq$ 100 mJ/molK^2), consistent with our observation of carrier mass renormalisation among the highest recorded in transition metal compounds [43]. We have discovered superconductivity in YFe_2Ge_2 [14, 15, 17], emerging out of an anomalous normal state with a $T^{3/2}$ power-law dependence of the resistivity. More recently, we also found superconductivity in the newest generation of high purity crystals of $LuFe_2Ge_2$ (Fig. 3a). These findings depended on our ability to produce ultra-high quality crystals of both materials, with purity levels in YFe_2Ge_2 exceeding those of the best samples grown outside Cambridge tenfold [16].

b) Moderate heavy fermion compounds such as $CeNi_2Ge_2$ and its relative $CePd_2Si_2$, which likewise straddle an antiferromagnetic quantum phase transition (Figs. 2 and 3b) and display superconducting transitions out of an anomalous normal state [22, 24]. Because $CeNi_2Ge_2$ ($C/T \simeq$ 400 mJ/molK^2) forms naturally close to the border of antiferromagnetism [24], it represents an ideal starting point for multi-probe studies in the immediate vicinity of a quantum critical point. New growth methods [16] for the first time deliver high quality crystals of $CeNi_2Ge_2$ of sufficient purity for quantum oscillation measurements.

c) Ultra-heavy fermion superconductors such as compressed $CeSb_2$, in which we have recently discovered superconductivity with a strongly enhanced upper critical field beyond the Pauli paramagnetic limit (Fig. 3c). $CeSb_2$ in its high pressure structure lacks inversion symmetry around the Ce sites, which connects it with recent findings in superconducting $CeRh_2As_2$ [37].

d) Quasiperiodic superconductors such as the pressure-induced incommensurate host-guest structures in Bi, Sb and Ba, which can host a unique low frequency sliding mode because the host and guest sub-lattices cannot lock into mutual alignment. We have discovered signatures of a sliding mode in high-pressure bismuth, Bi-III [30], including strong coupling superconductivity and a linear temperature dependence of the resistivity at low T (Fig. 3d), which is reminiscent of the non-Fermi liquid forms observed in strongly correlated electron systems (e.g. Fig. 3c). Similar aperiodic structures are found in Nowotny chimney-ladder systems [38] and misfit compounds [39], and they share the aperiodic nature of artificial twisted bilayer or multilayer systems.

Research questions

These four material families span a broad range of electronic energy scales, from the low Kelvin regime in high pressure $CeSb_2$ to thousands of Kelvin in Bi, Sb and Ba, with YFe_2Ge_2 and $CeNi_2Ge_2$ connecting the two extremes. Nevertheless, they share striking similarities, such as tunability by pressure or composition and proximity to magnetic or structural instabilities. Above their superconducting transitions, they all exhibit anomalous ‘normal’ states with a sub-quadratic temperature dependence of the electrical resistivity, signalling low-lying excitations. In Bi-III (Fig. 3d), this non-Fermi liquid form of the resistivity may be attributed to low-lying sliding phonon modes, which are a built-in consequence of its quasiperiodic structure. By contrast, magnetic excitations that become long-lived and long-range near the threshold of magnetism likely play the dominant role in the other three material families.

Exploiting these contrasts and similarities, we 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 as in high-pressure $CeSb_2$? What is the origin of residual C/T in the low T limit in unconventional superconductors such as YFe_2Ge_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 heavy fermion compounds such as $CeNi_2Ge_2$, but also in Fe-based systems such as YFe_2Ge_2, in which it exceeds density functional theory (DFT) values by up to 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. [40, 41]) 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 [34], 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 [42]? 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?

Programme and Methodology

The research programme capitalises on the group’s recent breakthroughs in the four material systems listed above. In three work packages, it combines in-house experiments, joint projects with external partners, and materials growth and discovery.

Work package 1 (WP1): in-house studies

These 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].

Quantum phase transitions and phase diagrams: the power of mapping out pressure and composition phase diagrams is illustrated in Fig. 2. We will examine the role of magnetic quantum phase transitions by joint pressure and composition tuning within the space spanned (i) by YFe_2Ge_2/$LuFe_2Ge_2$ and related compounds, such as $LaFe_2Ge_2$, YFe_2Si_2, and $CaFe_2Ge_2$, and (ii) by $CePd_2Si_2$/$CeNi_2Ge_2$. In $CeSb_2$, measurements at high pressure and in high magnetic fields will examine the interplay between superconductivity and a low-lying magnetic transition, and investigate the pressure dependence of the surprisingly high upper critical field, which far exceeds Pauli limiting. In quasiperiodic materials (high-pressure Bi, Sb and Ba), we will scout for structural instabilities such as chain-melting (disordering of one of the sublattices) or the incommensurate-to-commensurate Aubry transition, of which we have seen indications already in high-pressure Sb.

Non Fermi liquid signatures will be surveyed 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 [43]. 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.

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. 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. In YFe_2Ge_2 [43], $LuFe_2Ge_2$ and $CeNi_2Ge_2$, we have already observed de Haas-van Alphen quantum oscillations, extending to fields as low as 3 T in the latest generation of YFe_2Ge_2 crystals (Fig. 1). With further optimisation, quantum oscillations can be tracked to even lower fields, opening up the rare opportunity to investigate a range in which transport measurements suggest non-Fermi liquid behaviour (above).

Objective 1: *Explore the phase space surrounding YFe_2Ge_2, $CeNi_2Ge_2$ and $CeSb_2$ in high pressure and chemical substitution studies. Search for structural instabilities in quasiperiodic high-pressure phases of Bi, Sb and Ba.*

Objective 2: *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 3: *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.*

WP2: collaborative projects

Joint projects with expert project partners have been arranged (see also letters of support), and in most cases work has already begun.

1. Specific heat and dilatometry – Dr. Brando and Prof. Mackenzie (MPI CPfS Dresden, Germany)

2. Thermal conductivity – Prof. Hill (University of Waterloo, Canada)

3. Penetration depth using radio-frequency methods, ultra-high pressure transport and Raman spectroscopy – Profs. Carrington and Friedemann (University of Bristol)

4. Penetration depth using muon spin rotation spectroscopy, magnetic fluctuations using neutron scattering – Dr. Adroja (Rutherford Appleton Laboratory), beamtime awarded at MLZ Munich

5. Angle-resolved photoemission spectroscopy (ARPES) – Prof. Chang (Zurich University, Switzerland), beamtime already awarded at Swiss Light Source

6. Nuclear magnetic resonance – Prof. Ishida (Kyoto University, Japan)

7. Scanning tunneling spectroscopy – Prof. Suderow (Madrid University, Spain)

8. Quantum oscillation measurements at ultra-high magnetic fields – Dr. McCollam (HFML Nijmegen, Netherlands), magnet time already awarded at HFML

9. High resolution single crystal x-ray diffraction and electron microscopy to characterise crystalline disorder and defects – Prof. Grin (MPI CPfS Dresden, Germany)

10. High pressure x-ray diffraction – Dr. Grockowiak (LNLS Campinas, Brazil)

11. Theory of superconducting order parameter structure and anomalous normal state properties – Prof. Chubukov (University of Minnesota, USA)

12. High throughput numerical searches for new superconducting quantum materials – Prof. Pickard, Dr. Monserrat (University of Cambridge)

These projects complement in-house studies listed in WP 1 and address additional topic areas:

Superconducting states: combining a wide range of specialised experimental techniques (listed above) will help resolve the gap structures in YFe_2Ge_2, $LuFe_2Ge_2$, $CeNi_2Ge_2$ and high-pressure $CeSb_2$. 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 [44] and by varying the impurity level.

Excitations: neutron scattering studies will map out the <u>magnetic fluctuation spectrum</u> and thereby inform theories for the superconducting pairing mechanism in YFe_2Ge_2 and $CeNi_2Ge_2$ and for normal state heat capacity [45] and transport properties. Initial studies in YFe_2Ge_2 have already been completed on LET at ISIS/RAL and Thales at ILL Grenoble, and beamtime on PANDA at MLZ Munich has been approved. <u>Electronic excitations</u> will be probed by ARPES (beamtime at SLS awarded) and in quantum oscillation measurements to ultra-high magnetic fields at HFML Nijmegen (two weeks magnet time awarded). The <u>phonon spectrum</u> in quasiperiodic materials will be studied by high pressure Raman spectroscopy.

Theoretical and computational studies: Results arising in WP1 and 2 will feed into work by theorists in the UK and abroad, including project partner Chubukov listed above. The resulting insights will help refine the filters used to select new candidate materials in collaboration with Cambridge colleagues Pickard and Monserrat. A heuristic filter consisting of guiding principles (e.g. proximity to threshold of magnetism, layered materials, bad-metal behaviour in electrical resistivity indicating strong correlations) will be complemented by a computational filter. Without aspiring to an accurate description of unconventional superconductivity, this computational filter will boost the search success rate by combining ab initio calculations of the electronic structure with phenomenological models for the magnetic fluctuation spectrum to examine trends for magnetically mediated superconductivity within Eliashberg theory, as outlined in [34].

Objective 4: *Probe the superconducting states in all four material systems with complementary techniques in order to resolve the superconducting order parameter structure.*

Objective 5: *Resolve magnetic, electronic and vibrational excitations by neutron scattering, ARPES, ultra-high field quantum oscillation measurements and Raman spectroscopy.*

Objective 6: *Develop a theoretical understanding of superconductivity and of anomalous normal state properties in all four material systems.*

WP3: crystal growth and materials discovery

Crystal quality plays a central role in the discovery of new collective phenomena in quantum materials. Bulk superconductivity was only observed in YFe_2Ge_2 after systematic improvements in sample quality [15, 17], culminating in the introduction of a new growth method [16]. The resulting crystals exhibit a residual resistivity ratio $RRR \simeq 650$, an order of magnitude higher than the best values reported outside Cambridge. The same approach can be used for growing superior single crystals of $CeNi_2Ge_2$. Preliminary tests have produced samples with $RRR > 100$, exceeding the quality of the best previously grown $CeNi_2Ge_2$ crystals by at least a factor of five and clean enough to allow us, for the first time, to observe quantum oscillations in this key material.

Crystal growth: YFe_2Ge_2, $LuFe_2Ge_2$, and $CeNi_2Ge_2$ will be grown using our carefully optimised horizontal liquid transport method in a two-zone furnace [16]. We will further improve our flux growth protocol for high-quality crystals of $CeSb_2$, which already achieve $RRR > 100$. The elements Bi, Sb, and Ba for studies of quasiperiodic superconductors are available commercially.

We will widen our programme

1. to other Ce-based Kondo-lattice systems such as $CePd_2Si_2$ (Fig. 2), the ferromagnet $CeAgSb_2$ [46], of which we have recently grown crystals with $RRR > 180$ and the antiferromagnet $CeAl_2$.

2. to other Fe-based intermetallics such as $LaFe_2Ge_2$, YFe_2Si_2, and $CaFe_2Ge_2$ as well as their composition series with YFe_2Ge_2, and

3. to other material families of current interest, including the high pressure superconductor MnP [47] and the Kagomé lattice superconductors (K/Rb/Cs)V_3Sb_5 [48], which have already been grown in our lab, as well as the ruthenate high pressure superconductor Ca_2RuO_4 [49].

When flux growth is not productive, we use cold-crucible arc or 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 ^{3}He inserts). As part of WP2, more detailed investigation of the nature of disorder and impurities will be carried out in collaboration with project partner Juri Grin at MPI-CPfS Dresden, using high resolution single crystal x-ray diffraction and electron microscopy.

Materials discovery: we will follow up fresh opportunities in targeted searches for altogether new unconventional superconductors. For instance, can we expect to hit a quantum critical point in high pressure studies of YFe_2Si_2 or $LaFe_2Ge_2$, mentioned above? Can we extend insights from Fe-based systems to Mn, Ni, Co or Ru-based materials? Can we find relatives to high-pressure $CeSb_2$? 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 YFe_2Ge_2, $CeNi_2Ge_2$ and $CeSb_2$ 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).*

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 (WP 1) in-house measurements (Grosche, Sutherland, Worasaran, Alireza), (WP 2) collaborative measurements (Chen, Grosche) with associated theory (Chubukov) and numerical studies (Monserrat, Pickard), and (WP 3) crystal growth and materials discovery (Chen). Analysis and interpretation accompanying these activities will be coordinated by Grosche and Lonzarich. We will schedule in-house measurements according to urgency and sample availability. We will start with $CeSb_2$ and high pressure Sb-II, to be followed by the iron-based superconductors, then $CeNi_2Ge_2$/$CePd_2Si_2$, and then materials requiring higher pressures, such as Ba-IV or Ca_2RuO_4. Collaborative measurements follow the scheduling of our project partners, some of whom have already initiated exploratory studies.

Management: the core team is located in the same laboratory. 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) radiofrequency and microwave devices such as exceedingly sharp, low-noise filters for base stations of radio communications systems; (iv) ultrafast, 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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