

Science in high magnetic fields: probing and perturbing electrons in novel materials

Paul Goddard, University of Oxford p.goddard@physics.ox.ac.uk

Abstract – High magnetic fields are a powerful tool for exploring the properties of novel magnetic and electronic materials. Here I outline why high fields are so beneficial in this area, describing a number of different magnet technologies and the uses to which they can be put. In particular, I will discuss the quantum oscillation technique and how it is used to map out one of the most enlightening, if abstract, properties of a metallic system, the Fermi surface.

I. INTRODUCTION

The study of materials whose physical properties are dominated by strong electronic interactions is a highly active field of research. These systems often exhibit unexpected phenomena as a result of the precise interplay between their structural, magnetic and metallic degrees of freedom, with the ground state of the material being determined by the delicate balance of the competing interactions. Examples of such materials include high-temperature cuprate and iron-based superconductors, low-dimensional and molecular magnets, organic metals, topological insulators, and multiferroics. As well as the fundamentals of understanding quantum theories of matter, experimental studies of these systems are also significant with regard to the search for the next generation of functional materials.

One of the most powerful methods for revealing the complex behaviour found in materials with strong electronic interactions is the application of high magnetic fields. The reason for this is that magnetic fields couple to both the free, conduction electrons as well as the more localised electrons responsible for magnetism. The application of a magnetic field sets the electrons in motion, upsetting the balance of the interactions and hence perturbing the properties of the material. In this way fields are used to align magnetic moments, start conduction electrons orbiting, shift energy levels, promote excited states, relieve degeneracies and induce new phases of matter. By monitoring the change in physical properties as a result of the perturbation, the nature of the interactions can be deduced. Furthermore, because a magnetic field is directional, the topology of the interactions can also be investigated.

Superconducting magnet technology allows even small laboratories regular access to fields up to around 20 T. The scale of the perturbation produced by the magnetic field (B) is of the order of $g\mu_B B$, where g is the gyromagnetic ratio and μ_B is the Bohr magneton. Comparing energy scales using this expression we find that that 20 T is equivalent to about 30 K. However, many

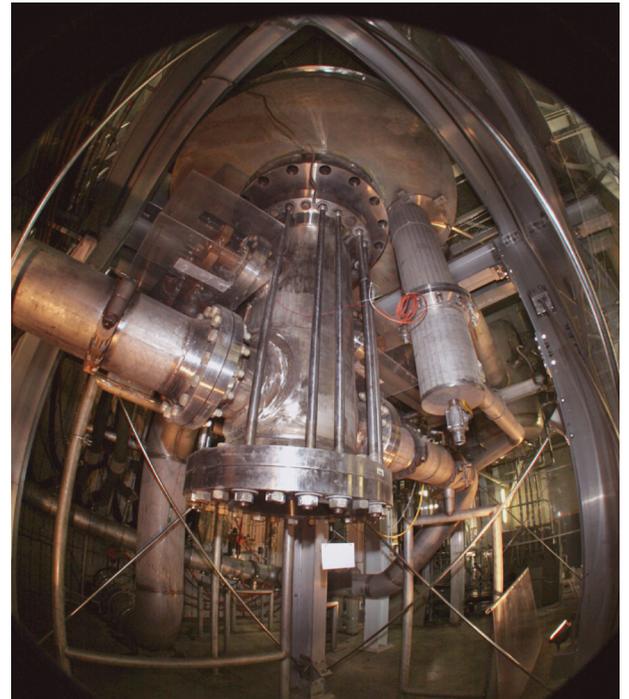


Fig. 1. The 45 T hybrid magnet located at the National High Magnetic Field Laboratory in Tallahassee, Florida [3].

materials of interest have interaction strengths far in excess of this range and so, in order to significantly perturb such systems, one needs access to much higher magnetic fields than are available in typical university science departments or industrial research centres.

II. ULTRA-HIGH FIELDS – A GLOBAL CONCERN

The versatility of ultra-high fields as a research tool means that there are a number of international facilities set up to meet the demands of a worldwide user base, with different laboratories specialising in different magnet technologies. Although all these technologies are essentially based on simple solenoids in which large currents produce large fields, they differ in the way in which they overcome the problems of heating and internal stresses that develop in the magnets themselves.

Large, continuous magnetic fields can be achieved through the use of resistive magnets - giant, water-cooled devices that use megawatts of power to achieve fields of up to 35 T. High field laboratories that provide access to such magnets include those located in Grenoble [1], Nijmegen [2] and Tallahassee [3]. The last is also home to

the world's most powerful continuous-field magnet, the 45 T hybrid magnet, which works via a combination of superconducting and resistive technologies (see Fig. 1).

By limiting the duration of the energising current, pulsed magnets grant access to much higher fields, regularly achieving up to 70 T, but only for a short space of time, typically of the order of milliseconds. Laboratories in Dresden [4], Los Alamos [5], Oxford [6] and Toulouse [7], among others, specialise in magnets of this type with the pulsed currents usually being delivered by high voltage capacitor banks. The world-record for a non-destructive magnet is just over 100 T achieved earlier this year in Los Alamos using a large generator-driven, multi-shot magnet [5].

Ultra-high fields are not generated for their own sake; cutting-edge condensed matter and materials physics experiments are performed year round in the facilities mentioned above, often combining high fields with cryogenic temperatures and applied hydrostatic pressures. Magnetometry, heat capacity, thermal and electronic transport, magneto-optics, magnetic resonance and acoustics are just some of the techniques employed to characterize the properties of new and novel materials in high fields. As mentioned above the perturbation caused by the field can also be used to adjust the balance of electronic interactions. In this way one can: induce new magnetic phases in quantum magnets [8]; close energy gaps in semiconducting nanostructures [9]; enhance quantum fluctuations by tuning phase transitions towards zero temperature [10]; and drive up bulk resistivity to better observe the conductive properties of topologically protected surface states [11]; to name just a few recent examples. Another illustration of why ultra-high fields are required comes courtesy of the high-temperature superconductors. Despite the technological and theoretical significance of these systems, a full understanding of the mechanisms behind this type of superconductivity remains elusive and much current research activity is directed to the problem and high fields are proving indispensable.

III. FERMI SURFING – LOOKING INSIDE METALS AND SUPERCONDUCTORS

A key measurement for understanding a conductive system is a measurement of its Fermi surface - the boundary in momentum space between full and empty electronic energy states. Although this may seem an abstract concept, it is an extremely useful tool for predicting or explaining the material's magnetic, electronic, optical and thermal properties as well as certain structural instabilities. In a solid metallic system the conduction electrons' crystal momentum is quantized into discrete states and, because of the Pauli exclusion principle, only one electron can occupy each one. This leads to the existence of a conceptual three-dimensional boundary in the ground state of the material that separates filled from empty electronic energy levels. In order to make an electron do something, such as carry a current in a particular direction, one needs to give it a little kick, i.e. shift it from one energy state to another. Herein lies the importance of the Fermi surface: in general only those electrons in the proximity of the boundary can make the hop to a nearby empty state, those far below are completely surrounded by full states and so are stuck with nowhere to go. This means that pretty much all the

physical properties of a metal depend crucially on the density of the states, both full and empty, that lie close to the Fermi surface. The precise topology of the Fermi surface is dictated by the symmetry of the crystal structure as well as the number of electrons present in the material. As an example, the Fermi surface of copper is shown in the left-hand side of Fig. 2. This represents the repeating unit of the material's electronic structure in the same way that the unit cell is the repeating entity of crystal structure. A knowledge of their Fermi surfaces can be used to explain why the electronic properties of the elements vary as one moves across the periodic table, not to mention what happens when one starts to construct compounds [12].

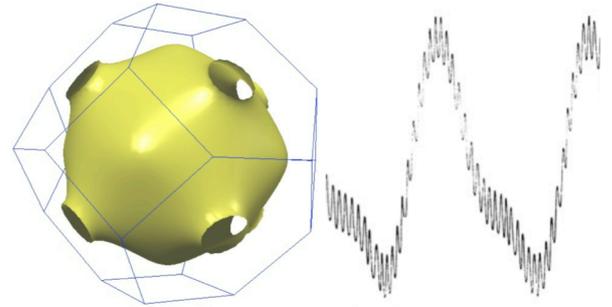


Fig. 2. Left: Fermi surface of copper [13]. Right: Copper quantum oscillations measured in torque magnetometry as a function of magnetic field applied along the [111] direction [14].

Measuring the Fermi surface is thus of great interest to materials scientists. Angle-resolved photoemission spectroscopy is a commonly used surface technique, but by far the best established method for mapping out the Fermi surface in the bulk of a material is to cool the material to liquid helium temperatures, apply a magnetic field and measure the resulting magnetic quantum oscillations. The applied field drives the charge carriers into orbital motion leading to another quantization, this time of the angular momentum, on top of that already imposed on the linear momentum by the crystal structure. At certain values of the magnetic field – periodically spaced in B^{-1} – the demands of the two quantization conditions converge at the Fermi surface, at which point there is a peak in the density of available energy states there. The result is that all those properties of the material that depend on this density-of-states oscillate as a function of the field. This effect, known as magnetic quantum oscillations, is observed typically in resistivity or magnetization, but can also be seen in heat capacity, sound velocity, elastic constants, among others. The ability of the magnetic field to affect so many of the physical properties of the material is a powerful demonstration of how the electrons and electronic interactions have a hand in almost every aspect of a metallic system.

In 1952 Lars Onsager predicted that each frequency of quantum oscillation is proportional to an extremal cross-sectional area of the Fermi surface in the direction perpendicular to the applied magnetic field. This result is illustrated in right-hand side of Fig. 2, which shows the quantum oscillations observed in the resistivity of copper as a function of applied field pointing along the [111] crystallographic direction. Two distinct frequencies are

seen, the smaller one corresponding to the small extremal cross-section near the thin “neck” of the Fermi surface, and the larger to the fat “belly” in the middle of the diagram on the left. The upshot of all this is that by measuring quantum oscillations with the magnetic field pointing in different directions with respect to the crystal axes one can completely map out the Fermi surface of a material. In this way many elemental Fermi surfaces were experimentally determined in the 1950s and 60s and compared to the results of theoretical predictions [15].

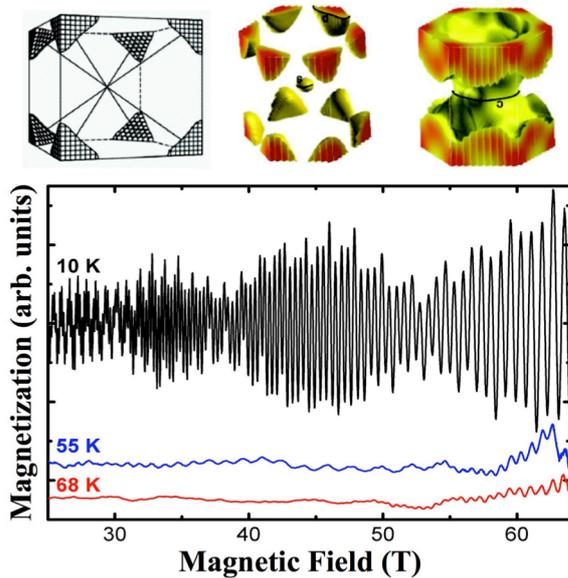


Fig. 3. Bottom: Quantum oscillations in the magnetization of AuZn on either side of the 67 K shape memory transition, measured using pulsed magnetic fields. Top: Fermi surface sections arising from different electronic bands in the high-temperature (black and white) and low-temperature (colour) phase [16].

In modern times interest has shifted from elemental metals and semiconductors to the strongly interacting electron systems described above. In these materials the effect of scattering as well as of the interactions themselves is to slow down the orbiting electrons and reduce the amplitude of the quantum oscillations making them difficult to measure. To counterbalance this problem the oscillation amplitudes grow quickly with increasing field so the solution is to go to higher and higher magnetic fields. Fig. 3 shows the quantum oscillations observed in the shape-memory alloy, AuZn, at temperatures above and below its shape memory transition at 67 K. Typically quantum oscillations can only be seen at liquid helium temperatures where the effects of thermal scattering are minimized, but the use of ultra-high fields, combined with the high mobility of the electrons in this material, enabled their observation up to 100 K. This allowed experimental verification of theoretical predictions for the Fermi surface and led to the suggestion that the shape memory transition, in this material at least, could be driven by an electronic effect [17].

An additional problem arises when trying to measure the electronic properties of the high-temperature superconductors. In this case the zero resistance state itself prevents the measurement of the underlying Fermi surface. To investigate the intrinsic properties of the electrons, and hopefully understand why they are

susceptible to forming the high-temperature

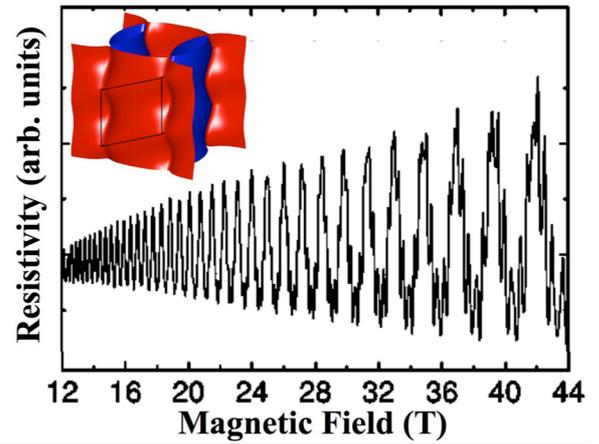


Fig. 4. Quantum oscillations seen in the resistivity of an organic superconductor at 600 mK using the 45 T hybrid magnet in Tallahassee. Inset: The experimentally derived Fermi surface [18].

superconducting state, one needs to suppress the superconductivity with high fields before one can even start looking for quantum oscillations. In the cuprate superconductors this typically means that fields in excess of 40 T must be used and access to one of the international high-field labs is a necessity.

Fig. 4 shows the quantum oscillations observed in the organic superconductor $\text{ET}_2\text{Cu}[\text{NCS}]_2$ along with the experimentally derived Fermi surface. The large slow oscillations are due to the blue quasi-cylindrical section of Fermi surface, while the faster oscillations occur when the magnetic field is high enough to cause the electrons to jump from one section of Fermi surface to another, a phenomenon known as magnetic breakdown [15].

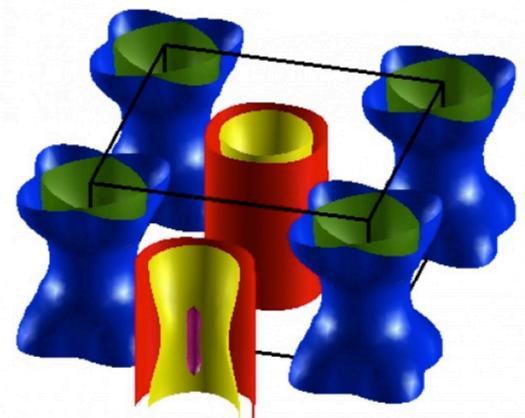
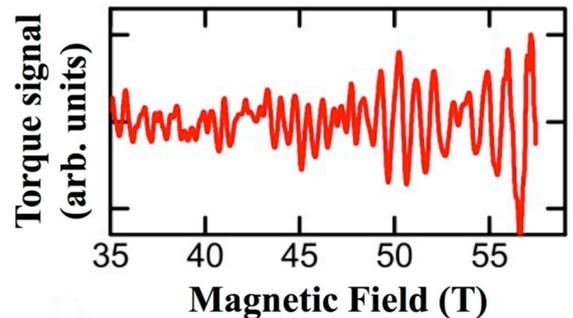


Fig. 5. Top: Quantum oscillations measured via torque magnetometry at 1.5 K in the iron-based superconductor LiFeAs [19]. Bottom: Fermi surface of the same as predicted by band-structure calculations [20].

As another example, Fig. 5 shows the quantum oscillations measured in one of the newly discovered iron-based superconductors, LiFeAs, together with the theoretically predicted Fermi surface. The properties of this family of superconductor are complicated by the fact that several bands can contribute to the Fermi surface, but the experimental results enable complex theoretical models to be ratified. In some cases the Fermi surface is found to indicate an instability, driven by the electrons, but magnetic in nature, that could explain how the superconductivity comes about.

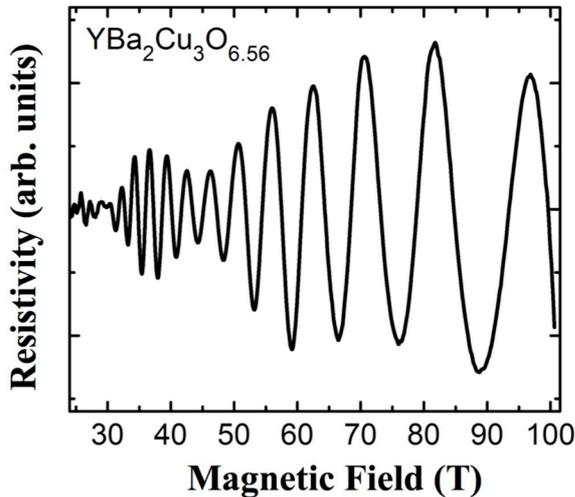


Fig. 6. Quantum oscillations measured in a cuprate superconductor at 1.6 K via a contactless conductivity technique using the 100 T multishot magnet in Los Alamos [21].

Fig. 6 shows the quantum oscillations observed recently in the cuprate superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, notice the very high fields needed to measure these signals. Notice also that no corresponding Fermi surface is displayed. This is because at this time its exact nature remains controversial. It took a long time from the discovery of the cuprate superconductors in 1986 to reach the point where the samples were sufficiently free from scattering that quantum oscillations could be observed [22]. Determining the Fermi surface of this family of superconductor and thereby gaining a better understanding of what makes them tick is very much a topical concern.

IV. CONCLUSIONS

In summary, high magnetic fields continue to be a vital measurement tool in the area of condensed matter physics and materials discovery. As well as a versatile characterization method, the application of high fields in many cases has a perturbative effect, closing energy gaps and inducing new phases. The newest and most interesting materials are frequently those that foster strong electronic interactions, leading to multifunctional properties. For such materials it is often necessary to use magnetic fields of a magnitude that can only be accessed in specialized user facilities. A particular application of high fields is to measure the Fermi surface of conductive systems via the quantum oscillation technique. This

information enables many of the properties of materials of interest to be explained and is currently being used to understand the high-temperature superconducting state.

REFERENCES

- [1] <http://ghmfl.grenoble.cnrs.fr>
- [2] <http://www.ru.nl/hfml/>
- [3] <http://www.magnet.fsu.edu>
- [4] <http://www.hzdr.de>
- [5] <http://www.lanl.gov/orgs/mpa/nhmfl/>
- [6] <http://www2.physics.ox.ac.uk/enterprise/high-field-magnet-facility/nicholas-kurti-magnetic-field-laboratory>
- [7] <http://lncmi.cnrs.fr>
- [8] “A cascade of magnetic field induced spin transitions in LaCoO_3 ” M. M. Altarawneh, *et al.*, *Physical Review Letters* **109**, 037201 (2012)
- [9] “Direct Observation of Band-Gap Closure for a Semiconducting Carbon Nanotube in a Large Parallel Magnetic Field” S. H. Jhang, *et al.*, *Physical Review Letters* **106**, 096802 (2011)
- [10] “Formation of a Nematic Fluid at High Fields in $\text{Sr}_3\text{Ru}_2\text{O}_7$ ” R. A. Borzi, *et al.*, *Science* **315**, 214 (2007)
- [11] “Two-dimensional surface state in the quantum limit of a topological insulator” J. G. Analytis, *et al.*, *Nature Physics* **6**, 960 (2010)
- [12] For a fuller introduction to the Fermi surface see, for example, “Band theory and electronic properties of solids” J. Singleton (OUP, 2001)
- [13] <http://www.phys.ufl.edu/fermisurface/> (This website offers beautiful pictures of the Fermi surfaces of the elements as derived from band-structure theory.)
- [14] “Low-Field de Haas-van Alphen Effect in Copper” A. S. Joseph *et al.*, *Physical Review* **148**, 569 (1966)
- [15] An in-depth treatment of the theory of quantum oscillations is given in “Magnetic oscillations in metals” D. Shoenberg (CUP, 2009)
- [16] “Catastrophic Fermi Surface Reconstruction in the Shape-Memory Alloy AuZn” P. A. Goddard, *et al.* *Physical Review Letters* **94**, 116401 (2005)
- [17] “Fermi surface as a driver for the shape-memory effect in AuZn” R. D. McDonald *et al.* *Journal of Physics: Condensed Matter* **17**, L69 (2005)
- [18] “Angle-dependent magnetoresistance of the layered organic superconductor $\kappa\text{-(ET)}_2\text{Cu(NCS)}_2$: Simulation and experiment” P. A. Goddard *et al.* *Physical Review B* **69**, 174509 (2004)
- [19] “de Haas–van Alphen Study of the Fermi Surfaces of Superconducting LiFeP and LiFeAs” C. Putzke *et al.* *Physical Review Letters* **108**, 047002 (2012)
- [20] Picture courtesy of Amalia Coldea. For more see <http://www2.physics.ox.ac.uk/research/quantum-matter-in-high-magnetic-fields/fermi-surfaces>
- [21] “Quantum Oscillations from Nodal Bilayer Magnetic Breakdown in the Underdoped High Temperature Superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ” S. Sebastian *et al.* *Physical Review Letters* **108**, 196403 (2012).
- [22] “Quantum oscillations and the Fermi surface in an underdoped high- T_c superconductor” N. Doiron-Leyraud *et al.*, *Nature* **447**, 565 (2007)