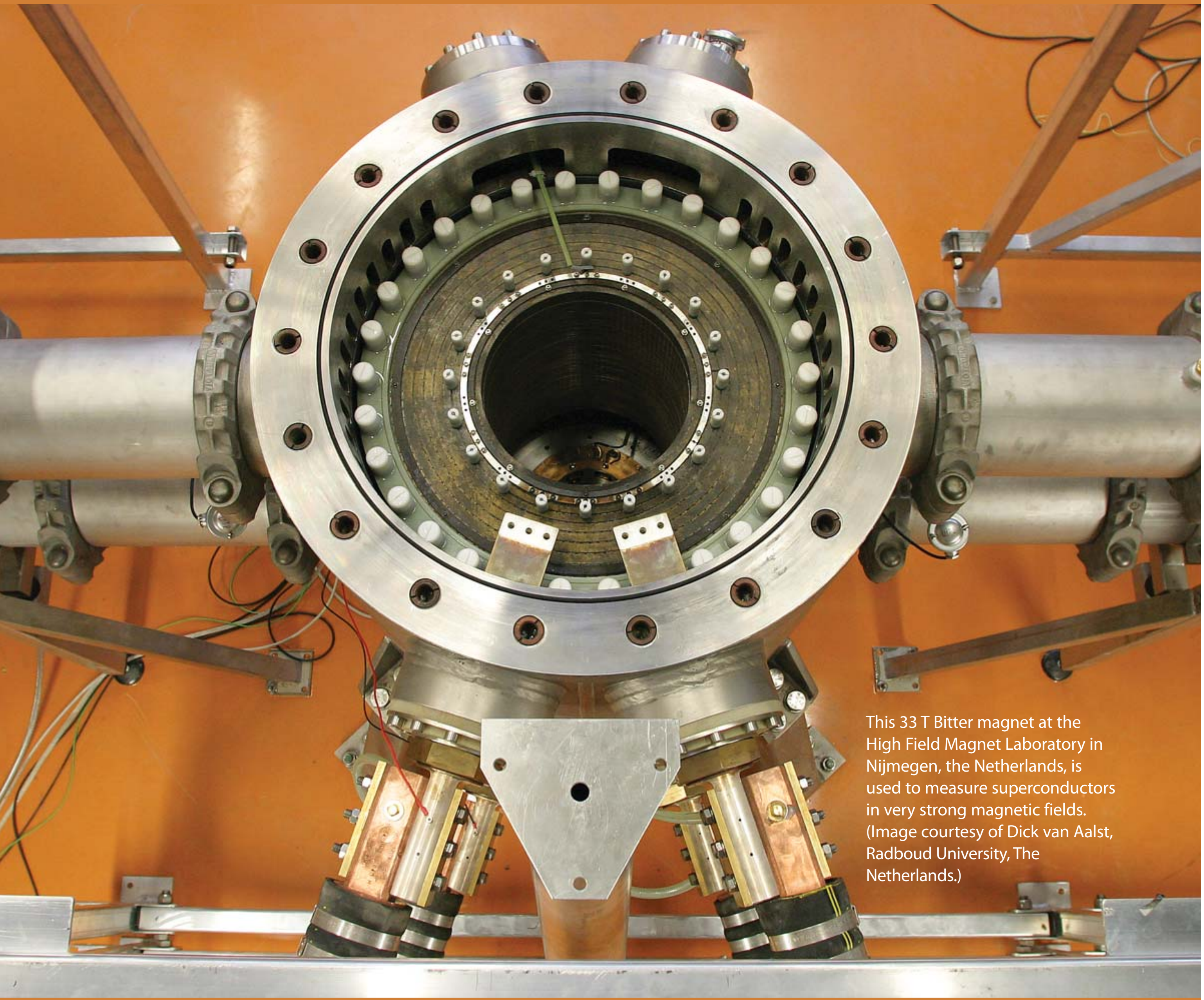
Magnetic field–boosted superconductivity

Anne de Visser

Citation: Physics Today **73**, 11, 44 (2020); doi: 10.1063/PT.3.4616
View online: https://doi.org/10.1063/PT.3.4616
View Table of Contents: https://physicstoday.scitation.org/toc/pto/73/11
Published by the American Institute of Physics



This 33 T Bitter magnet at the High Field Magnet Laboratory in Nijmegen, the Netherlands, is used to measure superconductors in very strong magnetic fields. (Image courtesy of Dick van Aalst, Radboud University, The Netherlands.)

Magnetic field–boosted SUPERCONDUCTIVITY

Anne de Visser is an associate professor at the Van der Waals–Zeeman Institute at the University of Amsterdam in the Netherlands.

Anne de Visser

Although a magnetic field gradually destroys the superconducting state in most materials, a small family of uranium compounds bucks the trend.

During his 1911 discovery of superconductivity Heike Kamerlingh Onnes made a simple observation: The electrical resistance of a metal—mercury in his experiment—dropped to zero below a critical temperature, T_c. (For a historical account, see reference 1 and the article by Dirk van Delft and Peter Kes, PHYSICS TODAY, September 2010, page 38.) Two decades later, in 1933, Walther Meissner and Robert Ochsenfeld discovered a second fundamental property of superconducting materials while they were investigating the magnetic properties of tin and lead. When the samples were cooled below T_c in a small magnetic field, the field was expelled from their interiors.

The Meissner effect is explained by screening currents that flow in a thin surface layer of a superconductor and produce a magnetic field that is directed opposite to the applied field. Therefore, the net magnetic field inside the superconductor is zero. With those two central properties—the complete loss of electrical resistance and the Meissner effect—at its heart, the phenomenon of superconductivity turned out to be a most difficult puzzle in condensed-matter theory. It wasn't until 1957, almost half a century after its discovery, that John Bardeen, Leon Cooper, and J. Robert Schrieffer developed the theory, now known as BCS, that resolved how it works. (See reference 1 and the article by Schrieffer, PHYSICS TODAY, July 1973, page 23.)

In the conventional BCS theory, lattice vibrations, or phonons, create an attractive interaction between electrons and bind them into so-called spin-singlet Cooper pairs. Composed of zero-angular-momentum states with spin-up and spin-down electrons, the pairs collectively lower the ground-state energy of the electron ensemble and form the superconducting condensate. The condensate is what gives rise to dissipationless transport. But the story does not end there.

Unconventional superconductivity

In the first 60 years after Kamerlingh Onnes's discovery, superconductivity research focused on materials that followed BCS behavior. Starting in the 1970s, though, superconductors were found that veered from that behavior.[2] Some of them, such as the Chevrel phases, borocarbides, and heavy-fermion superconductors, had a low T_c; others, such as the fullerenes, pnictides, and cuprates, exhibited a high T_c. More recently, metal hydrides at pressures up to 2 million atmospheres have brought the transition temperature to a record high of 250 K (see the article by Warren Pickett and Mikhail Eremets, PHYSICS TODAY, May 2019, page 52).

Some of those superconductors are genuinely unconventional. For example, in a conventional BCS superconductor below T_c, only the global phase symmetry of the wavefunction is broken; in an unconventional superconductor, spatial symmetry or time-reversal symmetry, or both, is broken as well.[3] The additional types of symmetry breaking allow exotic Cooper-pair states to emerge with finite angular momentum $L = 1$ (*p*-wave superconductivity) or $L = 2$ (*d*-wave superconductivity). Such unconventional superconductors have extraordinary properties: For instance, *p*-wave superconductors can sustain very strong magnetic fields, which is the topic of this article. *D*-wave superconductivity is found in the high-T_c cuprates, whose properties cannot be explained by BCS theory alone. The pairing mechanism is thought to be nonphononic,

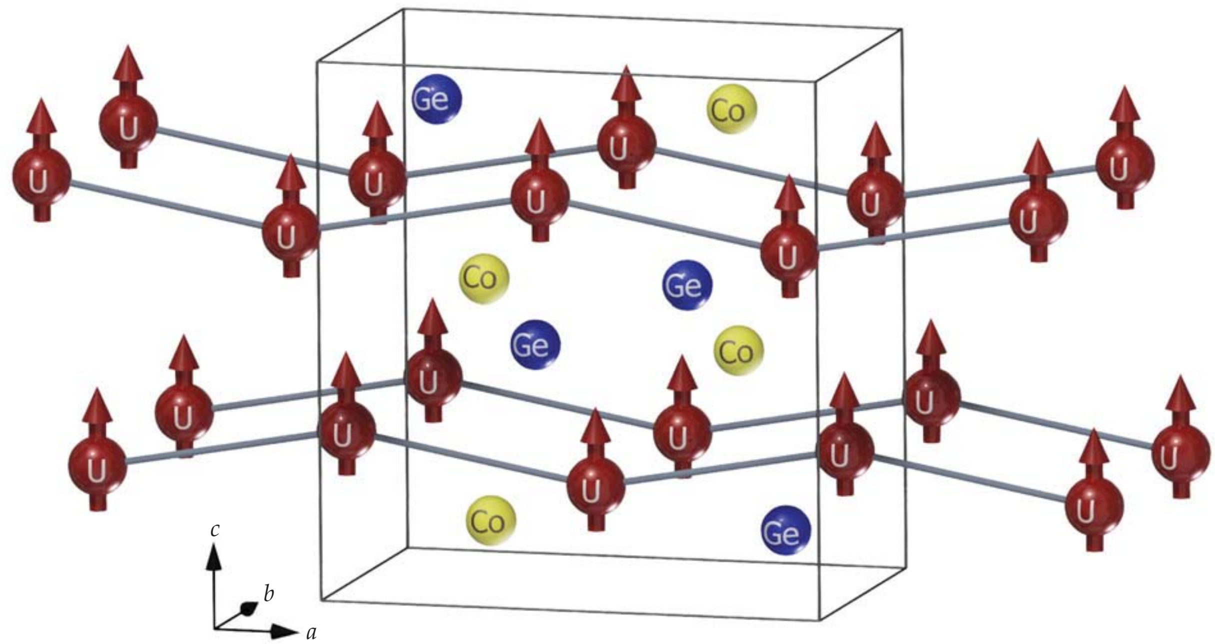


FIGURE 1. THIS VIEW OF THE CRYSTAL STRUCTURE of UCoGe shows the uranium atoms on zigzag chains. Red, yellow, and blue balls represent uranium, cobalt, and germanium atoms, respectively. The magnetic moments indicated by the arrows all point along the *c*-axis of the orthorhombic structure, an orientation that makes the magnetic structure uniaxial. Fluctuations of the moments along that direction are predicted to stimulate *p*-wave superconductivity. (Figure by Udo van Hes.)

and researchers are currently scrutinizing new theoretical scenarios that bear out that expectation.

An exceptional toolbox for probing unconventional superconductivity can be found in a small family of uranium-based metallic ferromagnetic compounds.[4] The first, UGe_2, was discovered by Siddharth Saxena and collaborators at Cambridge University in 2000. Dai Aoki and coworkers at the Atomic Energy Commission in Grenoble, France, reported a second, URhGe, one year later. And the third family member, UCoGe, was discovered by Nguyen Thanh Huy and colleagues at the University of Amsterdam in 2007. In all those compounds, the 5*f* electrons of the uranium atoms carry a magnetic moment. At high temperature, they are paramagnets—that is, the magnetic moments are oriented in random directions. However, they become ferromagnets below the transition temperature T_{Curie} of 53 K for UGe_2, 9.5 K for URhGe, and 3.0 K for UCoGe. Below T_{Curie}, the magnetic moments of each material point in the same direction and produce a net internal magnetic field.

In the BCS model, an internal magnetic field is incompatible with the Meissner effect, and it was long thought that ferromagnetism and superconductivity were competing ground states. The discovery of superconductivity in the three uranium-based ferromagnets below the Curie temperature was therefore unexpected. Those exceptions to the long-standing belief reveal an alternative route to explore in the field of superconductivity. Indeed, recent cutting-edge experiments that use nuclear magnetic resonance techniques[5] and strong magnetic fields[6] demonstrate that the superconducting condensate in those ferromagnets is unconventional.

As I explain in this article, the superconductivity involves spin-triplet Cooper pair states, which in their simplest form consist of two spin-up or two spin-down electrons or a linear combination of them. In a 2019 comprehensive review, Aoki, Kenji Ishida, and Jacques Flouquet collected compelling evidence that those pair states are mediated by quantum critical spin fluctuations rather than by the usual lattice vibrations.[7] Thus superconducting ferromagnets provide a rare case of superconductivity without phonons, an alternative route to superconductivity pioneered by Gilbert Lonzarich at Cambridge University.[8]

A peculiar order

Ferromagnetic order in these compounds has two special features that give rise to superconductivity. First, the magnetic order has a band character.[4] Prime examples of band ferromagnets are simple metals, such as iron, cobalt, and nickel. Band magnetism is caused not by the magnetic moments localized at atoms but by electrons occupying energy bands at the Fermi level. The exchange interaction splits the energy of electron states with different spins, which gives rise to an imbalance in the number of spin-up and spin-down electrons at the Fermi level. That imbalance produces a spontaneous magnetization associated with ferromagnetism.

In the case of band magnetism, it turns out that the Curie temperature and the ordered moment are highly tunable. For instance, in UCoGe, both the ordered moment $m_0 = 0.07\ \mu_B$/U-atom and $T_{Curie} = 3$ K are quite small and can easily be depressed[8] to 0 K by a moderate external pressure of 1.0 GPa. In UGe_2, an applied pressure[4] of 1.6 GPa suffices to reduce T_{Curie} from 53 K to 0 K. And for UTe_2, a recently discovered nearly ferromagnetic superconductor[9] with $T_c = 1.6$ K, T_{Curie} is already (accidentally) close to zero.

That proximity shows that those uranium-based alloys are close to a magnetic instability, a so-called quantum critical point on the phase diagram where T_{Curie} becomes 0 K. In the vicinity of such a critical point, quantum fluctuations of the order parameter—here, the magnetic moment—control the ground state. The band nature of the ferromagnetic order plays a pivotal role in triggering superconductivity. The energy bands at the Fermi level constitute the conduction bands that are involved in the superconducting state. Hence the same electrons that bring about ferromagnetism also produce superconductivity.

A second special feature of the materials lies in their orthorhombic crystal structure. In UCoGe, shown in figure 1, the U atoms form zigzag chains and their magnetic moments point in the same direction. Such magnetic order is called uniaxial. And its reduced dimensionality supports a special type of fluctuation in the magnetic moments along the moment direction, which favors superconductivity.[7]

An everyday, conventional superconductor, such as niobium, exhibits two distinct phases when exposed to a magnetic field, as shown in figure 2. If the field is small, Meissner currents on the surface screen the field from the material's interior. Above a certain critical field, called the lower critical field B_{c1}, the magnetic field starts to penetrate the superconductor in the form of flux lines, or vortices. Each vortex carries a quantum of flux $\Phi_0 = h/2e$, where e is the charge of an electron and h is Planck's constant. Because the flux lines repel each other, they arrange themselves on a triangular lattice—the well-known Abrikosov lattice—whose lattice constant a_Δ is proportional to $(\Phi_0/B)^{1/2}$. That second superconducting phase is known as the vortex, or mixed, state.

By raising the magnetic field higher still, more vortices penetrate the material at the expense of superconducting regions until a_Δ becomes so small that the vortices eventually touch, at which point superconductivity disappears. That suppression field is called the upper critical field B_{c2}. For niobium, which has a T_c of 9.25 K, $B_{c1} = 0.17$ T and $B_{c2} = 0.40$ T. Measuring the upper critical field provides a way to determine the strength of the superconductor. In the limit where T goes to 0 K, $B_{c2}(0) = \Phi_0/2\pi\xi^2$, where the coherence length ξ is the distance over which the two electrons in a Cooper pair are bound together. A large value of $B_{c2}(0)$ implies a relatively small value of ξ and thus a strongly bound Cooper pair.

In a superconducting ferromagnet well below the Curie temperature, the electrons at the Fermi level team up into Cooper pairs and superconductivity sets in. Upon cooling the material in the absence of an external magnetic field, something peculiar happens: the spontaneous creation of a vortex lattice. The flux lines are produced by the weak internal magnetic field, which is caused by magnetic moments. The existence of such a self-induced vortex lattice implies that the Meissner effect is absent. When an external magnetic field is applied, it penetrates the superconductor in the form of the usual vortex lattice. Thus superconducting ferromagnets only have a mixed state.

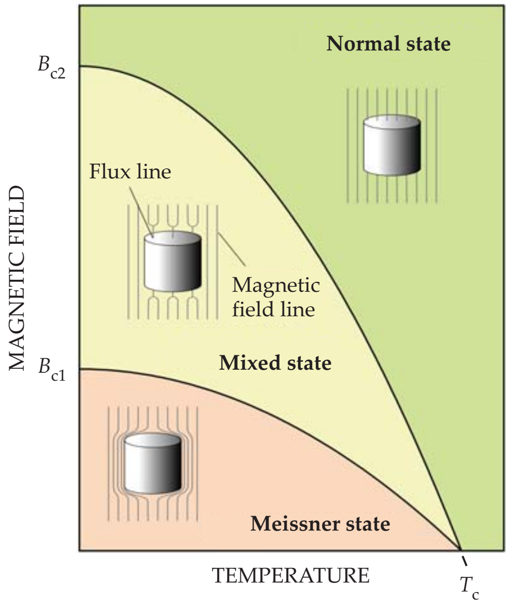


FIGURE 2. THE PHASE DIAGRAM OF A CONVENTIONAL BCS SUPERCONDUCTOR. In the Meissner phase, the magnetic field is expelled from the material's interior. In the mixed (or vortex) phase, the magnetic flux penetrates the material in the form of quantized vortices. In the normal phase, the magnetic field passes through the material uniformly. The fields B_{c1} and B_{c2} are known as the lower critical field and the upper critical field, respectively, and T_c refers to the superconductivity critical temperature. (Figure by Anne de Visser.)

Surprises in large fields

In superconductors, the temperature variation of the upper critical field $B_{c2}(T)$ is routinely measured. Normally, $B_{c2}(T)$ is a smooth, monotonous function that gradually drops to zero at T_c, as shown in figure 2. But in the case of superconducting ferromagnets, measurements of $B_{c2}(T)$ yield a surprise: Superconductivity is revived—that is, strengthened or reinforced by the magnetic field—and may persist up to the highest fields produced in the laboratory (see figure 3). In 2005, Florence Lévy and colleagues[6] found that in URhGe superconductivity is revived at fields between 10 T and 13 T. Four years later, Aoki and coworkers reported an exotic upward slant in the $B_{c2}(T)$ curve in UCoGe.[6] In that alloy, superconductivity strengthens above 6 T (in the form of subtly higher values of T_c) until it is again suppressed at 17 T. And this past year, Georg Knebel[10] and Shen Ran,[11] separately with their coworkers, discovered that T_c in UTe_2 suddenly increases above 16 T and that superconductivity survives up to a spectacularly high 35 T, above which it suddenly disappears.

It's important to note that very strong magnetic fields are required to completely suppress superconductivity. The $B_{c2}(0)$ values reached in figure 3 are much larger than one would expect in a conventional superconductor. To appreciate why, consider how the magnetic field interacts with the electrons of the Cooper pairs in a conventional spin-singlet superconductor. Figure 4 illustrates the situation schematically. The B field acts on the Cooper pair via the electrons' spin and charge. In the first case, presented in figure 4a, the field acts on the antiparallel spins of the electrons via the Zeeman effect. When the field is small, the antiparallel arrangement is unaffected and the spin-singlet state is stable. However, in a strong enough magnetic field, one of the spins flips and both spins then align with the field direction. At that point the spin-singlet Cooper pairs are broken and superconductivity is lost. The phenomenon is dubbed spin pair breaking.

The threshold field B^P where the pairs break is known as the Pauli limiting field,[1] and it's easy to show that $B^P(0) = 1.84 \times T_c$. That rule of thumb predicts the maximum critical magnetic field in which a spin-singlet superconductor may survive once T_c is known. The $B^P(0)$ values for UCoGe, URhGe, and UTe_2 are 0.5 T, 1.1 T, and 2.9 T, respectively, whereas the experimental $B_{c2}(0)$ values are 16 T, 14 T, and 35 T. The upshot is that $B_{c2}(0) > B^P(0)$ implies that the Cooper pairs cannot be of the spin-singlet type, but must instead be of the spin-triplet type. The spins of the two electrons in such Cooper pairs are parallel and cannot be broken by the Zeeman effect.

Besides bringing about spin pair breaking, the magnetic field acts on the momenta of paired electrons via the electron charge. As the magnetic field becomes larger, the resulting Lorentz force will eventually exceed the binding force between the two electrons and break the Cooper pair. That process, illustrated in figure 4b, is termed orbital pair breaking. Since the Lorentz force acts on the charge of the electrons but not their spin, orbital pair breaking will have a similar effect in spin-singlet and spin-triplet superconductors.

For a conventional spin-singlet superconductor, that behavior is well understood and captured by the Werthamer-Helfand-Hohenberg (WHH) model.[12] Its curve is a smooth function of temperature: In an applied field, T_c decreases and gradually drops to zero at the orbital critical field $B_{c2}^{orb}(T)$, as shown in figure 2. For most superconductors, $B_{c2}^{orb}(0) < B^P(0)$ and orbital pair breaking is the main reason that T_c is suppressed by the field.

The initial depression of T_c as a function of magnetic field for the three alloys shown in figure 3 is attributed to orbital pair breaking. But at higher fields, T_c is no longer depressed. Indeed, the revival of superconductivity in URhGe, the unusual upward slant in the B_{c2} curve for UCoGe, and the sudden increase of T_c in UTe_2 are at odds with the WHH model. That's mainly because an important parameter in the model—the electron–phonon coupling parameter λ_{ep}, a measure of the pairing strength—is a constant and does not depend on the magnetic field. But assuming a fixed value for λ_{ep} cannot lead to a revival of superconductivity in strong magnetic fields.

Spin fluctuations

In 2017 Beilun Wu and coworkers proposed an elegant solution to capture the revival of superconductivity.[13] They replaced λ_{ep} in the WHH model with a new field-dependent coupling parameter $\lambda_{sf}(B)$. By letting λ_{sf} increase in the magnetic field, which implies a stronger pairing interaction, T_c is increased. The subscript sf refers to spin fluctuations of the magnetic moments and reflects another important aspect of unconventional superconductivity in ferromagnets: The attractive interaction between electrons is mediated not by phonons but by spin fluctuations.

Magnetically mediated superconductivity has been a challenging research field in past decades, especially in the context of heavy-fermion superconductors and the high-T_c cuprates.[8] The reason is that close to the border of a magnetically ordered phase, the magnetic moments are not static but fluctuate in space and time. Those fluctuations can enhance the spin susceptibility, which also varies. The resulting dynamic, magnetic landscape—on the scale of tens of interatomic spacings—can induce, in special cases, an attractive potential and a binding force between neighboring electrons in the spin-triplet channel. If the interaction is strong enough to defeat the electrons'

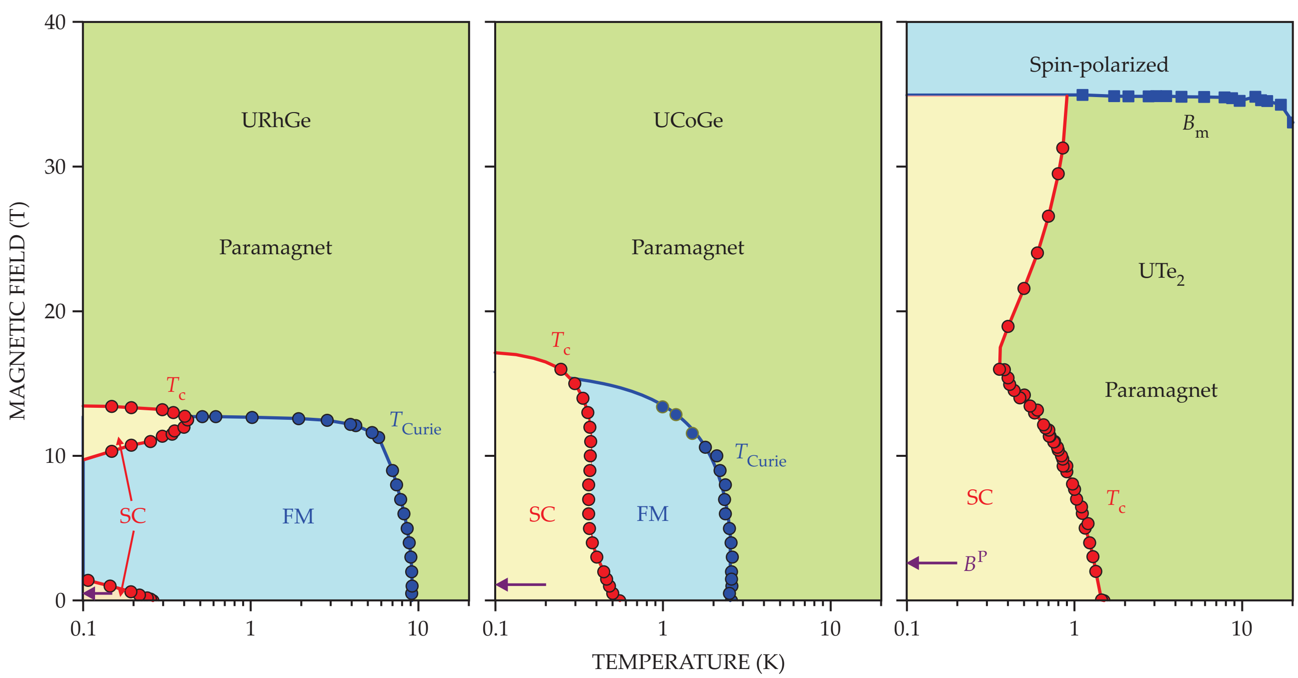


FIGURE 3. THE MAGNETIC FIELD–TEMPERATURE PHASE DIAGRAM OF THREE URANIUM-BASED ALLOYS. In these plots, the superconductivity (SC) phase is shown in yellow, the magnetic normal phase (FM or spin-polarized) is in blue, and the paramagnetic normal phase is in green. In URhGe (left), SC is revived between 10 T and 13 T. In UCoGe (middle) and UTe_2 (right), the superconducting critical temperature T_c (red dots) exhibits a pronounced upturn above 6 T and 16 T, respectively. SC persists for $B > B^P$, the Pauli-limiting field (purple arrows) at which spin-singlet Cooper pairs break apart. For URhGe and UCoGe, SC coexists with FM; the blue dots, which mark the Curie temperature T_{Curie}, delimit the border of the FM phase at which SC is strengthened by the abundance of spin fluctuations. For UTe_2, blue squares mark the transition to the spin-polarized phase above the metamagnetic transition field B_m. In all diagrams, the magnetic field is aligned along the *b*-axis, perpendicular to the direction of the magnetic moments. (Figure adapted from refs. 7 and 16.)

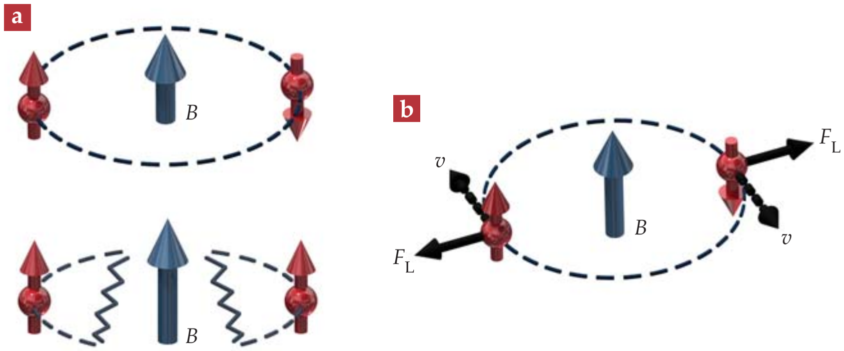


FIGURE 4. COOPER PAIR BREAKING, ILLUSTRATED. **(a)** In a small magnetic field B, the spins of the two electrons (red) in the Cooper pair are antiparallel (top). But in a large magnetic field, the electron spins will align due to the Zeeman effect and the pair breaks apart (bottom), a phenomenon known as spin pair breaking. **(b)** In orbital pair breaking, an applied magnetic field produces a Lorentz force F_L on two electrons with opposite momentum and velocity $\pm v$. When the magnetic field increases, the oppositely directed Lorentz forces eventually exceed the binding force of the electrons and the Cooper pair breaks. (Figure by Udo van Hes.)

Coulomb repulsion, Cooper pairs may form. Keep in mind that superconductivity here is driven by quantum fluctuations of the magnetic moments, and those fluctuations become most pronounced as the temperature approaches 0 K. An appealing way to induce magnetically mediated superconductivity is by tuning the magnetic ordering temperature to 0 K with hydrostatic pressure. That has been achieved[4] in UGe_2 with a modest pressure of 1.6 GPa.

Another way to push the border of a ferromagnetic phase to low temperatures is by field tuning the critical point T_{Curie}, an approach that was successfully achieved in UCoGe and URhGe. As the critical point approaches 0 K, the quantum-critical spin fluctuations at the magnetic phase boundary become more pronounced and revive the superconductivity. In the adapted WHH model, the intensity of the spin fluctuations is captured by the field-dependent coupling parameter λ_{sf}. The field variation of λ_{sf} can also be extracted from the normal-state properties, such as electrical transport and heat capacity. Spin fluctuations, which provide low-energy excitations, give an additional contribution to the low-temperature electronic heat capacity. Measurements of the heat capacity in a magnetic field confirm the direct link between the revived superconductivity and the strength of the ferromagnetic spin fluctuations.[13]

Magnetic-field direction

Magnetic field–boosted superconductivity is one of the most remarkable features of superconducting ferromagnets. But modeling new phenomena on the microscopic level is a great challenge, and no unifying model is yet at hand. For one thing, calculating λ_{sf} is notoriously difficult. All the current models use superconductivity stimulated by critical spin fluctuations at the border of a magnetic phase, but they differ in details.[7] An important ingredient in the models is the low dimensionality of the spin fluctuations along the direction of the magnetic moment (see figure 1). The uniaxial nature of the moments imposes a precise tuning of the magnetic field direction in single crystal samples. For UCoGe and URhGe, that tuning was demonstrated by magnetic-field angle-dependent transport measurements that probed the superconducting transition via electrical resistance.

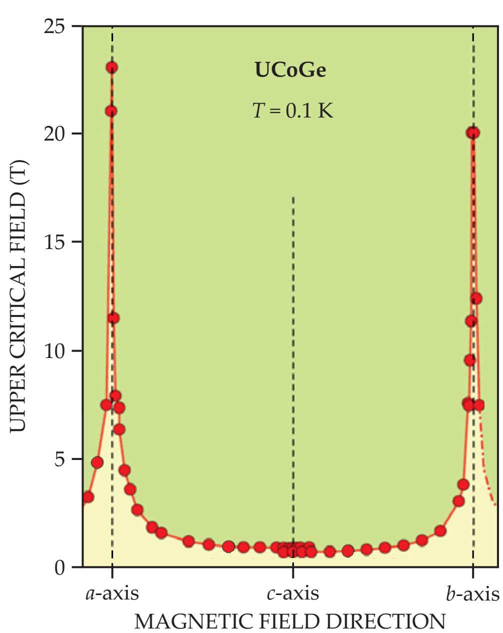


FIGURE 5. ANGULAR VARIATION OF THE UPPER CRITICAL FIELD B_{c2} of UCoGe at low temperature. When the field is aligned with the crystal's *a*- or *b*-axis, B_{c2} exhibits sharp peaks. As soon as the field is rotated toward the *c*-axis, the magnetic field–boosted superconductivity is suppressed. (Courtesy of Dai Aoki, adapted from ref. 14.)

Figure 5 illustrates those results in the case of UCoGe, as measured by Aoki and coworkers.[14] The critical field $B_{c2}(\theta)$ exhibits a sharp peak when the field angle θ aligns with either the *a*- or *b*-axis in the crystal. At either of those orientations, the magnetic field is perpendicular to m_0, which points along the *c*-axis. The strong reduction of B_{c2} when the magnetic field is not exactly aligned along the *a*- or *b*-axis confirms the uniaxial nature of the spin fluctuations, as any small component of the field along m_0 will relentlessly depress those fluctuations and hence superconductivity. For a field along the *c*-axis, $B_{c2}(0)$ amounts to just 1 T. On the microscopic scale, strong support for the key role of those spin fluctuations comes from NMR data.[5] When the field is applied along the *a*- or *b*-axis, pronounced longitudinal spin fluctuations along the *c*-axis stimulate superconductivity. But when the field is rotated toward the *c*-axis, the longitudinal mode is depressed, as is the superconductivity.

The experimental phase diagrams of the uranium-based alloys differ in important details. UCoGe presents the simplest case. The magnetic phase boundary, $T_{Curie}(B)$, bends toward lower temperatures for fields above about 6 T. At the same time, superconductivity becomes stronger—that is, T_c increases, as shown in figure 3. That result is in line with the scenario of spin-fluctuation-mediated superconductivity sketched above. In URhGe, the magnetic phase boundary at low temperatures is due to a rotation of the magnetic moments at a field of 12.7 T. At that field, the accompanying spin fluctuations cause a revival of the superconductivity and a maximum in T_c.

For UTe_2, a different theoretical treatment might be required because long-range ferromagnetic order is absent. And yet a ferromagnet-like spin-polarized phase is induced above the metamagnetic transition field, $B_m = 35$ T. The alloy holds other surprises as well, such as a second revived superconducting phase, reported to exist in the range of 35–65 T for a field directed between the *b*- and *c*-axes,[11] and multiple superconducting phases that were observed under pressure.[15]

All in all, the discovery of the family of superconducting ferromagnets has led to momentous progress in our understanding of unconventional superconductivity, with magnetic field–boosted superconductivity its ultimate litmus test. An obvious question is, Are there any other family members? Researchers are also considering what their strategy should be to unearth new ferromagnetic superconductors. Evidently, the necessary ingredients include band ferromagnetism, uniaxial magnetic moments, and strong spin fluctuations in close proximity to a quantum critical point. Critical transition temperatures reported so far are so low that new experimental tests are needed down to those very low temperatures.

Unraveling the superconducting and magnetic parameters of such complex superconducting materials also necessitates their preparation in high-quality, single-crystal form. So far, all materials in the family contain the element uranium. That apparent requirement restricts the research to dedicated laboratory space. Nonetheless, researchers with new, creative ideas will undoubtedly succeed in adding new superconductors to the toolbox. The unforeseen marriage of superconductivity and ferromagnetism has already produced unprecedented discoveries that have found their place in modern textbooks. Many more are likely on the horizon.

I thank Udo van Hes for preparing figures 1 and 4 and Dai Aoki, Kenji Ishida, Jacques Flouquet, George Knebel, Jean-Pascal Brison, Daniel Braithwaite, Andrew Huxley, and Nick Butch for fruitful discussions.

REFERENCES

1. M. Tinkham, *Introduction to Superconductivity*, McGraw-Hill (1996).
2. Special issue, "Superconducting Materials: Conventional, Unconventional and Undetermined," *Phys. C* **514** (2015).
3. J. Annett, *Superconductivity, Superfluids and Condensates,* Oxford U. Press (2004).
4. S. S. Saxena et al., *Nature* **406**, 587 (2000); D. Aoki et al., *Nature* **413**, 613 (2001); N. T. Huy et al., *Phys. Rev. Lett.* **99**, 067006 (2007).
5. Y. Ihara et al., *Phys. Rev. Lett.* **105**, 206403 (2010); T. Hattori et al., *Phys. Rev. Lett.* **108**, 066403 (2012).
6. F. Lévy et al., *Science* **309**, 1343 (2005); D. Aoki et al., *J. Phys. Soc. Jpn.* **78**, 113709 (2009).
7. D. Aoki, K. Ishida, J. Flouquet, *J. Phys. Soc. Jpn.* **88**, 022001 (2019).
8. P. Monthoux, D. Pines, G. Lonzarich, *Nature* **450**, 1177 (2007).
9. S. Ran et al., *Science* **365**, 684 (2019).
10. G. Knebel et al., *J. Phys. Soc. Jpn.* **88**, 063707 (2019).
11. S. Ran et al., *Nat. Phys.* **15**, 1250 (2019).
12. N. R. Werthamer, E. Helfand, P. C. Hohenberg, *Phys. Rev.* **147**, 295 (1966).
13. B. Wu et al., *Nat. Commun.* **8**, 14480 (2017).
14. D. Aoki, J. Flouquet, *J. Phys. Soc. Jpn.* **83**, 061011 (2014).
15. D. Braithwaite et al., *Commun. Phys.* **2**, 147 (2019).
16. D. Aoki et al., *JPS Conf. Proc.* **30**, 011065 (2020).