

IRON-BASED SUPERCONDUCTORS, seven years later

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Discovery of the new materials raised researchers' hopes of solving the riddle of high-temperature superconductivity. But the unexpectedly diverse systems turned out to possess their own treasure trove of unusual physics that continues to challenge condensed-matter theorists.

How electrons in high-temperature superconductors pair up and conduct electricity without dissipation is one of the most challenging issues in physics—and one of the most exciting. In that respect, the 2008 discovery of high-temperature superconductivity in a class of materials based on iron¹ was among the most significant breakthroughs in condensed-matter research in the past two decades.

Suddenly, in addition to the famous cuprate superconductors, researchers had a second class of materials exhibiting the macroscopic quantum phenomenon of superconductivity at high temperatures (see the article by Charles Day, *PHYSICS TODAY*, August 2009, page 36). The road to room-temperature superconductivity appeared smoother because of the chance to compare the two systems.

In conventional superconductors, such as lead or mercury, electrons Bose condense at temperatures of a few kelvin after binding into Cooper pairs. The polarization of the crystal lattice of positively charged ions provides the attractive force between the two electrons in a pair. The enduring fascination with the cuprates, and now with the iron-based superconductors (FeSCs), isn't just due to their high critical temperatures T_c . They are unconventional materials in which electrons somehow bind into Cooper pairs via the repulsive Coulomb interaction without significant help from the ionic lattice.

Understanding how superconductivity can possibly emerge from Coulomb repulsion is a notoriously difficult task. After nearly 30 years of re-

search, a universally accepted scenario for superconductivity in the cuprates still hasn't emerged. Researchers initially anticipated that the problem might be theoretically more tractable in FeSCs because the screened Coulomb interaction in those materials is generally weaker than in the cuprates. The hope was to find new insights about the pairing mechanism in FeSCs and then apply that knowledge to the cuprates. That idea is still alive. But seven years of collective effort by the condensed-matter community has revealed that the physics of FeSCs is far richer than anticipated and that they display some unique, highly nontrivial properties.

Materials

The large and growing list of FeSCs includes various Fe pnictides and Fe chalcogenides. Pnictogens are elements in group 15 of the periodic table, like arsenic, and chalcogens are elements from group 16, like selenium. Examples of Fe pnictides are so-called 1111 systems $R\text{FeAsO}$ (R represents a rare-earth element), 122 systems XFe_2As_2 (X represents an alkaline earth metal), and 111 systems such as LiFeAs . Examples of Fe chalcogenides are 11 systems FeSe and FeTe and 122 systems $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ (A can be alkali atoms). The crystallographic structures of various families of FeSCs are shown in figure 1. All the structures are layered, and the common units are planes made of Fe atoms, with pnictogen or chalcogen atoms above and below the planes.²

Theoretical band-structure calculations, together with angle-resolved photoemission spectroscopy (ARPES) and other measurements, have successfully established the electronic structures of FeSCs at low energies. Iron's six valence electrons occupy $3d$ orbitals. At least three of those orbitals



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(d_{xy} , d_{yz} , and d_{xz}) contribute to the electronic states near the Fermi surface (FS), and charge carriers hop between Fe sites primarily via a pnictogen or chalcogen ion.

As shown in figure 2, most FeSCs have energy bands that are hole-like near the center of the reciprocal-lattice unit cell (Brillouin zone) and electron-like near the zone boundary. Because electron and hole FSs are small and well separated in momentum space, they are often called hole and electron pockets.

The crystallographic unit cell actually contains two inequivalent Fe positions, so the FSs and Brillouin zone shown in figure 2b should be more properly viewed in a representation with two Fe atoms in a unit cell. However, the Brillouin zone corresponding to one Fe per unit cell used throughout this article allows for a more straightforward discussion without sacrificing the essential physics.

Figure 3 shows the phase diagram of a typical FeSC. The undoped parent compound is usually an antiferromagnet. The magnetic phase of the FeSC is often called a spin-density wave (SDW) to stress that the magnetism is of itinerant electrons rather than of localized electron spins. The superconducting state can be reached by substituting with elements that add holes or electrons (hole or electron doping), by applying pressure, or even by replacing one element with another that has the same valence. There is also another ordered phase, termed nematic, in which the electronic state is believed to spontaneously break the symmetry between the x and y spatial directions without displaying magnetic or superconducting order.

Magnetic and nematic phases

The magnetic, SDW phase is the best understood and least controversial part of the phase diagram of FeSCs. Figure 4a illustrates the magnetic structure of most undoped or weakly doped FeSCs, which is best described as stripe order, with spins aligning ferromagnetically in one direction and antiferromagnetically in the other. Such an order breaks not only spin rotational symmetry but also an additional twofold discrete symmetry, since the stripes align along either x or y . Spin-orbit coupling requires that the

lattice symmetry be simultaneously reduced from tetragonal to orthorhombic. In some doped systems, a small region of magnetic order that preserves tetragonal lattice symmetry has recently been discovered as well.

Both tetragonal-breaking and tetragonal-preserving magnetic orders are consistent with the theory of itinerant magnetism.³ In chromium metal, researchers have known for some time that the presence of hole and electron pockets enhances magnetic fluctuations, and that picture appears to hold for the FeSCs, where the wavevector $\mathbf{Q}$ of the magnetic order connects the Γ - and X - or Y -centered pockets (see figure 2).

Measurements of lattice parameters, DC resistivity, optical conductivity, magnetic susceptibility, and other probes have found that as the temperature is lowered, the stripe SDW order is often preceded by a phase with broken tetragonal structural symmetry but unbroken spin rotational symmetry (see figure 4b). Such a state has been called nematic, by analogy with liquid crystals, to emphasize that the order breaks rotational symmetry but preserves time-reversal and translational symmetry.

The debate about the origin of the nematic phase has been lively. One proposal is that the nematic order is a result of a conventional structural transition caused by phonons. Another possibility is spontaneous orbital order, specifically a difference in the occupation of d_{xz} and d_{yz} orbitals. Yet another is a so-called spin-nematic phase in which magnetic fluctuations along x and y are no longer equivalent but the long-range magnetic stripe order has not yet taken place.

The phonon-driven explanation seems unlikely, since the orthorhombic distortion of the crystal associated with nematic order is too tiny to account for the size of the observed anisotropy in electronic properties. Most researchers believe that nematic order is a spontaneous electronic order due to electron-electron interactions. However, structural order, orbital order, and spin-nematic order all break the same tetragonal symmetry, hence the

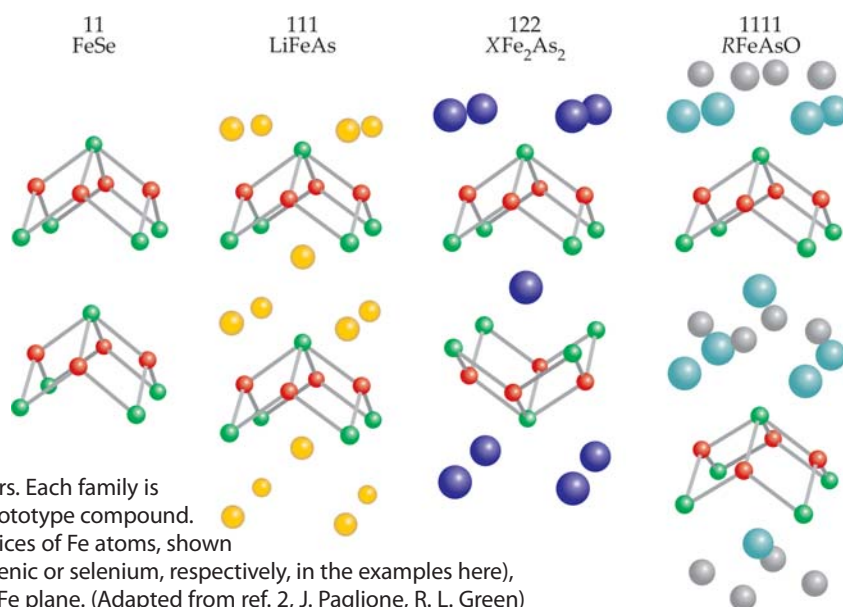
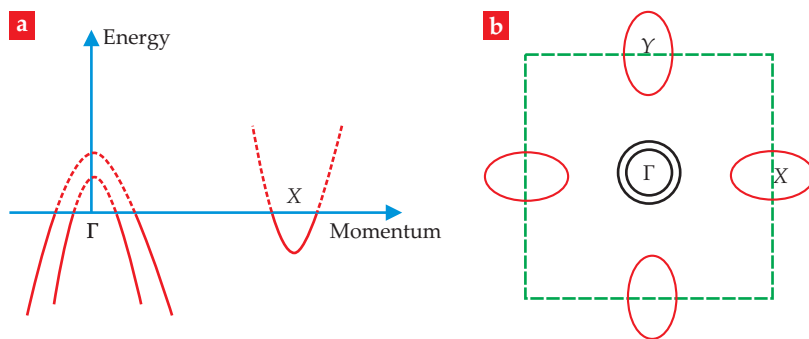


Figure 1. Crystallographic structures of various families of iron-based superconductors. Each family is labeled according to the stoichiometry of its prototype compound. Common to all systems is the set of square lattices of Fe atoms, shown in red, with pnictogen or chalcogen atoms (arsenic or selenium, respectively, in the examples here), shown in green, located above and below the Fe plane. (Adapted from ref. 2, J. Paglione, R. L. Green)

Figure 2. The electronic structure of iron-based superconductors (FeSCs). **(a)** This simplified picture of Fe valence bands illustrates the multiband nature of FeSCs. The momentum axis coincides with the Fermi energy, which separates filled (solid) and empty (dashed) states. Two hole-like bands appear as hills and an electron-like band appears as a valley.

(b) A two-dimensional slice through momentum space reveals the Fermi surface topology. The green dotted lines mark the edges of the reciprocal-lattice unit cell, or Brillouin zone.

At the zone center, labeled Γ , is a hole-like pocket, and at the midpoints of the zone edges, labeled X and Y , are electron-like pockets.



corresponding order parameters are linearly coupled. A spontaneous creation of one triggers the appearance of the other two.

Proponents of the spin-nematic scenario point to the observation that the SDW and nematic transition lines follow each other across all the phase diagrams of 1111 and 122 materials, perhaps even inside the superconducting dome.⁴ On the other hand, in some systems, like FeSe, nematic order emerges when magnetic correlations are still weak, which has fueled speculations that, at least in those systems, nematicity may be due to spontaneous orbital order.

Superconducting phase

Superconductivity with T_c up to nearly 60 K has been detected in 1111 FeSCs, and possibly even 100 K in 11 monolayer systems. The intriguing properties of monolayer FeSe grown on strontium titanate substrates are discussed in box 1. The goal of theorists is to understand the origin of such unusually high critical temperatures, which, for most researchers, is equivalent to understanding how and why electrons bind into Cooper pairs.

In the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity, which successfully describes

many conventional superconductors, two electrons effectively attract each other by emitting and absorbing a phonon. Figure 5a schematically depicts the conventional scenario: One electron polarizes a lattice of positively charged ions, and a second electron is attracted into the same area by the momentary accumulation of positive charge.

But before the second electron can move into that area, it must wait until the first electron is out of the way because of the Coulomb interaction between the two. For that reason, the electron-phonon interaction is said to be retarded in time. The retardation allows electrons to occupy the same point in space, so an isotropic pair wavefunction with *s*-wave symmetry, or zero angular momentum, turns out to be energetically the most favorable. In BCS theory, the pair wavefunction is directly related to a gap function $\Delta(\mathbf{k})$ that is isotropic in momentum space and gives the binding energy of a Cooper pair.

For FeSCs, first-principles studies showed that if superconductivity was phonon mediated, T_c should be around 1 K, much smaller than what is observed. That leaves a nominally repulsive screened Coulomb interaction as the most likely source of the pairing. The unconventional scenario for superconductivity is sketched roughly in figure 5b. The excitations that pair electrons are now those of the electronic medium itself, either spin or charge fluctuations.

Attraction from repulsion

The possibility of superconductivity arising from purely repulsive electron-electron interactions is based on the observation,⁵ first made by several scientists around 1960, that the BCS gap equation for an isotropic gas of interacting electrons decouples into independent equations for each pairing channel characterized by its own angular momentum $l = 0, 1, 2, 3, \dots$ Although the total interaction can be repulsive, components in one or more channels may be attractive.

All it takes is a single attractive channel for the system to undergo a superconducting transition at some nonzero temperature T_c^l . In the electron gas, the screened Coulomb interaction is repulsive at short distances but oscillates at large distances. Walter Kohn and Joaquin Luttinger showed explicitly in 1965 that the angular components of the interaction at large l are attractive.⁶

In a crystalline solid, the FS is not spherical, but one can still identify an orthonormal basis of angu-

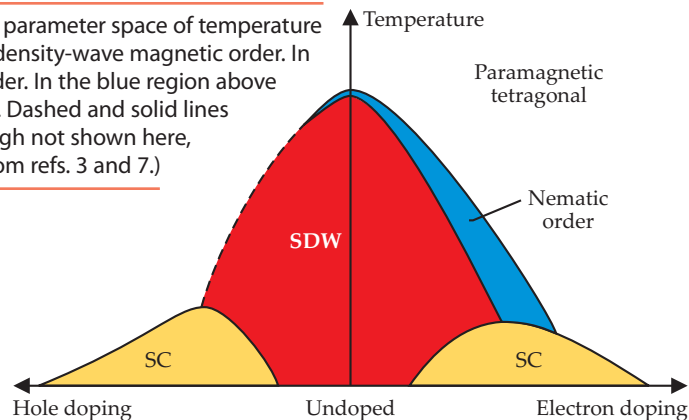
Box 1. Iron selenide monolayers

The most spectacular iron selenide-based material is certainly monolayer FeSe grown epitaxially on strontium titanate,¹⁷ first made by the Tsinghua University group of Qi-Kun Xue in 2012. After careful treatment of the substrate and annealing, that system exhibited signs of superconductivity at very high temperatures. Surprisingly, the two-layer film grown by the same technique was not superconducting at all, which indicated the importance of the active electronic layer's proximity to the substrate.

Whereas zero resistance in the initial monolayer films came only below 35 K (still much higher than the 8 K bulk T_c), angle-resolved photoemission spectroscopy (ARPES) measurements showed a large gap in the electronic spectrum that persisted up to about 65 K. Subsequent refinements have raised the ARPES gap-closing temperature to 75 K, not far from the benchmark temperature of 77 K where nitrogen liquefies. The ARPES measurements indicate that in the monolayers, the band normally responsible for the central hole pocket is located many tens of meV below the Fermi level, similar to alkali-intercalated FeSe systems.

The high-temperature superconductivity in monolayer films and the ARPES results were confirmed recently by Zhi-Xun Shen's group at Stanford University. In addition, when the Xue group performed *in situ* measurements,¹⁸ they found that the resistivity disappeared below 108 K. If confirmed, that would be a clear record for the critical temperature among Fe-based systems.

Figure 3. Schematic phase diagram of iron-based pnictides in the parameter space of temperature and doping. In the red region, labeled SDW, the system has a spin-density-wave magnetic order. In the yellow regions, labeled SC, the system has superconducting order. In the blue region above the SDW phase, the system develops a nematic order (see figure 4). Dashed and solid lines indicate second-order and first-order transitions, respectively. Though not shown here, at small dopings, SC, SDW, and nematic orders coexist. (Adapted from refs. 3 and 7.)



lar functions on a single FS. The crucial feature that allows an unconventional pair state to minimize the repulsive Coulomb interaction is the fact that its gap function changes sign, which usually requires an $l \neq 0$ pair function. For example, $l = 2$ in the cuprates.

Unlike the slow ionic motions that allow for retardation effects in the conventional case, the electronic fluctuations occur on the same time scales as the motions of the electrons they are trying to pair. The electrons partially escape the effects of the repulsive Coulomb interaction by avoiding each other in space rather than time; they tend to form gap functions that are highly anisotropic in momentum and often possess gap nodes—that is, $\Delta(\mathbf{k})$ changes sign at certain positions on the FS. In the cuprates, for example, $\Delta(\mathbf{k})$ vanishes when momentum components are $k_x = \pm k_y$.

In multiband systems like the FeSCs, a new possibility arises. The system can, in principle, retain the symmetry of an s -wave state, but the gap changes sign between the electron and hole FSs (see figures 4c and 5b). As an s -wave state, $\Delta(\mathbf{k})$ is invariant under symmetry operations of the crystal, and thus does not exhibit symmetry imposed nodes. Such a state,^{7–9} called s^{++} , is a multiband analog of the higher-angular-momentum pairings in iso-

tropic, single-band systems, such as $d_{x^2-y^2}$ superconductivity in the cuprates. Thus FeSCs are the first electronically driven s -wave superconductors.

To get s^{++} superconductivity, one needs the repulsion to be stronger between pockets than within them. That requires some additional mechanism, because the usual screened Coulomb interaction is larger within pockets than between them. The most popular scenario is that the enhancement of inter-pocket interactions is due to spin fluctuations (see figure 5b), because the magnetic ordering vector $\mathbf{Q}$ of the SDW state is the same as the momentum connecting hole and electron pockets.

The majority of researchers think that s^{++} is the right symmetry for most of the FeSCs. The most often cited evidence for s^{++} symmetry is the observation in neutron scattering experiments of the so-called resonance peak,¹⁰ which implies that $\Delta(\mathbf{k})$ at the hole pocket has the opposite sign relative to the electron pockets. Nonetheless, two other states have been proposed for at least some FeSCs.

One such state is a conventional s -wave.¹¹ Conventional s -wave superconductivity may be due to phonons, but it also emerges in the electronic scenario if the inter-pocket interaction again dominates over the intrapocket one but is attractive rather than repulsive.

Another alternative is $d_{x^2-y^2}$ superconductivity. Theoretical studies of the pairing show that the interaction in the $d_{x^2-y^2}$ channel is attractive and is comparable in

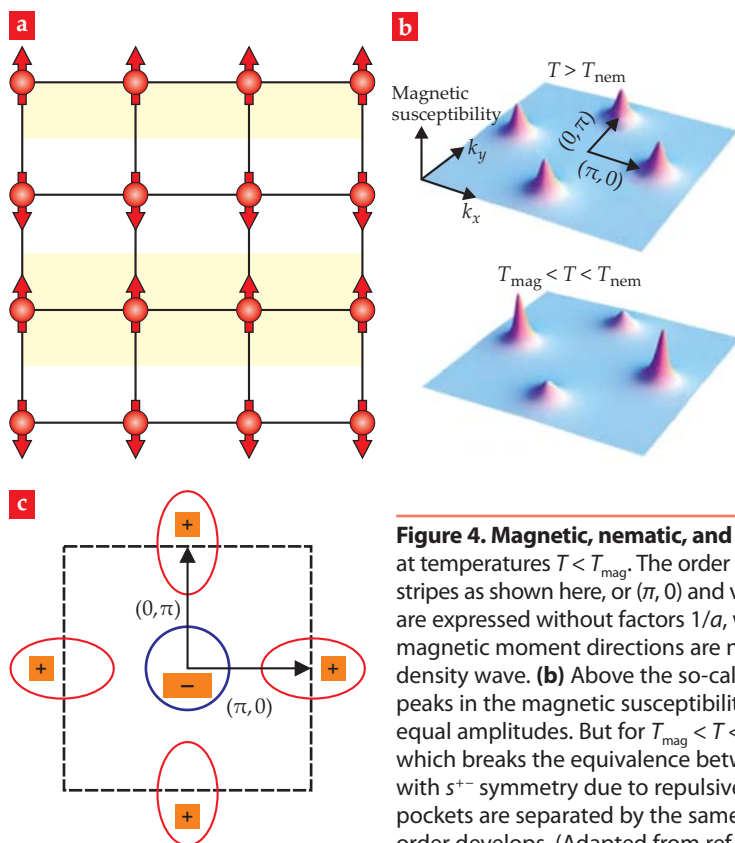


Figure 4. Magnetic, nematic, and superconducting order. (a) Stripe magnetic order develops at temperatures $T < T_{\text{mag}}$. The order is a spin-density wave with momentum $(0, \pi)$ and horizontal stripes as shown here, or $(\pi, 0)$ and vertical stripes. For convenience, positions in reciprocal space are expressed without factors $1/a$, where a is a lattice parameter. The arrows representing magnetic moment directions are not literal but serve to illustrate the periodicity of the spin-density wave. (b) Above the so-called nematic-ordering temperature T_{nem} , the two inelastic peaks in the magnetic susceptibility at positions $(\pi, 0)$ and $(0, \pi)$ in reciprocal space have equal amplitudes. But for $T_{\text{mag}} < T < T_{\text{nem}}$, one of the peaks becomes stronger than the other, which breaks the equivalence between the x and y directions. (c) For superconductivity with s^{++} symmetry due to repulsive interactions between hole and electron pockets, the two pockets are separated by the same momenta $(0, \pi)$ and $(\pi, 0)$, at which spin-density-wave order develops. (Adapted from ref. 4.)

strength to the one in the s^{+-} channel. One rationale for d -wave pairing comes from the consideration of a repulsive interaction between the two electron pockets. If that interaction is somehow enhanced and exceeds other interactions, we again obtain a plus-minus superconductivity, but this time the sign change is between the gaps on the two electron pockets.⁸ By symmetry, such a sign change results in a $d_{x^2-y^2}$ state, since the superconducting gap $\Delta(\mathbf{k})$ changes sign under a $\pi/2$ rotation in momentum space.

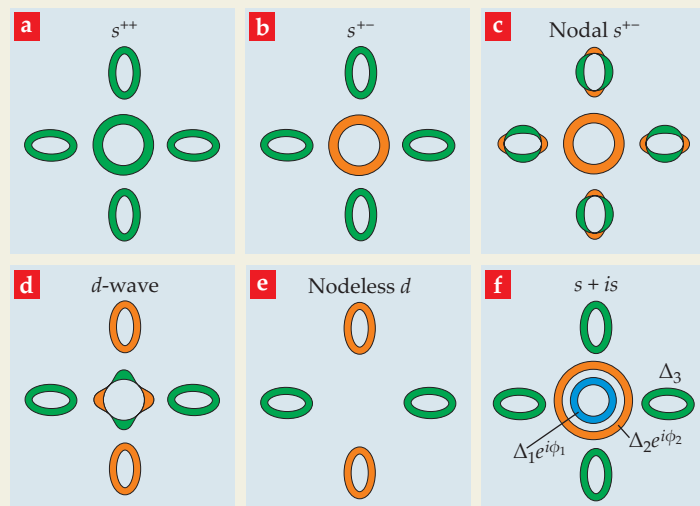
In weakly and moderately doped FeSCs, d -wave superconductivity comes as a close second

behind s^{+} , but it emerges as the leading superconducting instability in strongly electron-doped FeSCs, for which the electron-hole interaction is relatively small. It also has been proposed, for different reasons, for strongly hole-doped FeSCs.¹²

The possibility of observing a change in pairing symmetry in the same material upon doping is another reason why researchers are so excited about FeSCs. Several groups have argued that if the change in pairing symmetry with doping really happens, there must be an intermediate doping regime in which superconductivity has $s + id$ symmetry.¹³ In that case, both s^{+-} and $d_{x^2-y^2}$ gaps would be present, with a $\pm\pi/2$ relative phase between the two. Such a complex state would break time-reversal symmetry and exhibit a wealth of fascinating properties like circulating supercurrents near impurity sites.

Box 2. The s^{+-} state through a microscope

The symmetry of the superconducting gap function $\Delta(\mathbf{k})$ has turned out to be a subtle issue in iron-based superconductors (FeSCs). The figure schematically presents various possible scenarios with colors representing the phase of $\Delta(\mathbf{k})$. The conventional s -wave state (panel a) has a gap with the same sign everywhere on the Fermi surface (FS). The simplest scenario for FeSCs is the s^{+-} state (panel b) in which the gaps on hole and electron FSs are treated as constants and only differ in sign.



Theorists realized early on, however, that because of the multiorbital nature of FeSCs, an s^{+-} gap function on each pocket necessarily has an angular variation that may be quite substantial. In particular, in the one-Fe-per-unit-cell representation of figure 2 of the main text, the angular variation of the gaps on the two electron pockets is $\Delta(\mathbf{k}) = \Delta_e(1 \pm a \cos 2\theta)$, where Δ_e is the gap on the hole pocket, a is a dimensionless parameter, and θ is the angle measured from the x direction. If $|a| > 1$, $\Delta(\mathbf{k})$ has four nodes on each FS (panel c). Such nodes have been called accidental, since their position is not set by symmetry. In contrast, a d -wave gap (panel d), by symmetry, must have its nodes along certain directions in reciprocal space. But if there is no central hole pocket, a d -wave state need not have nodes (panel e). The presence or absence of the nodes is highly relevant, as it completely changes the low-temperature behavior of a system compared with a conventional s -wave superconductor.

An even more subtle issue is the actual structure of the gap function's phase in a generalized s^{+-} state. We considered the case when the phase changes by π between hole and electron pockets, but in multiband systems other cases are possible—for example, a sign change, as in s^{+-} , but now between different hole pockets, or phase differences which are not integer multiples of π (panel f). In the second case, superconducting order breaks time-reversal symmetry and is therefore dubbed $s + is$. (Figure adapted from ref. 8, P. J. Hirschfeld, M. M. Korshunov, I. I. Mazin.)

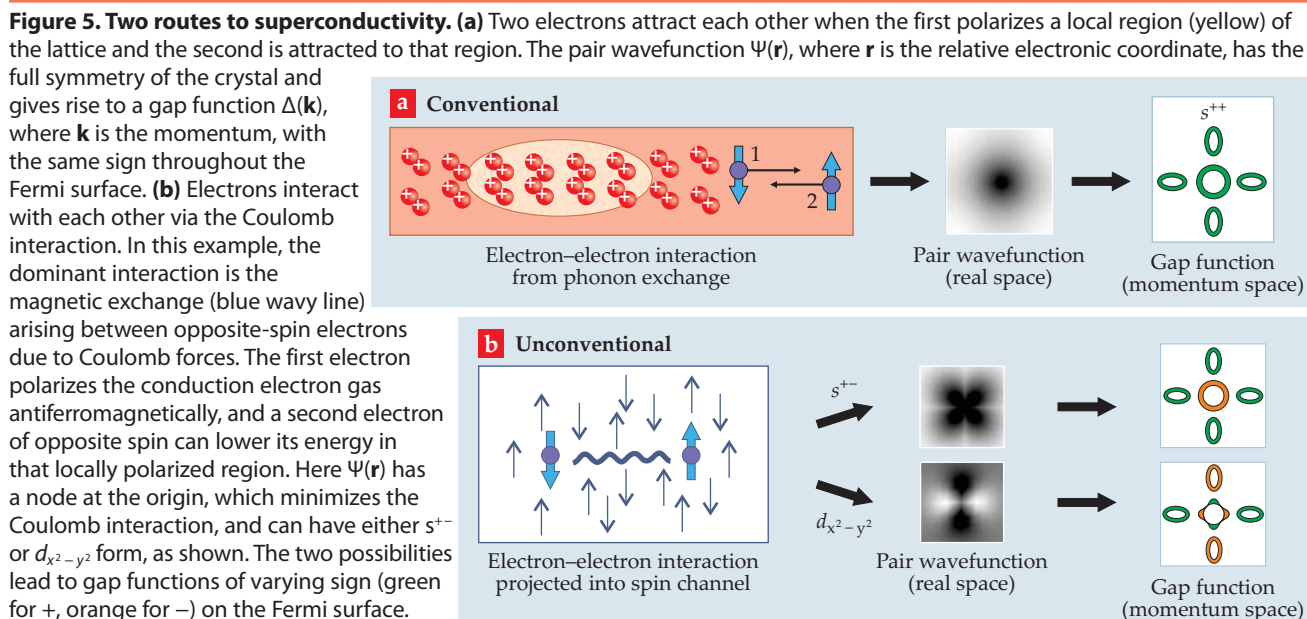
Pnictides versus cuprates

One of the main sources of initial excitement surrounding the FeSCs was the hope that comparing them with the cuprates might lead to a better understanding of the essential ingredients of high- T_c superconductivity. Discovered by Georg Bednorz and Alex Müller in 1986, the cuprate superconductors hold the current record for T_c at over 150 K. The proximity of the superconducting phase to an antiferromagnetically ordered one in both cuprates and FeSCs supported early suggestions that magnetic excitations mediate superconductivity in both cases.

On the other hand, the parent compounds of FeSCs are metals, whereas the parent compounds of the cuprates are invariably Mott insulators, systems in which strong Coulomb interactions localize the electronic states. Whether FeSCs display Mott physics was unclear after the initial discoveries. Many researchers, though not all,¹⁴ thought that good qualitative agreement between band structure calculations¹⁵ and ARPES experiments indicated moderate electron–electron interactions in FeSCs—strong enough to give rise to SDW magnetism and superconductivity at elevated temperatures but not strong enough to localize the electrons.

Two factors have suggested that a reexamination of that idea might be in order. First, density functional theory calculations consistently give bands that are more dispersive than the measured ones. Second, researchers have now created and studied Fe-based materials over a wide doping range—from close to 5.5 d electrons per Fe ion to 7 d electrons per Fe ion. Specific heat data consistently indicate that the electron–electron interaction grows stronger as the number of d electrons decreases toward five per Fe ion, corresponding to a half-filled d shell. An interesting new idea¹⁶ inspired by that observation is a phenomenon called orbital Mott selectivity, in which some orbitals show a stronger tendency to localize than others.

Another issue invites comparisons of FeSCs with cuprates. In both materials, resistivity shows a prominent linear temperature dependence above T_c near optimal doping (where T_c is the highest). There is no theory yet for linear-in- T resistivity down to $T = 0$ K. However, many researchers have attempted to link such behavior in the cuprates to



fluctuation effects associated with a possible quantum critical point, the end point of a phase boundary that is hidden by superconductivity.

The FeSCs may provide a simpler example, as the only two possible quantum critical points currently known in FeSCs are associated with either SDW magnetism or nematicity. Several groups are exploring the idea that fluctuations associated with one of those critical points may finally reveal the origin of linear-in- T resistivity in FeSCs and possibly in the cuprates and other unconventional systems like heavy fermion materials.

New systems, new paradigms?

The leading paradigm for FeSCs— s^{+-} pairing between central hole and outer electron pockets due to repulsive inter-pocket interactions—has recently been challenged in some outlying materials. The FeSCs are famously more variegated than their cuprate cousins, so it is not so easy to decide if those outliers, mostly systems with large hole or electron doping, represent a true challenge. Still, the outliers not only show the largest possible deviations from six d electrons per Fe atom of the parent compounds, but their low-energy electronic structures are quite different from those in weakly to moderately doped FeSCs.

In systems with strong electron doping, like $A_x\text{Fe}_{2-y}\text{Se}_2$, most ARPES data show that hole bands move below the Fermi level and only electron pockets remain. In systems with strong hole doping, like KFe_2As_2 , the opposite happens—the electron band moves above the Fermi level, and only hole pockets remain. In both cases, one of the two types of carriers that were apparently necessary for s^{+-} superconductivity disappears.

Because superconductivity normally involves electrons near the FS, one might expect T_c to disappear, or at least greatly decrease, if one type of FS pocket is removed. Yet that doesn't happen when hole pockets are removed, as evidenced by $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ with $T_c \geq 30$ K. One possibility is that the

interaction between the two electron pockets, one at X and one at Y in figure 2b, is strong enough to produce superconductivity without hole pockets.

The pairing symmetry should then be a d -wave. However, mixing between the pockets could produce exotic variants of the s^{+-} state. A conventional s -wave is also possible if the inter-pocket interaction is attractive (see box 2). Both scenarios fall outside the standard paradigm of s^{+-} superconductivity.

For strongly hole-doped KFe_2As_2 , $T_c \sim 3$ K, which is small and still may be due to the interaction between hole pockets and gapped electron states. Other possibilities are d -wave superconductivity or a different s^{+-} superconductivity due to interactions between electrons solely near hole pockets. If the interaction between hole pockets truly causes superconductivity, we have another example of a pairing mechanism outside the standard paradigm.

Finally, recent experimental results on FeSe have raised the question of whether that simplest of FeSCs is also an outlier. The compound is not magnetic, yet the nematic transition occurs at 80 K. The superconducting T_c is 8 K and is rather small, but it grows to nearly 40 K under pressure and is even higher in monolayer FeSe grown epitaxially on strontium titanate (see box 1).

What's next?

Perhaps the most amazing thing about the FeSCs is the unprecedented richness of the physics. Researchers have found practically all phenomena associated with strongly correlated electron systems in the Fe-based materials, sometimes all within a single family—magnetism, unconventional superconductivity, quantum criticality, linear-in- T resistivity, nematic order, and a tendency toward orbital selective Mottness, to name a few.

In addition, FeSCs, with their multiple FS pockets, are the most likely candidates to show a change in the pairing symmetry upon doping. Therefore, they are also the most likely to develop mixed superconducting order, which breaks time-reversal

symmetry—for example, $s + id$ or $s + is$. Such states have a rich phenomenology and strong potential for applications.

It is likely that the superconducting state in weakly to moderately doped FeSCs has s^{++} symmetry, and magnetic fluctuations are the primary suspects to mediate that kind of pairing. What happens at stronger hole and, particularly, electron doping is an important open question. The high transition temperature found in FeSe films, which apparently only have electron pockets, raises the possibility that the pairing mechanism in those materials may represent a completely new paradigm for superconductivity.

The number of FeSCs keeps growing, and materials with higher T_c and qualitatively new features will likely be found. But the volume of existing experimental data is sufficient to create enough puzzles for the community working on FeSCs and to keep the level of excitement, and the intensity level of the discussions, high for years to come.

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