Iron based superconductors: parent antiferromagnet goes insulating

Recommended and a Commentary by Qimiao Si, Rice University

Superconductivity in iron pnictides and chalcogenides arises by chemical doping of their antiferromagnetic parents compounds. By common usage, a parent compound is one which has a composition such that the nominal valence of Fe is 2^+ with maximum possible magnetic moment of $4\mu_B$/Fe. From the very beginning of this 3-year-old field, the relationship between superconductivity and parent antiferromagnetism has been a primary focus of attention. The materials have a layered structure, with each layer containing a square lattice of Fe atoms. Their electronic properties are primarily associated with the Fe 3d- orbitals; most, if not all, of the five 3d-orbitals are active at low energies. The appropriate microscopic Hamiltonian contains a kinetic-energy part involving both intra- and inter-orbital hoppings, as well as an interaction component featuring Coulomb repulsions and Hund's coupling. For the purpose of our discussion, it is adequate to label the kinetic energy t and interaction U.

A natural way to elucidate the microscopic physics is tuning the parent systems to increase U/t and see whether they become Mott insulating. An earlier study [1] along this direction considered $La_2O_3Fe_2Se_2$. Because the Fe square lattice is expanded compared to the iron pnictides, this parent system possesses a smaller kinetic energy. The enhanced U/t ratio makes the material a Mott insulator, which orders antiferromagnetically. The ordered moment is large, about 2.8 μ_B/Fe [2]. Unfortunately, it is not yet known whether superconductivity arises by carrier-doping this Mott insulator.

The new 122 iron selenides, $A_{1-y}Fe_{2-x}Se_2$, have changed the situation. Guo *et al.*, working on A=K, discovered superconductivity with a T_c above 30 K in this family of compounds. Fang *et al.*, using A=Tl,K, established that the parent compounds are insulating. The existence of antiferromagnetic order was initially suggested by measurements of magnetic susceptibility and has since been established by neutron scattering experiments. These and related works have triggered a flurry of activities over the past few months. A number of important properties have emerged, including:

- The insulating states occur at Fe contents corresponding to $x = 0.4$ and $x = 0.5$. Both have ordered vacancies in the Fe square lattices, such that the Fe valence is +2, with six electrons partially occupying the 3d orbitals at each Fe site, as in the metallic parent of the iron pnictides.

- For $x = 0.4$, elastic neutron scattering measurements [3] have identified its antiferromagnetic order with a large moment, about $3\mu_B$/Fe.

- The Fermi surfaces of the carrier-doped superconducting compounds have only electron pockets, and no hole pockets. This has been established by both ARPES measurements [4] and *ab initio* electronic-structure calculations.

To put things in perspective, we recall that two broadly based microscopic approaches have been taken. One uses the limit of non-interacting electrons as the starting point, with a particular emphasis placed on the Fermi surfaces comprising both electron and hole pockets. Because of the nesting between the electron and hole pockets, relatively weak electron-electron interactions would already produce an antiferromagnetic order in the parent compounds. When doped, the spin fluctuations due to the approximate nesting between the electron and hole pockets are taken as the mechanism for superconductivity because momentum transfer near the antiferromagnetic $\mathbf{Q}$ promotes attractive interactions for scattering of fermions of $\mathbf{k}$ in one band with states of $\mathbf{k} \pm \mathbf{Q}$ in the other band.

The other approach is to consider the iron pnictides as bad metals located not too far away from a Mott transition; in other words, U/t, the ratio between the combined Coulomb and Hund's interactions to the kinetic energy, is smaller than but close to U_c/t, the Mott transition point. The proximity to the Mott transition makes the coherent-electron spectral weight, as probed by, *e.g.*, the Drude component of the optical conductivity spectrum, to be only a small part of the total spectral weight. Instead, most of the electronic spectral weight belongs to the incoherent part, which leads to quasi-localized magnetic moments with short-range exchange interactions and hence antiferromagnetic order. In the doped systems, these short-range exchange interactions would give rise to superconductivity.

In this context, studies on the 122 iron selenides are bringing out major theoretical insights:

- The parent antiferromagnets are naturally described as Mott insulators: Ordered vacancies reduce the kinetic energy, thereby enhancing the effective U/t ratio and pushing it through the Mott transition threshold.

- The absence of hole Fermi-surface pockets removes Fermi-surface nesting. Therefore, it is not viable to understand the large-moment antiferromagnetic order of the parent insulators from the Fermi-surface nesting picture.

- In the weak-coupling description, the absence of particle-hole Fermi-surface nesting should lead to a considerably weaker superconductivity in the 122 iron selenides compared to their pnictide counterparts. However, experiments show that the superconductivity here is just as strong.

I now turn to several related pertinent issues:

- One may ask whether the structural modulation associated with the ordered Fe vacancies in the parent compounds can by itself be responsible for the insulating behavior. This is not the case: for the paramagnetic phase of the parent compounds in the presence of the vacancies, the ground states found in LDA calculations are metallic.

- By contrast, magnetically-constrained LDA calculations yield insulating antiferromagnetic ground states. The calculated ordered moment is over $3\mu_B/\mathrm{Fe}$, which is very large considering that the maximum possible atomic value is $4\mu_B/\mathrm{Fe}$. Hence, the underlying electron-electron interactions must be sufficiently strong. When combined with the fact that there are an integer number of electrons partially occupying the 3d orbitals, this suggests that Mott insulating is the underlying mechanism for the antiferromagnetic semiconducting behavior derived from the magnetically-constrained LDA calculations.

- Superconductivity has been seen to coexist with antiferromagnetic order. At the present time, there is evidence for both phase separation (between insulating AF phases and carrier-doped superconducting ones) and microscopic coexistence. While evidence for the former is rapidly growing, the issue still needs to be settled.

- More generally, as the Fe-content increases, how does the system evolve from the parent insulating antiferromagnet to the carrier-doped superconductor? Are there more phases in between?

Taken together, the salient properties that have already been established in the 122 iron selenides favor the strong coupling approach to the magnetism and superconductivity of both iron pnictides and chalcogenides. The identification of parent Mott insulators in the iron selenides provides an opportunity

to systematically study multi-orbital local-moment antiferromagnetism that is in proximity to superconductivity. It also offers the prospect to access a metal-insulator transition as a function of external or chemical pressure in these iron based compounds.

[1] J. X. Zhu et al, Phys. Rev. Lett. **104**, 216405 (2010).

[2] D. G. Free and J. S. O. Evans, Phys. Rev. B**81**, 214433 (2010).

[3] W. Bao et al, arXiv:1102.0830.

[4] T. Qian et al, Phys. Rev. Lett. **106**, 187001 (2011); Y. Zhang et al, Nature Mate. **10**, 273 (2011); D. Mou et al, Phys. Rev. Lett. **106**, 107001 (2011).