Iron pnictide superconductors: Electrons on the verge

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An optical analysis reveals that the electronic correlations in the ‘parent’ compounds of the iron pnictide superconductors are sufficiently strong to significantly impede the mobility of the electrons.

Superconductivity occurs in diverse materials. Until 1986, the highest observed superconducting transition temperature ($T_c$) was 23 K, prompting some researchers to suggest that the conventional mechanism based on electron-phonon coupling is incapable of yielding transition temperatures much higher. Then came the discovery of the copper oxide superconductors, which, after two decades, still hold the record $T_c$ of over 130 K at ambient pressure. The recent discovery of superconductivity at 26 K in LaFeAsO$_{1-x}$F$_x$ has generated much activity, and $T_c$ has since been pushed to over 55 K in related FeAs-based materials. Like the copper oxides, the iron pnictides have well-defined parent systems, such as LaFeAsO and BaFe$_2$As$_2$. Typically, superconductivity arises when charge carriers (either electrons or holes) are introduced into the parent compound, for example by substituting fluorine for oxygen in LaFeAsO. Fluorine donates its extra electron to the FeAs layer, thereby changing the layer’s carrier concentration.

Conventional superconductors are made from simple metals. Their electrons are weakly correlated, with mutual interactions that are small in comparison with their kinetic energy; it suffices to consider such essentially free electrons coupled to phonons. At the opposite limit, copper oxide superconductors involve strongly correlated electrons. The repulsive interaction among the electrons is so strong that the parent compounds are ‘Mott insulators’; energetically speaking, it pays for the electrons to stay localized. The question of how strong the electron correlations are in the iron pnictide superconductors is as important as it is delicate. On page 647 of this issue, Mumtaz Qazilbash et al. determine the strength of these correlations using optical measurements of the kinetic energy of the electrons.

The correlation question is not merely academic, as different answers lead to different points of departure for the study of the low-energy physics of magnetism and superconduc-
tivity. Early clues were ambiguous. The parent iron arsenides are antiferromagnetic, which suggests that some amount of electronic correlation must be present. However, the parents are always metallic, indicating that the correlations fall short of the Mott limit, in contrast to the copper oxides.

One perspective is that the parent iron arsenides are weakly correlated, and that the antiferromagnetism arises in the way it does in the standard itinerant antiferromagnet, chromium. This picture invokes a strong ‘nesting’ of the Fermi surfaces, which provides an enhanced phase space for exchange interactions among the well-defined electronic states near the Fermi surfaces, and leads to an antiferromagnetic ground state even though $U/t$, the ratio of the characteristic Coulomb repulsion, $U$, and the kinetic energy, or bandwidth, $t$, might be relatively small.

An alternative view is that the electronic correlations in the parent iron arsenides, although somewhat weaker than those in the copper oxides, are still sufficiently strong to place the system close to the boundary between itinerancy and interaction-induced electronic localization. In other words, they are ‘bad metal’ states in proximity to a Mott transition.

The work of Qazilbash and co-workers\textsuperscript{3} provides insight into this issue. The authors measure the optical conductivities of LaFePO, the phosphorus counterpart of the arsenic-based LaFeAsO, and BaFe$_2$As$_2$. They compare their room-temperature results with other electronic systems of varying degrees of correlation: parent and doped copper oxides; transition-metal compounds, such as V$_2$O$_3$, which are known to be bad metals close to Mott transition; and relatively simple metals such as copper, silver, chromium and MgB$_2$. They analyse the quantity

$$K_{\text{exp}} = \frac{2\hbar^2 c}{\pi e^2} \int d\omega \sigma_{\text{Drude}}(\omega),$$

where $\sigma_{\text{Drude}}$ is the measured Drude part of the optical conductivity, $\omega$ denotes frequency, and $c$ is the inter-FeAs-layer distance. This quantity has the physical meaning of the kinetic energy associated with the coherent part of the single-electron excitations in an interacting metallic system. It can be compared to $K_{\text{band}}$, which is the kinetic energy of the underlying non-interacting system, and is in practice extracted from \textit{ab initio} band-structure calculations. Coulomb repulsion impedes itinerancy and, hence, renders $K_{\text{exp}}$ smaller than $K_{\text{band}}$. The ratio $K_{\text{exp}}/K_{\text{band}}$ therefore provides a measure of the degree of correlation, as shown in Figure 1. It should take the value one in a non-interacting electron system; for a Mott
FIG. 1: Electronic correlations in the undoped iron pnictides. a. Optical conductivity as a function of frequency. The area under the $\omega = 0$ Drude peak is (for certain simplified energy dispersions) proportional to the kinetic energy of the coherent electrons near the Fermi energy, yielding $K_{\text{band}}$ and $K_{\text{exp}}$ for the non-interacting and interacting cases, respectively. The degree to which $K_{\text{exp}}/K_{\text{band}}$ is smaller than one is a measure of the interaction-induced reduction of the electrons’ mobility and, by extension, the normalized electron-electron repulsion, $U/t$. The interaction transfers spectral weight from low energies to high energies, up to order $U$. b. Different regimes of correlation. $U_c$ is the threshold interaction for a Mott localization transition in a system with an integer number of electrons partially occupying some electronic bands. That $K_{\text{exp}}/K_{\text{band}}$ is substantially smaller than one means that $U/t$ is smaller than but close to $U_c/t$, where the electrons are on the verge of losing their mobility.
insulator, it should be zero at zero temperature and remain a small number, much less than one, at room temperature.

Qazilbash et al. show that the value of $K_{\text{exp}}/K_{\text{band}}$ at room temperature is indeed close to one for simple metals and is considerably smaller than one for a Mott insulator. The value for the parent iron arsenide, BaFe$_2$As$_2$, is about 0.3 – the same order of magnitude as the optimally doped copper oxides or the incipient Mott insulator V$_2$O$_3$. For the parent iron phosphide, LaFePO, the value is about 0.45, which is somewhat larger than for BaFe$_2$As$_2$ but still not close to one.

One natural interpretation of these results is that the parent iron arsenides are indeed located near the boundary between itinerancy and localization. Further support for this interpretation can be inferred from the optical conductivity measurements of several other groups$^{4,5,6}$, which have shown that changes in temperature can induce transfer of optical spectral weight between the low-energy part of the spectrum and the part at high energies, greater than 1 eV. Such a spectral-weight transfer is a hallmark of bad metals close to Mott localization, which feature incoherent electronic excitations in the form of ‘precursor Hubbard bands’ (Figure 1) at some distance from the Fermi energy.

To fully establish that the incipient Mott localization operates in the iron pnictides, the precursor Hubbard bands need to be probed. One difficulty lies in there being several $d$ bands, for the precursor Hubbard bands can be embedded in other high-energy features associated with interband transitions. (This makes the temperature-induced spectral-weight transfer mentioned above especially illuminating.) In addition, it remains to be seen whether the system can be tuned across the Mott transition and into a Mott insulator state. The observations of Qazilbash et al. in fact provide a clue to this. The phosphides are shown to be less correlated than the arsenides, presumably because they have a larger chemical pressure (as the phosphorous ion is appreciably smaller). Will the replacement of arsenic by antimony or bismuth give rise to Mott insulators?

Despite these cautionary considerations, it is tempting to draw implications of the strong-coupling picture for low-energy physics. The existence of substantial incoherent weight in the excitation spectrum makes it meaningful to consider nearly localized magnetic moments, coupled by superexchange interactions, which have been shown to lead to the observed magnetic structure. This strong-coupling approach seems to provide a good basis for understanding the magnetic dynamics as well. Inelastic neutron scattering experiments$^{7,8}$ have recently
identified spin-wave-like excitations all the way to the antiferromagnetic zone boundary. The spin waves have a fairly high energy (\(\sim 200\) meV) that, incidentally, bodes well for a magnetic mechanism for superconductivity. They also have a very large spectral weight.

Superconductivity comes further down the energy hierarchy. The observation of sizeable electronic correlations supports strong-coupling approaches to superconducting pairings. More generally, it supports the widely held belief that the mechanism for superconductivity in the iron pnictides lies in electron-electron interactions, and not in the standard electron-phonon coupling.

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