Manifesto for a higher Tc – lessons from pnictides and cuprates

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(Dated: January 19, 2013)

We explore energy scales, features in the normal state transport, relevant interactions and constraints for the pairing mechanisms in the high-Tc cuprates and Fe-pnictides. Based on this analysis we attempt to identify a number of attributes of superconductors with a higher Tc. Expanded version of the article published in Nature Physics, 7, 271 (2011).

I. PNICTIDES VS OTHER EXOTIC SUPERCONDUCTORS

The discovery of superconductivity in Fe-based pnictides in 2008 (binary compounds of the elements from the 5th group: N, P, As, Sb, Bi) with \( T_c \) reaching almost 60K was, arguably, among the most significant breakthroughs in condensed matter physics during the past decade.\(^2\) The excitement was enormous and so were the efforts – the amount of data obtained for Fe-pnictides over the last three years is comparable to that collected for other known superconductors over several decades.\(^6\) A large number of new superconducting materials have been discovered not only within the Fe-pnictide family but also in the Fe-chalkogenides group: Fe-based compounds with elements from the 16th group: S, Se, Te.

Before 2008, the term “high-temperature superconductivity” (HTS) was reserved for Cu-based superconductors (CuSC), discovered in 1986. The transformation from the “Copper age” to the “Iron age” was swift and the term HTS now equally applies to both CuSC and Fe-based superconductors (FeSC).

Why FeSC are such a big deal? Even a cursory look at the phase diagram (Fig. 1) and the properties of FeSC reveal an intricate interplay between magnetism and superconductivity, also typical for other “exotic” superconductors discovered in the last three decades of the last millennium – heavy fermions, cuprates, ruthenates, organic and molecular conductors. Magnetism and superconductivity are antithetical in elemental superconductors, but in exotic superconductors magnetism associated with either d- or f-electrons is believed to be more a friend than a foe of the zero resistance state. However, with the exception of the cuprates the \( T_c \) of exotic superconductors known before 2008 were quite low, and many considered CuSC to being unique among exotic superconductors.\(^6\) The FeSC’s with \( T_c \)'s comparable to some CuSC’s, appear to undermine the uniqueness of the cuprates and have prompted the community to rethink what is important and what is not for the occurrence of high-\( T_c \) superconductivity in any material. Empowered by the two complementary perspectives on the high-Tc phenomenon one is well poised to address (and resolve) a number of outstanding issues such as: (i) do all high-Tc materials superconduct for the same reason? (ii) are the rather anomalous normal state properties of exotic superconductors a necessary prerequisite for high-Tc superconductivity? (iii) is there a generic route to increase \( T_c \)? Below we give our perspective on these three issues.

We argue that in CuSCs and FeSCs the answer to the first two questions is affirmative, and use the commonalities between the two classes of materials to detect the tools for the search for a higher \( T_c \).

We leave aside a number of interesting commonalities of CuSC and FeSC which only peripheral related to superconductivity, including the origin of magnetic order and Fermi Surface (FS) reconstruction in the magnetically ordered state, nematic order in FeSC above magnetic \( T_N \) and its relation to nematicity observed in the pseudogap phase in CuSC, and many others. A detailed review of the properties of Fe-based materials is given in Ref.\(^2\), which also contains an extensive list of references.

II. PHASE DIAGRAMS

From a distance, phase diagrams and relevant energy scales of FeSC and CuSC look amazingly similar (see Fig. 1 and Table 2). In both classes of systems there...
FIG. 2: Characteristic energy scales for fermionic and bosonic excitations, electronic kinetic energy $K_{\text{exp}}$, superfluid density and superconducting energy gap in cuprates (red boxes) and Fe-based materials (blue boxes). $K_{\text{LDA}}$ is the band structure kinetic energy discussed in the text.

is a region of a magnetic order near zero doping, and superconductivity emerges upon either hole or electron doping.

On a closer look, however, there are notable differences. Magnetic order in undoped CuSC is a conventional antiferromagnetism (spins of nearest neighbors are aligned antiparallel to each other), while in most of undoped FeSC the order is antiferromagnetic in one direction and ferromagnetic in the other (a stripe order). The superconducting order parameter in CuSC has $d$-wave symmetry, and the gap measured in momentum space as a function of the direction of the Fermi momentum has four nodes along the diagonals in the Brillouin zone (BZ). The nodes have been explicitly detected in angle-resolved photoemission (ARPES) measurements. FeSC have multiple FSs leading to complex doping trends and rather unconventional gap structures for a given symmetry. Still, ARPES measurements showed that the gap on the FSs centered at $\Gamma$ is near-isotropic, clearly inconsistent with $d$-wave symmetry.

Furthermore, the phase diagram of cuprates is richer than just antiferromagnetism and superconductivity – there is a region of Mott insulating behavior at the smallest dopings and the still mysterious pseudogap phase occupying a substantial portion of the normal state phase diagram, particularly in the underdoped regime (the term “pseudogap” is commonly used to describe a partial gap of fermionic excitations not accompanied by any obvious long range order). A comparison with FeSC again shows differences: there is no Mott phase in undoped pnictides, which show bad metallic, but still metallic behavior of resistivity. As of now, there is only sporadic evidence of the pseudogap.

The geometry of the FS and the low-energy excitations in CuSC and FeSC are also quite different. In CuSC, there is a single “open” cylindrical FS; its 2D cross-section uncovers four large segments (Fig. 3a). In FeSC, the FS has multiple quasi-2D sheets due to hybridization between all five $Fe$ $d$-orbitals – there are two small elliptical electron pockets centered at $(0, \pi)$ and $(\pi, 0)$ two small near-circular hole pockets centered at $\Gamma$-point (Fig. 3b), and, in some materials, additional hole pocket centered at $(\pi, \pi)$ The actual FS geometry is even more complex because of extra hybridization due to $Fe-Fe$ interaction via a pnictide/chalkogenide (Ref. 4). Given all these disparities in the FS structure, magnetism, and the order parameter symmetry, it is tempting to conclude that the phase diagrams of FeSC and CuSC are merely accidental lookalikes. We argue below that the actual situation is more involved. Differences apart, CuSC and FeSC reveal strikingly similar trends consistent with the notion of all-electronic scenario of electron pairing. By analyzing these trends we speculate on essential ingredients for unconventional and higher $T_c$ superconductivity.

III. ELEMENTS OF THE PAIRING MECHANISM FOR CuSC AND FeSC

A. gap symmetry and the pairing glue

A generic pairing scenario for moderately interacting itinerant systems assumes that fermions attract each other by exchanging quanta of bosonic excitations. A boson can be a phonon or it can be a collective density-wave excitation in either spin or charge channel. In the latter case, a direct interaction between the two given fermions is purely repulsive, but once it is renormalized by screening and by exchanges with other fermions, it acquires complex dependence on the angle along the FS. Its overall sign doesn’t change, but one or more angular momentum components may become attractive. The beauty of superconductivity is that it develops even if
just one angular component is attractive, no matter how strongly repulsive are the others.

In CuSC, there is no consensus on the pairing mechanism, but the most frequently discussed scenario for $d_{x^2-y^2}$ pairing in the optimally doped and overdoped regime is the collective exchange of collective excitations in the spin channel, commonly referred to spin fluctuations. Because antiferromagnetic phase is nearby, interaction mediated by spin fluctuations is peaked at momenta at or near $(\pi, \pi)$, which links fermions in different “hot regions” of the BZ near $(0, \pi)$, $(\pi, 0)$, and symmetry related points (Fig. 3a). The overall sign of such interaction is positive (repulsive), but its d-wave component is attractive, because a d-wave gap changes sign in between hot regions. In FeSC, “hot regions” are replaced by electron pockets centered at $(0, \pi)$ and $(\pi, 0)$. If the pairing interaction were to be peaked at $(\pi, \pi)$, it would give rise to a d-wave superconductivity with sign-changing gap between electron pockets, in complete analogy with CuSC. This may be the case for recently discovered strongly electron-doped Fe-chalkogenides $AFE_2Se_2$ ($A = K, Rb, Cs$), but for other FeSCs direct interaction between electron pockets is weak, and is overshadowed by the effective interaction through virtual hoppings to hole FS, which use a somewhat different assumption that the “glue” and superconductivity develop together, the “Debye frequency is the scale at which the pairing susceptibility become affected by the coupling to static fluctuations of a “glue”. In CuSC, magnetic fluctuations are peaked at or near $(\pi, \pi)$ up to $200 - 300 meV$, which is well above the pairing scale, so the “glue” may be considered as preformed, at least for a magnetic scenario. In FeSC magnetic fluctuations have been detected up to $200 meV$, but magnetic response at such energies is quite broad in $k$-space, and it still remain to be seen whether high-energy spin fluctuations contribute to a pairing glue or are featurless background which doesn’t affect the pairing.

### B. the gap structures

In CuSC, the geometry of the FS, which consists of a single sheet, and the $d$-wave symmetry predetermine the momentum dependence of the superconducting gap $\Delta(k)$ along the FS – it changes sign twice and has four nodes along the diagonal directions in the BZ. In FeSC, the multiple FSs and multi-orbital nature of excitations make the gap structure rather complex, even though from the symmetry perspective it is the simplest $s$-wave.

Let us elaborate on the above complexity. First, if the pairing glue are stripe spin fluctuations, an $s$-wave pairing adjusts to a repulsive $U_{eh}$ and changes sign between hole and electron pocket. Such a state is referred to as an extended $s$-wave, or an $s^\pm$ state. If the pairing is due to orbital fluctuations, the gap is a conventional $s$-wave, or $s^{++}$. Second, an inter-pocket electron-hole interaction competes with intra-pocket hole-hole and electron-electron repulsions which disfavor any gap with $s$-wave symmetry. Third, both intra-pocket and inter-pocket interactions generally depend on the angles along the FSs. Because of the last two effects, $\Delta(k)$ necessary acquires some angular dependence to minimize the effect of intra-pocket repulsion and to match angle de-
pendences of the interactions. If this angular dependence is sufficiently large, the gap develops “accidental” nodes at some points along the FSs. Calculations show that the nodes likely develop for electron-doped cuprates (the larger the doping, the more probable is that there are nodes), while for hole-doped FeSC the additional hole FS stabilizes a no-nodal gap.

Putting subtle issues aside, we see that there are two viable scenarios for the pairing in FeSC. First is \( s^\pm \) pairing due to attractive interaction between electron pockets and repulsive interaction between hole and electron pockets, and the second is \( s^{++} \) pairing due to attractive interaction between electron pockets and between electron and hole pockets. These scenarios yield different gap symmetry compared to that in CuSC, but the pairing mechanisms are essentially equivalent to spin-fluctuation and charge-fluctuation/phonon mechanisms for CuSC. A \( d - \)wave pairing in the FeSC is possible for very strongly electron and hole doped FeSCs, but it has been ruled out by the ARPES data for FeSCs which contain both hole and electron FSs. There are some hints for the \( p \)-wave gap in one of FeSC (LiFeAs) but solid evidence is still lacking.

The methods used to determine symmetry and structure of superconducting gaps in FeSC were for the most part developed or refined to address the symmetry issue in CuSC. A casual sampling of data acquired with all these techniques – neutron resonance, quasiparticle interference, penetration depth, thermal conductivity, [22] may signal a rather controversial situation on the issue of the gap structure in FeSC unlike the cuprates where the \( d_{x^2 - y^2} \) state has been ironed out. Yet, an in-depth query shows that seemingly conflicting results for FeSC are all in accord with the picture of the \( s^{++} \) gap by taking proper account of peculiarities of the multiband/multigap nature of this class of compounds. [10,11,13,19,20] Kontani and his collaborators argue, however, [23] that the data do not rule out an \( s^{++} \) gap.

Overall, an important lesson learned from the pnictides is that a high symmetry state, e.g., \( d_{x^2 - y^2} \) is not a necessity to overcome repulsion within an all-electronic mechanism, and that \( s \)-wave superconductivity is a viable option for an electronic pairing in a multi-band high-\( T_c \) superconductor.

IV. ESSENTIAL INGREDIENTS OF HIGH-TC SUPERCONDUCTIVITY

We now discuss a number of universal trends detected in Fe-based and Cu-based systems. We first point out that optimally doped FeSC and CuSC exemplify conductors in which electrons are neither fully itinerant nor completely localized. A way to quantify the tendency towards localization is to analyze the experimental kinetic energy \( K_{exp} \) that can be determined from ARPES or optical measurements in conjunction with the non-interacting value provided by band structure calculations, \( K_{LDA} \). The two extremes of the \( K_{exp}/K_{LDA} \) ratios are instructive. In conventional metals and electron-phonon superconductors \( K_{exp}/K_{LDA} \approx 1 \) signaling that there is not need to invoke localizing trends to explain electrons dynamics. In the opposite extreme of Mott insulators, localization arrest electron motion and \( K_{exp}/K_{LDA} \to 0 \). To the best of our knowledge, in all exotic superconductors, including both FeSC and CuSC, there is a noticeable renormalization of the kinetic energy, i.e., all these systems show some tendency towards localization (see Table I). Notably, for materials with the highest \( T_c \) in each family, \( K_{exp}/K_{LDA} \sim 0.3 - 0.5 \) implying a substantial distance away from both purely itinerant and Mott regimes. We therefore witness a remarkable tendency of materials with the highest \( T_c \) for a given series to strike the right balance between considerable strength of interactions and itinerancy. Because an estimate of \( K_{exp} \) is readily accessible from experiments at ambient condition, the above rather remarkable unifying aspect of a extremely diverse group of superconductors can be exploited for the search for new superconducting materials.

Why must \( K_{exp}/K_{LDA} \) be “right in the middle” to yield a high \( T_c \)? We believe the most generic reasoning for this “Goldie Locks law of superconductivity”, is rather straightforward. At weak coupling, itinerant fermions are ready to superconduct upon pairing, but \( T_c \) is exponentially small. In the opposite limit, when the interaction strength exceeds the fermionic bandwidth, fermions are completely localized and cannot move, even though the binding gap \( \Delta \) in this latter case is large.

A connection between the itinerancy-localization balance and the superconducting \( T_c \) can be appreciated by realizing that our measure of the interaction strength \( K_{exp} \) also sets the upper limit for the superfluid density \( \rho_s \). Once \( K_{exp} \) is diminished, so is \( \rho_s \). Without proper superfluid stiffness superconductivity becomes susceptible to the destructive role of phase and amplitude gap fluctuations and of competing orders. As a consequence, \( T_c \) is reduced compared to \( \Delta \). A necessity for substantial \( \rho_s \) (and therefore not too small \( K_{exp} \)) is epitomized through “the Uemura plot”:

\[
\rho_s \propto T_c^{-\alpha} \quad \text{with } \alpha = 0.2
\]

Is \( K_{exp}/K_{LDA} \) the only parameter essential for \( T_c \)? No. We argue that \( T_c \) can be further modified even when \( K_{exp}/K_{LDA} \) is fixed at “optimal” value by changing the structure of low-energy fermionic excitations inside the band. An important input for this consideration is the empirically determined “Homes scaling”:

\[
\rho_s \propto \sigma_{DC} \times T_c \quad \text{where } \sigma_{DC} \text{ is the DC conductivity}
\]

This scaling law holds for both FeSC and CuSC (see Fig. 1) and establishes a link between superconductivity and the normal state transport. Sum rules provide useful guidance to appreciate this link. According to the Ferrell-Glover-Tinkham sum rule, the superfluid density is given by the missing spectral weight in the real part of the conductivity. In a superconduc-
TABLE I: The ratio of the experimental kinetic energy $K_{exp}$ extracted from ARPES and optical measurements, as described in Ref.[22] and $K_{LDA}$ provided by band structure calculations.

| Superconductor | $T_{c, max}$ | $K_{exp} / K_{LDA}$ | Refs |
|----------------|--------------|---------------------|------|
| CuSCs          |              |                     |      |
| Nd$_2$-CeCu$_4$O$_4$ | 35           | 0.3                 | [23] |
| Pr$_2$-CeCu$_4$O$_4$ | 35           | 0.32                | [23] |
| La$_{2-x}$Sr$_x$CuO$_4$ | 40           | 0.25                | [22] |
| YBa$_2$Cu$_3$O$_7$ | 93.5         | 0.4                 | [2]  |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$ | 94           | 0.45                | [22] |
| FeSCs          |              |                     |      |
| LaFePO         | 7            | 0.5                 | [14] |
| Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ | 23          | 0.35-0.5            | [14,15] |
| Ba$_{1-x}$Co$_x$Fe$_2$As$_2$ | 39          | 0.3                 | [24] |
| Exotic SCs     |              |                     |      |
| CeCoIn$_5$     | 2.3          | 0.17                | [34,50] |
| Sr$_2$RuO$_4$  | 1.5          | 0.4                 | [14] |
| $\kappa - (BEDT - TF)$_2$Cu(SCN)$_2$ | 12          | 0.4                 | [25] |
| Electron-phonon SCs |            |                     |      |
| MgB$_2$        | 40           | 0.9                 | [14] |
| K$_3$C$_60$    | 20           | 0.96                | [46,47] |
| Rb$_3$C$_60$   | 30           | 0.9                 | [46,47] |

Strong dissipation suppresses fermionic coherence and, at a first glance, should also diminish the ability of fermions to (super)conduct. However, strong dissipation does not necessarily require the interaction to be larger than the bandwidth as it can be additionally enhanced by bringing the system to the vicinity of a quantum-critical point. In this latter case, the fermionic self-energy $\Sigma(\omega, k)$ at energies below (already renormalized) electronic bandwidth becomes predominantly frequency-dependent, what makes fermions incoherent but does not localize them. Furthermore, $T_c$ actually increases with increasing $\Sigma(\omega)$ because the suppression of fermionic coherence is overshadowed by the simultaneous increase of the dynamical pairing interaction. The increase is stronger in quasi 2D systems than in 3D. In CuSC, the effect of dissipation has been analyzed from various perspectives and the upper limit on $T_c$ was found to be around 2% of $E_F \sim 1 eV$, (Ref.[14]), in good agreement with the experimental $T_c$ values. Full scale calculations of $T_c$ in FeSC have not been done yet and are clearly called for.

We summarize this article by a brief outline of common characteristics of pnictides and cuprates that may teach us a lesson on generic attributes of a high-$T_c$ superconductor and thus may aid the search for new materials with even higher $T_c$. First, the screened Coulomb interaction should be strong, but not too strong to induce localization causing the reduction of $\rho_s$. The interaction of the order bandwidth appears to be optimal leading to $K_{exp} / K_{LDA} \simeq 0.5$. Second, the compliance of exotic and high-$T_c$ superconductors with the Homes law demands strong dissipation that can be registered through transport and spectroscopic methods. This dissipation predominantly comes from the effective dynamical electron-electron interaction within the band rather than from disorder. The same dynamical interaction gives rise to the pairing. Third, it is imperative that a system is able to avoid the repulsive nature of electron-electron interactions. Cuprates and pnictides have taught us that there is more than one way to deal with the repulsion problem (d-wave pairing in the cuprates and gap variations between multiple FSs in the pnictides). Finally, we stress significance of real space inhomogeneities that may in fact favor the increase of $T_c$ under optimal conditions[50]. Notably, ALL these effects are observed both in...
Fe-based and Cu-based systems thus identifying to a surprisingly consistent leitmotif of high Tc superconductivity driven through all-electronic interaction in these systems. A theoretical challenge is to accommodate these diverse effects in a microscopic theory with a predictive power. On a practical side, incorporating the above prerequisites into a viable protocol that facilitates the search for new superconductors is still an iron in the fire.

Acknowledgements

The authors wish to thank for valuable discussions large numbers of colleagues working on both cuprates and pnictides. This work was supported by NSF, DOE and AFOSR (D.N.B) and by NSF-DMR-0906953 (A.V.C).

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