Manifesto for a higher T_c

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The term 'high-temperature superconductor' used to refer only to copper-based compounds — now, iron-based pnictides have entered the frame. The comparison of these two types of superconductor is revealing, and suggestive of what might be needed to achieve even higher transition temperatures.

The 2008 discovery of superconductivity in Fe-based pnictides (compounds of the elements from Group V: N, P, As, Sb, Bi) with transition temperatures T_c reaching almost 60 K was, arguably, among the most significant breakthroughs in condensed-matter physics during the past decade[1]. The excitement was enormous and so were the efforts — the amount of data obtained for Fe-pnictides over the previous three years is comparable to that collected for other known superconductors over several decades[2–4]. A number of new superconducting materials have been discovered not only within the Fe-pnictide family but also in the Fe-chalcogenides group: Fe-based compounds with elements such as S, Se and Te. Before 2008, the term 'high-temperature superconductivity' (HTS) was reserved for Cu-based superconductors (CuSC or cuprates). 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 are FeSC such a big deal? Even a cursory look at the phase diagram (Fig. 1) and the properties[2–4] of FeSC reveals an intricate interplay between magnetism and superconductivity, also typical of 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 be unique among exotic superconductors[5]. The FeSC with their high transition-temperatures seem 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-T_c phenomenon we are well poised to address (and resolve) a number of outstanding issues such as: (1) Do all high-T_c materials superconduct for the same reason? (2) Are the rather anomalous normal-state properties of exotic superconductors a necessary prerequisite for high-T_c superconductivity? (3) Is there a generic route to increase T_c? We would argue that in the case of CuSC and FeSC the answer to the first two questions is affirmative, and also give our perspective on the third issue.

We will leave aside a number of interesting commonalities of CuSC and FeSC, which only peripherally relate to superconductivity, including the origin of magnetic order and Fermi surface (FS) reconstruction in the magnetically ordered state, nematic order in FeSC above the magnetic transition 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.)

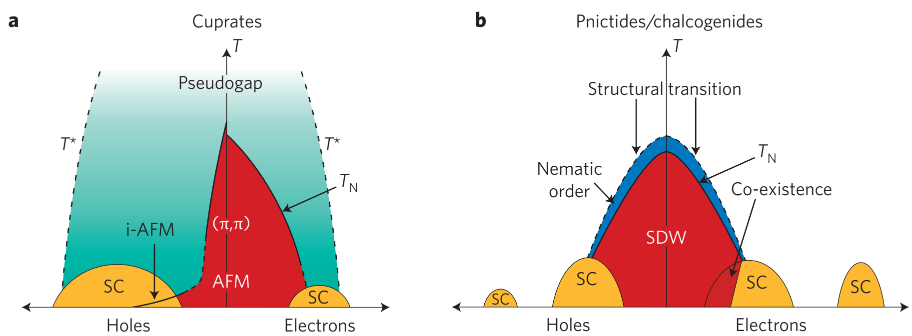

Figure 1 | Schematic phase diagrams of the cuprates and pnictides on hole- or electron-doping[2–4]. At relatively small dopings, superconductivity and antiferromagnetism co-exist. Not all details/phases are shown. Superconductivity in Fe-based systems can be initiated not only by doping but also by applying pressure and/or replacing one isovalent pnictide element by another[49]. The nematic phase in pnictides at $T > T_N$ is a subject of much debate. Superconductors at large doping are KFe_2As_2 for hole doping and AFe_2As_2 (A = K, Rb, Cs) for electron doping. Whether superconductivity in pnictides exists at all intermediate dopings is not yet clear. An additional superconducting dome in very strongly hole-doped cuprates has also been reported[50].

Phase diagrams

From a distance, the phase diagrams and relevant energy scales of FeSC and CuSC look amazingly similar (Figs 1 and 2). In both classes of system there is a region of magnetic ordering near zero doping, and superconductivity emerges on charge doping with either holes or electrons.

On a closer inspection, however, there are notable differences. Magnetic order in undoped CuSC is a conventional type of antiferromagnetism (spins of nearest neighbours are aligned antiparallel to each other), whereas in most undoped FeSC the order is antiferromagnetic in one direction and ferromagnetic in the other (a stripe order[2]). 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. The nodes have been explicitly detected in angle-resolved photoemission (ARPES) measurements[6]. FeSC have multiple FSs leading to complex doping trends and rather unconventional gap structures for a given symmetry. Still, ARPES measurements showed[7] that the gap on the FSs centred at the Γ point 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 behaviour at the smallest dopings[8] and the still mysterious pseudogap phase occupying a substantial portion of the normal-state phase diagram, particularly in the underdoped regime[9]. A comparison with FeSC again shows differences: there is no Mott phase in undoped pnictides, which instead show poor metallic — but still metallic nonetheless — behaviour in the resistivity. As of now, there is only sporadic evidence for a pseudogap in the pnictides[10].

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 two-dimensional 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 centred at (0,π) and (π,0), two small near-circular hole pockets centred at the Γ point (Fig. 3b), and, in some materials, an additional hole pocket centred at (π,π). The actual FS geometry is even more complex because of extra hybridization due to the Fe–Fe interaction through a pnictide/chalcogenide atom[4]. Given all these disparities in the FS structure, magnetism and order-parameter symmetry, it is tempting to conclude that the phase diagrams of FeSC and CuSc are merely accidental lookalikes. We think the actual situation is more involved. Differences apart, CuSC and FeSC reveal strikingly similar trends consistent with the notion of an all-electronic scenario of electron pairing.

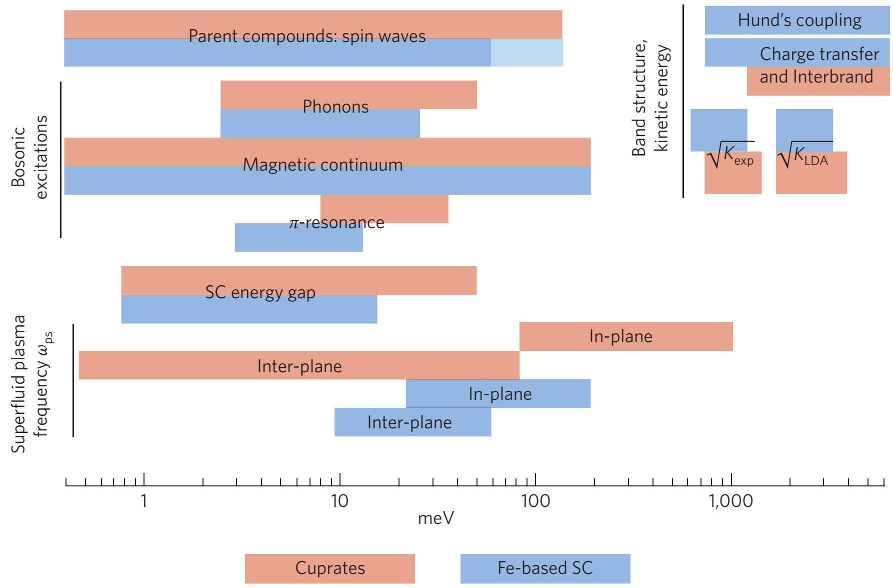

Figure 2 | Characteristic energy scales for fermionic and bosonic excitations, electronic kinetic energy K_{exp}, superfluid density and superconducting energy gap in cuprates (red boxes) and Fe-based materials (blue boxes). K_{LDA} is the band-structure kinetic energy discussed in the text.

Pairing mechanism for CuSC and FeSC

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 the 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 a 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 the others are.

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 exchange of collective excitations in the spin channel, commonly called spin fluctuations[11]. Because the antiferromagnetic phase is nearby, an interaction mediated by spin fluctuations is peaked at momenta at or near (π,π), which links fermions in different 'hot regions' of the Brillouin zone near (0,π), (π,0) and symmetry-related points (Fig. 3a). The overall sign of such an interaction is positive (repulsive), but its *d*-wave component is attractive, because a *d*-wave gap changes sign between 'hot regions'.

In FeSC, hot regions are electron pockets centred at (0,π) and (π,0). If the pairing interaction peaked at (π,π), it would give rise to *d*-wave superconductivity with a sign-changing gap between electron pockets, in complete analogy with CuSC. This may be the case for recently discovered strongly electron-doped Fe-chalcogenides[12] AFe_2Se_2 (A = K, Rb, Cs), but for other FeSC a direct interaction between electron pockets is weak, and is overshadowed by the effective interaction through virtual hoppings to hole FSs[4,13–16]. A simple exercise in quantum mechanics shows that such an effective interaction scales as U_{eh}^2, where U_{eh} is the electron–hole interaction and is attractive — that is, it gives rise to an *s*-wave pairing (no sign change of the gap between electron pockets), in clear distinction from $d_{x^2-y^2}$ pairing in CuSC.

The difference in the gap symmetry does not imply different pairing mechanisms. Indeed, the distance between hole and electron FSs is the same (π,0) or (0,π) as the momentum of the stripe magnetic order (this can be easily understood if magnetism is viewed as itinerant[17]). If U_{eh} is positive (repulsive), stripe magnetic fluctuations

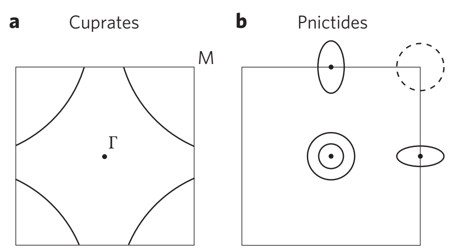

Figure 3 | Schematics of two-dimensional cross-sections of Fermi surfaces (FSs) for cuprates and pnictides. **a**, For weakly doped cuprates, the FS has a single sheet, and filled states (the ones closer to the Γ point) occupy about a half of the Brillouin zone. **b**, In the pnictides, the FS consists of multiple sheets: two hole pockets centred at Γ, two elliptical electron pockets centred at (0,π) and (π,0). There is another hole pocket at M = (π,π), which is cylindrical in some systems, and near-spherical around k_z = π in others[4].

Table 1 | The ratio of the experimental kinetic energy K_{exp} extracted from optical measurements, as described in ref. 27, and K_{LDA} provided by band-structure calculations.

Superconductor	$T_{c\,max}$	K_{exp}/K_{LDA} at $T_{c\,max}$	Refs
CuSCs			
$Nd_{2-x}Ce_xCuO_4$	25	0.3	31
$Pr_{2-x}Ce_xCuO_4$	25	0.32	31
$La_{2-x}Sr_xCuO_4$	40	0.25	31
$YBa_2Cu_3O_{7-x}$	93.5	0.4	8
$Bi_2Sr_2CaCu_2O_{8+\delta}$	94	0.45	*
FeSCs			
LaFePO	7	0.5	27
$Ba(Fe_{1-x}Co_x)_2As_2$	23	0.35–0.5	27,**
$Ba_{1-x}K_xFe_2As_2$	39	0.3	43
Exotic SCs			
$CeCoIn_5$	2.3	0.17	44,45
Sr_2RuO_4	1.5	0.4	27
κ-(BEDT-TTF)$_2$Cu(SCN)$_2$	12	0.4	46
Electron-phonon SCs			
MgB_2	40	0.9	27
K_3C_{60}	20	0.96	47,48
Rb_3C_{60}	30	0.9	47,48

*D. van der Marel *et al.*, unpublished; **A. Schafgans *et al.*, unpublished.

do enhance U_{eh} and from this perspective provide the glue for *s*-wave pairing in FeSC in the same way that (π,π) magnetic fluctuations provide the glue for *d*-wave pairing in CuSC. Essentially the same conclusion follows from renormalization group studies[14–16] that use a somewhat different assumption that the 'glue' and the superconductivity originate from the same set of interactions, and analyse how spin and pairing susceptibilities simultaneously grow on the system's flow towards low energies. We see therefore that pairing symmetries are different in CuSC and FeSC, but the pairing mechanism is likely to be the same — an exchange of spin fluctuations.

Alternative scenarios for pairing in both CuSC and FeSC postulate the dominance of charge fluctuations and/or phonons[18,19]. For FeSC this scenario can be realized[19] provided $U_{eh} < 0$ (this requires inter-orbital repulsion to be larger than the intra-orbital one). For $U_{eh} < 0$, stripe magnetic fluctuations are inactive, but charge (orbital) fluctuations and/or phonons do enhance $|U_{eh}|$ and therefore may act as a glue. We note, however, that the electron–phonon interaction in FeSCs can enhance U_{eh} but is too weak to produce a significant T_c on its own[20].

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 $\Delta(\mathbf{k})$ along the FS — it changes sign twice and has four nodes along the diagonal directions in the Brillouin zone. In FeSC, the multiple FSs and multi-orbital nature of the 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 is stripe spin fluctuations, an *s*-wave gap adjusts to a repulsive U_{eh} and changes sign between hole and electron pockets[4]. Such a state is referred to as an extended *s*-wave, or an *s*+− 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 that disfavour 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, $\Delta(\mathbf{k})$ necessarily acquires some angular dependence to minimize the effect of intra-pocket repulsion and to match angle dependencies of the interactions[13–16]. If this angular dependence becomes sufficiently large, the gap develops 'accidental' nodes at some points along the FSs. Calculations show[13–16] that the nodes probably develop for electron-doped cuprates (the larger the doping, the higher the probability for nodes), whereas for hole-doped FeSC the additional hole FS stabilizes the gap with no nodes.

Putting subtle issues aside, we see that there are two viable scenarios for the pairing in FeSC. First is *s*+− pairing due to the effective attractive interaction between electron pockets and the repulsive direct interaction between hole and electron pockets, and the second is *s*++ pairing when both interactions are attractive. These scenarios yield different gap symmetries compared with 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 FeSC, but it has been ruled out by ARPES data[7] for FeSC, which contain both hole and electron FSs. There are some hints for a *p*-wave gap in one system (LiFeAs) but solid evidence is still lacking (B. Buechner *et al.*, unpublished).

The methods used to determine the symmetry and structure of the 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[21], quasiparticle interference[22], penetration depth[23,24], thermal conductivity[25] — 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 the peculiarities of the multiband/multigap nature of this class of compound[13–16,26]. Some[19] argue, however, that the data do not rule out an *s*++ gap.

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

Essentials of high-T_c superconductivity

A number of universal trends have been detected in Fe-based and Cu-based systems. Optimally doped FeSC and CuSC exemplify conductors in which electrons are neither fully itinerant nor completely localized[27,28]. A way to quantify the tendency towards localization is to analyse 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})[27,29–31]. The two extremes of the K_{exp}/K_{LDA} ratios are instructive. In conventional metals and electron–phonon superconductors $K_{exp}/K_{LDA} \approx 1$, signalling that there is no need to invoke localizing trends to explain electron dynamics. In the opposite extreme of Mott insulators, localization arrests 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, that is, all these systems show some tendency towards localization (Table 1). Notably, for materials with the highest T_c in each family, having a $K_{exp}/K_{LDA} \sim 0.3$–0.5 implies a substantial distance 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 the considerable strength of interactions and itinerancy. Because an estimate of K_{exp} is readily accessible from experiments at ambient conditions[27], the above rather remarkable unifying aspect of an extremely diverse group of superconductors can be exploited for the search for new superconducting materials.

But why must K_{exp}/K_{LDA} be 'just right' to yield a high T_c? We believe the most generic reasoning for this 'Goldilocks law of superconductivity' is rather straightforward. At weak coupling, itinerant fermions are ready to superconduct on pairing, but T_c is exponentially small. In the opposite limit, when the interaction is larger than the fermionic bandwidth, fermions are completely localized and cannot move, even though the binding gap Δ 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 ρ_s. Once K_{exp} is diminished, so is ρ_s. Without proper superfluid stiffness superconductivity becomes susceptible to the destructive role of phase and gap amplitude fluctuations[32] and of competing orders. As a consequence, T_c is reduced compared with Δ. A necessity for substantial ρ_s (and therefore not too small K_{exp}) is epitomized by the 'Uemura plot': $T_c \propto \rho_s$, which holds for all exotic superconductors[33].

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 an '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'[34,35] $\rho_s \propto \sigma_{d.c.} \times T_c$ where $\sigma_{d.c.}$ is the d.c. conductivity just above T_c. This scaling law holds for both FeSC and CuSC (Fig. 4) 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 superconductor with strong dissipation the conductivity spectra in the normal state are broadened by scattering. In Fig. 4b we show schematically that in this case the magnitude of ρ_s can be reliably estimated by taking a product of $2\Delta \times \sigma_{dc}$. Because $\Delta \propto T_c$, Homes scaling holds.

In BCS-type superconductors the key source for dissipation is disorder. A common and highly non-trivial aspect of both FeSC and CuSC in this context is that strong dissipation has little to do with disorder, as shown by observations of quantum oscillations, which demand high purity of the specimens[36]. Instead, strong dissipation in both FeSC and CuSC is an inherent characteristic of charge dynamics at finite frequencies. The dissipation predominantly comes from the effective dynamical electron–electron interaction within the band. In many cases, dissipation is caused by the same interaction (for example, spin-fluctuation exchange) that gives rise to the pairing.

Infrared and ARPES measurements support the notion of strong dissipation by registering incoherent spectral features[6,10,37,38]. A transport counterpart of these effects is the linear temperature dependence of the resistivity above T_c and T^α behaviour of resistivity, with $1 < \alpha < 2$, down to $T = 0$ at the end point of superconductivity in the overdoped regime[39].

Strong dissipation suppresses fermionic coherence and, at first glance, should also diminish the ability of fermions to (super) conduct. However, strong dissipation does

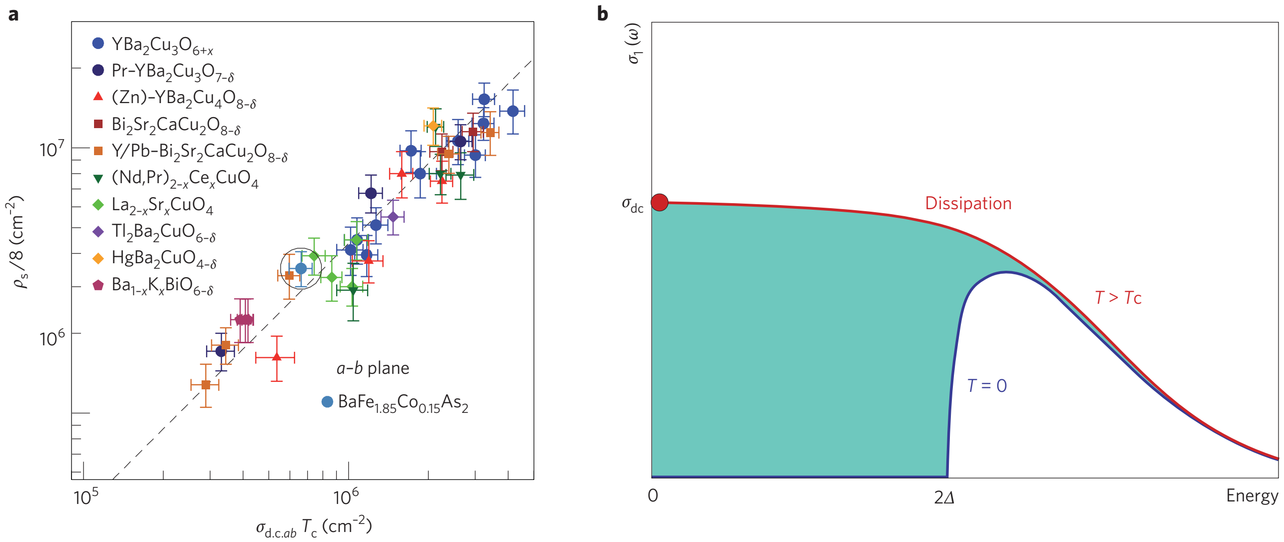

Figure 4 | Superfluid density in exotic superconductors. **a**, Homes scaling of the superfluid density ρ_s versus the product of the d.c. conductivity and the transition temperature $\sigma_{dc} \times T_c$ for several families of exotic superconductors including cuprates and pnictides. Reproduced with permission from ref. 51, © 2010 APS. **b**, Schematics of the condensate formation in a superconductor. The superfluid density is given by green-shaded area in the spectra of the real part of the conductivity $\sigma_1(\omega)$ and is accumulated primarily from the region given by the energy gap 2Δ. This area can be estimated from the product of $\sigma_{dc} \times 2\Delta$.

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 $\Sigma(\omega, \mathbf{k})$ at energies below the (already renormalized) electronic bandwidth becomes predominantly frequency dependent — which in most cases makes fermions incoherent but does not localize them and only modestly affects ρ_s. 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 three dimensions[40]. In CuSC, the effect of dissipation has been analysed from various perspectives and the upper limit on T_c was found to be around 2% of the Fermi energy $E_F \sim 1$ eV (ref. 41), in good agreement with experimental T_c values. Full-scale calculations of T_c in FeSC have not yet been done and are clearly called for.

In summary, a brief outline of common characteristics of pnictides and cuprates 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 ρ_s. The interaction of the order bandwidth seems to be optimal leading to $K_{exp}/K_{LDA} \leq 0.5$. Second, the compliance of exotic and high-T_c superconductors with Homes' law demands strong dissipation that can be registered through transport and spectroscopic methods. Third, it is imperative that a system is able to avoid the repulsive nature of electron–electron interactions. Cuprates and pnictides have instructed 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 the significance of real space inhomogeneities that may in fact favour the increase of T_c under optimal conditions[42]. Notably, all these effects are observed both in Fe-based and Cu-based systems thus identifying a surprisingly consistent leitmotif of high-T_c superconductivity driven by all-electronic interactions in these systems. A theoretical challenge is to accommodate these diverse effects in a microscopic theory with predictive power. On the practical side, incorporating the above prerequisites into a viable protocol that facilitates the search for new superconductors is still an iron in the fire. ❐

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