The intrinsic electronic phase diagram of iron-oxypnictide superconductors

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Abstract – We present the first comprehensive derivation of the intrinsic electronic phase diagram of the iron-oxypnictide superconductors in the normal state based on the analysis of the electrical resistivity $\rho$ of both LaFeAsO$_{1-x}F_{x}$ and SmFeAsO$_{1-x}F_{x}$ for a wide range of doping. Our data give clear-cut evidence for unusual normal-state properties in these new materials. In particular, the emergence of superconductivity at low doping levels is accompanied by distinct anomalous transport behavior in $\rho$ of the normal state which is reminiscent of the spin-density wave (SDW) signature in the parent material. At higher doping levels $\rho$ of LaFeAsO$_{1-x}F_{x}$ shows a clear transition from this pseudogap-like behavior to Fermi-liquid–like behavior, mimicking the phase diagram of the cuprates. Moreover, our data reveal a correlation between the strength of the anomalous features and the stability of the superconducting phase. The pseudogap-like features become stronger in SmFeAsO$_{1-x}F_{x}$ where superconductivity is enhanced and vanish when superconductivity is reduced in the doping region with Fermi-liquid–like behavior.

After the surprising discovery of iron pnictide superconductivity in LaFeAsO$_{1-x}F_{x}$ [1], strong evidence for unconventional superconductivity has rapidly emerged for this new material class. Particularly striking is a close interplay between superconductivity and magnetism: a commensurate spin-density wave (SDW) ground state has been observed in the undoped parent compounds [2–5], which is suppressed once superconductivity emerges upon doping [4,6–8]. It is important to note that up to now LaFeAsO$_{1-x}F_{x}$ is the only pnictide system which exhibits a homogeneous superconducting state [6]. Single crystals of intermetallic compounds [9–13] and even stoichiometric materials where superconductivity is induced by external pressure [14,15] show a spatially inhomogeneous magnetic state coexisting with superconductivity. Thus LaFeAsO$_{1-x}F_{x}$ is the only known FeAs-based material which allows the study of the intrinsic electronic properties of superconducting species both in the superconducting and in the normal state.

From extensive work on other unconventional superconductors with a similar antiferromagnetic parent state such as heavy-fermion and cuprate superconductors it is known that the exploration of the normal state is indispensable for the understanding of unconventional superconductivity. The electrical resistivity $\rho$ has been proven as a key experimental probe for this purpose [16–20]. In this letter we show that also the newly discovered pnictide superconductors show pronounced signatures of unusual normal-state properties. In particular, the analysis of $\rho$ reveals distinct anomalies in the underdoped superconducting doping regime, which appear as remnants of the anomalies that accompany the structural and magnetic phase transitions of the non-superconducting parent compounds, and which strongly resemble pseudogap signatures of underdoped-cuprate superconductors. Moreover, we find a transition from pseudogap-like to Fermi-liquid–like behavior at further increased doping, and thus a strong resemblance to the cuprate phase diagram.

Polycrystalline LaFeAsO$_{1-x}F_{x}$ (0 ≤ x ≤ 0.2) and SmFeAsO$_{1-x}F_{x}$ (0 ≤ x ≤ 0.1) were prepared and characterized by powder X-ray diffraction (XRD) [21] and further characterized by magnetization [22], nuclear magnetic resonance [23], and muon spin rotation (μSR)

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features. Upon further cooling, $\rho(T)$ shows a minimum at $\sim 70-90\,K$ followed by a strong low-$T$ upturn which is indicative of carrier localization, presumably arising from a SDW gap. When increasing the F doping level up to $x = 0.04$, the essential features of $\rho(T)$ remain qualitatively the same as in the undoped material. In particular, $T_{\text{max}}$ and $T_{\text{drop}}$ are slightly shifted towards lower $T$, and a little broadening of the corresponding anomalies occurs.

A further increase of $x$ leads to a sudden occurrence of superconductivity with rather high critical temperature $T_c$ [7] and drastic changes in $\rho(T)$ in the normal state. For $0.05 \leq x \leq 0.075$, a low-$T$ upturn ($T \leq 60\,K$) is still present before entering the superconducting state, which is reminiscent of the low-$T$ upturn of the low-doping compounds. At high $T$, however, the clear features at $\sim 150\,K$ of the non-superconducting samples have disappeared. Instead, $\rho$ increases monotonically for $T \geq 60\,K$ up to $300\,K$. A close inspection of this increase reveals a surprising feature: while $\rho(T)$ becomes linear at $T \geq 250\,K$, it drops below the low-$T$ extrapolation of this linearity (cf. fig. 1b) for a representative example). This drop is connected with an inflection point at $T_{\text{drop}} \approx 150\,K$, which can be conveniently extracted from the derivatives $d\rho/dT$. Thus, despite the suppression of the actual structural and magnetic transitions, a distinct reminiscent feature with the same energy scale persists in these superconducting samples.

Yet another drastic systematic change of $\rho(T)$ is observed when the doping level enters the regime $0.1 \leq x \leq 0.2$. Here, instead of the low-$T$ upturn, we find $\rho(T) = \rho_0 + AT^2$ ($\rho_0 = \text{const}$) from just above $T_c$ up to $\sim 200\,K$, i.e., a Fermi-liquid–like behavior [27] which indicates enhanced electron-electron interaction (cf. fig. 1c) for a representative example). A maximum $T_c = 26.8\,K$ is observed at $x = 0.1$ which quickly diminishes with further increasing $x$ [7]. Simultaneously, the anomaly connected with the inflection point becomes weaker at higher $T_{\text{drop}}$ and eventually vanishes completely for $x \geq 0.15$ where the quadratic low-$T$ increase shows a smooth crossover to a linear high-$T$ behavior (cf. fig. 1d).

At this point, a striking similarity to the electronic phase diagram of hole doped cuprates becomes apparent. In the underdoped region $0.05 \leq x \leq 0.075$, the overall temperature dependence, i.e., the low-$T$ upturn together with the feature at $150\,K$ and the linear increase at higher $T$, almost perfectly mimics the resistivity of underdoped cuprate superconductors [16]. In particular, the feature at $T_{\text{drop}}$ amazingly resembles well-known pseudogap signatures [16,17]. The finding of a Fermi-liquid–like behavior at higher doping levels $x \geq 0.1$ (hereafter called overdoped region) is a further similarity if compared with the normal state of overdoped cuprates where a qualitatively similar $\rho(T)$ is observed [16,17,28].

We summarize our major findings for LaFeAsO$_{1-x}$F$_x$, in the phase diagram shown in fig. 2. In the underdoped superconducting region the signatures of both the
The electronic phase diagram of iron-oxypnictide superconductors

![Phase Diagram of LaFeAsO\(_{1-x}\)F\(_x\)](image)

**Fig. 2:** (Colour on-line) Phase diagram of LaFeAsO\(_{1-x}\)F\(_x\) as a function of the doping level \(x\) and temperature \(T\), highlighting the unusual \(\rho(T)\) as compared to that of the (approximately) linear \(\rho(T)\) near 300 K. The latter \((\frac{d\rho}{dT}(T) = \frac{d\rho}{dT}(296 K) \approx 0)\) gives rise to the yellow areas. The blue regions \((\frac{d\rho}{dT} < 0)\) indicate carrier localization/fluctuation and Fermi-liquid-like behavior for \(x \leq 0.075\) and \(x \geq 0.1\), respectively. Across the whole phase diagram the red areas \((\frac{d\rho}{dT} > 0)\) are centered around \(T_{\text{drop}}\) and mark the signatures of the structural/magnetic transitions \((x \leq 0.04)\) and the corresponding remnant feature \((x \geq 0.05)\). The dark bars separate the non-superconducting, the underdoped and the overdoped superconducting regimes. The diagram shows also data points for \(T_\text{c} (\bigcirc), T_{\text{drop}} (\bigstar), T_{\text{max}} (\bigtriangledown)\), and, where available, \(T_N (\bigtriangleup)\) and \(T_S (\Box)\) from \(\mu\text{SR}\) and XRD experiments [7].

We now turn to \(\rho(T)\) of SmFeAsO\(_{1-x}\)F\(_x\), which cover the doping range \(0 \leq x \leq 0.1\) (see fig. 3). The resistivity drop and the low-\(T\) localization clearly “survive” despite the suppression of the structural/magnetic transitions and the occurrence of superconductivity. These anomalous features strongly suggest that fluctuations connected to the SDW are still present. They apparently lead to a renormalization of the charge carriers, thus playing a major role in the physics of the superconductivity in the system. The observation of \(T_{\text{drop}}\) in the underdoped region being about the same as at \(x \leq 0.04\) shows that these fluctuations are of a similar energy scale as the actual SDW state. Intriguingly, in the overdoped region the fluctuation features vanish and Fermi-liquid–like behavior becomes increasingly dominating over a large \(T\)-range. In view of this strong change towards less unconventional normal-state properties and the similarity to the cuprate phase diagram quantum critical behavior should be considered in this material [29,30].

One might speculate that the reinforcement of the pseudogap-like anomalies seen for the superconducting samples is related to substantial remnants of the structural and magnetic transitions at low doping. In fact, recent \(\mu\text{SR}\) data [4,8] provide clear-cut evidence for magnetic
order and/or slow magnetic fluctuation in all superconducting species of SmFeAsO\(_{1-x}\)F\(_x\), and thus corroborate this notion.

The direct comparison of the phase diagrams shown in figs. 2 and 4 allows important conclusions about the correlation of anomalous transport behavior and superconductivity. Interestingly, \(T_c\) is strongly enhanced in SmFeAsO\(_{1-x}\)F\(_x\) despite the reinforcement of the pseudogap-like features and static magnetism [4,8], and thus suggests the intimate connection between these phenomena. On the other hand, in LaFeAsO\(_{1-x}\)F\(_x\) the fading of the pseudogap-like anomalies at high doping levels is accompanied by a weakening of the superconducting state which is readily seen by the strongly reduced \(T_c\). Thus, a key approach to unravel the nature of superconductivity in the iron pnictide materials is the understanding of the nature of the electronic renormalization which is connected with the fluctuation of the SDW state.

To conclude, our data unambiguously show that the high temperature superconductivity in the iron pnictides is intimately correlated with anomalous transport properties in the normal state. This is clearly seen in the evolution of the superconducting dome as a function of doping, which is accompanied by strong changes of the electrical resistivity at higher \(T\). In particular, a rather conventional Fermi-liquid–like behavior occurs in the overdoped region where the critical temperature \(T_c\) is small. True high-\(T_c\) superconductivity with \(T_c > 20\) K is found only in the lower doped region of the phase diagram where the resistivity shows pronounced anomalies which are clearly related to the anomalous SDW state of the undoped parent compounds. Note, that the stronger manifestation of these anomalies in SmFeAsO\(_{1-x}\)F\(_x\) is connected with an enhancement of \(T_c\). Summing up all these anomalous features, the electronic phase diagram of the FeAs superconductors (cf. fig. 5) yields a striking resemblance to the generic phase diagram of cuprate superconductors [30,32] and that of other unconventional superconductors in the vicinity of a quantum critical point [19,20]. However, unlike the latter examples, the doping-driven transition from the non-superconducting magnetic ground state to superconductivity appears to be first-order–like and/or accompanied by inhomogeneity [4,8,9,14].

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