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Developments and Perspectives of Iron-based High-Temperature Superconductors**

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Since the first report on superconductivity at 26 K in F-doped LaOFeAs at the end of February 2008, the superconducting transition temperature has been quickly raised to about 55 K and many new superconductors have been found, and superconductors with a higher condensing temperature are expected in this system. In this brief Research News article, the author will relate the enormous interest that has been generated towards new superconductor exploration, the refinement and tailoring of the single phase of the available superconductors, the growth of single crystals, and the theoretical and experimental efforts toward targeting the mechanism. Finally, a preliminary comparison between the iron-based system and the cuprates is given, as well as perspectives on these new iron-based superconductors.

1. Introduction

Superconductivity is a quantum phenomenon that shows the vanishing of resistivity and exclusion of magnetic field because of the condensation of paired electrons. In a normal metal, the conducting electrons move on a background of all kinds of vibrations, such as the vibration of the atomic lattice with the elementary excitation, termed phonons. The electrons collide with these phonons and lose their kinetic energy, which leads to the consumption of energy. This is the origin of resistivity in a normal metal at a finite temperature. However, this energy consumption and thus resistivity becomes zero in a very pure system when the temperature goes to zero, because all kinds of thermal vibrations will cease at that limit (assuming the quantum fluctuation of atoms is weak). In a superconductor, the electrons will bind each other into so-called Cooper pairs. These Cooper pairs behave as bosons statistically and can condense at a finite condensing temperature (T_c), and their very long wavelength will overlook the local detailed scattering induced by atomic vibrations, which leads to transport without consuming energy. Therefore, the resistivity drops to zero when it is still at a finite temperature. Because of the excellent performance of a superconductor under a high current or magnetic field, new superconductors with high transition temperatures are highly desired.

The search for new superconductors has lasted for more than 100 years. In the early times, the explorations towards new superconductors were focused mainly on mono-element materials or multi-element alloys. Unfortunately, such materials exhibit superconductivity with a transition temperature below about 23 K; the limit held by Nb_3Ge until 1986.[1] A breakthrough occurred at the end of 1986, when superconductivity beyond 30 K was found by the IBM Zurich group led by K. A. Muller for the cuprate superconductor LaBaCuO.[2] Since then the pursuit of high-temperature superconductors has begun all over the world, and within several years the superconducting transition temperature was raised to 134 K (measured at ambient pressure) and 164 K (measured at high pressure). The cuprate superconductors exhibit a very short coherence length, very high anisotropy, and are brittle, all of which strongly hinder large-scale applications. Therefore, the discovery of a non-cuprate superconductor with

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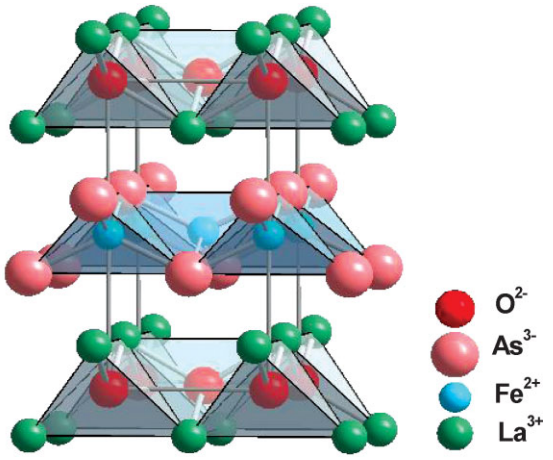

Figure 1. The atomic structure of LaFeAsO. It has a typical layered structure with $-(LaO)_2-(FeAS)_2-(LaO)_2-(FeAs)_2-$ alternative stacking along the *c*-axis. The Fe ions construct a square lattice with a near-neighbor distance of about 2.853 Å, but a diagonal distance of about 3.97 Å.

a higher T_c is strongly desired by the superconductivity community.

A turning point happened at the end of February 2008, when a Japanese group led by Prof. Hosono at the Tokyo Institute of Technology found superconductivity at 26 K in F-doped LaOFeAs.[3] This type of material can be traced back to 1974, when Jeitschko et al. were exploring new functional materials[4] for the DuPont company. Later on, a German group fabricated a series of materials with the same ZrCuSiAs structure.[5] They are generally referred to as quaternary oxypnictides and have the general formula LnOMPn (where Ln = La, Ce, Pr, Nd, Sm, Eu, and Gd, etc.; M = Mn, Fe, Co, and Ni, etc.; Pn = P and As, etc.). A typical structure for LaOFeAs is shown in Figure 1. The system has a layered structure and a tetragonal *P*4/*nmm* space group, with a stacking series of $-(LnO)_2-(MP)_2-(LnO)_2-$ along the *c*-axis. In one unit cell, there are two molecules of LnOMP, and its valence is self-balanced in the parent phase, that is, the charge of $(LnO)^{1+}$ is balanced by $(MP)^{1-}$. Some of the quaternary oxypnictides were shown to be superconductors at low temperatures and thus constructed a new family of layered superconductors without copper.[6,7] Very recently, Kamihara et al.[3] found that the material $LaO_{1-x}F_xFeAs$ ($x = 0.05$–0.12) becomes superconductive at 26 K. This interesting discovery has stimulated a new round of intense study in the field of superconductivity.

2. The Pursuit of Higher T_c

Soon after the report by Kamihara et al.,[3] several groups[8–10] repeated the preparation of the new superconductor $LaO_{1-x}F_xFeAs$ with $T_c = 26$ K. The early transport measurements revealed the existence of electron-like charge carriers with low density. The race for new superconductors then immediately started. Most results were posted on the Los Alamos website http://arxiv.org/list/cond-mat.supr-con/. By using a high pressure up to 3 GP, Takahashi et al. found that[11] the onset superconducting transition temperature can be increased from 26 to 43 K in $LaO_{0.89}F_{0.11}FeAs$. On March 20, 2008, Wen et al. reported the observation of superconductivity at about 25 K by substituting trivalent La with divalent Sr and without F-doping, and suggested that hole doping, instead of electron doping, can also possibly induce superconductivity in the Fe-based system.[12] This is supposed to be the first discovery of hole-doped superconductors in the iron-based system. Soon, superconductivity at 43 K was reported by X. H. Chen et al.[13] in $SmO_{0.85}F_{0.15}FeAs$. At almost the same time, G. F: Chen et al.[14] and Ren et al.[15] reported superconductivity close to or above 50 K in $NdO_{0.85}F_{0.15}FeAs$ and $PrO_{0.85}F_{0.15}FeAs$, respectively. During the competition of pursuing a higher T_c, it has been gradually found that the high-pressure synthesis technique seems to be very efficient: it needs only a short sintering time, normally less than two hours, and the fluorine element is more effectively maintained in the composite in the high-pressure capsule. This led to a quick improvement of superconducting transition temperatures up to 55 K by incorporating different light rare-earth elements, ranging from La to Gd in $ReO_{1-x}F_xFeAs$.[16] Furthermore, Ren et al. realized that the oxygen sites may be vacant since the nominal fluorine component will escape from the samples. They thus used the high-pressure technique to directly fabricate the F-free but oxygen-deficient superconducting samples, all of which show superconducting transition temperatures up to 55 K.[17] The interesting point is that the oxygen-deficient and F-free samples show no superconductivity when they are made at ambient pressures, although the phase is very pure.[18] A recent report shows that even the late (or so-called heavy) rare-earth elements, such as Tb and Dy, can induce superconductivity above 40 K by using the high-pressure synthesis technique.[19] In Figure 2, the discovery time dependence of superconductivity transition temperatures is shown, and one can see that T_c rises quickly. The figure implies that superconductors with a higher T_c can be anticipated.

In exploring new superconductors with higher T_c, there are several possible routes. In the following some of these possibilities are addressed. The first straight-forward path would be to fabricate materials with multilayers of FeAs in one unit cell. The idea is inspired from the case of cuprate superconductors and is actually very simple: the pairing strength reflected from the upper critical field value is very high, but the superfluid density is low in both systems.[9] A multilayer will generate a high superfluid density in a local region, as argued in the cuprate superconductors, and it may induce a higher T_c if the superconducting transition is governed by phase fluctuation, not by the pairing strength. However, it must be mentioned that the superconducting phase fluctuation does not seem very strong, or at least that is the case for the optimized samples (here, we hesitate to call them optimally doped samples). Recently, in transport measurements on single crystals, it was found that the critical fluctuations are not very strong; the anisotropy $\Gamma = (m_c/m_{ab})^{1/2}$ is below 5–6. It

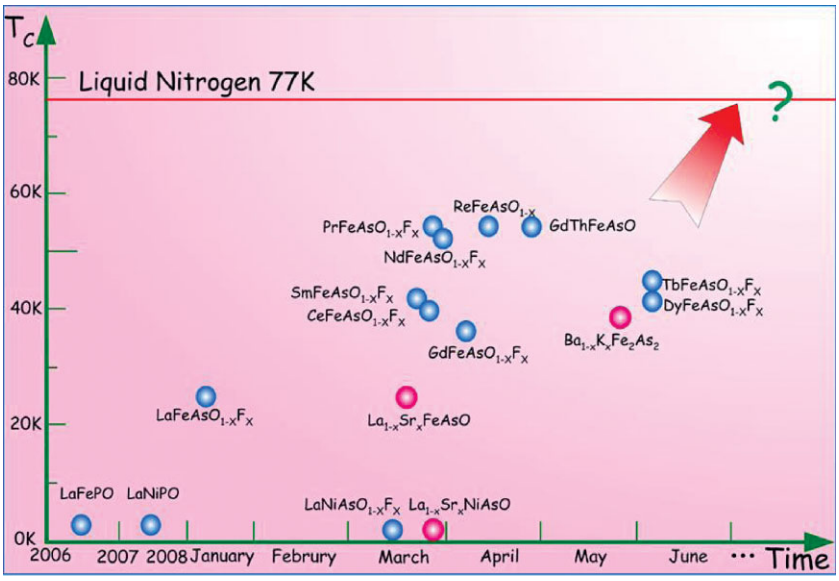

Figure 2. The superconducting transition temperatures of the iron-based superconductors versus the time of discovery. The blue dots represent the electron-doped samples, either by F-doping or by using oxygen vacancies. The red dots show the samples with hole doping. Superconductors with a higher T_c are expected. (This picture was composed by Lei Fang and Hai-Hu Wen on 6 June 2008.)

remains to be seen whether or not the pairing gap as well as the anisotropy will go up in less electron (or hole) doped samples. If this is not the case, but rather a BCS-like behavior is found with $2\Delta_{sc}/k_BT_c \approx 4$ in the underdoped samples,[20] the opportunity for increasing T_c by having multilayers would be significantly limited. However, a new structure will always induce a change of pairing strength as well as the density of states at the Fermi energy, and thus the T_c can be altered up or down randomly by having a multilayered structure.

The second route towards higher T_c would rely on finding materials with new structures. As far as we know, no explicit explanation has been given about why the T_c in the FeAs system is higher than in other systems (e.g., NiAs, FeP); however, any efforts to explore new As-free materials are worthwhile, especially when the new materials contain only non-toxic elements. If sticking to the FeAs system, hole doping would be another interesting and important way to impart superconductivity. According to Wen et al.,[12] superconductivity can be induced by hole doping, and the T_c exhibits a symmetric behavior in electron- and hole-doped sides. A similar behavior has been observed in F (electron) and Ca (hole) doped LaOFeP.[21] Recently, we also found symmetric behavior of T_c vs. doping in both electron- and hole-doped sides in $LaO_{1-x}F_xNiAs$ and $La_{1-x}Sr_xONiAs$ systems, respectively.[22] In the Sr-doped samples, the T_c increases monotonously with the lattice constant (the radius of Sr^{2+} is slightly larger than that of La^{3+}), which indicates a stability of the same structure with Sr doping. One step further for hole-doped superconductors was achieved by Rotter et al.[23] who observed superconductivity at 38 K when 40% of the Ba^{2+} in $Ba(FeAs)_2$ was substituted by K^+. The parent phase of $Ba(FeAs)_2$ is a poor metal, with a spin density wave anomaly at about 140 K. This is very similar to the properties of the undoped LaOFeAs phase. This work clearly proves the earlier work by Wen et al.[12] that introducing holes into the system can also lead to superconductivity. In this sense the hole-doped side remains a wide area to be explored.

3. Tailoring the Materials

In order to get a precise, correct, and deep insight into the physical properties, good-quality samples are highly desirable. So far the single-phase superconductors $ReFeAsO_{1-x}F_x$ (Re = La, Nd, Ce, Pr, Sm, and Gd) have been made by many groups. The major technique is the solid-state reaction method. This technique can be carried out with one step, or more steps, but the highest sintering temperatures are about 1150 to 1260 °C with the materials sealed in quartz tubes. Caution should be taken when handling the rare-earth metals because they oxidize easily. Normally, it is necessary to cut or grind these rare-earth elements in a glove-box filled with inert gas. In addition, any treatment of As should be strictly confined within the glove box because of its toxicity. It seems that the two-step method suggested by our group is quite good for obtaining the pure phase.[9] The second method to grow a phase-pure sample is the high-pressure synthesis technique. As mentioned previously, this technique is fast, with F and As well-sealed within the pressure capsule, but because of the short sintering time it is difficult to avoid the existence of some unreacted components, such as FeAs and others. The third way to grow very pure single-phase samples is to use a flux method at a relatively low sintering temperature.[24] By using this method, we have grown single-phase samples with only plate-like crystals, with sizes ranging from 5 to 50 μm.

In order to know the intrinsic properties of the superconductors, measurements on single crystals are necessary. Up to date, there are only two reports about the fabrication of single crystals. One is based on the high-temperature/high-pressure synthesis technique (ETH, Zurich). For the growth of single crystals they used NaCl/KCl as flux. The ReFeAsOF material to flux ratio varied between 1:1 and 1:3. The thoroughly ground materials were pressed into pellets and were placed in a BN crucible together with the flux inside a pyrophyllite cube. In the experiment, a pressure of 3 GPa was applied and the temperature was ramped up within 1 h to the maximum value of 1350–1450 °C, maintained for 4–10 h, and decreased over 5–24 h to room temperature for the crystal growth. The crystals obtained in this way have a maximum size of 300 μm.[25] In fact, we have successfully grown single crystals at ambient pressures. The crystals were also made by a flux method using NaCl as the flux. First the starting materials Nd

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grains (purity 99.95%) and As grains (purity 99.99%) were mixed in a 1:1 ratio, ground, and pressed into a pellet shape. The pellet was sealed in an evacuated quartz tube followed by burning at 800 °C for 10 h. The resultant pellet was smashed and ground together with NdF_3 powder (purity 99.95%), Fe_2O_3 powder (purity 99.9%), and Fe powder (purity 99.9%) in a stoichiometry of formula $NdO_{0.82}F_{0.18}FeAs$. Again, the powder was pressed into a pellet and together with a NaCl powder of mass ratio NaCl:NdOFFeAs = 10:1) was sealed in an evacuated quartz tube and burned at about 1050 °C for 10 days. The product was cooled at a rate of 3 °C per hour to 850 °C, followed by rapid cooling by shutting off the power of the furnace. The resulting product is a pellet with a dark color. It contains mainly plate-like small crystals. By tuning the sintering temperature, larger crystals can be obtained. Clearly, with the passage of time, an optimized method will be found in the future to grow larger crystals.

4. Theoretical and Experimental Approaches to Understand the Mechanism

Soon after the report of superconductivity in the iron-based system, researchers were attracted to understand the physics and try to resolve the pairing mechanism. From a first-hand calculation, the electron–phonon coupling constant $\lambda_{\text{el-ph}}$ is found to be only about 0.21[26] on the FeAs planes; this is certainly too small to interpret a transition temperature as high as 55 K. The band structure calculation had already been done for LaOFeP,[27] which superconducts at about 4 K and shows a metallic behavior up to 300 K. This kind of temperature dependence of resistivity is in sharp contrast with the undoped LaOFeAs system, which exhibits a sharp drop of resistivity at about 150 K,[3,28] and a slight upturn appears when the temperature approaches zero. This sharp drop was first attributed to the spin density wave (SDW) transition induced by the nesting between the hole pockets and the electron pockets. The neutron diffraction, reported later, proved that there is an antiferromagnetic order with a commensurate ordering wave vector of (0.5 0.5 0.5) below 136 K, but the structural phase transition occurs at about 150 K. Therefore, it seems that the structure transition occurs first and then the SDW order is established.[29] However, among the spin or lattice distortions, it is difficult to say which is the driving force. This structural transition and SDW order was later proven by Mossbauer measurements,[30] which show at least a consistency between the data from different experiments. By doping the system with fluorine, the sharp drop of resistivity at about 150 K will be gradually smeared away, and the slight upturn in the low-temperature region is strongly suppressed. At a certain amount of doped electrons or holes, the superconductivity sets in. It is not yet clear at which doping level the superconductivity sets in and whether the T_c increases monotonically with the doping level in the underdoped region. This point is obscure as a result of the true fluorine content being unknown, because some fluorine will escape from the sample during preparation. So far the highest reported doping level is about 50%, which always seems to increase the T_c slightly or stabilize it at about 55 K. Thus, phase separation may occur in the heavily doped samples: the fluorine content is simply not uniform.

According to band structure calculations by Lebègue[27] and by Singh and Du,[31] it is known that all five Fe-derived d-bands cross the Fermi energy in the parent phase LaOFeP or LaOFeAs. The Fermi surface consists of five sheets: two electron cylinders centered around the M–A line with high velocity, two hole cylinders around the Γ–Z line with lower velocity, and an additional heavy three-dimensional hole pocket that is centered at Z. It is supposed that the electron cylinders have a higher velocity and make the dominant contribution to the in-plane electrical conductivity. However, these calculations have not taken the correlation effect into account. Kotliar et al.[32] estimated the correlation energy to be about 4 eV, and concluded that the parent compound is a bad metal at the verge of the metal–insulator transition. After taking the correlation effect as well as the super-exchange energy between Fe–Fe bonds (J1, near neighbor), and that by the Fe–As–Fe bonds (J2 next near neighbor) into account, it is possible to determine that the lowest-energy state is the one with an antiferromagnetic state[33] with an interpenetrated AF state. The system can lower its energy by accommodating this state by having a structural phase transition at a slightly higher temperature. To date, it is still difficult to say whether there is a close relationship between the SDW order and the structure phase transition, or which is the driving force if they do correlate with each other.

Concerning the pairing mechanism/symmetry of superconductivity, there have been many theoretical proposals, and only a number of them are mentioned here. A few theories suggest the so-called extended s-wave pairing, in which the gaps on the different Fermi surface sheets[34,35] have different signs, as a result of the particular structure of the pairing function $V_{k\acute{k}}$. Some proposals focus on the two-electron elliptic Fermi pockets around the M point. For example, based on a first-principles calculation Zhang et al.[36] suggest that the doping dependence of the superconducting transition temperature is driven by the orbital degeneracy of the two electron pockets. The prominent role of the ferromagnetic Hund's coupling, which is believed to be strong in Fe d-orbitals, is emphasized by a number of theories. For example, Dai et al. suggested a novel way of pairing: two electrons of a Cooper pair may reside in different electron pockets, which result in a pairing state of spin triplet, orbital singlet, and even parity in momentum space.[37] On the other hand, Lee and Wen suggested that the Hund's coupling may result in intraband p-wave pairing, which is believed to be more stable in the presence of band splitting.[38] Other mechanisms are more or less rooted in the electron–hole symmetry and assume the inter- or intraband fluctuation as the pairing media.[39,40] With limited information on the microscopic details of the system from the experimental side, it is still too early to have a conclusive answer to the pairing mechanism question. On the

other hand, based on symmetry considerations, a number of theories can already derive certain general properties of superconductivity, even without microscopic knowledge of the system.[41–43]

Experimentally, the first piece of indirect evidence for a nodal gap came from our low-temperature specific heat measurements, which exhibit a non-linear field dependence of the specific heat coefficient γ.[20] The point contact tunneling data show a strong zero bias conductance peak (ZBCP), which diminishes with both increasing temperature or magnetic field. This ZBCP is normally interpreted as the electron tunneling through a surface between a normal metal and a nodal superconductor.[44] The muon-spin-relaxation (μSR) experiments commonly point to the fact that the data may be interpreted as a ''dirty'' d-wave.[45,46] Another interesting discovery that is consistent with the early Hall effect measurement is that the superfluid density is indeed low and it seems to follow the well-known Uemura plot,[47] that is, $T_c \propto \rho_s$. If this is truly the case, the T_c can be further increased by having double layers. Recent NMR measurements suggest that the spin-singlet pairing, and $1/T_1T$ which measures the quasiparticle density of states in the superconducting state, exhibits a power law behavior that suggests a line node.[48,49] Therefore, from an experimental point of view, the d-wave pairing symmetry may gradually win more weight. However, recent measurements on the (Ba, K)Fe_2As_2 superconductors seem to support an s-wave gap symmetry.

5. A Brief Comparison between Iron-based and Cuprate Superconductors

Table 1 gives a preliminary comparison between the cuprates and the iron-based superconductors. All the parameters are quoted from the recent literature or from our measurements.

One can see that the iron-based system has many similarities with the cuprate system. Although it is still too early to say that they have the same mechanism, this may be confirmed when more refined data are obtained from single crystal samples.

Table 1. Comparison between iron-based and cuprate superconductors.

Features	Iron-based	Cuprate	Remark
H_{c1}	ca. 50–800 Oe	ca. 100 Oe	Only the ReFe AsO phase has low H_{c1} and follows the Uemura plot. The Ba-122 phase has a high H_{c1}
H_{c2}	$H_{c2}(H\|\|c)$: ca. 70 T $H_{c2}(H\|\|ab)$: ca. 300 T [a]	$H_{c2}(H\|\|c)$: 100 T $H_{c2}(H\|\|ab)$: 300–600 T for YBCO	The high H_{c2} values in both systems indicate a strong pairing gap.
Anisotropy $\Gamma = (m_c/m_{ab})^{1/2}$	4–5[a]	7–15 for YBCO 100 for Bi-2212	The single-layered iron-based system thus far exhibits an anisotropy even smaller than YBCO.
Gap	ca. 4 meV for LaFeAsOF ca. 7–10 meV for Nd,Pr,SmFeAsOF $2\Delta/k_BT_c = 3.5$–4.5	$2\Delta/k_BT_c = 8$? Not clear yet, complicated by the pseudogap	The phase fluctuation seems not that strong in the iron-based system, but this contradicts the fact of low superfluid density.
Gap symmetry	more experiments support singlet and nodal gap, less for s-wave or p-wave gaps	Mainly d-wave	Experimental data so far are highly controversial: some support gap with line nodes, some support s-wave gap.
Superfluid density	Low	Low	Phase fluctuation is clear in cuprates, seems not that strong in iron-based superconductors.
Coherence length	$\xi_{ab} = 15$–25 Å $\xi_c = 5$–10 Å for NdFeAsOF ($T_c = 52$ K)	$\xi_{ab} = 10$ Å $\xi_c = 2$–3 Å for YBCO ($T_c = 90$ K)	In both systems ξ is small, reflecting a small condensation energy-induced vortex pinning effect.
Penetration depth	$\lambda_{ab} = 1000$–2000 Å $\lambda_c = 3000$–5000 Å	$\lambda_{ab} = 1000$–2000 Å $\lambda_c = 3000$–6000 Å for YBCO	In both systems, λ is large, reflecting a small superfluid density.
Parent phase	AF or SDW order	AF order with a magnetic moment of 0.5 μ_B and J of ca. 0.13 meV	Values share some similarities. Superconductitivity can be induced by suppressing AF order.
Band	Multiband	Single-band for hole doping. Multiband for electron doping.	The iron-based superconductor more resembles the electron-doped cuprate

[a] Evaluated from $NdFeAsO_{0.88}F_{0.12}$ data near T_c.

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6. Perspectives and Concluding Remarks

Clearly, the new iron-based superconductors provide a new platform for research of superconductivity that is probably unconventional in nature. The high upper critical field, relatively small anisotropy, and longer coherence length (compared with the cuprate superconductors) make the materials very encouraging for applications. From the presented data we already know that the critical fields are much larger than that of MgB_2. Concerning the mechanism, it is highly desirable to know whether the AF order is a common feature for all systems in the undoped case, and whether or not the AF fluctuation has anything to do with the superconductivity. From the experimental data obtained up to now, d-wave pairing or a complex pairing symmetry that contains d-wave components may get more weight. More refined data from single crystals will clarify all these and illustrate what the Fermi surface looks like and how it evolves with the doping. An exploration into hole-doped materials and multilayer systems is encouraged to find new superconductors with higher T_c values.

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