Least momentum space frustration as a condition for a ‘high $T_c$ sweet spot’ in iron-based superconductors

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Received 2 April 2012, in final form 8 May 2012
Published 17 July 2012
Online at stacks.iop.org/SUST/25/084004

Abstract
In the present paper, we describe how the band structure and the Fermi surface of iron-based superconductors vary as the Fe–As–Fe bond angle changes. We discuss how these Fermi surface configurations affect the superconductivity mediated by spin fluctuations, and show that, in several situations, frustration in the sign of the gap function arises due to the repulsive pairing interactions that requires a sign change of the order parameter. Such a frustration can result in nodes or very small gaps, and generally works destructively against superconductivity. Conversely, we propose that the optimal condition for superconductivity is realized for the Fermi surface configuration that gives the least frustration while maximizing the Fermi surface multiplicity. This is realized when there are three hole Fermi surfaces, where two of them have $d_{x^2−y^2}$ orbital character and one has $d_{x^2−y^2}$ for all $k_z$ in the three-dimensional Brillouin zone. Looking at the band structures of various iron-based superconductors, the occurrence of such a ‘sweet spot’ situation is limited to a narrow window.

(Some figures may appear in colour only in the online journal)

1. Introduction

The discovery of high temperature superconductivity in the iron-based superconductors [1] has attracted much attention in many aspects. Not only the high $T_c$ itself, but also a number of experiments indicating non-universality of the superconducting gap function, such as sign reversal, anisotropy, or the presence of nodes, suggest an unconventional pairing mechanism. The most probable candidate for such an unconventional mechanism is the pairing mediated by spin fluctuations, where the superconducting gap changes sign between the disconnected Fermi surfaces, namely, the so-called s± pairing [2, 3].

Back in 2001, one of the present authors proposed that spin fluctuation mediated pairing in systems with nested disconnected Fermi surfaces may give rise to a very high $T_c$ superconductivity [4, 5]. The idea is that the repulsive pairing interaction mediated by spin fluctuations can be fully exploited without introducing nodes of the superconducting gap on the Fermi surfaces. Although the Fermi surface of the iron-based superconductors does resemble the proposed Fermi surface configuration, there are some important differences. The first is that the Fermi surface in the iron-based superconductors has multiple orbital character, and the second is that there can be frustration in the sign of the gap function, which can give rise to gap nodes on the Fermi surface. In the present paper, we focus on this gap-sign frustration problem, and propose that the Fermi surface configuration that gives the least frustration provides the optimal condition for high $T_c$ in the iron-based superconductors. In [6], two of the present authors pointed out that maximizing the Fermi surface multiplicity leads to the optimization for $T_c$, but considering the frustration problem studied in the present paper, the ‘sweet spot’ for high $T_c$ is further limited to a narrow window of the Fermi surface configuration.
2. Typical band structure and Fermi surfaces

Let us first describe the band structure of LaFeAsO. LaFeAsO takes a layered structure, where Fe atoms form a square lattice in each layer, sandwiched by As atoms with tetrahedral coordination. We use the band structure obtained from first principles [7] to construct the maximally localized Wannier functions [8]3. These Wannier functions have five orbital symmetries (d\text{xy}, d\text{yz}, d\text{zx}, d\text{yz}, d\text{zx}), where X, Y, Z refer to those for the unit cell with two Fe sites as shown in figure 1(a). The two Wannier orbitals in each unit cell are equivalent in that each Fe atom has the same local arrangement of other atoms. We can then take a unit cell that contains only one orbital per symmetry by unfolding the Brillouin zone, and an effective five-band model on a square lattice is obtained, where x and y axes are rotated by 45° from X–Y.

In figure 1(b) (right), the Fermi surface for 10% electron doping is shown in the two-dimensional unfolded Brillouin zone. The Fermi surface consists of four pieces: two concentric hole pockets (denoted here as \(\alpha_1\), \(\alpha_2\)) centered around \((k_x, k_y) = (0, 0)\), and two electron pockets around \((\pi, 0)(\beta_1)\) or \((0, \pi)(\beta_2)\), respectively. Besides these pieces of the Fermi surface, there is a portion of the band near \((\pi, \pi)\) that touches the \(E_F\), so that the portion acts as a ‘quasi-Fermi surface’ \((\gamma)\) around \((\pi, \pi)\). In regards to the orbital character, \(\alpha\) and portions of \(\beta\) near Brillouin zone edge have mainly \(d\text{xy}\) and \(d\text{yz}\) orbital character, while the portions of \(\beta\) away from the Brillouin zone edge and \(\gamma\) have mainly \(d\text{xz}\) orbital character (see also figure 1(c)).

3 The Wannier functions are generated by the code developed by Mostofi A A, Yates J R, Marzari N, Souza I and Vanderbilt D.

3. Fermi surface appearance/disappearance with the bond angle

The band structure and the Fermi surfaces of the iron-based superconductors are sensitive to the lattice structure. In this section, we consider hypothetical lattice structures of LaFeAsO, where we fix the bond length at its original length and vary the bond angle \(\alpha\) (figure 2(a)) [6]. We first neglect the three-dimensionality (out-of-plane hoppings) and consider a two-dimensional model. The Fermi surface is obtained for 10% electron doping. When the bond angle is large, two hole Fermi surfaces, \(\alpha_1\) and \(\alpha_2\), are present around the wavevector \((0, 0)\), while the \(\gamma\) around \((\pi, \pi)\) is missing. As \(\alpha\) decreases, the \(\gamma\) Fermi surface appears around \((\pi, \pi)\) and there are now three hole Fermi surfaces. This appearance of the additional Fermi surface has been noticed as an effect of increasing the pnictogen height [9, 10, 16, 11, 12]. When \(\alpha\) is further reduced, the \(\alpha_1\) Fermi surface disappears, and again the Fermi surface multiplicity reduces to two, but in this case one around \((0, 0)\) and another around \((\pi, \pi)\). Such a disappearance of the \(\alpha_1\) hole Fermi surface was first noticed in the band calculation of Ca$_2$Al$_4$O$_6$Fe$_2$As$_2$ [13] by Miyake et al in [14, 15]. This material indeed has very small bond angle of about 102°.

Figure 3 explains schematically the band structure/Fermi surface variation with the bond angle reduction [14]. As the bond angle is reduced, the \(X^2–Y^2\) band below the Fermi level around \((\pi, \pi)\) rises up, while another \(X^2–Y^2\) portion of the band above the Fermi level around \((0, 0)\) comes down. This band deformation can be understood from figure 1(c), where the \(X^2–Y^2\) portion of the band for the original LaFeAsO is extracted. In the tight binding picture, the energy difference between the wavevectors \((0, 0)\) and \((\pi, \pi)\) is roughly equal to \(8t_{X^2–Y^2}\), where \(t_{X^2–Y^2}\) is the nearest neighbor hopping of the \(X^2–Y^2\) orbital. As the bond angle is reduced, the contribution to \(t_{X^2–Y^2}\) from the Fe – As – Fe path decreases, while that from the direct Fe – Fe path increases. The two contributions have opposite signs, so that the reduction of the bond angle results in a decrease of \(t_{X^2–Y^2}\) [14].

When the bottom of the upper \(X^2–Y^2\) portion sinks below the \(XZ/YZ\) bands around \((0, 0)\), a band structure reconstruction takes place, and now the two bands that are degenerate at \((0, 0)\) repel each other (one going up, the
Figure 2. The band structure and the Fermi surface of hypothetical lattice structures of LaFeAsO with varying Fe–As–Fe bond angle while fixing the Fe–As bond length to its original value. The position of the bands having $X^2-Y^2$ and $XZ/YZ$ orbital characters at (0, 0) is indicated.

other going down) as the wavevector moves away from (0, 0) (figure 3(c)). In this situation, the band below these two bands has $X^2-Y^2$ character near (0, 0), and changes its character to $XZ/YZ$ as the wavenumber increases. Therefore, just before the inner hole ($\alpha_1$) Fermi surface disappears, it has strong $X^2-Y^2$ character.

4. Frustration in the superconducting gap in the absence of the $\gamma$ Fermi surface: a brief review

Having understood the Fermi surface variation with the bond angle, we now consider how this should affect the gap function of the superconductivity mediated by spin fluctuations. The pairing interaction mediated by spin fluctuations is repulsive, so the superconducting gap has an tendency to change its sign between the initial and final wavevectors of the pair scattering. Another important point is that spin fluctuations develop at wavevectors that bridge the portions of the Fermi surface having the same orbital character [16].

In the presence of the $X^2-Y^2$ originated $\gamma$ and $XZ/YZ$ originated $\alpha_1$ and $\alpha_2$ Fermi surfaces (case (b) in figure 3), it is known that the $\gamma-\beta$ interaction among portions having $X^2-Y^2$ character and the $\alpha-\beta$ interactions among portions with $XZ/YZ$ character dominate, and the superconducting gap is fully open on all the Fermi surfaces while changing its sign as $+, -$, $+$ along $\alpha$, $\beta$, $\gamma$ Fermi surfaces. This is the fully gapped $s\pm$ gap [16–18].

On the other hand, in the absence of the $\gamma$ Fermi surface (case (a) in figure 3), the pairing interaction between the $X^2-Y^2$ portions of the $\beta$ Fermi surfaces and that between the $XZ/YZ$ portions of the $\alpha$ and $\beta$ Fermi surfaces results in a frustration in the sign of the gap function. This situation has been studied in several previous papers [19, 16–18]. This can result in either nodal $s\pm$ wave or $d$ wave pairings, where the nodes of the gap go into the $\beta$ Fermi surface in the former, and $\alpha$ in the latter. A schematic figure summarizing the above is shown in figure 4 [16]. As was shown in [16], this frustration effect degrades $T_c$ of the superconductivity, so that the lattice structure acts as a switch between high $T_c$ nodeless and low $T_c$ nodal superconductivity.

5. Frustration in the case of a nearly vanishing $\alpha_1$ Fermi surface

Here we discuss another situation where frustration arises in the sign of the superconducting gap function. As mentioned in section 3, the $X^2-Y^2$ orbital character strongly mixes into the $\alpha_1$ Fermi surface just before the Fermi surface vanishes as the bond angle is reduced. In this situation, there are now $X^2-Y^2$ components on $\alpha_1$, $\beta$, and $\gamma$ (if present) Fermi surfaces. Since these Fermi surfaces interact with repulsive pairing interactions, once again, a frustration arises in the sign of the superconducting gap (figure 5(a)). In addition to this, there can also be some $XZ/YZ$ component remaining in the $\alpha_1$ Fermi surface, and this portion tends to change the sign from
Figure 3. A schematic figure for the band structure variation with the bond angle $\alpha$. The black (red) portions indicate the bands with strong $X^2−Y^2$ ($XZ/YZ$) orbital character.

Brillouin zone. In this angle regime, the $\alpha_1$ Fermi surface is barely present, and it is indeed composed of mixed $X^2−Y^2$ and $XZ/YZ$ orbital components. We apply the fluctuation exchange approximation to this model [20], and obtain the eigenfunction (gap function) of the linearized Eliashberg equation, as was done in [6]. In figure 6, we show the gap function for the two angles for 10% electron doping and temperature $T=0.01\text{ eV}$. It can be seen that the magnitude of the gap on the $\alpha_1$ Fermi surface is very small, and its sign actually changes as the bond angle is varied, reflecting the frustration.

6. Effect of three-dimensionality

So far, the argument has been based on a purely two-dimensional system, where the band structure does not depend on the wavevector along the $z$ direction, $k_z$. When the systems exhibit some three-dimensionality, the above mentioned variation of the Fermi surface configuration against the bond angle depends on $k_z$. This is shown schematically in figure 7. In systems with moderate three-dimensionality, the position of the $X^2−Y^2$ band at $(k_x,k_y,k_z)=(0,0,\pi)$ is lower than at $(0,0,0)$, namely, there is a large dispersion along $(0,0,0)−(0,0,\pi)$. This is because of the strong hybridization between the Fe $d_{X^2−Y^2}$ and the As $p$ orbitals. (See for example the ‘upper $X^2−Y^2$’ along $Z−\Gamma$ in the band structure of LiFeAs given in figure 8.) Therefore, the orbital character change of the $\beta$ Fermi surfaces (figure 5(b)), making it another possible factor for the frustration.

To actually see this frustration effect, here we consider hypothetical lattice structures of Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$ [13], where we set the bond angle to $108^\circ$ and $109^\circ$, and construct a two-dimensional five orbital model in the unfolded Brillouin zone.
Figure 6. The gap function obtained by FLEX for the hypothetical lattice structures of Ca₄Al₂O₆Fe₂As₂. The bond angle $\alpha$ is set to 108° or 109°, while the bond length is fixed to the original value.

Figure 7. A schematic figure representing the band structure and the hole Fermi surface variation against the bond angle for moderately three-dimensional systems. (a)–(d) correspond to the configurations shown in figure 3. The ‘sweet spot’ is restricted to the regime where the $X^2-Y^2$ originated $\gamma$ Fermi surface is effective and the $\alpha_1$ Fermi surface has $XZ/YZ$ character for all $k_z$. The lower panels are schematic figures of the $\alpha_1$ Fermi surface for the bond angle depicted by the dashed line (right) or the dash-dotted line (left) in the upper panel. See the text for details.

the $\alpha_1$ Fermi surface and its disappearance first starts at $k_z = \pi$ (around Z point) as the bond angle is reduced, and ends at $k_z = 0$ (around $\Gamma$ point). For instance, let us first consider the bond angle indicated by the dashed line in figure 7. Here, $k_z$ around $\pi$ is in ‘regime (c)’, which means that the orbital character of the $\alpha_1$ hole Fermi surface has changed to $X^2-Y^2$. 
Figure 8. First principles band structure for various iron-based superconductors. $k_z = 0$ and $\pi$ planes are shown. Note that the original (folded) Brillouin zone is adopted here, as opposed to the unfolded Brillouin zone taken in figures 2 and 3. The red solid circles (or ellipse) indicates the portion where the two bands degenerate at wavevector $(k_x, k_y) = (0, 0)$ repel with each other as the wavenumber increases, while the dashed blue circles are the portions where the two bands degenerate at $(0, 0)$ both form a hole Fermi surface. For LaFeAsO, the three-dimensional band along $\Gamma$-$Z$ having $X^2-Y^2$ character (green dashed ellipse) stays above the degenerate $XZ/YZ$ bands (red solid). For LiFeAs, the $X^2-Y^2$ band corresponding to that around $(0, 0)$ in figure 3 is the upper $X^2-Y^2$, while that around $(\pi, \pi)$ in figure 3 is the lower $X^2-Y^2$. 
On the other hand, the \( k_z \) region smaller than \( k_z \approx \pi/2 \) is in ‘regime (b)’, so that the \( \alpha_1 \) Fermi surface still has \( XZ/YZ \) character for this \( k_z \) region. A schematic sketch of the \( \alpha_1 \) Fermi surface in this case is shown in the lower right panel of figure 7. Although this Fermi surface is still two-dimensional in the topological sense, we can call it a ‘three-dimensional’ Fermi surface in the sense that the orbital character changes with \( k_z \). If we reduce the bond angle to that indicated by the dash-dotted line, \( k_z \) around \( \pi \) is now in ‘regime (d)’, which indicates that the \( \alpha_1 \) Fermi surface is lost there. The \( \alpha_1 \) is still present around \( k_z = \pi/2 \), but the orbital character there is now \( X^2-Y^2 \), and the \( \alpha_1 \) with mainly \( XZ/YZ \) character remains only around \( k_z = 0 \). In this case, the Fermi surface is now truly three-dimensional, as shown in the lower left panel of figure 7. In the situations described above, the frustration effect should be present around the top and the bottom portions of the three-dimensional Fermi surface because the orbital character has changed to \( X^2-Y^2 \) around those portions.

In fact, the above-mentioned three-dimensionality of the band structure is rather common for the iron-based superconductors. This has been discussed in detail in [12]. In figure 8, we show the band structure of various iron-based superconductors for \( k_z = 0 \) and \( \pi \) planes (in the original folded Brillouin zone) calculated using the Wien2k package [21]. One can see that, except for LaFeAsO, the two bands degenerate at \( Z \) point \((0,0,\pi)\) repel each other as the wavevector moves away from \( Z \) in the \( k_z = \pi \) plane. This means that configurations (c) or (d) in figure 3 are realized near the \( k_z = \pi \) plane. On the other hand, in FeSe, LiFeAs and BaFe\(_2\)As\(_2\), the band structure near the \( k_z = 0 \) plane takes configuration (b), so that in these materials, the band structure near the Fermi level is indeed \( k_z \) dependent, namely, three-dimensional. To be more precise, the three-dimensionality of the Fermi surface appears in various ways depending on the materials. For the case of LiFeAs, the ‘upper’ \( X^2-Y^2 \) band crosses the Fermi level between \( Z-\Gamma \), indicating that the \( \alpha_1 \) Fermi surface has the form given in the lower left panel of figure 7. In the case of 122 systems such as BaFe\(_2\)As\(_2\), the \( Z^2 \) band hybridized with the \( X^2-Y^2 \) band gives rise to a three-dimensional Fermi surface around the \( Z \) point (shown by the dashed curve in the lower right panel of figure 7), and this, continuously connected to the \( XZ/YZ \) portion, gives the well known warped hole Fermi surface.

Conversely, the 1111 systems can be considered as somewhat exceptional in that, although the three-dimensional \( X^2-Y^2 \) band does exist (dashed green circle in LaFeAsO in figure 8), it does not come down too rapidly before the \( X^2-Y^2 \) -\( \Gamma \) Fermi surface appears. In fact, NdFeAsO, one of the materials having the highest \( T_c \), seems to have the least frustration from the above mentioned viewpoint. Namely, as seen in the band structure shown in figure 9 in the unfolded Brillouin zone [16], the \( \gamma \) Fermi surface is present but the three-dimensional \( X^2-Y^2 \) band still lies above the \( XZ/YZ \) bands for all \( k_z \). On the other hand, from the comparison to the band structure of LaFeAsO (figure 1(b)), it can be seen that as a trade-off for the appearance of the \( \gamma \) Fermi surface around \((\pi,\pi)\), the three-dimensional \( X^2-Y^2 \) band along \((0,0,\pi)\) -\( (0,0,\pi) \) has certainly come down very close to the \( XZ/YZ \) band, and for smaller bond angle or higher pnictogen position, the band reconstruction and thus the frustration starts to take place. Therefore, from the present viewpoint, the ‘sweet spot’ for high \( T_c \) is restricted to a narrow window, which may explain the experimental observations [22, 23].

7. Conclusion

As described in the present paper, the multiplicity of the Fermi surfaces and even their orbital characters change as the lattice structure is varied. Because of the presence of multiple Fermi surfaces and the repulsive pairing interaction mediated by the spin fluctuations, frustration can arise in the sign of the gap on the disconnected Fermi surfaces. This can result in nodal structures in the gap function, and should generally work destructively against superconductivity. Conversely, from the present viewpoint, the optimal situation for the spin fluctuation mediated superconductivity is when the \( X^2-Y^2/\gamma \) Fermi surface is effective and the \( \alpha_1 \) Fermi surface has \( XZ/YZ \) orbital character for all \( k_z \). As far as the first-principles band calculations are concerned, this situation is realized in

![Figure 9. The band structure of NdFeAsO in the unfolded Brillouin zone. The Fermi level is for 10% electron doping. The two conditions for least frustration are satisfied: (i) the \( X^2-Y^2 \) Fermi surface is present around the wavevector \((\pi,\pi)\), and (ii) the three-dimensional \( X^2-Y^2 \) band from \((0,0,\pi)\) to \((0,0,\pi)\) does not intersect the \( XZ/YZ \) bands for all \( k_z \).](https://example.com/figure9.png)
limited materials such as NdFeAsO and SmFeAsO, where the highest $T_c$ among the iron-based superconductors is observed experimentally. From this viewpoint, further band structure calculation studies may give useful information for obtaining new related superconductors with higher $T_c$.

Acknowledgments

We acknowledge C H Lee, T Miyake, O K Andersen, H Mukuda, H Kinouchi, and Y Kitaoka for illuminating discussions.

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