Maximizing Fermi surface multiplicity optimizes superconductivity in iron pnictides

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We study the condition for optimizing superconductivity in the iron pnictides from the lattice structure point of view. Studying the band structure of the hypothetical lattice structure of LaFeAsO, the hole Fermi surface multiplicity is found to be maximized around the Fe-As-Fe bond angle regime where the arsenic atoms form a regular tetrahedron. Superconductivity is optimized within this three hole Fermi surface regime, while the stoner factor of the antiferromagnetism has an overall tendency of increasing upon decreasing the bond angle. Combining also the effect of the varying the Fe-As bond length, we provide a guiding principle for obtaining high Tc.

The discovery of superconductivity in the iron pnictides and its Tc up to 55K has given great impact to the field of condensed matter physics. To attain even higher Tc, the investigation on the condition for optimizing the superconductivity is highly desired. In this context, much attention has been paid to the correlation between Tc and the lattice structure from the early stage. In particular, Lee et al. have shown that Tc systematically varies with the Fe-Pn-Fe (Pn=pnictogen) bond angle, and takes its maximum around 109 degrees, at which the pnictogen atoms form a regular tetrahedron (“Lee’s plot”). Theoretically, we have previously explained this lattice structure effect within a spin fluctuation mediated pairing theory on a five orbital model, and pointed out that superconductivity is strongly affected by the Fermi surface around the wave vector (, ) in the unfolded Brillouin zone. This Fermi surface has been found to be controlled by the pnictogen height hPn measured from the iron plane. When the pnictogen is at high positions, a hole Fermi surface originating from the X - Y2 orbital appears around (, ) and the spin fluctuation arising from the interaction between electron and hole Fermi surfaces gives rise to a high Tc s±-wave paring, where the gap is fully open but changes sign between electron and hole Fermi surfaces as was first proposed in ref.6. When the pnictogen position is low, the X - Y2 band sinks below the Fermi level, and a low Tc nodal s±-wave paring or d-wave paring takes place. Thus, the appearance of the X - Y2 Fermi surface around (, ) (which we call ) is favorable for superconductivity, so that the higher pnictogen height results in a higher Tc within this scenario. In ref.4, the saturation of Tc in LnFeAsO in the bond angle regime of less than 109 degrees has been attributed to the decrease of the lattice constant a, which is found to be unfavorable for superconductivity due to the decrease of the density of states.

However, it is becoming clearer that materials having very large hPn of > 1.5Å do not have high Tc. Recently, for example, CaAl2O6Fe2As2 and CaAl2O6Fe2P2, which are variations of the materials having perovskite block layers (21311 systems), have been found to exhibit superconductivity of 28.3K and 17.1K, respectively. These materials have very large hPn (~1.5Å), small lattice constant a (~3.71Å), and consequently a very small bond angle (~102 degrees). Such a high pnictogen position should give a robust Fermi surface around (, ) and thus strong spin fluctuations, and it seems difficult to understand the low Tc even if the effect of the small lattice constant is taken into account. In this context, a recent band structure calculation of CaAl2O6Fe2As2 have found an interesting feature; one of the hole Fermi surfaces around (0,0) is missing, resulting in two hole Fermi surfaces. This is found to be due to the very small bond angle.

Given this background, in the present Letter, we study the condition for optimizing superconductivity in the iron pnictides, varying hypothetically the lattice structure of LaFeAsO. In varying the bond angle α in a wide range while fixing the bond length, the number of hole Fermi surfaces changes from two to three as α is decreased (as discussed as the effect of the pnictogen height in previous studies), but as α is decreased even further, one of the hole Fermi surfaces around (0,0) in the unfolded Brillouin zone disappears just as in CaAl2O6Fe2As2. Consequently, the number of hole Fermi surfaces is maximized around the bond angle where the arsenic atoms form a regular tetrahedron. We note that the disappearance of the hole Fermi surface was not considered in our previous study since such a small bond angle regime was not considered there. Applying fluctuation exchange (FLEX) method to the model, we find that the superconductivity is optimized in the bond angle regime where there are three hole Fermi surfaces. Interestingly, the tendency toward magnetism (the strength of the spin fluctuation at zero energy) is not necessarily correlated with the hole Fermi surface multiplicity, and therefore the correlation between superconductivity and the strength of the spin fluctuations is complicated. We also vary the Fe-As bond length while fixing the bond angle, and find that Tc and the bond length is positively correlated mainly due to the increase of the density of states without affecting the Fermi surface.

We first calculate the band structure of hypo-
length and vary the bond angles where we fix the bond length at its original theoretical lattice structures of LaFeAsO using the Quantum-ESPRESSO package [13], and construct five band models [9] from maximally localized Wannier functions [16], where we fix the bond length at its original length [1] and vary the bond angle $\alpha$ (Fig. 1a) as has been done for Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$ by Miyake et al. [13]. The Fermi surface is obtained for the band filling (=number of electrons per site) of $n = 0.1$. When the bond angle is large, two hole Fermi surfaces, $\alpha_1$ and $\alpha_2$ are present around the wave vector $(0, 0)$. As $\alpha$ decreases, the $\gamma$ Fermi surface appears around $(\pi, \pi)$, and we now have three hole Fermi surfaces. This is what has been noticed as an effect of increasing the pnictogen height [4, 17, 18]. As we decrease $\alpha$ even further, the $\alpha_1$ Fermi surface disappears, and again there are only two hole Fermi surfaces but in this case one around $(0, 0)$ and another around $(\pi, \pi)$. So the tendency found in Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$ holds also for LaFeAsO. However, the range of the bond angle in which there are three hole Fermi surfaces (three hole Fermi surface zone in Fig. 1c) turns out to be much larger for LaFeAsO than for Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$. We find that the three hole Fermi surface zone tends to be small for materials with large $c$-axis length. Another point that should be mentioned is that the total band width is almost unchanged as the bond angle is varied.

Having understood the effect of the bond angle on the band structure, we next fix $\alpha = 108$ and vary the bond length (Fig. 1b). The band width is reduced upon increasing the bond length as expected, but interestingly, the Fermi surface is barely affected. In fact, we find that multiplying the bands for different bond length by certain factors gives nearly the identical band structures, meaning only the band width, but not the shape, is affected by the bond length.

We now move on to the FLEX calculation. We consider the standard multi-orbital interactions (intraorbital and interorbital repulsion, Hund’s coupling, and the pair hopping interaction), and apply FLEX. In FLEX, bubble and ladder type diagrams consisting of renormalized Green’s functions are summed up to obtain the susceptibilities, which are used to calculate the self energy. The renormalized Green’s functions are then determined self-consistently from the Dyson’s equation. The obtained Green’s function is plugged into the linearized Eliashberg equation, whose eigenvalue $\lambda$ reaches unity at the superconducting transition temperature $T = T_c$. Also, in order to investigate the correlation between superconductivity and magnetism, we obtain the Stoner factor $a_S$ of the antiferromagnetism at the wave vector $(\pi, 0)$ in the unfolded Brillouin zone, which is defined as the largest eigenvalue of the matrix $U\chi(0) = (\pi, 0), i\omega_n = 0$, where $U$ is the interaction and $\chi(0)$ is the irreducible susceptibility matrices, respectively. This value monitors the tendency towards stripe type antiferromagnetism and the strength of the spin fluctuations at zero energy. Since the three dimensionality is not strong in LaFeAsO, we take a two dimensional model where we neglect the out-of-plane hopping integrals, and take $32 \times 32$ $k$-point meshes and 4096 Matsubara frequencies.

As for the electron-electron interaction values, we adopt the orbital-dependent interactions as obtained from first principles calculation in ref. [19] for LaFeAsO, but multiply all of them by a constant reducing factor $f$. The reason for introducing this factor is as follows. As has been studied in refs. [20, 22] the FLEX for models obtained from LDA calculations tend to overestimate...
the effect of the self-energy because LDA already partially takes into account the effect of the self-energy in the exchange-correlation functional. When the electron-electron interactions as large as those obtained from first principles are adopted in the FLEX calculation, this double counting of the self-energy becomes so large that the band structure largely differs from its original one. In such a case, the spin fluctuations will develop around the wave vector \((\pi, \pi)\) rather than \((\pi, 0)\) in the unfolded Brillouin zone, which is in disagreement with the experiments. In the present study, we therefore introduce the factor \(f\) so as to reduce the electron-electron interactions. We vary \(f\) within the range where the spin fluctuations develop around \((\pi, 0)\) and not \((\pi, \pi)\). This approach would have problems in accuracy if we try to estimate the absolute values of \(T_c\) (the approach should underestimate \(T_c\)), but instead here we calculate the eigenvalue of the Eliashberg equation and the stoner factor at a fixed temperature, thereby comparing the relative strength towards superconducting/magnetic instability among various lattice structures.

In Fig.2(a), we plot the eigenvalue of the Eliashberg equation \(\lambda\) for the \(s\pm\)-wave superconductivity and the antiferromagnetic stoner factor \(\alpha_s\) at \((\pi, 0)\) as functions of the bond angle while fixing the bond length at its original value. In the large angle regime, the eigenvalue of the Eliashberg equation remains relatively small. As the angle is decreased, the \(\gamma\) Fermi surface around \((\pi, \pi)\) becomes effective and the eigenvalue becomes large. This enhancement of superconductivity is correlated with the presence/absence of nodes in the superconducting gap on the electron Fermi surface (not shown) as has been studied previously [4, 7, 23, 24]. As the angle is decreased further, superconductivity tends to be suppressed, although the variation of the eigenvalue is not monotonic. Thus, the superconductivity is optimized around 110 degrees, which is in agreement with the Lee’s plot [3]. The fact that superconductivity is optimized for maximum Fermi surface multiplicity can be considered as natural since larger number of Fermi surfaces gives rise to larger number of pair scattering channels as shown in Fig.3(a). We stress here that due to the quasi-two-dimensionality, the density of states is not affected by the size of the Fermi surface, so two small Fermi surfaces is more favorable than one large Fermi surface regarding the number of pair scattering channels.

On the other hand, the situation is not so simple since the stoner factor at \((\pi, 0)\), which measures the spin fluctuations and thus should also affect \(T_c\), is found to be not necessarily correlated with the multiplicity of the Fermi surface. In the large angle regime of \(115 < \alpha < 120\), superconductivity begins to grow as the bond angle is decreased, but the change in the stoner factor is small. This can be interpreted as follows. When the \(X^2 - Y^2\) band around \((\pi, \pi)\) sits somewhat away from the Fermi level, the interaction between this band and the electron Fermi surfaces gives rise to spin fluctuations with finite energy that contributes to the pairing interaction but has small contribution to spin fluctuations with nearly zero energy. This can be related to the NMR experiments in LaFeAsO, which do not find any enhancement in the low energy spin fluctuations for the optimally doped systems [25–28]. In the intermediate angle regime (105 < \(\alpha\) < 115),
where the three hole Fermi surfaces coexist, superconductivity and the Stoner factor is correlated, but in the smaller angle regime ($\alpha < 105$), where $\alpha_1$ Fermi surface disappears, there is an complicated correlation between the two. In this regime, the effect of the $\gamma$ Fermi surface becomes even larger, overcoming the disappearance of the $\alpha_1$ Fermi surface and resulting in an increase of the Stoner factor. This works positively for superconductivity, and in fact, $\lambda$ is slightly enhanced in the angle regime $97 < \alpha < 100$ for $T = 0.005\text{eV}$. However, in the smaller bond angle regime, $\alpha_S$ continues to increase, but $\lambda$ is degraded. This is mainly because all the Fermi surfaces, including the electron ones, become large in this regime. Namely, the relative size of the Fermi surface with respect to the spread of the spin fluctuations $\Delta Q$ becomes large, so that the pair scattering processes occur only between restricted portions of the Fermi surfaces, while when the size of the Fermi surfaces is comparable to $\Delta Q$, the pair scattering can occur between the entire regime, as shown in Fig.3(b). Thus, in the small angle regime, although the spin fluctuations at $(\pi,0)$ becomes stronger, superconductivity is degraded due to the disappearance of the $\alpha_1$ and the enlargement of other Fermi surfaces.

Finally we turn to the bond length dependence. In Fig.2(b), we plot the eigenvalue of the Eliashberg equation as a function of the bond length. The eigenvalue monotonically increases with the increase of the bond length. Since the change in the Fermi surface is small here, the enhancement of the superconductivity is mainly due to the increase of the density of states originating from the narrowing of the band width.

In Fig.2(c), we present a schematic $T_c$ diagram in the bond angle and bond length space. High $T_c$ is obtained when the bond angle is around 110 deg. (the regular tetrahedron angle) so that there are (nearly) three hole Fermi surfaces and also when the bond length is large so that the band width is narrow.

To summarize, we have studied the condition for optimizing superconductivity in the iron pnictides from the lattice structure point of view. As found in Ca$_4$Al$_2$O$_6$Fe$_2$AsO$_2$, the band structure of the hypothetical lattice structure of LaFeAsO also exhibits a disappearance of one of the hole Fermi surface in the small bond angle regime, and therefore, the hole Fermi surface multiplicity is maximized around the bond angle regime where the arsenic atoms form a regular tetrahedron. Superconductivity is optimized within this three hole Fermi surface regime, while the $(\pi,0)$ Stoner factor has an overall increasing tendency upon decreasing the bond angle. Consequently, the correlation between superconductivity and the spin fluctuations becomes rather complicated. Combining also the effect of varying the Fe-As bond length, we have provided a schematic $T_c$ diagram, which may give a guiding principle for obtaining higher $T_c$ materials.

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[30] This monotonic enhancement of $\lambda$ is restricted to the regime of $f$ where the spin fluctuations develop at $(\pi, 0)$. When large $f$ is taken, the spin fluctuation wave vector changes from $(\pi, 0)$ to $(\pi, \pi)$ as the bond length is increased, and in that case $\lambda$ takes a maximum value at a certain bond length.
[31] A recently discovered superconductor $K_xFe_2Se_2$ (J. Guo et al, Phys. Rev. B 82, 180520(R) (2010)) is also interesting from this viewpoint since the top of the three hole bands near $E_F$ are nearly degenerate and the band width is narrow.