Even Parity, Orbital Singlet and Spin Triplet Pairing for Superconducting $\text{La}(O_{1-x}F_x)\text{FeAs}$

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In the present paper, we propose the parity even, orbital singlet and spin triplet pairing state as the ground state of the newly discovered superconductor $\text{La}(O_{1-x}F_x)\text{FeAs}$. The pairing mechanism involves both the special shape of the electron fermi surface and the strong ferromagnetic fluctuation induced by Hund’s rule coupling. The special behavior of the Bogoliubov quasi-particle spectrum may lead to “Fermi arc” like anisotropy super-conducting gap, which can be detected by angle resolved photo emission (ARPES). The impurity effects are also discussed.

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Recent discovery of superconductivity in layered Fe-based compounds has attracted much attention. It has been reported that the transition temperatures $T_c$ of $\text{La}(O_{1-x}F_x)\text{FeAs}[1]$ is much lower than the other Fe-based superconductors with $\text{Sr}_2\text{RuO}_4$. There is a perfect nesting between the hole Fermi surface (FS) centered at Γ point and electron FS centered at M point. The density wave state at low temperature $T_c = 41$ K in $\text{CeO}_1-x\text{F}_x\text{FeAs}[2]$ and $T_c = 43$ K in $\text{SmO}_{1-x}\text{F}_x\text{FeAs}[3]$. This class of superconductors shows highly unusual properties, indicating possible unconventional non-BCS superconductivity.[4, 5]

The electronic band structure calculations for $\text{LaOF}_{\text{FeAs}}$ suggest the compound to be a semi-metal[6, 7, 8]. There is a perfect nesting between the hole Fermi surface (FS) centered at Γ point and electron FS centered at M point, which leads to a spin density wave state at low temperature $T_c = 26$ K in $\text{LaO}_{1-x}\text{F}_x\text{FeAs}[1]$. Superconductivity occurs when part of Fe$^{2+}$ ions are replaced by Fe$^+$, which removes the nesting. The layered Ni based compound is also superconducting (SC) although $T_c$ is much low[11]. This implies the importance of ferromagnetic (FM) fluctuation to the superconductivity. The similarity of Fe-based superconductors with $\text{Sr}_2\text{RuO}_4$ has suggested a possible spin triplet pairing. While the conventional s-wave BCS state is robust against non-magnetic disorder due to the Anderson theorem[11], the p-wave superconductivity of $\text{Sr}_2\text{RuO}_4$ is only observed in clean samples and is strongly suppressed by the non-magnetic impurity[12, 13]. The Fe-based superconductivity, on the other hand, does not require a clean sample and appears to be robust against disorder. Together with their high transitional temperatures, this raises an important and interesting question on the symmetry of the newly discovered Fe-based superconductivity.

In this Letter, motivated by the approximate two-fold degenerate electron FS revealed in the electronic structure calculations, we propose a spin triplet pairing with even parity for SC $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$. The pairing is due to the FM fluctuation between electrons in two different orbitals with almost degenerate bands. Our theory explains the robustness of superconductivity to the disorder in a spin triplet SC state, similar to the disorder effect to the spin singlet s-wave BCS superconductor. The splitting of the orbital degeneracy strongly suppresses the superconductivity. The high pressure reduces the splitting, and may further increase $T_c$ in $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$. The splitting of the degeneracy also leads to a pronounced $k$-dependence in the isotropic s-wave SC state, which may be tested in angle resolved photoemission spectra (ARPES).

We start from the special electronic structure of layered compound $\text{LaOF}_{\text{FeAs}}$. The Fe-ions forms a square lattice with two atoms in each unit cell. The distance of neighboring Fe atoms is rather short, so that the electron direct hoppings between Fe ions are important, similar to the elemental Fe, which is a FM metal. Due to the multiple d-orbitals, there are five FS with three hole-like cylinders around the Γ point and two electron-like cylinders around the M point of the Brillouine zone. Upon doping of F-atoms, the three hole-like FS shrink rapidly, while the two electron FS expand their areas. Therefore, it is reasonable to expect that the two bands of electron-like states are responsible for the superconductivity in $\text{La}(O_{1-x}F_x)\text{FeAs}$. Competing spin fluctuations exist in this compound. One is the anti-ferromagnetic spin fluctuations due to the nesting between the electron and the hole FS, which are connected by a commensurate $q$-vector. The other is the FM spin fluctuation likely due to Hund’s coupling. The presence of the nesting between electron and hole FS will induce a spin density wave instability. This is the 150K anomaly observed experimentally[4]. Doping F-ions destroys the spin density wave state and opens the door for superconductivity.

The itinerant ferromagnetism is an interesting but difficult problem in the condensed matter theory with a long history[12, 13, 14, 15]. One of the important issues is if the multi-band nature is necessary for the itinerant ferromagnetism. Both the analytical and numerical studies indicate that itinerant ferromagnetism is very difficult to ob-
tain in the single band system, unless the Fermi energy is close to a van hove singularity\cite{5,6}. Usually the mult bands are necessary to stabilize the FM phase, and the Hund’s rule coupling plays a crucial role. In LaOFeAs, the density of state is very low near the FS, as evidenced in both optical conductivity measurement\cite{10} and first principle calculation by several groups\cite{2,3}. Therefore, the Hund’s rule coupling is likely to be the main reason for the FM fluctuation here. It is thus reasonable to speculate that the pairing gluie of the superconducting in LaO$_{1-x}$Fe$_x$As be the inter-band FM fluctuation and the Cooper pair is formed by the spin triplet pairs of the electrons on two different bands.

In what follows we examine the SC properties of a model Hamiltonian consisting of two approximately degenerate bands denoted by orbital 1 and orbital 2 and a pairing field between the two orbitals with parallel spins. The band structure of the model reproduces the two electron bands obtained in the local density approximation for LaO$_{1-x}$Fe$_x$As. The pairing interaction is from the inter-band ferromagnetic fluctuation induced by the Hund’s coupling. We consider a tight binding model for electrons $C_{k\sigma}$ in a square lattice given by

$$H = \sum_{k\sigma} \left( \varepsilon_{k,\sigma} - \mu \right) C_{k\sigma}^\dagger C_{k\sigma} - J_{k-k'} \sum_{kk'm} \Delta_{km}^\dagger \Delta_{k'm}$$  \hspace{1cm} (1)$$

where $\alpha = 1, 2$ are orbital indices, and

$$\varepsilon_{k,1} = t\gamma_k + t_1 \gamma_k^{(1)} + t_2 \gamma_k^{(2)},$$

$$\varepsilon_{k,2} = t\gamma_k + t_2 \gamma_k^{(2)} + t_1 \gamma_k^{(1)},$$  \hspace{1cm} (2)$$

with $\gamma_k = \cos k_x + \cos k_y$, $\gamma_k^{(1)} = \cos (k_x + k_y)$ and $\gamma_k^{(2)} = \cos (k_x - k_y)$.

In the calculations below, we choose $t = 0.3 eV$, $t_1/t = 0.267$, which are obtained by approximately fitting the shape of the two electron FS and the overall band width with the first principle calculations for LaOFeAs\cite{3}. We consider $t_2$ as a tuning parameter to study the effect of the FS anisotropy, with $t_2/t_1 = 1$ corresponding to the isotropic case, and $t_2/t_1 = 0.6$ for undoped LaOFeAs, $t_2/t_1 = 0.8$ for LaO$_{0.9}$Fe$_{0.1}$As under the normal pressure. $\mu$ is the chemical potential.

The second term in $H$ describes an inter-band pairing interaction with $J_{\vec{k}}$ the pairing strength, and $m = 1, 0, -1$ the three components in the spin triplet state.

$$\begin{align*}
\hat{\Delta}_{k,1}^\dagger &= C_{k1,1}^\dagger C_{k1,2}^\dagger, \\
\hat{\Delta}_{k,-1}^\dagger &= C_{k1,1}^\dagger C_{k1,2}^\dagger, \\
\hat{\Delta}_{k,0}^\dagger &= \frac{1}{\sqrt{2}} \left( C_{k1,1}^\dagger C_{k1,2}^\dagger + C_{k1,1}^\dagger C_{k1,2}^\dagger \right). 
\end{align*}$$  \hspace{1cm} (3)$$

We note that the spin triplet Cooper pairs described by $\Delta$ above are singlets in orbital sector. Therefore, in order to obey the Fermi statistics, the spatial part of the wave function must be of even parity, such as $s$-wave, or extended $s$-wave, or $d$-wave.

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We now turn to the discussion of the inter-band pairing strength $J_{\vec{k}}$. In general $J_{\vec{k}}$ can be expanded by crystal harmonics as

$$J_{\vec{k}} = J_0 + J_1 \left( \cos k_x + \cos k_y \right) + ...,$$  \hspace{1cm} (4)$$

where $J_0$ may be viewed as the on-site Hund’s coupling between two Wannier orbitals centered on the same site and $J_1$ is the magnetic coupling between the neighboring sites, which may be induced by the Coulomb exchange interaction between the Wannier orbitals in the itinerant electron systems. If $J_0$ is strong enough to overcome the on-site direct Coulomb interaction $U$ between the two orbitals, an $s$-wave spin-triplet pairing state is favored. If $J_1$ is large or if $U$ is large, extended $s$-wave or $d$-wave pairing states could be stabilized to avoid the cost in $U$. Note that the possibility of the spin triplet pairing state induced by FM fluctuation was previously proposed to explain the superconductivity in Sr$_2$RuO$_4$\cite{19} and the inter-band pairing was discussed by a number of authors previously\cite{18,20}.

Before we discuss the properties of the superconducting phase, we first examine the special shape of the Fermi surfaces obtained by the above Hamiltonian with the electron filling $\delta = 0.08$ for each band, which are shown in Figure 1. The two Fermi surfaces match each other perfectly by rotating with 90 degrees and the degeneracy along the M-X line is guaranteed by the four-fold rotational symmetry. Because of the metallic nature of the system, the crystal anisotropy is not so strong, and the two Fermi surfaces overlap to each other quite well, which gives the system relatively large phase space for the inter-band pairing.

The above Hamiltonian can be solved by mean field decoupling assuming that only $\sum_{k'} J_{k-k'} \langle \hat{\Delta}_{k'}^{\dagger} \rangle = \Delta_{k,0} \neq 0$ and $\Delta_{k,1} = \Delta_{k,-1} = 0$, which is the natural choice for the phase with time reversal symmetry. Therefore, the mean field Hamiltonian reads.
order parameter $\Delta_s$-wave, the "Fermi arc" only appears for very small angles, there will be no gap at the FS along $k_z$ faces to match exactly. The Cooper instability does not occur with infinitesimal coupling $J$. Instead there is a quantum phase transition with critical value of $J_c$, above which the inter-band spin triplet pairing state has lower energy. First we assume the on-site inter-orbital repulsion is not strong enough to suppress the on-site triplet pairing. In that case, we only consider the on-site term of the effective pairing strength $J_0$, and applied a mean field theory in $s$-wave channel to solve the Hamiltonian and calculate the super-conducting order parameter $\Delta_0$ as a function of $J$ for various ratios of $t_2/t_1$, characterizing the crystal anisotropy. The results are plotted in Fig. 3. As we can see, the critical $J_c$ depends strongly on the crystal anisotropy. For the case in LaOF$_{2}$Fe$_{2}$As, where $t_2/t_1$ is around 0.8, the critical $J_c$ is found to be around 0.4 eV, which is quite feasible for iron compounds. Our mean field theory suggests that the high sensitivity of the super-conducting gap hence the transition temperature $T_c$ to the anisotropy or the deviation of the approximately degenerate bands. $T_c$ may be raised dramatically if the anisotropy is reduced. We speculate the high pressure measurement may reduce the anisotropy and hence increase $T_c$.

When the on-site repulsion is strong, the on-site inter-orbital triplet pairing will be suppressed. In that case, the nearest neighbor Hund's coupling $J_1$ will be important and the spacial paring symmetry may be extended s-wave or d-wave. For the LaO$_{1-x}$F$_x$Fe$_2$As compounds, from the LDA calculation the effective filling factor for the two electron pocket is around 10%, which strongly favors the extended s-wave against the d-wave pairing. While if the effective filling factor is increased by either further doping the system or correlation effect, the d-wave pairing state may also be stabilized.

Below we examine the impurity effect to the proposed pairing state. As it is well known, in the absence of the orbital degrees of freedom, we have even parity with spin singlet or odd parity with spin triplet. The spin singlet $s$-wave superconductivity is unaffected by nonmagnetic

\[
H_{mf} = \Psi_k^\dagger \left( \begin{array}{cc} \hat{h}_k & 0 \\ 0 & \hat{h}_k \end{array} \right) \Psi_k
\]

with $2 \times 2$ matrix $\hat{h}_k = -\delta_k \mathbb{1} + (\varepsilon_{k,1} - \mu + \delta_k) \hat{\sigma}_z + \Delta_{k,0} \hat{\sigma}_x$, $\delta_k = 1/2 (\varepsilon_{-k,1} + \varepsilon_{-k,2} - \varepsilon_{k,1})$ and $\Psi_k^\dagger = (C_{k,1}^\dagger, C_{-k,2}^\dagger, C_{k,1}^\dagger, C_{k,2}^\dagger)$ to be the Nambu representation. The Bogoliubov quasi-particle spectrum can be obtained by solve the above Hamiltonian, which can be written as $E_{k\sigma \pm} = -\delta_k \pm \sqrt{(\varepsilon_{k,1} - \mu + \delta_k)^2 + \Delta_{k,0}^2}$. The above Bogoliubov quasi-particle spectrum gives the minimum gaps sizes detected by angle resolved photo emission to be $E_{min}^{gap} = \max(0, |\Delta_0(\theta)| - |\delta_{kF}(\theta)|)$, where $\theta$ denotes the angle around the Fermi surface. The band splitting $\delta_{kF}$ has strong angle dependence, which vanishes at the four crossing points with $\theta = \frac{n\pi}{2}$ and reaches the maximum at $\theta = \frac{(2n+1)\pi}{4}$. Thus even the order parameter $\Delta_{k0}$ itself is isotropic, i.e. the s-wave or extended s-wave case, the super-conducting gap can have strong angle dependence if $\delta_{kF}$ is compatible to $\Delta_{k0}$. Further, if the absolute value of $\delta_{kF}$ is bigger than the amplitude of the order parameter $\Delta_{k,0}$ at some specific angle, there will be no gap at the FS along that direction. Therefore a "Fermi arc" may appear. If the spacial pairing symmetry is s-wave or extended s-wave, the "Fermi arc" only appears for very small order parameter $\Delta_{k,0}$. While for d-wave case, since $\delta_{kF}(\theta)$ takes the maximum value along the d-wave nodal direction where the order parameter vanishes, the "Fermi arc" will be always there.

In Fig. 2 we plot the angle dependence of the gap function on the Fermi surface with four different values of the order parameter for both s-wave (a) and d-wave case (b). For the s-wave case, the "Fermi arc" appears only when the order parameter is small. While for d-wave case, it always exists. The strange behavior of the Bogoliubov quasi-particles indicate that it is possible to have low lying excitations in this orbital singlet, spin triplet state even with s-wave or extended s-wave pairing symmetry.

Note that there are four points on the two Fermi surfaces to match exactly. The Cooper instability does not

\[
H_{mf} = \Psi_k^\dagger \left( \begin{array}{cc} \hat{h}_k & 0 \\ 0 & \hat{h}_k \end{array} \right) \Psi_k
\]
impurities due to Anderson’s theorem\cite{11}, but is strongly affected by magnetic impurities\cite{12}. On the other hand, a p-wave superconductor with spin triplet is very sensitive to both non-magnetic and magnetic impurities\cite{12}. This explains why spin triplet p-wave superconducting state \(Sr_2RuO_4\) requires clean sample. For the orbitally paired state, the impurity effect to the spin triplet state is very different.

We consider the proposed even parity, orbital singlet and spin triplet state. We shall focus on the s-wave pairing. The case for extended s-wave case will be similar. We follow Balian and Werthamer to apply a perturbation theory to calculate the change of the free energy due to the impurity for the proposed state. In the weak coupling limit, the change of free energy is \(\delta(F_s - F_n) \propto (1-\gamma)\) due to impurity scattering, where \(\gamma\) is a coherence factor determined by both the impurity scattering and the superconducting state. \(\gamma = 1\) corresponds to vanishing effect, while \(\gamma = -1\) corresponds to the strongest suppression. In the conventional pairing state, an s-wave state scattered by nonmagnetic impurities leads to \(\gamma = 1\), hence the change of free energy is zero at leading order, while such an s-wave state scattered by magnetic impurities will result in \(\gamma = -1\), indicating a very strong suppression. A p-wave state scattered by either nonmagnetic or magnetic impurities will lead to \(\gamma = 0\) by average over k-space, indicating strong suppression. We have found that for the proposed orbital singlet state, the s-wave with spin triplet pairing state has \(\gamma = 1\) for nonmagnetic impurities, and \(\gamma = 1/3\) for the magnetic impurities. Therefore the state is robust against non-magnetic impurity and is relatively weakly suppressed by magnetic impurity.

We may speculate the effect of ”orbital impurity” which flip the orbitals as the magnetic impurity flips the spin. Such orbital impurities would strongly suppress the proposed superconductivity in a way similar to the magnetic impurity to suppress conventional s-wave state. While since the orbital degeneracy along the M-X line (which is crucial for inter-band pairing) is protected by four-fold rotational symmetry within the Fe-As plane, impurities which do not break the local four-fold symmetry (such as the off plane impurities) will only generate very weak ”orbital flip” scattering terms. Therefore for this system, the off-plane impurities act like the nonmagnetic impurities in the traditional spin singlet superconductor, which has very little effect for s-wave or extended s-wave. While the in-plane impurities, which will induce the local lattice distortion and generate the inter-band scattering, act like the magnetic impurities for the traditional spin singlet super-conductor, which will kill the superconductivity very efficiently.

In summary, in the present letter we have proposed the parity even, orbital singlet but spin triplet pairing state for the newly discovered super-conductor \(LaO_{1-x}F_xFeAs\). The pairing glue of the SC phase is the strong ferromagnetic fluctuation induced by the Hund’s rule coupling in the iron compound. The pairing state is insensitive to the non-magnetic disorder in contrary to the p-wave spin-triplet state. The Bogoliubov quasiparticle spectrum has quite different behavior with the conventional s-wave spin singlet superconducting phase, which leads to the possible anisotropy in the gap function and can be detected by angle resolved photoemission spectral.

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