Even Parity, Orbital Singlet, and Spin Triplet Pairing for Superconducting $LaFeAsO_{1-x}F_x$

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We examine the spin-triplet superconducting state of even parity mediated by ferromagnetic Hund's coupling between electrons in two almost degenerate orbital bands. This state may be realized in the recently discovered $LaFeAsO_{1-x}F_x$. It is robust against orbital-independent disorder. The splitting of the orbital degeneracy suppresses superconductivity and leads to an anisotropic spectrum in the Bogoliubov quasiparticle. The former predicts a strong pressure dependence of T_c and the latter predicts Fermi pockets, which may be tested in angle resolved photoemission spectra.

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The recent discovery of superconductivity in layered Fe-based compounds has attracted much attention. It has been reported that the transition temperatures are $T_c = 26$ K in $LaFeAsO_{1-x}F_x$ [1], $T_c = 41$ K in CeF_xFeAsO_{1-x} [2], and $T_c = 43$ K in SmF_xFeAsO_{1-x} [3]. This class of superconductors shows highly unusual properties, indicating possible unconventional non-BCS superconductivity [4,5].

The electronic band structure calculations for LaFeAsO suggest the compound to be a semimetal [6–8]. There is a perfect nesting between the hole Fermi surface (FS) centered at the Γ point and the electron FS centered at the M point, which leads to a spin density wave (SDW) state at low temperatures [9,10]. 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 Sr_2RuO_4 suggests a possible spin-triplet pairing. While the conventional s-wave BCS state is robust against nonmagnetic disorder due to the Anderson theorem [12], the p-wave superconductivity of Sr_2RuO_4 is only observed in clean samples and is strongly suppressed by the nonmagnetic impurity [13,14]. The Fe-based superconductivity 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 twofold degenerate electron FS revealed in the electronic structure calculations, we propose a spin-triplet pairing with even parity for SC $LaFeAsO_{1-x}F_x$. The pairing is due to the FM fluctuation between electrons with different orbitals. Our theory explains the robustness of superconductivity to the disorder in a spin-triplet SC state. The splitting of the orbital degeneracy strongly suppresses the superconductivity, and leads to a pronounced $\vec{k}$-dependence in the isotropic s-wave SC state, which may be tested in angle resolved photoemission spectra (ARPES). The high pressure reduces the splitting, and may increase T_c.

We start from the special electronic structure of the layered compound LaFeAsO. The Fe-ions form a square lattice with two atoms in each unit cell. The distance of the neighboring Fe atoms is rather short, so that the electron direct hoppings between the Fe ions are important, similar to the metallic FM elemental Fe. Because of the multiple d-orbitals, there are five FS with three holelike cylinders around the Γ point and two electronlike cylinders around the M point of the Brillouine zone. Upon doping of F-atoms, the three holelike FS shrink rapidly, while the two electron FS expand. Therefore, it is reasonable to expect that the two bands of the electronlike states are responsible for the superconductivity. Competing spin fluctuations exist in this compound. One is antiferromagnetic due to the nesting between the electron and hole FS, connected by a commensurate q vector. The other is FM likely due to the Hund's coupling. The presence of the nesting between the electron and hole FS will induce a SDW instability, which has been observed in experiments [4]. Doping F ions destroys the SDW state and opens the door for superconductivity.

The itinerant ferromagnetism is an interesting but difficult problem in condensed matter physics with a long history [15–17]. One of the important issues is if the multiband nature is necessary for the itinerant ferromagnetism. Both the analytic and numerical studies indicate that itinerant ferromagnetism requires multibands, and is very difficult to obtain within a single band system, unless the Fermi energy is close to a van Hove singularity [15,16]. The density of state in LaFeAsO is very low near the FS, as evidenced in both optical conductivity measurement [10] and first principle calculations [7,8]. Therefore, the Hund's rule coupling is likely to be the main reason for the FM fluctuation here and the multiorbital nature should play an important role in determining the SC properties [18]. It is thus reasonable to speculate that the pairing glue of the SC $LaFeAsO_{1-x}F_x$ is the interband 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 orbitals 1 and 2 and a pairing field between two electrons with parallel spins and different orbitals. The band structure of the model mimics the two electron bands obtained in the local density approximation for $LaFeAsO_{1-x}F_x$. We consider a tight binding model in a square lattice,

$$H = \sum_{k\sigma\alpha} (\varepsilon_{k,\alpha} - \mu) C^{\dagger}_{k\sigma\alpha} C_{k\sigma\alpha} - J_{k-k'} \sum_{kk'm} \hat{\Delta}^{\dagger}_{km} \hat{\Delta}_{k'm}, \quad (1)$$

where $C^{\dagger}_{k\sigma\alpha}$ is to create an electron of crystal momentum k, spin σ and orbital $\alpha = 1, 2$, and

$$\begin{aligned} \varepsilon_{k,1} &= t\gamma_k + t_1\gamma_k^{(+)} + t_2\gamma_k^{(-)}, \\ \varepsilon_{k,2} &= t\gamma_k + t_2\gamma_k^{(-)} + t_1\gamma_k^{(+)}, \end{aligned} \quad (2)$$

with $\gamma_k = \cos k_x + \cos k_y$, $\gamma_k^{(\pm)} = \cos(k_x \pm k_y)$.

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 [8]. We consider t_2 to be 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 the undoped LaFeAsO, $t_2/t_1 = 0.8$ for $LaFeAsO_{0.9}F_{0.1}$ under the normal pressure. μ is the chemical potential. The second term in H describes an interband pairing interaction with J_k the pairing strength, and $m = 1$, 0, -1 the three components in the spin-triplet state.

$$\begin{aligned} \hat{\Delta}^{\dagger}_{k,1} &= C^{\dagger}_{k\uparrow,1} C^{\dagger}_{-k\uparrow,2}, \qquad \hat{\Delta}^{\dagger}_{k,-1} = C^{\dagger}_{k\downarrow,1} C^{\dagger}_{-k\downarrow,2}, \\ \hat{\Delta}^{\dagger}_{k,0} &= (C^{\dagger}_{k\downarrow,1} C^{\dagger}_{-k\uparrow,2} + C^{\dagger}_{k\uparrow,1} C^{\dagger}_{-k\downarrow,2})/\sqrt{2}. \end{aligned} \quad (3)$$

We note that the spin-triplet Cooper pairs described by Δ above are singlets in orbital sector. Because of the Fermi statistics, the spatial part of the wave function must then be of even parity.

We now turn to the discussion of the interband pairing strength J_k, which can in general be expanded in terms of crystal harmonics,

$$J_k = J_0 + J_1(\cos k_x + \cos k_y) + \ldots \quad (4)$$

where J_0 is the effective on-site Hund's coupling between the 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 large, an s-wave spin-triplet pairing states will be favored. If J_1 is large, an extended s-wave or d-wave pairing states will be realized. Note that the possibility of the spin-triplet pairing state induced by FM fluctuation was proposed to explain the superconductivity in Sr_2RuO_4 [19] and the interband pairing was discussed by a number of authors [20,21].

In Fig. 1 we show the FS of Eq. (1) in the normal state at electron filling $\delta = 0.08$ per band. The two FS are related by a rotation of 90 degrees, and the degeneracy along the $M - X$ line is guaranteed due to the fourfold rotational symmetry. Because of the metallic nature of the system, we do not expect the crystal anisotropy to be strong, and the two FS overlap to each other, giving rise of a large phase space for the interband pairing.

Hamiltonian (1) can be solved by using standard mean field theories. We consider a SC state with time reversal symmetry, and assume $\sum_{k'} J_{k-k'} \langle \hat{\Delta}_{k'0} \rangle = \Delta_{k,0} \neq 0$, and $\Delta_{k,1} = \Delta_{k,-1} = 0$. Our choice is based on the group theory analyses and the simplification. The state corresponds to the $\vec{d}$-vector commonly adopted in the study of spin-triplet states to have the form of $\vec{d}(k) = \Delta_{k,0}\hat{z}$, which forms the $A_{1g}(\Delta_{k,0} = \cos k_x - \cos k_y)$ or $B_{1g}(\Delta_{k,0} = \cos k_x + \cos k_y)$ representation in terms of the group analysis. It is the allowed state within the one-dimensional representation of the group theory classification for point group D_{4h} [22]. The other states with two-dimensional representation are more complicated and may further break fourfold rotational symmetry. The mean field Hamiltonian then reads

$$H_{mf} = \sum_{\sigma} \Psi^{\dagger}_{k\sigma} \begin{pmatrix} \hat{h}_k & 0 \\ 0 & \hat{h}_k \end{pmatrix} \Psi_{k\sigma},$$

where $\Psi^{\dagger}_k = (C^{\dagger}_{k\uparrow,1}, C_{-k\downarrow,2}, C^{\dagger}_{k\downarrow,1}, C_{-k\uparrow,2})$ and the 2×2 matrix $\hat{h}_k = -\delta_k \hat{1} + (\varepsilon_{k,1} - \mu + \delta_k)\hat{\sigma}_z + \Delta_{k,0}\hat{\sigma}_x$, in the Nambu representation, and $\delta_k = 1/2(\varepsilon_{-k,2} - \varepsilon_{k,1})$. The Bogoliubov quasiparticle spectrum can be obtained by solving H_{mf} and is given by

$$E_{k\sigma\pm} = \sqrt{(\varepsilon_{k,1} - \mu + \delta_k)^2 + \Delta^2_{k,0}} \pm \delta_k.$$

The minimum gap value measured in ARPES is $E^{\text{gap}}_{\text{min}} = \max(0, |\Delta_0(\theta)| - |\delta_{k_F}(\theta)|)$, where θ is the angle around the FS. As we can see from Fig. 1, the band splitting δ_{k_F} has a strong angle dependence. It vanishes at the four crossing points on the FS with $\theta = \frac{n\pi}{2}$ and is maximum at

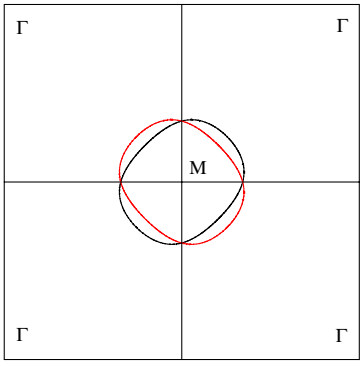

FIG. 1 (color online). The Fermi surface (FS) derived from the two-band tight binding model H of Eq. (1) in the absence of the pairing, which reproduces the electron FS obtained in the first principle calculations [8] for the undoped LaFeAsO.

$\theta = \frac{(2n+1)\pi}{4}$. Therefore, the SC gap may show a strong angle dependence even in the isotropic s-wave or extended s-wave states provided that δ_{k_F} is compatible to $\Delta_{k,0}$, and we may even have "Fermi pockets" [23]. In a state with Δ fully gapped, the "Fermi pocket" will only appear if $\Delta_{k,0}$ is very small. In a SC state with nodal lines in $\Delta(k, 0)$, "Fermi pocket" always appears since $\delta_{k_F}(\theta)$ takes the maximum value along the nodal direction where the order parameter vanishes.

In Fig. 2, we plot the angle dependence of the gap function on the FS for four different values of the order parameter for the full gapped s-wave case in (a) and for the $d_{x^2-y^2}$-wave case with nodal lines in (b). We note that the "Fermi pocket" appears along the directions where the SC gap vanishes.

If the two FS are not completely degenerate, the Cooper instability does not occur with infinitesimal pairing strength, and there is a critical value $J = J_c$, above which the interband spin-triplet pairing state has a lower energy. Let us first consider the on-site term of the effective paring strength J_0. We use a mean field theory to study the s-wave pairing and to calculate Δ_0 for various ratios of t_2/t_1, characterizing the crystal anisotropy. The results are plotted in Fig. 3. As we can see, J_c depends strongly on the crystal anisotropy. For $t_2/t_1 = 0.8$, suitable to the doped LaFeAsO, we find $J_c = 0.4$ eV, which is quite feasible for the iron compounds. Our mean field theory suggests that the SC gap hence the transition temperature T_c are sensitive to the anisotropy. We speculate the high pressure measurement may reduce the anisotropy hence increase T_c.

When the on-site Coulomb repulsion is strong, the on-site interorbital triplet pairing will be suppressed. In this case, the nearest neighbor Hund's coupling J_1 will be important and the spatial paring symmetry may be extended s wave or d wave. For the $LaFeAsO_{1-x}F_x$ compounds, from the LDA calculations the effective filling factor enclosed in each electron FS is around 10%. With that filling, the extended s wave is found to be more favorable than the d-wave pairing. But the d-wave pairing may be favored by correlations or at large doping.

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 impurities due to Anderson's theorem [12], but is strongly affected by magnetic impurities [13]. On the other hand, a p-wave superconductor with spin triplet is very sensitive to both nonmagnetic and magnetic impurities [13]. This explains why spin-triplet p-wave SC state Sr_2RuO_4 requires clean sample. As we will see, for the interorbital 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 the extended s-wave case will be similar. We follow Balian and Werthamer [13] 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 the impurity scattering, where γ is a coherence factor determined by both the impurity scattering and the SC state. $\gamma = 1$ corresponds to null impurity effect, while $\gamma = -1$ corresponds to the strongest suppression of the superconductivity. In the conventional pairing state, an s-wave

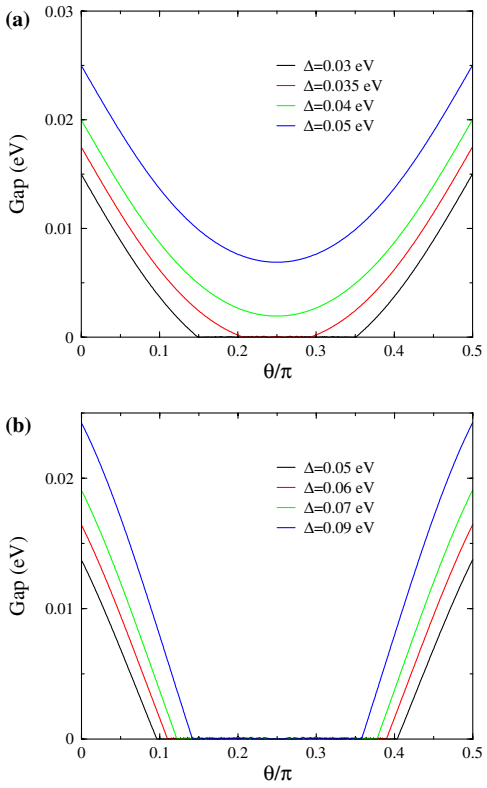

FIG. 2 (color online). The angle dependence of the minimum gap value at the Fermi surface for four values of the SC order parameter.

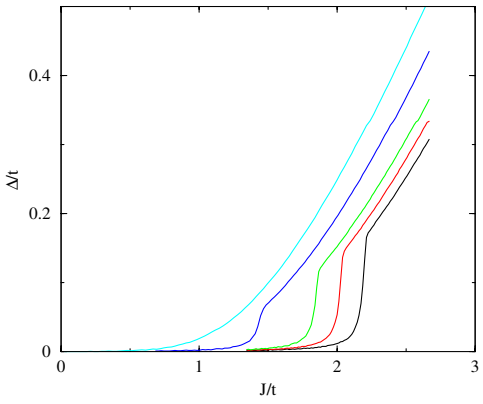

FIG. 3 (color online). The SC order parameter as a function of effective FM exchange coupling J_0. From left to right, the tight binding parameter t_2/t_1 is 100%, 75%, 50%, 37.5%, 25%, respectively.

state scattered by nonmagnetic impurities leads to $\gamma = 1$; hence, the change of free energy is zero at the leading order in perturbation, 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 averaging over the 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 and orbital-independent impurities, and $\gamma = 1/3$ for the magnetic and orbital-independent impurities. Therefore the SC state is robust against nonmagnetic impurity and is relatively weakly suppressed by magnetic impurity provided that the impurity does not couple to the two degenerate orbitals. We note that both the nonmagnetic or magnetic impurity effects to the d wave are severe and $\gamma = 0$ for the d-wave state.

The doping in $LaFeAsO_{1-x}F_x$ is outside the FeAs plane, which is believed to be responsible for the appearance of the superconductivity. It is thus reasonable that the impurities introduced upon the doping is orbital independent and nonmagnetic. 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 states. This is consistent with the insensitivity of the disorder to the SC state in the preliminary experiments on this class of the superconductors.

We remark that the in-plane impurities, which may induce the local lattice distortion and generate the interband scattering, act like the magnetic impurities to the conventional spin singlet superconductor, which will strongly suppress the superconductivity.

In summary, in the present Letter we have proposed an even parity, orbital singlet and spin-triplet SC state for the newly discovered Fe-based superconductors. The pairing glue of the SC phase is the strong ferromagnetic fluctuation induced by the Hund's rule coupling. The pairing state is insensitive to the nonmagnetic disorder in contrary to the p-wave spin-triplet state. The Bogoliubov quasiparticle spectrum of isotropic s-wave or extended s-wave states shows possible anisotropy in k space in the gap function and can be detected in angle-resolved photoemission spectra. The symmetry analysis outlined here may be applied to SC materials with valley degeneracy or other forms of pseudospin degree of freedoms.

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Note added.—Since the submission of the manuscript, there have been a number of new experiments reported on this compound, and doped $BaFe_2As_2$ was found also to be a superconductor. Most of the experiments indicate nonconventional BCS SC properties, in support of the general scenario considered in our theory. Recent muon experiment [24] showed some similarity of the LaFeAsO SC state to the physics of He-3, where the solid phase is antiferromagnetic and the liquid phase is a spin-triplet superfluid. This analogy is in favor of the present theory. There are experiments indicating either full gap [25] or line nodals [26] in the SC gaps, which remains to be an issue for further study. The NMR knight shift data of PrOFeAs showed a strong suppression in the SC state, which was interpreted for a spin singlet state [27]. However, other groups have not found any obvious suppression of the knight shift in Fe-based SC compounds with La or other non–rare earth ions. Since the knight shift reported in PrOFeAs is several times larger than that in LaFeAsO, it may be possible that the suppression is due to the contribution from the rare earth element.

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