Fractional violation of Anderson’s Theorem for Sign-Reversing $s$-Wave State of Iron-Pnictide Superconductors

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Based on the five-orbital model, we study the effect of local impurity in iron pnictides, and find that the interband impurity scattering is promoted by the $d$-orbital degree of freedom. This fact means that the fully-gapped sign-reversing $s$-wave state, which is predicted by spin fluctuation theories, is very fragile against impurities. In the BCS theory, only 1% impurities with intermediate strength induce huge pair-breaking, resulting in the large in-gap state and prominent reduction in $T_c$, contrary to the prediction based on simple orbital-less models. The present study provides a stringent constraint on the pairing symmetry and the electronic states in iron pnictides.

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Since the discovery of high-$T_c$ superconductors with FeAs layers $^1$, the symmetry of the superconducting (SC) gap has been studied very intensively. NMR studies had revealed that the singlet SC state is realized in iron pnictides $^{2–4}$. The realized gap function is isotropic. The fully-gapped state is also supported by the rapid suppression in $T_1$ $\propto$ $T^{6}$ below $T_c$, contrary to the prediction based on simple orbital-less models. The present study provides a stringent constraint on the pairing symmetry and the electronic states in iron pnictides.

Figure 1 shows the $\pi - \pi - \pi$ and $\pi - \pi - \pi$ hole-pockets (FSs) in the unfolded Brillouin zone $^{11}$. Hereafter, we fix the electron number as 6.1 (10% electron-doped case). Then, the total density of states (DOS) per spin is $N(0) = 0.66$ eV$^{-1}$ at the Fermi level. In the presence of the Coulomb interaction, antiferromagnetic (AF) fluctuations with $Q \approx (\pi,0)$ is expected to emerge due to the nesting between FS1,2 and FS3,4 $^{12}$. Based on this fact, a fully gapped $s$-wave state with sign reversal ($\Delta_{1,2} > 0$, $\Delta_{3,4} < 0$), which is called the $s_{\pm}$-wave state, had been predicted theoretically $^{11,12}$. However, no conclusive evidence for the $s_{\pm}$-wave state has been obtained experimentally so far.

Historically, the study of impurity effects had offered us significant information in determining the pairing symmetry in many superconductors. In iron pnictides, the SC state survives against high substitution of Fe sites by other element (more than 10%), like Co, Rh, Ni, Zn, Ru, and Ir $^{2, 13–20}$. In Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ $^{19, 20}$, the AF ordered state is removed at $x = 0.07$, and SC state with $T_c = 24$ K appears. $T_c$ gradually decreases as $x$ increases, and it reaches zero at $x = 0.17$. This phase diagram suggests the relation $-\Delta T_c \sim 2K$ per 1% Co substitution, and cannot be understood if the impurity pair-breaking gives $-\Delta T_c > 20$ K/\% like in high-$T_c$ cuprates. According to the first principle calculation, the impurity potential due to Co substitution for $xz$, $yz$-orbitals is 1.52 eV, and its radius is only 1 Å $^{21}$. Also, a bulk superconducting state with $T_c = 24$ K is realized in Sr(Fe$_{1-x}$Ir$_x$)$_2$As$_2$ for $x \sim 0.25$ $^{18}$. These experimental facts would eliminate the possibility of nodal gap state.

To explain these experiments, simple multiband BCS models with constant impurity potential $I^b_{\alpha,\beta}$ in the band-diagonal basis ($\alpha, \beta$ being the FSs), which we call the “orbital-less multiband model”, had been studied intensively. In the Born regime ($I^b \ll (\pi N(0))^{-1}$), the $s_{\pm}$-wave state is suppressed by interband scattering $^{22–24}$. In the unitary regime, in contrast, the $s_{\pm}$-wave state is very robust since the interband (intraband) scattering is renormalized to zero (finite) if $I^b$ is constant and $\det(I^b) \neq 0$ $^{25, 26}$. That is, Anderson’s theorem $^{27}$ is recovered in the unitary regime. However, the latter result should be re-examined since $I^b$ has complex momentum-dependence in usual multiorbital systems.

In this letter, we present the first study of the impurity effect on the bulk DOS and $T_c$ based on the five-orbital model given in Ref. $^{11}$. By treating the multiorbital effect correctly, we reveal that the Anderson’s theorem is seriously violated for the $s_{\pm}$-wave state, due to the inter-

\[ \text{FIG. 1: Hole-pockets (FS1,2) and electron-pockets (FS3,4) in iron pnictides. } |T_{k,k'}^{b,32}|^2 \text{ represents the impurity induced pair-hopping amplitude between (FS2, } \pm k') \text{ and (FS3, } \pm k'). \]
band scattering in Fig. 1, mainly via $xz$ and $yz$ orbitals. For this reason, only 1% impurities with a moderate or strong potential induce large in-gap DOS and prominent reduction in $T_c$ ($\Delta T_c \sim 20$ K/$\%$) in the BCS theory, which are comparable to those in nodal gap SC states. The present study suggests a reasonable possibility that a conventional $s$-wave state without sign reversal ($s_{++}$-wave state) would be realized in dirty iron pnictides.

Hereafter, we study the following $10 \times 10$ Nambu BCS Hamiltonian in the real $d$-orbital basis; $t_{2g}$ ($xy$, $yz$, $zx$) and $e_g$ ($x^2$-$y^2$, $z^2$):

$$\hat{H}_k = \left( \begin{array}{cc} \hat{H}_k^0 & \Delta_k \\ \Delta_k^\dagger & -\hat{H}_k^0 \end{array} \right),$$

where $\hat{H}_k^0$ is the $5 \times 5$ hopping matrix of the five-orbital tight-binding model, which was introduced in Ref. [11]. $\Delta_{k,\alpha}^{\ell,l} = \sum_{\alpha} U_{j,l}^{\alpha,\alpha} \Delta_{k,\alpha} U_{j,l}^{\alpha,\alpha\ast}$ is the singlet gap function in the $d$-orbital basis, where $\Delta_{k,\alpha}$ is the gap function for $FS\alpha$ in the band-diagonal basis, and $U_{j,l}^{\alpha,\alpha\ast}$ is the $5 \times 5$ transformation unitary matrix between two representations. Then, the Green function is given by

$$\hat{G}_{k}(i\omega_n) = \left( \hat{G}_{k}^0(i\omega_n) - \hat{F}_{k}(i\omega_n) \right)^{-1} \hat{G}_{k}^0(i\omega_n),$$

$$= (i\omega_n \mathbb{1} - \hat{\Sigma}_{k}(i\omega_n) - \hat{H}_k)^{-1},$$

where $\omega_n = \pi T(2n + 1)$ is the fermion Matsubara frequency, $\hat{G}_k$ ($\hat{F}_k$) is the $5 \times 5$ normal (anomalous) Green function, and $\hat{\Sigma}_k$ is the self-energy in the $d$-orbital basis.

First of all, we calculate the impurity effect on the DOS in the SC state using the $T$-matrix approximation, which is reliable when the impurity concentration $n_{imp}$ is much smaller than unity. We consider the local impurity potential due to the substitution of Fe by other 3$d$ elements: In the present $d$-orbital basis, the impurity potential is momentum-independent and diagonal. Then, the $T$-matrix for a single impurity, which is $k$-independent in the $d$-orbital basis, is given as

$$\hat{T}(i\omega_n) = (1 - \hat{I}\hat{G}_{loc}(i\omega_n))^{-1}\hat{I},$$

where $\hat{G}_{loc}(i\omega_n) = \frac{1}{\pi} \sum_{k} \hat{G}_{k}(i\omega_n)$. Neglecting the crystalline-field splitting between $t_{2g}$ and $e_g$, the impurity potential $\hat{I}$ is simply given as $I_{j,l} = I\delta_{j,l}$ for $1 \leq j \leq 5$, and $I_{j,l} = -I\delta_{j,l}$ for $6 \leq j \leq 10$. In the $T$-matrix approximation, the self-energy matrix in the $d$-orbital basis is $k$-independent. It is given as

$$\hat{\Sigma}(i\omega_n) = n_{imp}\hat{T}(i\omega_n).$$

The gap function $\hat{\Delta}_{k}$ in eq. (1) is given by the solution of the Eliashberg equation:

$$\Delta_{k}^{j,l}(i\omega_n) = -\frac{T}{N} \sum_{q,m} \sum_{l,l'} V_{k,q}^{j,l,\ell,l'}(i\omega_n, i\omega_m) F_{q}^{l,l'}(i\omega_m),$$

where $V_{k,q}^{j,l,\ell,l'}$ is the pairing potential. In the fully self-consistent $T$-matrix approximation, we solve eqs. (2)-(5) self-consistently. In calculating the DOS, however, we solve only eqs. (2)-(4) self-consistently, assuming the isotropic $\Delta_{n}$ for $FS \sim 4$. As we will show below, the reduction in $T_c$ in the $s_\pm$-wave state at $n_{imp} = 0.01$ exceeds 20 K for $I \gg W_{band}$. Then, the reduction in the SC gap is $\sim 36$ K if the relation $\Delta/T_c = 1.8$ is supposed. Hereafter, we assume that this reduction in $\Delta$ had been included from the beginning.

![FIG. 2: (color online) Obtained DOS in the $s_\pm$-wave state per spin for $n_{imp} = 0.01$. (a) $|\Delta_{1,2,3,4}| = 0.05$ eV, and (b) $|\Delta_{1,3,4}| = 0.025$ eV.](image)

Figure 2 shows the obtained DOS in the $s_\pm$-wave SC state per spin for $n_{imp} = 0.01$ at $T = 0$, assuming the isotropic gap in the band-diagonal basis. The number of $k$-meshes is $N = 512 \times 512$. The DOS is given by the normal Green function in eq. (2) as $N(\omega) = \frac{1}{\pi \omega} \sum_{k} \text{Im}\{\text{Tr}\hat{G}_{k}(\omega - i\delta)\}$. In Fig 2 (a), we put $\Delta_{1,2} = -\Delta_{3,4} = 0.05$ eV. We also study the case where only $\Delta_2$ is changed to 0.025 eV in Fig 2 (b), consistently with ARPES measurements [6, 7]. Since these gaps are a few times larger than experimental values, the obtained impurity effect is underestimated.

In Fig. 2, prominent in-gap state due to impurity pair-breaking emerges even for small value of $I = 0.5$, which means that only 1% impurity concentration induces strong pair-breaking effect in the $s_\pm$-wave state. This result is inconsistent with the analysis in the orbital-less model, where pair-breaking due to unitary impurity is absent in the $s_\pm$-wave state. Now, we explain that this discrepancy arises from the presence or absence of orbital degree of freedom: In the band-diagonal basis, the
In this case, pair-breaking due to interband scattering is the most prominent at the state with $\Delta_n$ates from the fact that the quasiparticle damping effect strongly depends on the sign of $\Delta$. The residual resistivity in overdoped cuprates \cite{29}. For the same reason, we find that $T_{b,k,p}$ has large offdiagonal elements \cite{28}. For this reason, the impurity pair-breaking for $s_+$-wave state is as large as that in the nodal gap state.

As recognized in Fig. 2, the impurity pair-breaking effect strongly depends on the sign of $I$ except for $I = \infty$: The impurity effect is stronger for $I > 0$, and it is the most prominent at $I \sim 1.5$ eV. This result originates from the fact that the quasiparticle damping $\gamma_{\alpha} = n_{imp} \text{Im} T_{\alpha,\alpha}^{b}(-i0)$, which works as the depairing effect when the Anderson’s theorem is not satisfied, depends on the sign of $I$ in the presence of strong particle-hole asymmetry. \cite{29,30}. For the same reason, we find that the residual resistivity in the $T$-matrix approximation, which is given by $\rho^{imp} = (2e^2/h) \sum_k \text{Tr}\{\partial H^b_{\alpha,k}/\partial k_x \cdot \partial H^b_{\alpha,k}/\partial k_x \}$, is more than 6 A for positive $I$; $\rho^{imp} = 22, 14$, and 10 [\mu\Omega cm] at $I = 1, 4.5$, and $\infty$ [eV], respectively. For $I < 0$, $\rho^{imp} = 3$ and 7 [\mu\Omega cm] at $I = -1$ and $-4.5$ [eV], respectively. In Ref. \cite{31}, $\rho^{imp}$ for 1% Co doping in polycrystalline Nd1111 exceeds 120 $\mu\Omega cm$, which corresponds to 30 $\mu\Omega cm$ in single crystal. $\rho^{imp}$ in Co-doped La1111 is still larger. Near AF quantum-critical-point, $\rho^{imp}$ can exceed the value given by the $T$-matrix approximation since the many-body correlations are enhanced around the impurity, as discussed in Ref. \cite{29}. In underdoped high-$T_c$ cuprates, for example, $\rho^{imp}$ due to 1% Zn impurity reaches 100 $\mu\Omega cm$, which is 10 times of the residual resistivity in over-doped cuprates \cite{29}.

We also study the impurity effect on the $s_+$-wave state with $\Delta_{1,3,4} = 0.05$ eV and $\Delta_2 = 0.025$ eV in Fig. 3. In this case, pair-breaking due to interband scattering is very small since the sign of $\Delta_n$ on all the FSs are the same. As $n_{imp}$ increases, the structure of DOS is gradually smoothened. The absence of zero-energy state is consistent with many NMR measurements \cite{2-4,9,10}.

Next, we calculate the impurity effect on $T_c$. To demonstrate the qualitative difference between $s_1$-wave and $s_+$-wave states, we introduce the following BCS-type pairing interaction in the band basis \cite{32}:

$$V_{\alpha,\beta}^{(\omega_n, \omega_{m})} = g_{\alpha, \beta} \cdot \phi_{\alpha, \beta} \cdot \xi(\omega_n) \xi(\omega_{m})$$

(8)

where $\phi_{\alpha, \beta} = 1$ for $s_+$-wave, and $\phi_{\alpha, \beta} = \text{sgn}\{\cos k_x \cos k_y\}$ for $s_1$-wave. We also put $\xi(\omega_n) = \omega_D/(\omega_n^2 + \omega_D^2)$, where $\omega_D$ is the cutoff energy. $V_{\alpha,\beta}^{(\omega_n, \omega_{m})}$ in eq. (5) is given by $\sum_{\alpha, \beta} V_{\alpha, \beta}^{(\omega_n, \omega_{m})} U_{\alpha, \beta}^{k} \leftrightarrow U_{\alpha, \beta}^{k'} \leftrightarrow U_{\alpha, \beta}^{k} \leftrightarrow U_{\alpha, \beta}^{k'}$. Then, $T_c$ is obtained by solving the eqs. (2)-(5) fully self-consistently. Here, we do not consider the mass-enhancement since the energy derivative of the self-energy $\Sigma_{\alpha, \beta}^{\omega_n, \omega_{m}}$ vanishes for $\omega_n \rightarrow 0$ since eq. (8) is a separate function of $\omega_n$ and $\omega_m$ \cite{32}.

To consider the superconductivity due to interband interaction between FS1,2 and FS3,4, we put $g_{\alpha, \beta} = \text{sgn}\{\cos k_x \cos k_y\}$ for $s_+$-wave. We also put $\xi(\omega_n) = \omega_D/(\omega_n^2 + \omega_D^2)$, where $\omega_D = 0.03$, the obtained $T_c$'s are shown in Fig. 4 for (a) $g_1 = g_2 = 2$ eV ($T_{c0} = 46$ K at $n_{imp} = 0$) and (b) $g_1 = 3g_2 = 3$ eV ($T_{c0} = 40$ K). In the present choice of $\phi_{\alpha, \beta}$, $T_{c0}$ is equal for both $s_+$- and $s_1$-wave states. In case (a), the obtained $\Delta_{\alpha, \beta}$ in the band diagonal basis is almost isotropic and the same for $\alpha = 1 \sim 4$: In contrast, $\Delta_1 : \Delta_2 : \Delta_{3,4} \approx 3 : 1 : 1.5$ in case (b). In the BCS theory, $-\Delta_{\alpha} \equiv T_{c0} - T_{c\alpha}$ is independent of $T_{c0}$, and a qualitative relation $-\Delta_{Tc} \sim \gamma_{\alpha}$ holds when the Anderson’s theorem is violated \cite{25,32}. Since $\gamma_{\alpha}$ takes the largest value at $I \sim 1$ eV \cite{30}, $T_c$ for the $s_+$-wave state vanishes at $n_{imp} \sim 0.005$ for $I = +1$ eV. In the $s_+$-wave state, $T_c$ in (b) slowly decreases with $n_{imp}$ with downward convex, since weak pair breaking occurs unless $\Delta_{\alpha} = \Delta_{\beta}$.

Until now, we have neglected the mass-enhancement, which is $z^- = m_e/m \approx 2$ by ARPES \cite{6,7}. Since the
FIG. 4: (color online) Obtained $T_c$ for the $s_\pm$-wave and $s_{++}$-wave states as functions of $n_{imp}$. (a) $g_{1,2} = 2$ eV ($T_c = 46$ K) and (b) $g_1 = 3$ eV ($T_c = 40$ K).

The depairing effect is renormalized by $z$, $-\Delta T_c$ is reduced to $-\Delta T_c \sim z \approx z \gamma_0$ [32]. When $z^{-1} = 2$, $T_c$ for the $s_{++}$-wave state vanishes at $n_{imp} \approx 0.01$, 0.02, and 0.066 for $I = 1$ eV, $I = -1$ eV, respectively. Although $-\Delta T_c$ ranges from 46 K/\% to 7 K/\%, $T_c$ for $s_{++}$-wave vanishes when $\rho_{imp} \approx 20 \mu\Omega cm$ for any $I$. However, Sato et al. studied the impurity effects in polycrystalline La1111 and Nd1111, and found that $T_c$ vanishes when $\rho_{imp} \approx 3 \mu\Omega cm$, which corresponds to $\approx 750 \mu\Omega cm$ in single crystal [31]. Also, $\rho$ in single-crystal Fe(Se,Te) just above $T_c = 15$ K exceeds 50 $\mu\Omega cm$ in ref. [33], mainly due to elastic scattering. Then, the estimated mean-free-path $l_{mpf}$ is as short as $\alpha_{Fe-Fe} = 2.8\AA$ [34] that is about $1/100$ of the coherence length in Ba122 [35]. (Note that $\rho_{mpf} \propto l_{mpf}^{-1}$ is independent of $z$.) When $l_{mpf} \sim 0.4\AA$, even $s_{++}$-state will be broken by localization [31].

In summary, we have studied the effect of Fe-site substitution in iron pnictide superconductors. Due to the presence of orbital degree of freedom, the $s_\pm$-wave state is as fragile as nodal gap states against nonmagnetic impurities. The critical residual resistivity for vanishing $T_c \sim 40$ K for $s_+\pm$-wave state is only $\rho''_{mp} \sim 20 \mu\Omega cm$. The corresponding mean-free-path is $\sim 25\alpha_{Fe-Fe}$, which is longer than the experimental coherence length.

Considering the robustness of superconductivity against impurities or randomness, the $s_{++}$-state would be a promising candidate for iron pnictide superconductors. However, $s_\pm$-wave state will become stable when (i) $|I| \ll 1$ eV, or the potential radius is comparable to the lattice spacing and the large momentum scattering is suppressed. To reveal this possibility, we need more systematic first principle calculations for impurity potentials or measurements of $\rho_{mpf}$. Also, the $s_\pm$-wave state can be stable when (ii) the $d$-orbital weight on the FS is completely modified by many-body effect, or (iii) strong coupling SC state like in heavily under-doped high-$T_c$ cuprates is realized. Thus, it is important to study the many-body electronic states to clarify these possibilities.

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[1] Y. Kamihara et al.: J. Am. Chem. Soc. 130, 3296 (2008).
[2] A. Kawabata et al., J. Phys. Soc. Jpn. 77, 103704 (2008); S. C. Lee et al., J. Phys. Soc. Jpn. 78, 043703 (2009).
[3] H. J. Grafe et al., Phys. Rev. Lett. 101, 047003 (2008).
[4] K. Matano et al., Europhys. Lett. 83, 57001 (2008).
[5] K. Hashimoto et al., Phys. Rev. Lett. 102, 017002 (2009); K. Hashimoto et al., Phys. Rev. Lett. 102, 207001 (2009).
[6] D. V. Evtushinsky et al., New J. Phys. 11, 055006 (2009).
[7] K. Nakayama et al., Europhys. Lett. 85, 67002 (2009).
[8] Y. Kobayashi et al., J. Phys. Soc. Jpn. 78, 073701 (2009).
[9] Y. Nakai et al., J. Phys. Soc. Jpn. 77, 073701 (2008).
[10] H. Mukuda et al., J. Phys. Soc. Jpn. 77, 093704 (2008).
[11] K. Kuroki et al., Phys. Rev. Lett. 101, 087004 (2008); K. Kuroki et al., Phys. Rev. B 79, 224511 (2009).
[12] I. L. Mazin et al., Phys. Rev. Lett. 101, 057003 (2008).
[13] A. S. Sefat et al., Phys. Rev. Lett. 110, 177004 (2013); A. Leithe-Jasper et al., Phys. Rev. Lett. 101, 207004 (2008).
[14] F. Han et al., Phys. Rev. B 80, 024506 (2009).
[15] L. J. Li et al., New J. Phys. 11, 025008 (2009).
[16] K. Li et al., New J. Phys. 11 053008 (2009).
[17] W. Schnelle et al., Phys. Rev. B 79, 214516 (2009).
[18] F. Han et al., Phys. Rev. B 80, 024506 (2009).
[19] N. Ni et al., Phys. Rev. B 80, 024511 (2009).
[20] L. Fang et al., arXiv:0903.2418.
[21] A. F. Kemper et al., arXiv:0904.1257v2.
[22] A. V. Chubukov et al., Phys. Rev. B 78, 134512 (2008).
[23] D. Parker et al., Phys. Rev. B 78, 134524 (2008).
[24] V. Cvetkovic, and Z. Tesanovic, Europhys. Lett. 85, 37002 (2009).
[25] Y. Senga and H. Kontani, New J. Phys. 11, 035005 (2009).
[26] Y. Bang et al., Phys. Rev. B 79, 054529 (2009).
[27] P. W. Anderson, J. Phys. Chem. Solids 11, 26 (1959).
[28] The formal solution of eq. (6) can be expressed as $\tilde{T}_{u,v} = \tilde{\mathbf{P}}^{(b)}(\mathbf{b}^{(a)} - 1 \tilde{\mathbf{P}}^{(0)})^{-1}$, where $u = (\alpha, k)$ and so on. Since $\det(\tilde{\mathbf{P}}) = 0$ in this representation in the present model, $\tilde{T}_{u,v}$ is not band-diagonal even for $I = \infty$.
[29] H. Kontani, Rep. Prog. Phys. 71, 026501 (2008).
[30] In a single band model, $\gamma/n_{imp} = \text{Im}T(-i\omega_0) = (1 + 2a(\pi N(0))^{-1/2}I^{-1})^{-1} + O(I^{-2})$ for $a \approx \text{Re}(g_{R0}(0)) \approx N(0)$, which depends on the sign of $I$ when $\alpha \neq 0$ (particle-hole asymmetric case). We find that $a > 0$ in the present model.
[31] M. Sato et al., arXiv:0907.3007.
[32] P. B. Allen, and B. Mitrovic, Solid State Physics 37, 1 (1983).
[33] J. Liu et al., arXiv:0904.0824.
[34] Using the $T$-matrix approximation, we numerically derived the relation $\rho_{mpf} \approx 500 \times (\alpha_{Fe-Fe}/l_{mpf}) \mu\Omega cm$, where $l_{mpf}$ is the averaged mean-free-path. It is also obtained based on the 2D electron gas model; the conductivity due to four FSs is $4 \times (e^2/h)k_F l_{mpf}$ and $k_F \sim 0.2\pi/\alpha_{Fe-Fe}$.
[35] Y. Yin et al., Phys. Rev. Lett. 102, 097002 (2009).