Violation of Anderson's Theorem for the 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 and band dependent according to the penetration depth measurement [5] and the angle-resolved photoemission spectroscopy (ARPES) [6,7]. This result is contrasting to high-T_c cuprates, where the nodal d-wave state is realized. The fully gapped state is also supported by the rapid suppression in $1/T_1$ ($\propto T^n$; $n \sim 6$) below T_c in several compounds [8]. On the other hand, the relation $1/T_1 \propto T^3$ had been reported [9,10] in different compounds.

Figure 1 shows the Fermi surfaces (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 $N(0) = 0.66\ \mathrm{eV}^{-1}$ at the Fermi level. In the presence of the Coulomb interaction, antiferromagnetic (AF) fluctuations with $\mathbf{Q} \approx (\pi, 0)$ 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 elements (more than 10%), like Co, Rh, Ni, Zn, Ru, and Ir [2,13–20]. In $Ba(Fe_{1-x}Co_x)_2As_2$ [19,20], the AF ordered state is removed at $x = 0.07$, and the 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 $-\Delta T_c \sim 2$ K 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 $Sr(Fe_{1-x}Ir_x)_2As_2$ for $x \sim 0.25$ [18]. These experimental facts would eliminate the possibility of a nodal gap state.

To explain these experiments, simple multiband BCS models with constant impurity potential $I^b_{\alpha,\beta}$ in the band-diagonal basis (α, β 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 $\hat{I}^b$ is constant and $\det\{\hat{I}^b\} \neq 0$ [25,26]. That is, Anderson's theorem [27] is recovered in the unitary regime. However, the latter result should be reexamined since $\hat{I}^b$ 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

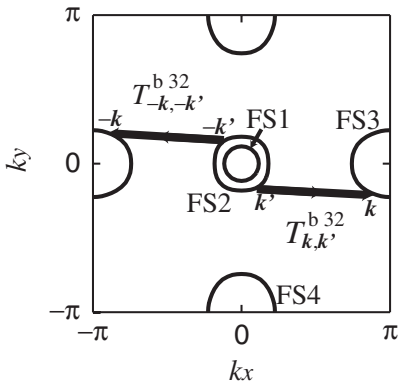

FIG. 1. Hole-pockets (FS1,2) and electron-pockets (FS3,4) in iron pnictides. $|T^{3,2}_{\mathbf{k},\mathbf{k}'}|^2$ represents the impurity induced pair-hopping amplitude between (FS2, $\pm\mathbf{k}'$) and (FS3, $\pm\mathbf{k}$).

correctly, we reveal that Anderson's theorem is seriously violated for the $s_{\pm}$-wave state, due to the interband 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.

Now, we study the 10×10 Nambu BCS Hamiltonian in the d-orbital basis; t_{2g} (xy, yz, zx) and e_g (x^2-y^2, z^2):

$$\hat{\mathcal{H}}_{\mathbf{k}} = \begin{pmatrix} \hat{H}^0_{\mathbf{k}} & \hat{\Delta}_{\mathbf{k}} \\ \hat{\Delta}^{\dagger}_{\mathbf{k}} & -\hat{H}^0_{\mathbf{k}} \end{pmatrix}, \tag{1}$$

where $\hat{H}^0_{\mathbf{k}}$ is the 5×5 hopping matrix of the five-orbital tight-binding model, which was introduced in Ref. [11]. $\Delta^{j,l}_{\mathbf{k}} = \sum_{\alpha} U^{j,\alpha}_{\mathbf{k}} \Delta_{\mathbf{k},\alpha} U^{l,\alpha *}_{\mathbf{k}}$ is the singlet gap function in the d-orbital basis, where $\Delta_{\mathbf{k},\alpha}$ is the gap function for FSα in the band-diagonal basis, and $U^{j,\alpha}_{\mathbf{k}} = \langle \mathbf{k}; j | \mathbf{k}; \alpha \rangle$ is the 5×5 transformation unitary matrix between two representations. Then, the Green function is given by

$$\hat{\mathcal{G}}_{\mathbf{k}}(i\omega_n) \equiv \begin{pmatrix} \hat{G}_{\mathbf{k}}(i\omega_n) & \hat{F}_{\mathbf{k}}(i\omega_n) \\ \hat{F}^{\dagger}_{\mathbf{k}}(i\omega_n) & -\hat{G}_{\mathbf{k}}(-i\omega_n) \end{pmatrix}^{-1} = (i\omega_n \hat{1} - \hat{\Sigma}_{\mathbf{k}}(i\omega_n) - \hat{\mathcal{H}}_{\mathbf{k}})^{-1}, \tag{2}$$

where $\omega_n = \pi T(2n+1)$ is the fermion Matsubara frequency, $\hat{G}_{\mathbf{k}}$ ($\hat{F}_{\mathbf{k}}$) is the 5×5 normal (anomalous) Green function, and $\hat{\Sigma}_{\mathbf{k}}$ 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 $n_{\rm imp}$ is much smaller than unity. We consider the local impurity potential due to the substitution of Fe by other $3d$ 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 $\mathbf{k}$ independent in the d-orbital basis, is given as

$$\hat{\mathcal{T}}(i\omega_n) = [\hat{1} - \hat{I}\hat{\mathcal{G}}_{\rm loc}(i\omega_n)]^{-1}\hat{I}, \tag{3}$$

where $\hat{\mathcal{G}}_{\rm loc}(i\omega_n) \equiv \frac{1}{N}\sum_{\mathbf{k}} \hat{\mathcal{G}}_{\mathbf{k}}(i\omega_n)$. 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 $1 \le j \le 5$, and $I_{j,l} = -I\delta_{j,l}$ for $6 \le j \le 10$. In the T-matrix approximation, the self-energy matrix in the d-orbital basis is $\mathbf{k}$ independent. It is given as

$$\hat{\Sigma}(i\omega_n) \equiv n_{\rm imp}\hat{\mathcal{T}}(i\omega_n). \tag{4}$$

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

$$\Delta^{j,j'}_{\mathbf{k}}(i\omega_n) = -\frac{T}{N}\sum_{\mathbf{q},m}\sum_{l,l'} V^{j,j';l,l'}_{\mathbf{k},\mathbf{q}}(i\omega_n, i\omega_m) F^{l,l'}_{\mathbf{q}}(i\omega_m), \tag{5}$$

where $V^{j,j';l,l'}_{\mathbf{k},\mathbf{q}}$ 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 Δ_{α}. As we will show below, the reduction in T_c in the $s_{\pm}$-wave state at $n_{\rm imp} = 0.01$ exceeds 20 K for $I \gg W_{\rm band}$. Then, the reduction in the SC gap is $\sim$36 K if $\Delta/T_c = 1.8$. Hereafter, we assume that this reduction in $\hat{\Delta}$ had been included from the beginning.

Figure 2 shows the obtained DOS in the $s_{\pm}$-wave SC state per spin for $n_{\rm imp} = 0.01$ at $T = 0$, assuming the isotropic gap in the band-diagonal basis. The number of $\mathbf{k}$ meshes is $N = 512 \times 512$. The DOS is given by the normal Green function in Eq. (2) as $N(\omega) = \frac{1}{\pi N}\sum_{\mathbf{k}} {\rm Im}\{{\rm Tr}\hat{G}_{\mathbf{k}}(\omega - i\delta)\}$. In Fig. 2(a), we put $\Delta_{1,2} = -\Delta_{3,4} = 0.05$ eV. We also study the case where only Δ_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, the 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 T-matrix in the normal state is given by the solution of

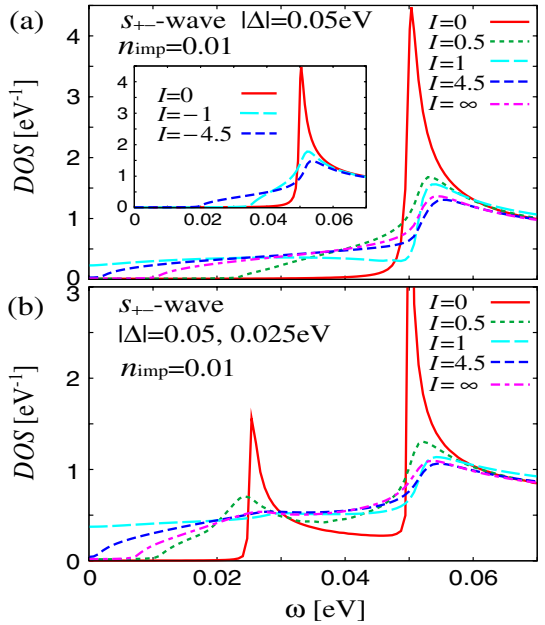

FIG. 2 (color online). Obtained DOS in the $s_{\pm}$-wave state for $n_{\rm imp} = 0.01$. (a) $|\Delta_{1,2,3,4}| = 0.05$ eV, and (b) $|\Delta_{1,3,4}| = 2|\Delta_2| = 0.05$ eV.

$$\hat{T}^{b}_{\mathbf{k},\mathbf{q}} = \hat{I}^{b}_{\mathbf{k},\mathbf{q}} + \frac{1}{N}\sum_{\mathbf{p}} \hat{I}^{b}_{\mathbf{k},\mathbf{p}} \hat{G}^{b}_{\mathbf{p}} \hat{T}^{b}_{\mathbf{p},\mathbf{q}}, \tag{6}$$

where $\hat{I}^{b}_{\mathbf{k},\mathbf{q}} = I\hat{U}^{\dagger}_{\mathbf{k}}\hat{U}_{\mathbf{q}}$ is the impurity potential in the band basis. $\hat{G}^{b}_{\mathbf{p}}$ is the normal Green function that is band diagonal. When $\mathbf{k}$ dependence of $\hat{U}_{\mathbf{k}}$ is small on each FS, $\hat{I}^{b}$ becomes constant by replacing $U^{l,\alpha}_{\mathbf{k}}$ with $\langle U^{l,\alpha}_{\mathbf{k}} \rangle_{\mathbf{k}\in \mathrm{FS}\alpha}$, which corresponds to the orbital-less model [22–26]. Using $\hat{g}^{b}_{\mathrm{loc}} \equiv \frac{1}{N}\sum_{\mathbf{k}} \hat{G}^{b}_{\mathbf{k}}$, the solution is given as

$$\hat{T}^{b} = (\hat{1} - \hat{I}^{b}\hat{g}^{b}_{\mathrm{loc}})^{-1}\hat{I}^{b}, \tag{7}$$

which becomes band diagonal as $T^{b}_{\alpha,\beta} = -1/g^{b}_{\mathrm{loc},\alpha}\delta_{\alpha,\beta}$ when $I \to \infty$, as far as $\det\{\hat{I}^{b}\} \neq 0$. Thus, the pair breaking due to interband scattering, which is described in Fig. 1, is absent for $I = \infty$ in the orbital-less model.

However, the above discussion does not hold in iron pnictides since $\hat{U}_{\mathbf{k}}$ strongly depends on $\mathbf{k}$: For example, $|U^{zx,1}_{\mathbf{k}}|^2 \sim \cos^2\theta_{\mathbf{k}}$ and $|U^{yz,1}_{\mathbf{k}}|^2 \sim \sin^2\theta_{\mathbf{k}}$ for FS1 (where $\theta_{\mathbf{k}} = \tan^{-1}(k_y/k_x) - \pi/4$), and xz, yz orbitals also constitute a large part of FS3,4 [11]. In fact, we have verified numerically that $\hat{T}^{b}_{\mathbf{k},\mathbf{p}}$ has large off diagonal elements [28]. For this reason, the impurity pair breaking for $s_{\pm}$-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 $\gamma_{\alpha} = n_{\mathrm{imp}}\mathrm{Im}T^{b}_{\alpha,\alpha}(-i0)$, 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. [29,30]. For the same reason, we find that the residual resistivity in the T-matrix approximation, which is given by $c/\rho_{\mathrm{imp}} = (2e^2/h)\sum_{\mathbf{k}}\mathrm{Tr}\{\partial\hat{H}^{0}_{\mathbf{k}}/\partial k_x \cdot \hat{G}_{\mathbf{k}}(+i0) \cdot \partial\hat{H}^{0}_{\mathbf{k}}/\partial k_x \cdot \hat{G}_{\mathbf{k}}(-i0)\}$ for $\Delta = 0$ ($h/e^2 = 6.45$ kΩ, and $c \approx 6$ Å is the interlayer spacing) [29], is larger for positive I: $\rho_{\mathrm{imp}}(n_{\mathrm{imp}} = 0.01) = 22$, 14, and 10 [$\mu\Omega$ cm] at $I = 1$, 4.5, and ∞ [eV], respectively. For $I < 0$, $\rho_{\mathrm{imp}}(n_{\mathrm{imp}} = 0.01) = 3$ and 7 [$\mu\Omega$ cm] at $I = -1$ and -4.5 [eV], respectively. In Ref. [31], ρ_{imp} for 1% Co doping in polycrystalline Nd1111 exceeds 120 $\mu\Omega$ cm, which corresponds to 30 $\mu\Omega$ cm in the single crystal. ρ_{imp} in Co-doped La1111 is still larger. Near AF quantum critical point, ρ_{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. [29]. In under-doped high-T_c cuprates, for example, ρ_{imp} due to 1% Zn impurity reaches 100 $\mu\Omega$ cm, which is 10 times of that in over-doped cuprates [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 Δ_{α} on all the FSs are the same. As n_{imp} increases, the structure of DOS is gradually smoothened.

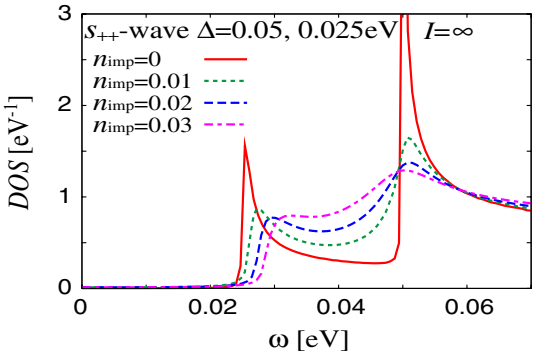

FIG. 3 (color online). Obtained DOS in the s_{++}-wave state for $I = \infty$. We put $\Delta_{1,3,4} = 2\Delta_2 = 0.05$ eV.

The absence of zero-energy state is consistent with many NMR measurements [2–4,9,10].

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

$$V^{\alpha,\beta}_{\mathbf{k},\mathbf{q}}(i\omega_n, i\omega_m) = g_{\alpha,\beta}\phi_{\mathbf{k}}\phi_{\mathbf{q}}\xi(\omega_n)\xi(\omega_m) \tag{8}$$

where $\phi_{\mathbf{k}} = 1$ for s_{++} wave, and $\phi_{\mathbf{k}} = \mathrm{sgn}\{\cos k_x \cos k_y\}$ for $s_{\pm}$ wave. We also put $\xi(\omega_n) = \omega_D^2/(\omega_n^2 + \omega_D^2)$, where ω_D is the cutoff energy. $V^{j,j';l,l'}_{\mathbf{k},\mathbf{q}}$ in Eq. (5) is given by $\sum_{\alpha,\beta} V^{\alpha,\beta}_{\mathbf{k},\mathbf{q}} U^{j,\alpha}_{\mathbf{k}} (U^{j',\alpha}_{\mathbf{k}})^* (U^{l,\beta}_{\mathbf{q}})^* U^{l',\beta}_{\mathbf{q}}$. 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 $\Sigma^{V}_{\mathbf{k}}(i\omega_n) = T\sum_{\mathbf{q},m} V_{\mathbf{k},\mathbf{q}}(i\omega_n, i\omega_m) G_{\mathbf{q}}(i\omega_m)$ vanishes for $\omega_n \to 0$ since Eq. (8) is a separate function of ω_n and ω_m [32].

To consider the superconductivity due to interband interaction between FS1,2 and FS3,4, we put $g_{\alpha,\beta}(= g_{\beta,\alpha}) = -g_1$ for $(\alpha, \beta) = (1, 3)$, $(1, 4)$, $g_{\alpha,\beta} = -g_2$ for $(\alpha, \beta) = (2, 3)$, $(2, 4)$, and $g_{\alpha,\beta} = 0$ for others. When $\omega_D = 0.03$, the obtained T_c's are shown in Fig. 4 for (a) $g_1 = g_2 = 2$ eV ($T_{c0} = 46$ K at $n_{\mathrm{imp}} = 0$) and (b) $g_1 = 3g_2 = 3$ eV ($T_{c0} = 40$ K). In the present choice of $\phi_{\mathbf{k}}$, T_{c0} is equal for both $s_{\pm}$- and s_{++}-wave states. In case (a), the obtained $|\Delta_{\alpha}|$ 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, $-\Delta T_c \equiv T_{c0} - T_c$ is independent of T_{c0}, and a qualitative relation $-\Delta T_c \sim \gamma_{\alpha}$ holds when Anderson's theorem is violated [25,32]. Since γ_{α} takes the largest value at $I \sim 1$ eV [30], T_c for the $s_{\pm}$-wave state vanishes at $n_{\mathrm{imp}} \sim 0.005$ 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 $z^{-1} = m^*/m \approx 2$ by ARPES [6,7]. Since the depairing effect is renormalized by z, $-\Delta T_c$ is reduced to $-\Delta T_c \cdot z(\sim z\gamma_{\alpha})$ [32]. When $z^{-1} = 2$, T_c for $s_{\pm}$-wave state vanishes at $n_{\mathrm{imp}} \approx 0.01$, 0.02, and 0.066 for $I = 1$ eV,

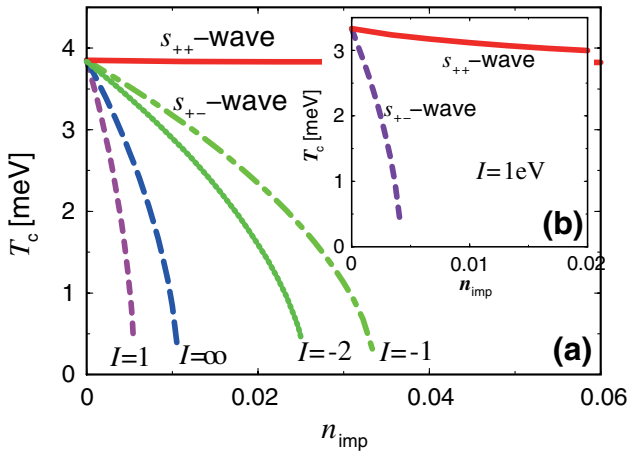

FIG. 4 (color online). Obtained T_c for the $s_{\pm}$-wave and s_{++}-wave states as functions of $n_{\rm imp}$. (a) $g_{1,2} = 2$ eV ($T_{c0} = 46$ K) and (b) $g_1 = 3g_2 = 3$ eV ($T_{c0} = 40$ K).

$I = \infty$, and $I = -1$ eV, respectively. Although $-\Delta T_c$ ranges from 46 K/% to 7 K/%, T_c for $s_{\pm}$ wave vanishes when $\rho_{\rm imp} \approx 20\ \mu\Omega\,$cm for any I. However, Sato *et al.* studied the impurity effects in polycrystalline La1111 and Nd1111, and found that T_c vanishes when $\rho_{\rm imp} \sim 3\ \mathrm{m}\Omega\,$cm, which corresponds to $\sim 750\ \mu\Omega\,$cm in the single crystal [31]. Also, ρ in single-crystal Fe(Se,Te) just above $T_c = 15$ K exceeds 450 $\mu\Omega\,$cm in Ref. [33], mainly due to elastic scattering. Then, the estimated mean free path $l_{\rm mfp}$ is as short as $a_{\rm Fe-Fe} = 2.8$ Å [34] that is about 1/10 of the coherence length in Ba122 [35]. (Note that $\rho_{\rm imp} \propto l_{\rm mfp}^{-1}$ is independent of z.) When $l_{\rm mfp} \sim a_{\rm Fe-Fe}$, even the s_{++} state will be broken by localization [31].

In summary, we have studied the effect of Fe-site substitution in iron-pnictide superconductors. Because of 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 the $s_{\pm}$-wave state is only $\rho_{\rm imp}^{\rm cr} \sim 20\ \mu\Omega\,$cm. The corresponding mean free path is $\sim 25 a_{\rm Fe-Fe}$, 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, the $s_{\pm}$-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 $\rho_{\rm imp}$. 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) the strong coupling SC state like in heavily under-doped high-T_c cuprates is realized. Thus, it is important to study many-body electronic states to clarify these possibilities.

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[34] Using the T-matrix approximation, we numerically derived the relation $\rho_{\rm imp} \approx 500(a_{\rm Fe-Fe}/l_{\rm mfp})\ \mu\Omega\,$cm, where $l_{\rm mfp}$ is the averaged mean free path. It is also obtained based on the 2D electron gas model; the conductivity due to four FSs is $4(e^2/h)k_F l_{\rm mfp}$ and $k_F \sim 0.2\pi/a_{\rm Fe-Fe}$.
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