Emergence of Fully-Gapped $s_{++}$-wave and Nodal $d$-wave States Mediated by Orbital- and Spin-Fluctuations in Ten-Orbital Model for KFe$_2$Se$_2$

Tetsuro Saito$^1$, Seiichiro Onari$^2$, and Hiroshi Kontani$^1$

$^1$ Department of Physics, Nagoya University and JST, TRIP, Furo-cho, Nagoya 464-8602, Japan.
$^2$ Department of Applied Physics, Nagoya University and JST, TRIP, Furo-cho, Nagoya 464-8602, Japan.
(Dated: January 12, 2013)

We study the superconducting state in recently discovered high-$T_c$ superconductor K$_x$Fe$_2$Se$_2$ based on the ten-orbital Hubbard-Holstein model without hole-pockets. When the Coulomb interaction is large, spin-fluctuation mediated $d$-wave state appears due to the nesting between electron-pockets. Interestingly, the symmetry of the body-centered tetragonal structure in K$_x$Fe$_2$Se$_2$ requires the existence of nodes in the $d$-wave gap, although fully-gapped $d$-wave state is realized in the case of simple tetragonal structure. In the presence of moderate electron-phonon interaction due to Fe-ion optical modes, on the other hand, orbital fluctuations give rise to the fully-gapped $s_{++}$-wave state without sign reversal. Therefore, both superconducting states are distinguishable by careful measurements of the gap structure or the impurity effect on $T_c$.

PACS numbers: 74.20.-z, 74.20.Fg, 74.20.Rp

The pairing mechanism of high-$T_c$ iron-based superconductors has been significant open problem. The main characters of FeAs compounds would be (i) the nesting between electron-pockets (e-pockets) and hole-pockets (h-pockets), and (ii) the existence of orbital degree of freedom. By focusing on the intra-orbital nesting, fully-gapped sign-reversing $s$-wave state ($s_{++}$-wave state) had been predicted based on the spin fluctuation theories. On the other hand, existence of moderate electron-phonon (e-ph) interactions due to Fe-ion optical phonons and the inter-orbital nesting can produce large orbital fluctuations. Then, orbital-fluctuation-mediated $s$-wave state without sign reversal ($s_{++}$-wave state) had been predicted by using the random-phase-approximation (RPA) or the fluctuation-exchange (FLEX) approximation. According to the analysis in Refs. 6,7 the $s_{++}$-wave state is consistent with the robustness of $T_c$ against randomness as well as the “resonance-like” hump structure in the neutron inelastic scattering. Non-Fermi liquid transport phenomena in can be explained by the development of orbital fluctuations.

Recently, iron-selenium 122-structure compound A$_2$Fe$_2$Se$_2$ (A= alkaline metals) with $T_c \sim 30$ K was discovered. This heavily electron-doped superconductor has been attracting great attention since both the band calculations and angle-resolved-photoemission-spectrum (ARPES) measurements indicate the absence of h-pockets. NMR measurements reports the weakens of spin fluctuations, and both ARPES and specific heat measurements indicate the isotropic SC gap. Thus, study of A$_2$Fe$_2$Se$_2$ will give us important information to reveal the pairing mechanism of iron pnictides.

The unit-cell of iron-based superconductors contains two Fe atoms. However, except for 122-systems, one can construct a simple “single-Fe model” from the original “two-Fe model” by applying the gauge transformation on $d$-orbitals. By this procedure, the original Brillouin zone (BZ) is enlarged to the “unfolded BZ”. Based on the single-Fe model, spin-fluctuation-mediated $d$-wave state ($B_{1g}$ representation) “without nodes” had been proposed by focusing on the nesting between e-pockets. However, we cannot construct a “single-Fe model” for 122 systems since finite hybridization between e-pockets prevents the unfolding procedure. Therefore, theoretical study based on the original two-Fe model is highly desired to conclude the gap structure.

In this paper, we study the ten-orbital (two Fe atoms) Hubbard-Holstein (HH) model for KFe$_2$Se$_2$ using the RPA. When the Coulomb interaction is large, we obtain the $d$-wave SC state due to the spin fluctuations, as predicted by the recent theoretical studies in the single-Fe Hubbard models. However, the gap function on the Fermi surfaces (FSs) inevitably has “nodal structure” in the two-Fe model, due to the symmetry requirement of the body-centered tetragonal lattice. On the other hand, orbital-fluctuation-mediated $s_{++}$-wave state is realized by small e-ph coupling; the dimensionless coupling constant $\lambda = gN(0)$ is just $\sim 0.2$. Since the nodal SC state is fragile against randomness, study of impurity effect will be useful to distinguish these SC states.

We perform the local-density-approximation (LDA) band calculation for KFe$_2$Se$_2$ using Wien2k code based on the experimental crystal structure. Next, we derive the ten-orbital tight-binding model that reproduces the LDA band structure and its orbital character using Wannier90 code and Wien2Wannier interface. The dispersion of the model and the primitive BZ are shown in Figs. 1 (a) and (b). Based on a similar ten-orbital model, Suzuki et al. studied the $s_{++}$-wave gap structure for BaFe$_2$As$_2$.

In Fig. 1 we show the FSs of KFe$_2$Se$_2$ for (c) $k_z = 0$ and (d) $k_z = \pi$ planes when the electron number per Fe-ion is $n = 6.5$: On each plane, there are four large and heavy e-pockets around X and Y points, and one small and light e-pockets around Z point. For $n = 6.5$, the energy of the h-band at $\Gamma$ point from the Fermi level,
that the reciprocal wave vector on the T' points and P and P' points are equivalent, meaning the strong evidence for the hybridization in wide momentum space. This hybridization disappears when inter-layer hoppings are neglected: Then, both xy(FS1) and xy(FS2) show cusps at $\theta = \pi/4$, and $xz(FS2)$ suddenly drops to almost zero for $\theta \geq \pi/4$. In (f), we explain the origin of nodal gap based on the fully-gapped $d$-wave solution in the single-Fe model. By introducing inter-layer hoppings, two elliptical e-pockets with positive and negative $\Delta$ in the unfolded BZ are hybridized to form FS1 and 2 with four-fold symmetry. As a result, nodal lines inevitably emerge on FS1 and 2, at least near the $|k_z| = \pi$ plane.

Here, we study the ten-orbital HH model using the RPA. As for the Coulomb interaction, we consider the intra-orbital term $U$, the inter-orbital term $U'$, Hund’s coupling or pair hopping $J$, and assume the relation $U = U' + 2J$ and $J = U/6$. In addition, we consider the $e$-electron interaction due to Fe-ion optical phonons; the phonon-mediated $e$-$e$ interaction ($-g$) and its matrix elements are presented in Ref. [4]. Hereafter, we perform the RPA on the two-dimensional planes for $k_z = 0$, $\pi/2$, and $\pi$.

For $n = 6.5$ and $k_z = 0$, the critical value of $g$ for the orbital-density-wave (ODW) is $g_c = 0.23$ eV for $U = 0$, and the critical value of $U$ for the spin-density-wave (SDW) is $U_c = 1.18$ eV for $g = 0$. These values change only $\approx 2\%$ for different $k_z$. The obtained $U-g$ phase diagram is very similar to Fig. 2 in Ref. [4], irrespective of the absence of h-pockets in KFe$_2$Se$_2$. The reason would be (i) the density-of-states (DOS) in KFe$_2$Se$_2$ is about $1\text{eV}^{-1}$ per Fe, which is comparable with other iron pnictides, and (ii) the nesting between e-pockets is rather strong because of their square-like shape. Figure 2 (a) shows the spin susceptibility $\chi^s(q, \mathbf{0})$ at $U = 1.1$ eV and $g = 0$ for $k_z = 0$ plane. $\chi^s$ is given by the intra-orbital nesting, and its peak position is $q \approx (\pi, 0, 0.4\pi)$, consistently with previous studies [22-24]. The obtained incommensurate spin correlation is the origin of the $d$-wave SC gap. Figure 2 (b) shows the off-diagonal orbital susceptibility $\chi^o_{xz,yz,xy}(q, \mathbf{0})$ for the $k_z = 0$ plane at $U = 0$ and $g = 0.22$ eV; its definition is given in Refs. [4, 25]. It is derived from the inter-orbital nesting between $xz$ and $xy$, and its peak position is $q \approx (0.7\pi, 0.4\pi)$. Note that the peak position of $\chi^o_{xz,yz-zx,xy}$ is $q \approx (0.4\pi, 0.7\pi)$. The obtained strong spin- and orbital-correlations are the origin of the $d$-wave and $s_{++}$-wave SC states.

In the following, we solve the linearized gap equation to obtain the gap function, by applying the Lanczos algorithm to achieve reliable results. In the actual calculation results shown below, we take $64 \times 64 \mathbf{k}$-point meshes and 512 Matsubara frequencies. First, we study the spin-fluctuation-mediated SC state for $U \lesssim U_c$ by putting $g = 0$. Figures 3 (a)-(c) show the gap functions of the $d$-wave solution at $T = 0.03$ eV for $k_z = 0$, $\pi/2$, and $\pi$.
and π, respectively. In case of \( U = 1.1 \text{ eV} \), the eigenvalue \( \lambda_E \) is 0.61 for (a), 0.63 for (b), and 0.62 for (c); the relation \( \lambda_E \geq 1 \) corresponds to the SC state. They are relatively small since the SC condensation energy becomes small when the SC gap has complicated nodal line structure. On the (c) \( k_z = \pi \) plane, the nodal lines are along \( \theta = \pi/4 \) and \( 3\pi/4 \) directions, consistently with the basis of \( B_{1g} \) representation in Fig. 1(d). These nodes move to near the BZ boundary, \( \theta = 0 \) and \( \pi \), on the (b) \( k_z = \pi/2 \) plane, and they deviate from the FSs on the (a) \( k_z = 0 \) plane. As results, the nodal gap appears for \( \pi/2 < |k_z| < 3\pi/2 \) in the whole BZ \(|k_z| \leq 2\pi\).

We also obtain the \( s_\pm \)-wave state, with the sign reversal of the SC gap between \( e \)-pockets and the “hidden \( h \)-pockets below the Fermi level” given by the valence bands 5, 6. The obtained solution is shown in Fig. 3(d) for \( k_z = \pi \). Interestingly, the obtained eigenvalue is \( \lambda_E = 0.99 \) for \( U = 1.1 \text{ eV} \), which is larger than \( \lambda_E \) for \( d \)-wave state in Fig. 3(a)-(c). Such large \( \lambda_E \) originates from the scattering of Cooper pairs between \( e \)-pockets and the “hidden \( h \)-pockets”, which was discussed as the “valence-band Suhl-Kondo (VBSK) effect” in the study of \( \text{Na}_x\text{CoO}_2 \) in Ref. 20.

Here, we analyze the \( T \)-dependence of \( \lambda_E \) based on a simple two-band model with inter-band repulsion: The set of gap equations is given by20  

\[
\lambda_E \Delta_h = -V N_c L_h \Delta_e \quad \text{and} \quad \lambda_E \Delta_e = -V N_h L_e \Delta_h, \tag{1}
\]

where \( V > 0 \) is the repulsive interaction between \( e \) and \( h \)-pockets, and \( N_{e,h} \) is the DOS near the Fermi level. When (i) the top of the \( h \)-pocket is well above the Fermi level, \( L_e = L_h = \ln(1.13 \omega_c / T) \), where \( \omega_c \) is the cutoff energy. Thus, the eigenvalue is given as

\[
\lambda_E = V \sqrt{N_e N_h} \ln(1.13 \omega_c / T) \propto -\ln T,
\]

similar to single-band BCS superconductors. On the other hand, when (ii) \( h \)-pocket is slightly below the Fermi level, \( L_h = (1/2) \ln(\omega_c / |E_h|) \), where \( E_h < 0 \) is the energy of the top of \( h \)-band25. Thus, the eigenvalue is given as

\[
\lambda_E = V \sqrt{N_e N_h} L_h \ln(1.13 \omega_c / T) \propto -\sqrt{-\ln T}.
\]

Therefore, in case (ii), the \( T \)-dependence of \( \lambda_E \) is much moderate. In fact, as shown in Fig. 3(a), \( \lambda_E \) for \( d \)-wave state increases monotonically with decreasing \( T \), while \( \lambda_E \) for \( s_\pm \)-wave state saturates at low temperatures. This result suggests that the \( d \)-wave state overcomes the \( s_\pm \)-wave state at \( T_c \sim 30 \text{K} \) in \( \text{K}_x\text{Fe}_2\text{Se}_2 \). Although \( T_c \) in the \( s_\pm \)-wave state is \( \sim 0.06 \text{ eV} \) in Fig. 3(a), it is greatly reduced by the self-energy correction that is absent in the RPA2.

We discuss the VBSK effect for \( s_\pm \) wave state in more detail: According to inelastic neutron scattering measurement of \( \text{Ba(FeCo)}_2\text{As}_2 \), the characteristic spin-fluctuation energy is \( \omega_{sf} \sim 100 \text{K} \) just above \( T_c \sim 30 \text{K} \). If we assume a similar \( \omega_{sf} \) in \( \text{KFe}_2\text{Se}_2 \) since \( T_c \) is close, we obtain the relation \( \omega_c \sim \omega_{sf} \ll |E_h| \) in \( \text{KFe}_2\text{Se}_2 \). Since \( L_h \) is a monotonic decrease function of \( |E_h|/\omega_c \) and \( L_h < 1 \) for \( -E_h/\omega_c > 0.15 \), we consider that \( d \)-wave state occupies the \( s_\pm \)-wave state in \( \text{KFe}_2\text{Se}_2 \), as far as the spin-fluctuation mediated superconductivity is considered. Although high-\( T_c \) \( s_\pm \)-wave state might be realized for \( |E_h|/\omega_c < 0.1 \), then the realized \( T_c \) will be very sensitive to \( E_h \) or the filling n20.

Now, we study the \( s_{++} \)-wave state due to orbital fluctuations on the \( k_z = 0 \) plane with \( n = 6.5 \). In Fig. 4(b), we show the \( \alpha_c \)-dependence of \( \lambda_E \) at \( T = 0.03 \) for the \( s_{++} \)-wave state with \( U = 0 \), and the \( \alpha_c \)-dependence of \( \lambda_E \) for the \( d \)-wave state with \( g = 0 \). Here, \( \alpha_c \) (\( \alpha_s \))
FIG. 4: (color online) (a) $T$-dependence of $\lambda_E$ for $d$- and $s_\pm$-wave states. $\lambda_E$ at $T = 0.01$ eV is underestimated because of the shortage of $k$- and Matsubara-meshes. (b) $\alpha_s$ ($\alpha_c$) dependence of $\lambda_E$ for $d$-wave ($s_{++}$-wave) state at $T = 0.03$ eV. (c) SC gap functions for $s_{++}$-wave state.

is the charge (spin) Stoner factor introduced in Ref. [3]. $\alpha_c = 1$ ($\alpha_s = 1$) corresponds to the ODW (SDW) state. In calculating the $s_{++}$-wave state, we use rather larger phonon energy: $\omega_D = 0.15$ eV, considering that the calculating temperature is about ten times larger than the real $T_c$. The SC gap functions for $s_{++}$-wave state are rather isotropic, as shown in Fig. 4 (c). However, the obtained SC gap becomes more anisotropic in case of $U > 0.5$.

We stress that the RPA is insufficient for quantitative study of $\lambda_E$ since the self-energy correction $\Sigma$ is dropped: In Ref. [5] we have studied the present model based on their FLEX approximation, and found that the critical region with $\alpha_c \gtrsim 0.95$ is enlarged by the inelastic scattering $\gamma = \text{Im}\Sigma$. Also, the $\gamma$-induced suppression in $\lambda_E$ for $d$- or $s_\pm$-wave states is more prominent than that for $s_{++}$-wave state, since $\gamma$ due to spin fluctuations is larger than that due to orbital fluctuations.

Recently, we found the paper by Mazin[27], in which the $k_z$ dependence of the nodal $d$-wave gap in Fig. 4 corresponds to Figs. 3 (a)-(c) in the present work.

In summary, we studied the mechanism of superconductivity in KFe$_2$Se$_2$ based on the ten-orbital HH model without h-pockets. Similar to iron-pnictide superconductors, orbital-fluctuation-mediated $s_{++}$-wave state is realized by small dimensionless $e$-ph coupling constant $\lambda = gN(0) \sim 0.2$. We also studied the spin-fluctuation-mediated $d$-wave state, and confirmed that nodal lines appear on the large e-pockets, due to the hybridization between two e-pockets that is inherent in 122 systems. Therefore, careful measurements on the SC gap anisotropy is useful to distinguish these different pairing mechanisms. Study of impurity effect on $T_c$ is also useful since $d$-wave (and $s_\pm$-wave) state is fragile against impurities.

Acknowledgments

We are grateful to D.J. Scalapino, P. Hirschfeld, A. Chubukov, Y. Matsuda, and other attendances in the international workshop “Iron-Based Superconductors” in KITP 2011 for useful and stimulating discussions. This study has been supported by Grants-in-Aid for Scientific Research from MEXT of Japan, and by JST, TRIP. Numerical calculations were performed using the facilities of the supercomputer centers in ISSP and Institute for Molecular Science.

1. I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, Phys. Rev. Lett. 101, 057003 (2008).
2. K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, Phys. Rev. Lett. 101, 087004 (2008).
3. H. Kontani and S. Onari, Phys. Rev. Lett. 104, 157001 (2010).
4. T. Saito, S. Onari, and H. Kontani, Phys. Rev. B 82, 144510 (2010).
5. S. Onari and H. Kontani, arXiv:1009.3882
6. S. Onari and H. Kontani, Phys. Rev. Lett. 103 177001 (2009).
7. S. Onari, H. Kontani, and M. Sato, Phys. Rev. B 81, 060504(R) (2010)
8. A. Kawabata, S. C. Lee, T. Moyal, Y. Kobayashi, and M. Sato, J. Phys. Soc. Jpn. 77 (2008) Suppl. C 103704; M. Sato, Y. Kobayashi, S. C. Lee, H. Takahashi, E. Satomi, and Y. Miura, J. Phys. Soc. Jpn. 79 (2010) 014710; S. C. Lee, E. Satomi, Y. Kobayashi, and M. Sato, J. Phys. Soc. Jpn. 79 (2010) 023702.
9. Y. Nakajima, T. Taen, Y. Tsuchiya, T. Tamegai, H. Kita-
mura, and T. Murakami. [arXiv:1009.2848]

10 A. D. Christianson, E. A. Goremychkin, R. Osborn, S. Rosenkranz, M. D. Lumsden, C. D. Malliakas, I. S. Todorov, H. Claus, D. Y. Chung, M. G. Kanatzidis, R. I. Bewley, and T. Guidi, Nature 456, 930 (2008); Y. Qi, W. Bao, Y. Zhao, C. Broholm, V. Stanev, Z. Tesanovic, Y. C. Gasparovic, S. Chang, J. Hu, B. Qian, M. Fang, and Z. Mao, Phys. Rev. Lett. 103, 067008 (2009); D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Haug, C. T. Lin, B. Keimer, and V. Hinkov, Nature Physics 6, 178 (2010).

11 S. Kasahara, T. Shibauchi, K. Hashimoto, K. Ikada, S. Tonegawa, R. Okazaki, H. Shishido, H. Ikeda, H. Takeya, K. Hirata, T. Terashima, and Y. Matsuda, Phys. Rev. B 81, 184519 (2010).

12 J. Guo, S. Jin, G. Wang, S. Wang, K. Zhu, T. Zhou, M. He, and X. Chen, Phys. Rev. B 82, 180520(R) (2010).

13 I. R. Shein and A. L. Ivanovskii, [arXiv:1012.5164]

14 I. A. Nekrasov, and M. V. Sadovskii, Pisma ZhETF, 93, 182 (2011).

15 Y. Zhang, L. X. Yang, M. Xu, Z. R. Ye, F. Chen, C. He, H. C. Xu, J. Jiang, B. F. Xie, J. J. Ying, X. F. Wang, X. H. Chen, J. P. Hu, M. Matsumani, S. Kimura, and D. L. Feng, Nature Materials, doi:10.1038/nmat2981.

16 T. Qian, X.-P. Wang, W.-C. Jin, P. Zhang, P. Richard, G. Xu, X. Dai, Z. Fang, J.-G. Guo, X.-L. Chen, and H. Ding, [arXiv:1012.6017]

17 L. Zhao, D. Mou, S. Liu, X. Jia, J. He, Y. Peng, L. Yu, X. Liu, G. Liu, S. He, X. Dong, J. Zhang, J. B. He, D. M. Wang, G. F. Chen, J. G. Guo, X. L. Chen, X. Wang, Q. Peng, Z. Wang, S. Zhang, F. Yang, Z. Xu, C. Chen, X. J. Zhou, [arXiv:1102.1057]

18 W. Yu, L. Ma, J. B. He, D. M. Wang, T.-L. Xia, G. F. Chen, [arXiv:1101.1017]

19 B. Zeng, B. Shen, G. Chen, J. He, D. Wang, C. Li, H.-H. Wen, [arXiv:1101.5117]

20 T. Miyake, K. Nakamura, R. Arita, M. Imada, J. Phys. Soc. Jpn. 79, 044705 (2010).

21 F. Wang, F. Yang, M. Gao, Z.-Y. Lu, T. Xiang, D.-H. Lee, [arXiv:1101.4390]

22 T. A. Maier, S. Graser, P. J. Hirschfeld, and D. J. Scalapino, [arXiv:1101.4988]

23 T. Das and A. V. Balatsky, [arXiv:1101.6055]

24 J. Kunes, R. Arita, P. Wissgott, A. Toschi, H. Ikeda, K. Held, Comp. Phys. Commun. 181, 1888 (2010).

25 K. Suzuki, H. Usui, and K. Kuroki, J. Phys. Soc. Jpn. 80, 013710 (2011).

26 K. Yada and H. Kontani, Phys. Rev. B 77, 184521 (2008); K. Yada and H. Kontani, J. Phys. Soc. Jpn. 75, 033705 (2006).

27 I. I. Mazin, [arXiv:1102.3655]