In theoretical studies of high $T_c$ superconductors, one of the most important challenges is to extract the minimal essence of the material that leads to the strong pairing state. After the discovery of the iron-based superconductors,[1] the nesting between electron and hole Fermi surfaces has been considered as such an essential feature, and therefore, the identity of the family. In fact, several theoretical studies suggested a possibility of spin fluctuation mediated pairing, where the spin fluctuation arises around the nesting vector ($\pi,0$).[2][3]. The spin fluctuation mediates s±-wave pairing, where the gap function has s-wave symmetry, but its sign is reversed between the electron and hole Fermi surfaces.

However, recent experiments suggest that high $T_c$ materials, obtained when the nesting is degraded, or even in the absence of the nesting,[10][14]. Then, a question of great interest is “what is the key ingredient for high $T_c$ peculiar to the iron-based superconductors?” In this context, the so-called hydrogen-doped 1111 systems, $ LnFeAsO_{1-x}H_x$ ($ Ln = Gd, Sm, Ce, La$)[14] where a large amount of electrons can be doped by O $\rightarrow$ H substitution, provide us with some important clues. For $Ln = La$, the $T_c$ vs $x$ (doping rate) phase diagram exhibits a double dome feature, and the second dome has higher $T_c$ than the first (see Fig. 2). The normal state properties above $T_c$ such as the temperature dependence of the resistivity are also different between the two domes. On the other hand, for $Ln = Sm, Ce, Gd$, the phase diagram exhibits a single dome feature, and very high $T_c$‘s close to or exceeding 50 K are observed. These single dome materials share commonalities with the second $T_c$ dome of $LaFeAsO_{1-x}H_x$[14], so that understanding the origin of the second dome directly leads to the origin of the very high $T_c$ in the iron-based superconductors.

One can easily expect that the Fermi surface nesting is degraded in the second dome compared to that in the first due to the large amount of doped electrons. In fact, the present study reveals that while the first $T_c$ dome originates from the spin fluctuation induced by the nesting of the Fermi surface having $d_{x^2-y^2}$ (and also $d_{xy}$ in some cases) orbital components, the second $T_c$ dome is due to the spin fluctuation enhanced by a peculiar motion of electrons within the $d_{xy}$, where the second neighbor diagonal hoppings are larger than the nearest neighbor ones. Such an electron motion is specific to the tetrahedral coordination of the pnictogen atoms, and we conclude that this prioritized diagonal motion is a key factor giving rise to the high $T_c$. In the single dome $T_c$ materials, “the nesting dominating” and the “prioritized diagonal motion” regimes are not well separated, and the highest $T_c \sim 50K$ is attained around the crossover regime. Then, another important key ingredient for high $T_c$ is that the $d_{xy}$ and $d_{xz/yz}$ orbitals both act as driving forces of the same pairing state, namely, s±-wave pairing. Among various multiorbital systems, this is an unparalleled feature peculiar to the iron-based superconductors.

In 1111 systems, electrons are doped into the FeAs layer by substituting O$^2-$ with F$^-$ or H$^-$ . The doping actually affects the electronic band structure in two ways; i.e., the increase of positive and negative charges in the LnO and FeAs layers, respectively, and the reduction of the As-Fe-As bond angle. The bond angle reduction occurs linearly with doping as shown in Fig.S1 of the Supplemental Material[15]. Changing the rare earth element appears as a parallel shift of the bond angle variance against the doping rate. Quite recently, this trend has further been confirmed by partially replacing As by P in SmFeAsO$_{1-x}$H$_x$[22], where a double dome phase diagram is found for sufficient amount of phosphorous substitution. To model these effects, band structure calculations are performed using the VASP code[22] for hypothetical variations of LaFeAsO, where we (i) adopt the virtual crystal approximation replacing the oxygen potential by a $1-x:x$ mixture of oxygen and fluo-
rime potentials [14], (x = 0.05 ~ 0.5 with an increment of 0.05), and (ii) vary the bond angle linearly according to x as α(x) = −7.48x + 114.36 + ∆α, where ∆α is the amount of parallel shift made with respect to the actual bond angle variance of LaFeAsO1−xHx. We consider ∆α = −3, −2, −1, 0, +1, +2°, as shown in Fig.S1 in the Supplemental Material [15]. Varying ∆α corresponds to considering materials with different rare earth (Ln) or anion (As partially replaced by P) elements [24]. To capture the essence, we vary only the bond angle, while fixing the Fe-As bond length. By extracting the bands near the Fermi level using the Wannier90 package [25], we construct models consisting of dxy, dyz, dxz, dxz−y2, and d3z2−r2 Wannier orbitals [3]. Considering that the three dimensionality is not essential to the single vs. double dome issue, we omit the interlayer hopping, and concentrate on two dimensional models in which the Brillouin zone can be unfolded to obtain a five orbital model [3].

Figure 1 shows the Fermi surface evolution with doping for ∆α = +1° and −1°. The main difference between the two cases is the presence or absence of the Fermi surface around the wave vector ((π, π), which originates from the dxy orbital [26]. The volume of the electron Fermi surfaces around (π, 0) and (0, π) increases with doping, and the hole Fermi surfaces around (0, 0), arising from the dzz/dyz orbitals, shrink. On the other hand, the volume of the dxy hole Fermi surface around (π, π) remains nearly unchanged due to the band structure variation with doping [27, 28]. In any case, the volume difference between electron and hole Fermi surfaces increases with doping, so that the nesting becomes ill-conditioned.

Considering intra- and interorbital electron-electron interactions on top of the five orbital band structure, we apply the fluctuation exchange (FLEX) approximation [6, 29, 30], to each model, and obtain the eigenvalue of the Eliashberg equation λ (at a fixed temperature of T = 0.005eV), which is taken as a measure of Tc. We take intraorbital U = 1.3eV, the interorbital U′ = U − 2J, Hund’s coupling, and the pair hoppings J = J′ = U/6. In a previous study, we adopted random phase approximation, where the self energy correction was neglected [28]. There, the eigenvalue of the Eliashberg equation was found to be monotonically enhanced with electron doping, which does not agree with the experimental observations. Also, the origin of the material dependence of the phase diagram was not clarified.

The calculated eigenvalues of the Eliashberg equation λ for ∆α = −1° ~ +2° are shown in Fig. 2(a). For ∆α = −1°, the λ against x plot shows a “single dome” variance. This is already quite interesting in that the magnitude of λ (and, hence, Tc) is maintained in such a large doping range. Even more interestingly, for ∆α = 0°, there appears a slight dip in the lightly doped regime, and this feature becomes more pronounced for ∆α = +1° and +2°. Also, the maximum Tc is obtained at a larger doping rate when ∆α is increased. Similar results are obtained also for orbital dependent interactions (Supplemental Material [15] Fig. S3). In Fig. 2(b), we

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### FIG. 1. Fermi surfaces for x = 0.05, x = 0.15, x = 0.25, x = 0.35 with (a) ∆α = −1° or (b) ∆α = +1°.

### FIG. 2. Eigenvalue of the Eliashberg equation against doping. (a) λ against doping for ∆α = −1, 0, +1, +2°. Similar in (a) for ∆α = −3, −2, −1°. (c) λ for simplified models in which only t1 is varied in conjunction with x so as to maintain the volume of the (π, π) Fermi surface. The numbers are the values of t1 (in eV). Here, t2 is fixed at 0.106 (0.113) eV for ∆α = −1 (+1). See text for more details. Upper right panel: Experimental result of Tc vs x for LnFeAs(O,H) with Ln=Gd, Sm, Ce, and La (from Ref. [14]).
show the doping dependence of $\lambda$ for $\alpha = -1^\circ \sim -3^\circ$. It can be seen that the increase of $\lambda$ with $x$ in the lightly doped regime becomes more rapid with decreasing $\Delta \alpha$. These results are overall in good agreement with the trend observed experimentally in Ref. 14 (Fig. 2 upper right panel) and also Ref. 24.

In Fig. 3 we show the doping dependence of the intraorbital spin susceptibility $\chi_{xy}$ and $\chi_{xz/yz}$ within the $d_{xy}$ and $d_{yz/d_{yz}}$ orbitals $\lambda$ for $\Delta \alpha = +1^\circ$ and $-1^\circ$. Let us first focus on $\Delta \alpha = -1$. For $x = 0.05$, there appear peaks around $(\pi,0)$ and $(0,\pi)$ in both $\chi_{xy}$ and $\chi_{xz/yz}$ reflecting the Fermi surface nesting in the lightly doped system. These peak structures are suppressed by electron doping because the nesting is degraded. However, $\chi_{xy}$ is unexpectedly reenhanced beyond $x \sim 0.2$. The reason for this cannot be the Fermi surface nesting in its original sense because the nesting is monotonically degraded by doping. For $\Delta \alpha = +1^\circ$, on the other hand, the variance of $\chi_{xy}$ is different in that there is no enhancement in the lightly doped regime. The absence of enhanced $\chi_{xy}$ here is natural because the $d_{xy}$ hole Fermi surface around $(\pi, \pi)$ is absent for $\Delta \alpha = +1^\circ$, so that there is no Fermi surface nesting. Conversely, it is surprising to find an enhancement in the largely doped regime. Interestingly, an inelastic neutron scattering experiment for LaFeAsO$_{1-x}$H$_x$ actually observes the suppression of the spin fluctuation around $x \sim 0.2$ and its reenhancement in the largely doped regime $\lambda$. Also, comparing $\Delta \alpha = -1^\circ$ and $\Delta \alpha = +1^\circ$, the spin fluctuation grows more rapidly with doping for the former than for the latter.

The doping dependence of the Eliashberg equation eigenvalue $\lambda$ and the intraorbital spin fluctuations are strongly correlated, so that understanding the latter directly leads to the understanding of the former. Since the Fermi surface evolution does not seem to be correlated with the doping dependence of the spin fluctuation, we now focus on the real-space hopping integrals within the $d_{xy}$ orbitals. In Fig. 4 we plot the doping dependence of the nearest ($t_1$) and the next nearest ($t_2$) neighbor hoppings within the $d_{xy}$ orbitals for $\Delta \alpha = -1^\circ$ and $+1^\circ$. The nearest neighbor hopping $t_1$ decreases rapidly with doping, and becomes smaller than $t_2$ at a certain doping rate $x_c \sim 0.17$ for $\Delta \alpha = -1^\circ$ and $x_c \sim 0.28$ for $\Delta \alpha = +1^\circ$. We also show in Fig. 4(b) the calculation result for the actual La1111 and Sm1111 using the experimentally determined lattice parameters. It is indeed seen that $x_c$ is larger for La than for Sm corresponding to the larger bond angle in the former. We will refer to this peculiar hopping relation $t_2 > t_1$ as “prioritized” diagonal motion (or hopping) of electrons.

The rapid decrease of $t_1$ by doping as compared to $t_2$ can be understood as a combined effect of (i) the increased positive charge in the LaO layer, (ii) reduction of the Fe-Fe distance, and (iii) increase of the pnicogen height, where (ii) and (iii) are the effects of the bond angle reduction. In the five orbital model, we consider Wannier orbitals, which implicitly take into account the Fe 3d and the hybridized As 4p atomic orbitals. If we consider these atomic orbitals explicitly, the present $t_1$ can be expressed as $t_1 = t_1^{\text{direct}} + 2t_1^{\text{indirect}}$, where $t_1^{\text{direct}}$ and $t_1^{\text{indirect}}$ are contributions from the direct hopping between Fe 3d$_{xy}$ orbitals and the indirect hopping via As 4p, respectively, as shown in Fig. 4. The two contributions have opposite signs, and $t_1^{\text{indirect}}$ dominates in the lightly doped regime. This cancellation of the direct and indirect hoppings has been discussed in Refs. 33, 34. On the other hand, the next nearest neighbor $t_2$ is mainly governed by $t_2^{\text{indirect}}$ because of the larger Fe-Fe distance.

As electrons are doped, the energy level of the As 4p orbital is lowered and moves away from the Fe 3d level due to the effect of (ii), so that the indirect hoppings decrease. The indirect contribution is also reduced because of (iii). By contrast, the direct hopping increases due to (ii). The combined effect of increasing $t_1^{\text{direct}}$ and decreasing $|t_1^{\text{indirect}}|$ results in a rapid decrease of $t_1$ with doping. The effect is weak for $t_2$ because it is mainly dominated by $t_2^{\text{indirect}}$. As can be understood from this explanation, $x_c$ is larger for materials with larger $\Delta \alpha$.

Intuitively, $t_1 < (>)t_2$ corresponds to $J_1 < (>)J_2$ in the limit of strong electron correlation $\xi, \tilde{\beta}$ since $J_1 \propto t_1^2/U$, where $U$ is the on-site intraorbital repulsion. Therefore, $t_1 < (0)t_2$ is naively expected to
be in favor of the \((\pi, 0) \left[ (\pi, \pi) \right] \) spin fluctuations. More precisely, however, the enhancement of the spin fluctuation in the largely doped regime should be traced back to the band structure (not just the Fermi surface) since we are adopting FLEX, which is essentially a weak coupling approach. In fact, as shown in the Supplemental Material\(^{[15]} \) (Fig.S2), the shape of the band changes with doping, which is mainly due to the reduction of \(t_1\). The disappearance of the van Hove singularity around the wave vector \((\pi, 0)\) (reminiscent of those commonly seen in the cuprates) works in favor of the \((\pi, 0)/(0, \pi)\) spin fluctuations over \((\pi, \pi)\).

In the models adopted above, not only \(t_1\), but also other parameters vary with electron doping. To more directly single out the reduction of \(t_1\) as the key factor, we have done the following analysis using simplified models. Namely, we start with five orbital models derived from a first principles band calculation performed with \(\Delta \alpha = -1\) or +1, both with 15\% fluorine doping. Within these models, we vary the electron density in the range of \(0.05 \leq x \leq 0.25\). If all the hoppings were fixed (rigid band), the hole Fermi surface would monotonically shrink as \(x\) increases. As seen above, however, the Fermi surface around \((\pi, \pi)\) is almost unchanged with electron doping if the variance of the lattice parameters and the O → F substitution effect is taken into account in the first principles calculation. To simulate this effect, in the simplified models, we vary only \(t_1\) by hand in conjunction with the electron doping so that the volume of the \((\pi, \pi)\) Fermi surface is the same as that for the original model. For \(\Delta \alpha = +1\), where the \((\pi, \pi)\) hole Fermi surface is absent, the energy difference between the chemical potential and the top of the \(d_{xy}\) band at \((\pi, \pi)\) is kept to be the same as that in the original model. \(\lambda\) calculated this way as a function of \(x\) for \(\Delta \alpha = \pm 1\) is shown in Fig.2(c), where a trend similar to that in Fig.2(a) is seen; when \(t_1\) is significantly larger than \(t_2\), \(\lambda\) decreases with doping, while when \(t_1\) is comparable to or smaller than \(t_2\), \(\lambda\) increases with doping.

In Fig.3 we show a schematic figure of the spin fluctuation contribution to superconductivity. For the \(d_{xz}/d_{yz}\) orbital, there is spin fluctuation mediated pair-arising from good nesting in the lightly doped regime, which is suppressed by doping because the nesting is degraded. In the \(d_{xy}\) orbital, there can be moderate Fermi surface nesting in the lightly doped regime depending on the absence or presence of the \(d_{xy}\) hole Fermi surface around \((\pi, \pi)\). Therefore, for materials with small \((\text{i.e., negative})\) \(\Delta \alpha\), the \(d_{xy}\) spin fluctuation crosses over from the nesting to the prioritized diagonal motion regime. On the other hand, in materials with large \((\text{positive})\) \(\Delta \alpha\), there is no nesting regime in the \(d_{xy}\) orbital, so that the \(d_{xy}\) spin fluctuation monotonically increases with doping.

For materials with small bond angle, the crossover from the nesting to the prioritized diagonal motion regime occurs smoothly because \(x_c\) is small. Therefore, the \(T_c\) phase diagram consists of a single dome. \(x_c\) is large for materials with large bond angle, so that the two regimes are separated, resulting in a double dome structure of the phase diagram. Interestingly, we have also come to realize a relation between the spin fluctuation and the resistivity, which is explained in the Supplemental Material\(^{[13]} \) (Fig.S4).

To conclude, our study has revealed the importance of the peculiar motion of electrons in the \(d_{xy}\) orbital, especially in cases with very high \(T_c\). Further tests for the present conclusion can be performed by examining the pressure effect. In Ref.\(^{[14]} \), it was found that apply-
ing pressure to LaFeAs(O,H) makes the double dome $T_c$ phase diagram turn into a single dome one. Our preliminary theoretical study on this problem shows that applying pressure enhances the $t_2/t_1$ ratio, and hence has an effect similar to that of replacing La by, say, Ce. Detailed analysis on this problem will be presented elsewhere. Also, it would be interesting to experimentally investigate $LnFeAs_{1-y}P_yO_{1-x}H_x$ other than $Ln$=Sm[12] as another test for the present conclusion. A surprisingly interesting feature of the iron-based superconductors is that the prioritized diagonal motion in the $d_{xy}$ orbitals and the nesting within $d_{xy}$ or $d_{x^2-y^2}$ Fermi surfaces can all be driving forces of the $s\pm$-wave superconductivity. This coherent cooperation among various components is indeed the unparalleled identity of the iron-based superconductors.

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Model of the Electronic Structure of Electron-Doped Iron-Based Superconductors: Evidence for Enhanced Spin Fluctuations by Diagonal Electron Hopping

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I. METHOD

Here we describe the details of the adopted method. In the actual materials, the bond angle reduction occurs almost linearly with doping as shown as αLa and αSm in Fig. S1. In our calculation, we vary the bond angle linearly according to x as \( \alpha(x) = -7.48x + 114.36 + \Delta\alpha \), where \( \Delta\alpha \) is the amount of parallel shift made with respect to the bond angle variance of the actual La1111.

First principles band calculation is performed by using the VASP package¹. We consider hypothetical materials LaFeAsO\(_{1-x}\)F\(_x\), where the As-Fe-As bond angle is determined as mentioned in the main text. Values of all the other lattice parameters are those determined experimentally for LaFeAsO\(_{0.95}H\(_{0.08}\). The effect of partially substituting O by F is taken into account through the virtual crystal approximation. Here, we adopt GGA-PBEsol exchange-correlation functional². The wave functions are expanded by plane waves up to cut-off energy of 550eV and 10\(^5\) k-point meshes are used. Five orbital tight-binding models are derived from the first principles band calculation exploiting the maximally localized Wannier orbitals³. The Wannier90 code is used for generating the Wannier orbitals⁴. We neglect the hopping in the \( z \) direction and concentrate on the two dimensional model for simplicity.

We consider two sets of electron-electron interactions, orbital independent and dependent ones. In standard notations, the orbital independent interactions are taken as follows: the intraorbital interaction is \( U = 1.3\) eV, the interorbital \( U’ = U - 2J \), Hund’s coupling and the pair hoppings \( J = J’ = U/6 \). The orbital dependent interactions are obtained by multiplying a constant factor (0.55 here) to all of the interactions obtained for LaFeAsO in Ref. 5. The results for the orbital independent interactions are shown in the main text, while those for the orbital dependent interactions are shown below.

We apply the FLEX⁶ method to the obtained five orbital model. In FLEX, bubble and ladder type diagrams are collected to calculate the spin \( \chi_{ijkl}^S(q, i\omega_n) \) and charge susceptibilities \( \chi_{ijkl}^C(q, i\omega_n) \), from which the self energy is calculated. Here, \( i, j, k, l \) are orbital indices, and the intraorbital susceptibilities displayed in the main text are obtained as \( \chi_{iiii}(q, i\omega_n = 0) \). The renormalized Green function is obtained from the Dyson equation. This process is repeated until a self consistent solution is obtained.

As was done in Ref. 7, we subtract the \( \omega = 0 \) component of the self energy for each iterations, which is considered to be taken into account already in the first principles band calculation. The resulting Green function and the susceptibilities are plugged into the linearized Eliashberg equation, whose eigenvalue \( \lambda \) reaches unity at \( T = T_c \), the superconducting transition temperature. Instead of going down to \( T_c \), which is generally a tedious calculation, we obtain \( \lambda \) at a fixed temperature \( (T = 0.005eV \) here), and use it as a qualitative measure for \( T_c \). In the FLEX calculation, we take 32 \( \times 32 \) k-point meshes and 4096 Matsubara frequencies. We also perform calculations for 64 \( \times 64 \) k-point meshes, or 8192 Matsubara frequencies for comparison for some of the parameter sets.

II. BAND STRUCTURE VARIATION WITH DOPOING

The band structure varies with doping as shown in Fig. S2, which is mainly due to the reduction of \( t_1 \). One effect is that the \( d_{xy} \) portion around \((\pi, \pi)\) is pushed up, which cancels out with the increase of the electron number to result in an unchanged Fermi surface. Another variation with doping occurs around the wave vector \((\pi, 0)\). For small doping rate (large \( t_1 \)), a saddle point is present in the \( d_{xy} \) band around the wave vector \((\pi, 0)/(0, \pi)\), which gives a diverging density of states.

FIG. S1. Doping dependence of the As-Fe-As bond angle. The dots are the actual experimental data.
Although this van Hove singularity itself is somewhat away from the Fermi level, it does affect the spin susceptibility, which is obtained by summing up contributions including those away from the Fermi level. This kind of band shape is reminiscent of the high $T_c$ cuprates, where the spin fluctuation is enhanced around $(\pi, \pi)$. On the other hand, the saddle point disappears (actually it moves away from $(\pi, 0)$) for small $t_1$, and becomes a local minimum. Such a band shape no longer enhances the $(\pi, \pi)$ spin fluctuation, and therefore has better matching with the Fermi surface configuration with holes and electrons around $(\pi, \pi)$ and $(\pi, 0)/(0, \pi)$, respectively, which itself is in favor of the $(\pi, 0)$ spin fluctuation. Since $x_c$ for $\Delta \alpha = -1$ is smaller than for $\Delta \alpha = +1$, the difference between $\sim (\pi, 0)$ and $(\pi, \pi)$ spin fluctuations grows faster with doping for $\Delta \alpha = -1$, as seen in Fig. 4 of main text. The present analysis shows the importance of the prioritized diagonal motion of the electrons for the enhanced $\sim (\pi, 0)$ spin fluctuation and thus $s\pm$ superconductivity.

### III. ADDITIONAL ANALYSIS REGARDING THE EIGENVALUE OF THE ELIASHBERG EQUATION AND THE SPIN FLUCTUATION

We show in Fig. S3 the doping dependence of $\lambda$ obtained by adopting orbital dependent electron-electron interactions proportional to those evaluated in Ref. 5 for LaFeAsO. It can be seen that the overall trend is the same as in the case for the orbital independent interactions presented in the main text.

In the main text, we have not put much focus on the variance of $\lambda$ against $\Delta \alpha$ (corresponding to the rare earth element dependence of $T_c$) for a fixed doping rate. As seen in Fig.2 of the main text (and also partially Fig. S3 here), $\lambda$ first increases with decreasing $\Delta \alpha$, then tends to saturate, and finally is reduced for smaller $\Delta \alpha$. This is similar to the trend observed experimentally as well as the results obtained in a previous theoretical study. In Ref. 9, the reduction of the number of hole Fermi surfaces in the small $\Delta \alpha$ regime has been proposed as the origin of the $T_c$ suppression there, but the more detailed present analysis shows that the $\lambda$ suppression occurs even when the Fermi surface multiplicity is maximized. Within the present study, the maximum $\lambda$ is attained when the $(\pi, \pi)$ $d_{xy}$ band is touching the Fermi level, i.e., when the $d_{xy}$ Fermi surface nesting appears to be ill-conditioned. In addition to $t_2 > t_1$, this condition also has to be fulfilled for the highest $T_c$. This is probably because the finite energy spin fluctuation that is responsible for pairing is enhanced when the top of the $d_{xy}$ hole band is touching

**FIG. S2.** Band structure variance against doping for $\Delta \alpha = 0^\circ$. (a) $x = 0.0$, (b) $x = 0.15$, (c) $x = 0.35$, (d) $x = 0.5$. The thickness represents the strength of the $d_{xy}$ orbital character. The $d_{xy}$ portion of the band around $(\pi, \pi)$, $(\pi, 0)$, and $(0, 0)$ are denoted as (i), (ii), and (iii), respectively.

**FIG. S3.** Eigenvalue of the Eliashberg equation against doping for orbital dependent electron-electron interactions.
the Fermi level. More detailed analysis on this issue is underway.

Regarding the doping dependence of the spin fluctuation, it is interesting to note that if we turn up side down the $d_{xy}$ orbital curves in Fig. S4, they are reminiscent of the $n$ vs. $x$ relation observed experimentally (right panel of Fig. S4)\textsuperscript{10}, where $n$ is the exponent of the temperature dependence of the resistivity. This means that the exponent is probing the strength of the spin fluctuation within the $d_{xy}$ orbital. This is natural since the portion of the Fermi surface with light mass determines the transport properties, and the light portion (the portion with large group velocity) is indeed the $d_{xy}$ orbital part of the electron Fermi surface.

**IV. RE-ENHANCEMENT OF THE $d_{xz}/d_{yz}$ SPIN FLUCTUATION**

In addition to the re-enhancement of the $d_{xy}$ spin fluctuation, the $d_{xz}/d_{yz}$ spin fluctuation is also enhanced in a even more heavily doped regime as shown in Fig. S5(a). This can again be traced back to the band structure variance with doping. The $d_{xy}$ portion of the band around $(0,0)$ above the Fermi level comes down with doping, and eventually sinks below the $d_{xz}/d_{yz}$ bands, as shown in Fig. S2 (a)~(d). This is once again due to the reduction of $t_1$. A band structure reconstruction occurs, and one of the $d_{xz}/d_{yz}$ bands changes into a concave dispersion as shown schematically in Fig. S5 (b)\textsuperscript{11–13}. Therefore, the density of states just above the Fermi level (the hole density of states) increases, thereby enhancing the electron-hole interaction between $\sim (0,0)$ and $\sim (\pi,0)$. Reflecting this re-enhancement of the $d_{xz}/d_{yz}$ spin fluctuations, there appears a “kink” in the $\lambda$ vs. $x$ plot in the heavily doped regime of $x = 0.35 \sim 0.45$ (Fig.2 of the main text). It should be noted, however, that this effect may be somewhat obscured in the actual materials due to the three dimensionality of the system, which is neglected here.

![FIG. S4. Left panel : a figure obtained by turning upside down the upper panel of Fig.5 in the main text. Right panel : the experimentally observed doping dependence of the $n$, where $n$ is the exponent of the temperature dependence of the resistivity $\rho \sim T^n$ (taken from Ref. 10).](image)

![FIG. S5. (a) Intraorbital spin susceptibilities $\chi_{xy}$ and $\chi_{xz/yz}$ for $x = 0.35$, $\Delta\alpha = -1^\circ$ (b) schematic figure of the band structure variance around $(0,0)$ with doping. Namely, this band structure reconstruction actually occurs at $k_z$ that varies with the doping rate\textsuperscript{11–13}.](image)

**V. CORRESPONDENCE BETWEEN REAL AND MOMENTUM SPACES**

In the cuprates, it is known that the nearest neighbor hopping $t_1$ dominates. This is likely to favor nearest neighbor pairing in real space, but there are two forms of nearest neighbor singlet pairing, the $d_{xz} - d_{yz}$ and the extended $s$-wave pairings. The former and the latter have sign reversing and conserving pair wave functions in real space, which corresponds to the gap forms $\cos(k_x) - \cos(k_y)$ and $\cos(k_x) + \cos(k_y)$, respectively, in momentum space. In the case of the cuprates, the density of states is large around the wave vector $(\pi,0)$ and $(0,\pi)$, and this is in favor of the $d_{xz} - d_{yz}$-wave pairing, whose gap is large around those wave vectors. On the other hand, systems with $t_2 \gg t_1$ can be considered as similar to the cuprates, but with a doubled unit cell, if we neglect $t_1$. In momentum space, this gives the gap of the form $\sin(k_x)\sin(k_y)$, namely the $d_{xy}$-wave pairing, if the density of states is large around the wave vectors $(\pm\pi/2,\pm\pi/2)$, which corresponds to $(\pi,0)/(0,\pi)$ in the halved Brillouin zone (dash dotted line). In real space, $d_{xy}$ pairing is a next nearest neighbor pairing whose wave function changes sign with 90 degrees rotation. Our present study have revealed that high $T_c$ iron-based superconductors tend to have large $t_2$, but there are no Fermi surfaces around $(\pm\pi/2,\pm\pi/2)$, which is unfavorable for $d_{xy}$ pairing. Instead they have Fermi surfaces (or bands near the Fermi level) around $(0,0)$, $(\pi,0)$, $(0,\pi)$, $(\pi,\pi)$, which are the points shifted by $(\pm\pi/2,\pm\pi/2)$ from
FIG. S6. Correspondence of pairing in real (upper) and momentum (lower panels) spaces. The signs in the upper panels represent the sign of the pair wave function. The circled areas in the lower panels are those where the density of states near the Fermi energy is large.

the wave vectors $(\pm \pi/2, \pm \pi/2)$. The $(\pm \pi/2, \pm \pi/2)$ shift transforms the $d_{xy}$ gap into $\sin(k_x - \pi/2) \sin(k_y - \pi/2) = \cos(k_x) \cos(k_y)$, i.e., the $s\pm$ form. This gap corresponds to next nearest neighbor pairing in real space, which goes hand in hand with large $t_2$, but in a sign conserving form. This relation is schematically depicted in Fig. S6.

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