A High-T_c Mechanism of Iron Pnictide Superconductivity due to Cooperation of Ferro-orbital and Antiferromagnetic Fluctuations

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The electronic states and superconductivity in iron pnictides are studied on the basis of the 16 band d-p model which includes both the onsite Coulomb interaction between Fe d electrons and the intersite one between Fe d and pnictogen p electrons. The model well accounts for experimentally observed two fluctuations: the d-d interaction-enhanced antiferromagnetic (AFM) fluctuation and the d-p interaction-enhanced ferro-orbital (FO) fluctuation responsible for the C_{66} elastic softening. The AFM fluctuation induces the repulsive pairing interaction for $q \sim Q_{\mathrm{AF}}$ while the FO does the attractive one for $q \sim 0$ resulting in the $s_\pm$-wave superconductivity where the two fluctuations cooperatively enhance the superconducting transition temperature T_c without any competition by virtue of the q-space segregation.

KEYWORDS: iron pnictide superconductors, d-p model, spin fluctuation, orbital fluctuation, elastic softening

Since the discovery of superconductivity with high transition temperature T_c in LaFeAsO$_{1-x}$F$_x$,[1] the pairing mechanisms of the iron pnictide superconductors have attracted much attention.[2,3] Two significant fluctuations: the stripe-type antiferromagnetic (AFM) fluctuation[2] which diverges towards the AFM transition and the ferro-orbital (FO) fluctuation corresponding to the O_{xy} ferro-quadrupole one which is responsible for the softening of the elastic constant C_{66}[4–6] and diverges towards the tetragonal-orthorhombic structural transition,[7] have been discussed as key ingredients for the pairing mechanisms. Theoretically, the AFM fluctuation for $q \sim Q_{\mathrm{AF}}$ corresponding to the nesting wave vector between electron and hole Fermi surfaces (FSs) was found to be enhanced by the onsite Coulomb interaction between Fe d electrons and to induce the repulsive pairing interaction for $q \sim Q_{\mathrm{AF}}$ resulting in the $s_\pm$-wave superconductivity where the gap function changes its sign between the electron and the hole FSs.[8,9]

On the other hand, the FO fluctuation for $q \sim 0$ was found to be enhanced by the electron-phonon interaction[10] and/or the mode-coupling,[11,12] where the antiferro-orbital (AFO) fluctuation for $q \sim Q_{\mathrm{AF}}$ was also enhanced due to the nesting as similar to the AFM fluctuation.[10–14] When the attractive pairing interaction mediated by the AFO fluctuation overcomes the repulsive one by the AFM fluctuation for the same wave vector $q \sim Q_{\mathrm{AF}}$, the s_{++}-wave superconductivity without the sign change of the gap function was found to be realized with the help of the attractive one for $q \sim 0$ by the FO fluctuation.[10–14] At the moment it is not clear which fluctuation is dominant for $q \sim Q_{\mathrm{AF}}$ that is crucial in determining whether the $s_\pm$- or the s_{++}-wave takes place, since the AFO fluctuation has not been explicitly observed in experiments so far.[15] In either case, the AFM and the AFO fluctuations compete with each other for the pairing interaction resulting in suppression of T_c as compared to the case with either fluctuation alone.

In this letter, we propose another mechanism of the FO fluctuation enhancement due to the intersite Coulomb interaction between Fe d and pnictogen p electrons. A recent experiment has actually provided evidence for strong coupling of Fe and pnictogen orbital polarizations (OPs).[16] Then, we employ the 16 band d-p model which explicitly includes Fe $3d$ and As $4p$ orbitals reproducing the band structure of LaFeAsO and has been extensively studied focusing on the effects of the d-d interaction[17,18] and/or the electron-phonon interaction.[10,14,19] The effect of the d-p interaction has also been investigated and found to enhance the charge fluctuation which mediates the $s_\pm$- or the s_{++}-wave pairing depending on the parameters.[20,21] However, the interaction between the Fe and As OPs depending on relative direction of d and p orbitals has not been considered there. We find that the d-p OP interaction enhances the FO fluctuation responsible for the C_{66} softening without enhancing the AFO one resulting in the $s_\pm$-wave superconductivity in collaboration with the AFM fluctuation enhanced by the d-d interaction. In this case, the experimentally observed two fluctuations cooperatively enhance T_c without any competition by virtue of the q-space segregation.

Our 16 band d-p model consists of 16 orbitals in each unit cell: five $3d$ orbitals of two Fe atoms and three $4p$ orbitals of two As atoms and is given by

$$H = H_0 + H_{\mathrm{int}}^{dd} + H_{\mathrm{int}}^{pp} + H_{\mathrm{int}}^{dp}, \qquad (1)$$

where H_0 is the non-interacting tight-binding Hamiltonian derived so as to reproduce the band structure of LaFeAsO[17,18] and H_{int}^{dd}, H_{int}^{pp} and H_{int}^{dp} represent the Coulomb interaction between the onsite Fe d electrons, the onsite As p electrons and the intersite Fe d and neighboring As p electrons, respectively. From the first-principles downfolding scheme, Miyake *et al.*[22] revealed that the Coulomb and the exchange integrals $U_{ll'}$ and $J_{ll'}$ in H_{int}^{dd} are orbital (l, l') dependent and the average of U_{ll} is $U_d = 4.2$eV for LaFeAsO, while H_{int}^{pp} and H_{int}^{dp} are also large: $U_p = 2.5$eV (LaFePO) and $U_{pd} = 1.2$eV
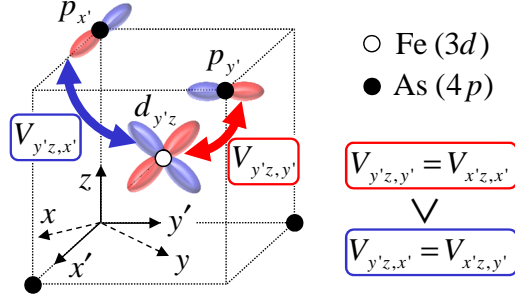
Fig. 1. (Color online) The origin of the d-p orbital polarization interaction $V' = (V_{x'z,x'} - V_{x'z,y'})/2$ due to the orbital dependence of Coulomb integrals between neighboring Fe and As sites with $V_{y'z,y'}(= V_{x'z,x'}) > V_{y'z,x'}(= V_{x'z,y'})$, where x', y' (x, y) refer to the direction along the nearest (second nearest) Fe-Fe bonds.

(LaFeAsO). As the explicit orbital dependence of H_{int}^{pp} and H_{int}^{dp} is not obtained so far, we set $U_{ll'} = U_p$ and $J_{ll'} = 0$ in H_{int}^{pp} for simplicity, and we assume that

$$H_{\text{int}}^{dp} = V \sum_{\langle i,j \rangle} \hat{n}_i^d \hat{n}_j^p + V' \sum_{\langle i,j \rangle} (\hat{n}_{ix'z}^d - \hat{n}_{iy'z}^d)(\hat{n}_{jx'}^p - \hat{n}_{jy'}^p),$$

(2)

where $\hat{n}_{il}^d$ ($\hat{n}_{jm}^p$) is the d (p) electron number operator for orbital l (m), $\hat{n}_i^d = \sum_l \hat{n}_{il}^d$ ($\hat{n}_j^p = \sum_m \hat{n}_{jm}^p$) and $\langle i,j \rangle$ are nearest-neighbor Fe and As sites. In eq. (2), V is the d-p charge transfer interaction which was found to enhance the charge fluctuation as originally discussed for the cuprate superconductors[23,24] and also for the iron pnictides,[20,21] and $V' = (V_{x'z,x'} - V_{x'z,y'})/2$ is the d-p OP interaction derived from the orbital dependence of the Coulomb integrals between Fe d_l and As p_m orbitals $V_{l,m}$ for $l = d_{x'z}$ or $d_{y'z}$ and $m = p_{x'}$ or $p_{y'}$ as shown in Fig. 1, where x', y' (x, y) refer to the direction along the nearest (second nearest) Fe-Fe bonds. A rough estimation with the use of hydrogenlike atomic wave functions yields a considerably large value of V' with several tenth percent of V. Here, we focus only on the $d_{x'z}$-$d_{y'z}$ type OP interaction crucial for the FO fluctuation responsible for the C_{66} softening but the effects of the other types of OP interaction will be discussed later. We also neglect the interaction between Fe charge and As OP which was found to enhance the charge fluctuations for $\boldsymbol{q} \neq \boldsymbol{0}$[21] but is almost irrelevant for the FO fluctuation.

Now, we investigate the Hamiltonian eq. (1) within the random phase approximation (RPA), where the spin and charge-orbital susceptibilities are given in the 68×68 matrix representation as[20]

$$\hat{\chi}^s(q) = \left[\hat{1} - \hat{\chi}^0(q)\hat{\Gamma}^s \right]^{-1} \hat{\chi}_0(q),$$

(3)

$$\hat{\chi}^c(q) = \left[\hat{1} + \hat{\chi}^0(q)\hat{\Gamma}^c(\boldsymbol{q}) \right]^{-1} \hat{\chi}_0(q)$$

(4)

with the noninteracting susceptibility $\chi^0_{\mu_1\mu_2\mu_3\mu_4}(q) = -\frac{T}{N} \sum_k G_{\mu_3\mu_1}(k + q)G_{\mu_2\mu_4}(k)$, where μ represents Fe d or As p orbitals and $\hat{G}(k) = [i\varepsilon_m \hat{1} - \hat{H}_0(\boldsymbol{k})]^{-1}$ is the noninteracting Green's function (16×16 matrix); $k = (\boldsymbol{k}, i\varepsilon_m)$ and $q = (\boldsymbol{q}, i\omega_n)$ with the wave vectors $\boldsymbol{k}, \boldsymbol{q}$ and the Matsubara frequencies $\varepsilon_m = 2(m + 1)\pi T$, $\omega_n = 2n\pi T$, and $\hat{1}$ is the unit matrix in μ basis. In

eqs. (3) and (4), $\hat{\Gamma}^{s(c)}$ is the spin (charge-orbital) vertex (68×68 matrix) in which the nonzero elements are as follows: $\Gamma_{llll}^{s(c)} = U_{ll}$ (U_{ll}), $\Gamma_{ll'll'}^{s(c)} = U_{ll'}$ ($-U_{ll'} + 2J_{ll'}$), $\Gamma_{ll'l'l}^{s(c)} = J_{ll'}$ ($2U_{ll'} - J_{ll'}$), $\Gamma_{ll'l'l}^{s(c)} = J_{ll'}$ ($J_{ll'}$) in the 25×25 d-d submatrix for each Fe atom: Fe^1, Fe^2, and $\Gamma_{mmmm}^s = \Gamma_{mm'mm'}^s = \Gamma_{mmmm}^c = -\Gamma_{mm'mm'}^c = U_p$, $\Gamma_{mmm'm'}^c = 2U_p$ in the 9×9 p-p submatrix for each As atom: As^1, As^2, where $l(\neq l') = d_{x^2-y^2}, d_{3z^2-r^2}, d_{xz}, d_{yz}, d_{xy}$ and $m(\neq m') = p_x, p_y, p_z$. The nonzero elements in the 50×18 d-p submatrix are: $\Gamma_{llmm}^c(\boldsymbol{q}) = 2V\phi_{\alpha\beta}(\boldsymbol{q})$ for all $l = d_l$ and $m = p_m$, $\Gamma_{ll'mm'}^c(\boldsymbol{q}) = 2V'\phi_{\alpha\beta}(\boldsymbol{q})$ for $l(\neq l') = d_{xz}, d_{yz}$ and $m(\neq m') = p_x, p_y$ with the $\boldsymbol{q}$ dependent factor $\phi_{\alpha\beta}(\boldsymbol{q}) = 1 + e^{iq_\nu}$ due to intersite Fe-As contributions, where $q_\nu = -(+)q_x$ for $(\alpha, \beta) = (Fe^{1(2)}, As^{1(2)})$ and $q_\nu = -(+)q_y$ for $(\alpha, \beta) = (Fe^{1(2)}, As^{2(1)})$. Here, we note that the longitudinal $d_{x'z}$-$d_{y'z}$ and $p_{x'}$-$p_{y'}$ polarizations coupled via V' with each other (see eq. (2)) are transformed into the transverse d_{xz}-d_{yz} and p_x-p_y ones by $45°$ rotation as shown in Fig. 1, respectively.

When the largest eigenvalue $\alpha_{s(c)}(\boldsymbol{q})$, called the spin (charge-orbital) Stoner factor, of the matrix $(-)\hat{\chi}^0(q)\hat{\Gamma}^{s(c)}$ in eq. (3) (eq. (4)) for a wave vector $\boldsymbol{q}$ with $i\omega_n = 0$ reaches unity, the instability towards the magnetic (charge-orbital) order with the corresponding $\boldsymbol{q}$ takes place. To examine the superconductivity, we solve the linearized Eliashberg equation

$$\lambda\Delta_{\mu\mu'}(k) = -\frac{T}{N} \sum_{k'} \sum_{\mu_1\mu_2\mu_3\mu_4} V_{\mu\mu_1\mu_2\mu'}(k - k')$$
$$\times G_{\mu_3\mu_1}(-k')\Delta_{\mu_3\mu_4}(k')G_{\mu_4\mu_2}(k'), \quad (5)$$

and obtain the superconducting gap function $\hat{\Delta}(k)$ (16×16 matrix) with the eigenvalue λ which becomes unity at the superconducting transition temperature T_c, where the effective pairing interaction for the spin-singlet state is given in the 68×68 matrix representation as

$$\hat{V}(q) = \frac{3}{2}\hat{\Gamma}^s\hat{\chi}^s(q)\hat{\Gamma}^s - \frac{1}{2}\hat{\Gamma}^c(\boldsymbol{q})\hat{\chi}^c(q)\hat{\Gamma}^c(\boldsymbol{q}) + \frac{1}{2}\left(\hat{\Gamma}^s + \hat{\Gamma}^c(\boldsymbol{q})\right).$$

(6)

The RPA calculations are performed with 32×32 $\boldsymbol{k}$-point meshes and 512 Matsubara frequencies for the temperature $T = 0.02\text{eV}$ and the number of electrons per unit cell $n = 24.2$ corresponding to 10% electron doping. We employ the d-d interaction parameters $U_{ll'}$ and $J_{ll'}$ obtained in Ref.[22] by multiplying a reduction factor $f_d = 0.37 - 0.40$ as done for the 5 orbital Hubbard model with $f = 0.42$[25] since the RPA overestimates the magnetic order and fluctuation as explicitly shown in Ref.[26] The p-p interaction $U_p = 2.5\text{eV}$ is taken from Ref.,[22] and the d-p charge transfer interaction $V = 0.5\text{eV}$ is assumed to be smaller than $U_{pd} = 1.2\text{eV}$ from Ref.[22] in order to avoid the instability towards the phase separation that occurs for $V \gtrsim 0.57\text{eV}$[27] but disappears with taking proper account of the long-range Coulomb interaction[28] which suppresses the uniform charge fluctuation due to charge screening effect[21] but not the FO fluctuation enhanced by the d-p OP interaction V' as mentioned below.
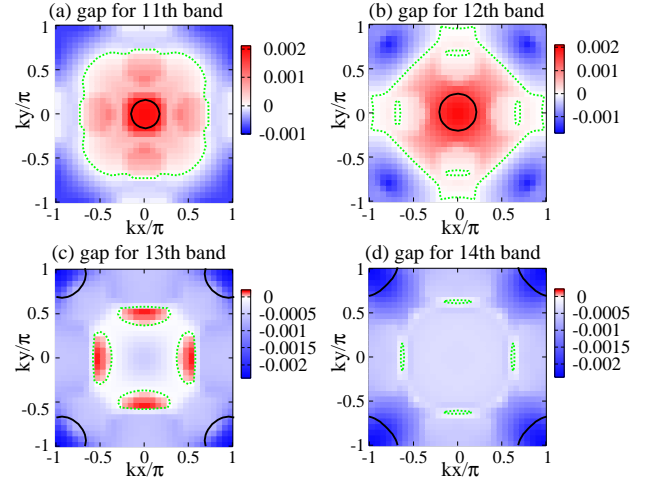
In Fig. 2 (a), we plot the spin (charge-orbital) Stoner
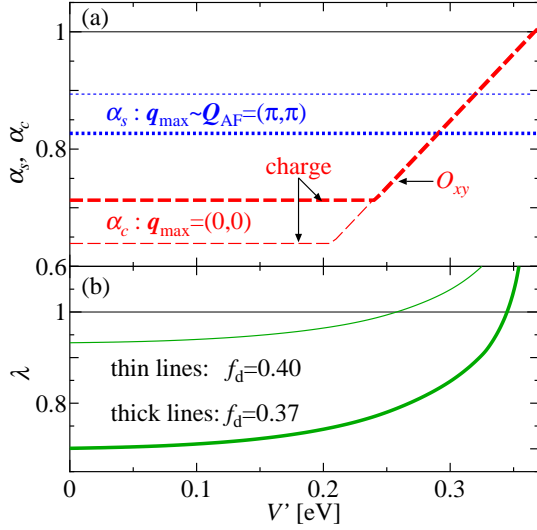
Fig. 2. (Color online) The spin and charge-orbital Stoner factors α_s (dotted lines) and α_c (dashed lines) for $\boldsymbol{q}_{\max}$ (a), and the eigenvalue of the Eliashberg equation λ (b) for the d-d interaction reduction factor $f_d = 0.37$ (thick lines) and $f_d = 0.40$ (thin lines) as functions of the d-p orbital polarization interaction V'.

Fig. 3. (Color online) The band representation of the gap function $\hat{\Delta}(\boldsymbol{k}, i\pi T)$ for the 11th (a) and 12th (b) (hole) bands and the 13th (c) and 14th (d) (electron) bands in the Brillouin zone corresponding to two FeAs per unit cell for $f_d = 0.37$ and $V' = 0.345$eV with $\lambda = 1$, where black solid and green dotted lines represent the FSs and the nodes of the gap functions, respectively.

factor $\alpha_{s(c)}$ for $\boldsymbol{q} = \boldsymbol{q}_{\max}$ at which $\alpha_{s(c)}$ shows a maximum as a function of the d-p OP interaction V' for the d-d interaction reduction factor $f_d = 0.37$ and 0.40. As V' appears only in the d_{xz}-d_{yz} off-diagonal elements of $\hat{\Gamma}^c(\boldsymbol{q})$ which takes a maximum value of $4V'$ at $\boldsymbol{q} = \boldsymbol{0}$ due to the $\boldsymbol{q}$ dependent factor $\phi_{\alpha\beta}(\boldsymbol{q})$, V' enhances the transverse d_{xz}-d_{yz} (longitudinal $d_{x'z}$-$d_{y'z}$) susceptibility which contributes to the O_{xy} quadrupole one $\chi_{O_{xy}}(\boldsymbol{q}) = \sum_{l_1l_2l_3l_4}[\hat{O}_{xy}]_{l_1l_2}[\hat{O}_{xy}]_{l_4l_3}\chi^c_{l_1l_2l_3l_4}(\boldsymbol{q},0)$ for $\boldsymbol{q} \sim \boldsymbol{0}$ responsible for the C_{66} softening,[7] while the charge susceptibility $\chi_c(\boldsymbol{q})$ which is enhanced by V for $\boldsymbol{q} \sim \boldsymbol{0}$[20, 21] is independent of V'. Therefore, $\chi_{O_{xy}}(\boldsymbol{0})$ monotonically increases with increasing V' and dominates over $\chi_c(\boldsymbol{0})$ at a certain V' at which α_c for $\boldsymbol{q}_{\max} = \boldsymbol{0}$ shows a kink as shown in Fig. 2 (a). Above the kink, α_c linearly increases with increasing V' and finally reaches unity at a critical value $V'_c = 0.365$eV where the instability towards the FO order with different occupations of $d_{x'z}$ and $d_{y'z}$ orbitals observed in the orthorhombic phase[29] takes place. We see from comparison between the results with $f_d = 0.37$ and 0.40 that the d-d interaction enhances the stripe-type AFM susceptibility for $\boldsymbol{q} \sim \boldsymbol{Q}_{AF} = (\pi, \pi)$, which is independent of V and V' included only in $\hat{\Gamma}^c(\boldsymbol{q})$, and suppresses $\chi_c(\boldsymbol{0})$ while keeping $\chi_{O_{xy}}(\boldsymbol{0})$ almost unchanged.

Fig. 2 (b) shows the eigenvalue λ of the Eliashberg equation (5) as a function of V' for $f_d = 0.37$ and 0.40. For $f_d = 0.37$ (0.40), λ reaches unity at $V' = 0.345$ (0.255)eV where $\alpha_c = 0.955$ ($\alpha_s = 0.894$) is larger than $\alpha_s = 0.827$ ($\alpha_c = 0.750$) and then the FO (AFM) fluctuation gives a larger contribution to the superconductivity relative to the AFM (FO) one. In both cases, $V' \sim 0.3$eV is a realistic value of the parameter as compared to the roughly estimated value mentioned before. In the FO fluctuation-dominated case with $f_d = 0.37$, the gap function on the hole FSs (Figs. 3 (a) and (b)) and the electron FSs (Figs. 3 (c) and (d)) has almost the same absolute value without nodes and changes its

sign between the hole and electron FSs, i.e., the fully gapped $s_\pm$-wave state which is obtained also for the AFM fluctuation-dominated case with $f_d = 0.40$ (not shown). In the orbital representation, the d-d component $\Delta^{dd}_{ll'}$ with $l = l' = d_{xz}$ or d_{yz} has the largest value but the d-p component Δ^{dp}_{lm} with $l = d_{xz}$ (d_{yz}) and $m = p_y$ (p_x) is also large $\sim 40\%$ of Δ^{dd}_{ll}, while the p-p component $\Delta^{pp}_{mm'}$ is small $\sim$ less than $1/10$ of Δ^{dd}_{ll}. Then, the d-p correlation effects are important not only for the FO fluctuation enhancement inducing the pairing interaction but also for the superconducting state itself. To see the latter effect explicitly, we solve the Eliashberg equation (5) in the absence of Δ^{dp}_{lm} and find that the obtained λ is about 0.2 smaller than that in the presence of Δ^{dp}_{lm}.

The $\boldsymbol{q}$ dependence of several susceptibilities and pairing interactions with $i\omega_n = 0$ are shown in Fig. 4 for the same parameters in Fig. 3. The spin susceptibility is enhanced by d-d interaction for $\boldsymbol{q} \sim \boldsymbol{Q}_{AF} = (\pi, \pi)$ due to the nesting effect (Fig. 4 (a)), while the transverse d_{xz}-d_{yz} orbital susceptibility (Fig. 4 (c)) contributing to the O_{xy} quadrupole one (Fig. 4 (b)) and the transverse p_x-p_y orbital one (Fig. 4 (d)) are simultaneously enhanced by the d-p OP interaction V' for $\boldsymbol{q} \sim \boldsymbol{0}$ due to the (intersite) $\boldsymbol{q}$ dependent factor $\phi_{\alpha\beta}(\boldsymbol{q})$. As seen from eq. (6) for the pairing interaction $\hat{V}(q)$, the FO fluctuation induces a large attractive $\hat{V}(q)$ for $\boldsymbol{q} \sim \boldsymbol{0}$ (Fig. 4 (f)) which mediates the s-wave pairing within each of the electron and the hole FSs almost independently of each other resulting in nearly degenerate s_{++} and $s_\pm$-wave pairings, while the AFM fluctuation induces a repulsive $\hat{V}(q)$ for $\boldsymbol{q} \sim \boldsymbol{Q}_{AF}$ (Fig. 4 (e)) which causes pair scattering between the electron and hole FSs and enhances the gap function with the sign change between the FSs, i.e., the $s_\pm$-wave pairing as shown in Fig. 3. In this case, the FO and the AFM fluctuations cooperatively enhance T_c without any competition by virtue of the $\boldsymbol{q}$-space segregation, although T_c is not explicitly shown here but has the same tendency
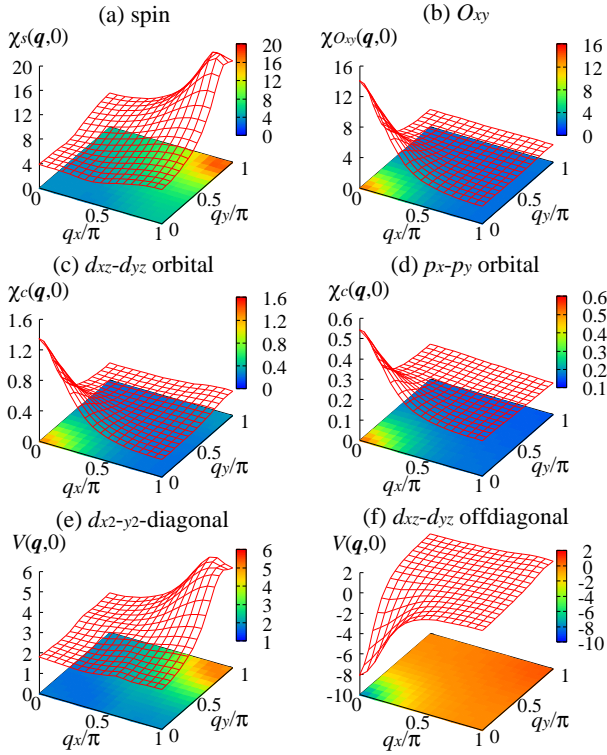
Fig. 4. (Color online) The susceptibilities for (a) spin χ_s, (b) O_{xy} quadrupole $\chi_{O_{xy}}$, (c) transverse d_{xz}-d_{yz} orbital $\chi^c_{xz,yz,xz,yz}$, (d) transverse p_x-p_y orbital $\chi^c_{x,y,x,y}$ and the pairing interactions for (e) $d_{x^2-y^2}(\equiv 1)$ diagonal element V_{1111} and (f) d_{xz}-d_{yz} off-diagonal element $V_{xz,yz,yz,xz}$ as functions of q with $i\omega_n = 0$ for $f_d = 0.37$ and $V' = 0.345$eV with $\lambda = 1$.

as λ that is a monotonically increasing function of f_d and V' as shown in Fig. 2 (b).

In summary, we have found that the d-p OP interaction derived from the orbital dependence of the Coulomb integrals between Fe d and As p orbitals is crucial for enhancing the FO fluctuation responsible for the C_{66} softening. The FO fluctuation induces the attractive pairing interaction for $q \sim 0$ which mediates the s-wave pairing within each of the electron and the hole FSs, while the AFM fluctuation enhanced by the d-d interaction induces the repulsive one for $q \sim Q_{AF}$ resulting in the $s_{\pm}$-wave pairing, where the experimentally observed FO and AFM fluctuations cooperatively enhance T_c without any competition by virtue of the q-space segregation.

In the present study, we have focused only on the $d_{x'z}$-$d_{y'z}$ ($p_{x'}$-$p_{y'}$) type OP interaction responsible for the C_{66} softening, but a preliminary examination with including the other types shows that the $d_{xz(yz)}$-$d_{x^2-y^2}$ (p_z-$p_{x(y)}$) type OP interaction enhances the $O_{3z^2-r^2}$ quadrupole fluctuation responsible for the C_{33} softening which was recently observed by the ultrasonic experiment[30] and was discussed by the mode-coupling theory.[31] More recently, an electron diffraction experiment revealed the strong coupling of Fe and As orbital polarizations along the c-axis[16] which can be also explained by the d-p OP interaction between Fe $d_{xz(yz)}$-$d_{x^2-y^2}$ and As p_z-$p_{x(y)}$ polarizations. Hence, detailed calculations with including the complete matrix elements of the d-p interaction together with the electron-phonon interaction crucial for

explicit description of the phonon softening[7,15] are important future problems.

Finally, we briefly discuss the impurity effect on T_c which is robust against nonmagnetic impurities[32] and is not consistent with the $s_{\pm}$-wave pairing.[33] As for the $s_{\pm}$-wave pairing obtained in the present study, however, the impurity effect is considered to largely depend on the parameters: it is large for the AFM fluctuation-dominated case where T_c is mainly determined by the repulsive pairing interaction for $q \sim Q_{AF}$ while small for the FO fluctuation-dominated case where T_c is mainly determined by the attractive one for $q \sim 0$. Explicit calculations of the impurity effect on T_c are now under way and will be reported in a subsequent paper.

Acknowledgments

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