

Self-Consistent Vertex Correction Analysis for Iron-Based Superconductors: Mechanism of Coulomb-Interaction-Driven Orbital Fluctuations

Seiichiro ONARI¹ and Hiroshi KONTANI²

¹ *Department of Applied Physics, Nagoya University and JST, TRIP, Furo-cho, Nagoya 464-8602, Japan.*

² *Department of Physics, Nagoya University and JST, TRIP, Furo-cho, Nagoya 464-8602, Japan.*

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We study the mechanism of orbital/spin fluctuations due to multiorbital Coulomb interaction in iron-based superconductors, going beyond the random-phase-approximation. For this purpose, we develop a self-consistent vertex correction (SC-VC) method, and find that multiple orbital fluctuations in addition to spin fluctuations are mutually emphasized by the “multimode interference effect” described by the VC. Then, both the antiferro-orbital and ferro-orbital (=nematic) fluctuations simultaneously develop for $J/U \sim 0.1$, both of which contribute to the s -wave superconductivity. Especially, the ferro-orbital fluctuations give the orthorhombic structure transition as well as the softening of shear modulus C_{66} .

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Since the discovery of iron-based superconductors, the mechanism of high- T_c superconductivity has been studied very actively. Theoretically, both the spin-fluctuation-mediated s_{\pm} -wave state (with sign reversal of the gap between hole-pocket (h-pocket) and electron-pocket (e-pocket)) [1–5] and the orbital-fluctuation-mediated s_{++} -wave state (without sign reversal) [6, 7] had been proposed. The latter scenario is supported by the robustness of T_c against impurities in many iron-pnictides [8–12]. Possibility of impurity-induced crossover from s_{\pm} to s_{++} states had been discussed theoretically [3, 6]. Also, orbital-independent gap observed in $\text{BaFe}_2(\text{As,P})_2$ and $(\text{K,Ba})\text{Fe}_2\text{As}_2$ by laser ARPES measurement [13, 14] as well as the “resonance-like” hump structure in the neutron inelastic scattering [15] are consistent with the orbital fluctuation scenario.

Nature of orbital fluctuations has been studied intensively after the discovery of large softening of the shear modulus C_{66} [16–18] and renormalization of phonon velocity [19] observed well above the orthorhombic structure transition temperature T_S . Consistently, a sizable orbital polarization is observed in the orthorhombic phase [20, 21]. Moreover, the “electronic nematic state” with large in-plane anisotropy of resistivity or magnetization well above T_S and T_c [22, 23], also indicates the occurrence of (impurity-induced local) orbital order [24].

Origin of orbital order/fluctuation had been actively discussed, mainly based on the multiorbital Hubbard model with intra (inter) orbital interaction U (U') and the exchange interaction $J = (U - U')/2 > 0$ [6, 25]. We had focused attention to a good *inter-orbital nesting* of the Fermi surfaces shown in Fig. 1 (a): Although moderate orbital fluctuations are induced by U' in the random-phase-approximation (RPA), the spin susceptibility due to the intra-orbital nesting, $\chi^s(\mathbf{q})$, is the most divergent for $J > 0$ (*i.e.*, $U > U'$). Since $J/U \approx 0.12 - 0.15$ according to the first-principle study [26], the RPA fails to explain experimental “nonmagnetic” structure transition.

This situation is unchanged even if the self-energy correction is considered in the fluctuation-exchange (FLEX) approximation [27].

To explain the strong development of orbital fluctuations, we had introduced a quadrupole interaction [6]:

$$H_{\text{quad}} = -g \sum_i \left(\hat{O}_{xz}^i \hat{O}_{xz}^i + \hat{O}_{yz}^i \hat{O}_{yz}^i \right) \quad (1)$$

where g is the coupling constant, and \hat{O}_{γ} is the charge quadrupole operator ; $\gamma = xz, yz, xy, x^2 - y^2, 3z^2 - r^2$. (Hereafter, x, y -axes (X, Y -axes) are along the nearest Fe-Fe (Fe-As) direction.) This term is actually caused by the electron-phonon (e -ph) coupling due to in-plane Fe-ion oscillations [6, 14, 27]. Since $\hat{O}_{xz(yz)}$ induces the inter-orbital scattering, strong antiferro (AF) orbital fluctuations develop for $g \gtrsim 0.2\text{eV}$ owing to a good inter-orbital nesting. We also studied the vertex correction (VC) beyond the RPA [28], and obtained strong enhancement of ferro-quadrupole ($\hat{O}_{x^2-y^2} \propto \hat{n}_{xz} - \hat{n}_{yz}$) susceptibility $\chi_{x^2-y^2}^c(\mathbf{0})$, which causes the orthorhombic structure transition and the softening of C_{66} [28]. This “nematic fluctuation” is derived from the interference of two AF orbitons due to the symmetry relation $\hat{O}_{x^2-y^2}(\mathbf{0}) \sim \hat{O}_{XZ}(\mathbf{Q}) \times \hat{O}_{YZ}(-\mathbf{Q})$, where $\hat{O}_{XZ(YZ)} = [\hat{O}_{xz} + (-)\hat{O}_{yz}]/\sqrt{2}$. Then, it was natural to expect that such multi-orbital interference effect, which is given by the VC while dropped in the RPA, induces large “Coulomb-interaction-driven” orbital fluctuations.

In this letter, we study the orbital and spin fluctuations in iron-based superconductors by considering the multiorbital Coulomb interaction with $U = U' + 2J$ and $J/U \sim O(0.1)$. We develop the self-consistent-VC (SC-VC) method, and find that both ferro- $O_{x^2-y^2}$ and AF- $O_{xz/yz}$ fluctuations strongly develop even for $J/U \sim 0.1$, due to the inter-orbital nesting and the positive interference between multi-fluctuation (orbital+magnon) modes. This result leads to a conclusion that RPA *underestimates* the orbital fluctuations in multiorbital sys-

tems. The present study offers a unified explanation for both the superconductivity and structure transition in many compounds.

Here, we study the five-orbital Hubbard model introduced in Ref. [1]. We denote d -orbitals $m = 3z^2 - r^2$, xz , yz , xy , and $x^2 - y^2$ as 1, 2, 3, 4 and 5, respectively. The Fermi surfaces are mainly composed of orbitals 2, 3 and 4 [28]. Then, the susceptibility for the charge (spin) channel is given by the following 25×25 matrix form in the orbital basis:

$$\hat{\chi}^{c(s)}(q) = \hat{\chi}^{\text{irr},c(s)}(q)(1 - \hat{\Gamma}^{c(s)}\hat{\chi}^{\text{irr},c(s)}(q))^{-1}, \quad (2)$$

where $q = (\mathbf{q}, \omega_l = 2\pi lT)$, and $\hat{\Gamma}^{c(s)}$ represents the Coulomb interaction for the charge (spin) channel composed of U , U' and J given in Refs. [6, 14]. The irreducible susceptibility in Eq. (2) is given as

$$\hat{\chi}^{\text{irr},c(s)}(q) = \hat{\chi}^0(q) + \hat{X}^{c(s)}(q), \quad (3)$$

where $\chi_{ll',mm'}^0(q) = -T \sum_p G_{lm}(p+q)G_{m'l'}(p)$ is the bare bubble, and the second term is the VC (or orbiton or magnon self-energy) that is neglected in both RPA and FLEX approximation. In the present discussion, it is convenient to consider the quadrupole susceptibilities:

$$\begin{aligned} \chi_{\gamma,\gamma'}^c(q) &\equiv \sum_{ll',mm'} O_{\gamma}^{l,l'} \chi_{ll',mm'}^c(q) O_{\gamma'}^{m',m} \\ &= \text{Tr}\{\hat{O}_{\gamma}\hat{\chi}^c(q)\hat{O}_{\gamma'}\}. \end{aligned} \quad (4)$$

Non-zero matrix elements of the quadrupole operators for the orbital 2 \sim 4 are $O_{xz}^{3,4} = O_{yz}^{2,4} = O_{x^2-y^2}^{2,2} = -O_{x^2-y^2}^{3,3} = 1$ [28]. Because of the symmetry, the off-diagonal susceptibilities ($\gamma \neq \gamma'$) are zero or very small for $\mathbf{q} = \mathbf{0}$ and the nesting vector $\mathbf{Q} \approx (\pi, 0)$ or $\mathbf{Q}' \approx (0, \pi)$ [28]. We do not discuss the angular momentum (dipole) susceptibility, $\chi_{\mu}^c(\mathbf{q}) \sim \langle \hat{l}_{\mu}(\mathbf{q})\hat{l}_{\mu}(-\mathbf{q}) \rangle$, since it is found to be suppressed by the VC. Note that $\hat{O}_{\mu\nu} \propto \hat{l}_{\mu}\hat{l}_{\nu} + \hat{l}_{\nu}\hat{l}_{\mu}$.

To measure the distance from the criticality, we introduce the charge (spin) Stoner factor $\alpha_{\mathbf{q}}^{c(s)}$, which is the largest eigenvalue of $\hat{\Gamma}^{c(s)}\hat{\chi}^{\text{irr},c(s)}(q)$ at $\omega_l = 0$: The charge (spin) susceptibility diverges when $\alpha_{\text{max}}^{c(s)} \equiv \max_{\mathbf{q}}\{\alpha_{\mathbf{q}}^{c(s)}\} = 1$. In a special case $J = 0$, the relation $\alpha_{\text{max}}^s = \alpha_{\text{max}}^c$ holds at the momentum \mathbf{Q} in the RPA; see Fig. 1 (b). That is, both spin and orbital susceptibilities are equally enhanced at $J = 0$, which is unchanged by the self-energy correction in the FLEX approximation [27]. For $J > 0$, the spin fluctuations are always dominant ($\alpha_{\text{max}}^s > \alpha_{\text{max}}^c$) in the RPA or FLEX. However, because of large $\hat{X}^c(q)$, the opposite relation $\alpha_{\text{max}}^s \lesssim \alpha_{\text{max}}^c$ can be realized even for $J/U \lesssim 0.1$ in the SC-VC method.

First, we perform the RPA calculation for $n = 6.1$ and $T = 0.05$, using 32×32 \mathbf{k} -meshes: The unit of energy is eV hereafter. Figure 1 (c) shows the diagonal quadrupole susceptibilities for $J/U = 0.088$; $\chi_{\gamma}^c(q) \equiv \chi_{\gamma\gamma}^c(q)$. (The spin susceptibility is shown in Ref. [1].) The Stoner

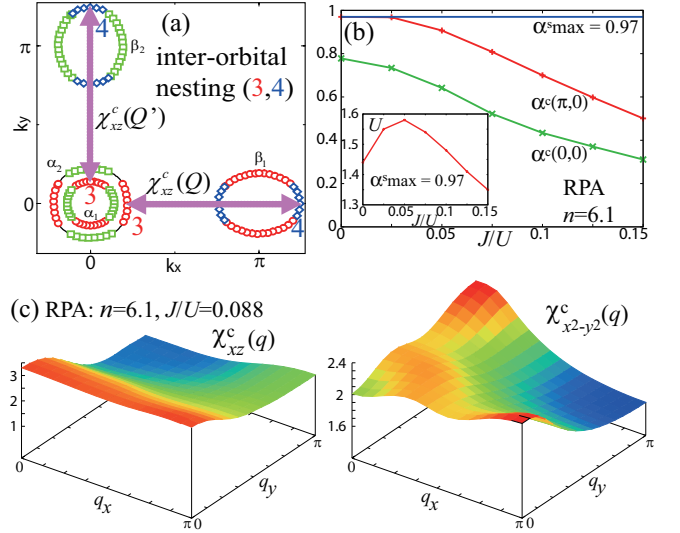


FIG. 1: (color online) (a) Fermi surfaces of iron pnictides. The colors correspond to 2 = xz (green), 3 = yz (red), and 4 = xy (blue), respectively. (b) $\alpha_{\mathbf{Q}}^c$, α_0^c and U as function of J/U in RPA under the condition $\alpha_{\text{max}}^s = 0.97$. (c) $\chi_{xz}^c(\mathbf{q})$ and $\chi_{x^2-y^2}^c(\mathbf{q})$ given by the RPA for $(J/U, U) = (0.088, 1.53)$.

factors are $\alpha_{\text{max}}^s = 0.97$, $\alpha_{\mathbf{Q}}^c = 0.76$, and $\alpha_0^c = 0.47$; see Fig. 1 (b). In the RPA, $\chi_{xz}^c(\mathbf{Q})$ [$\chi_{yz}^c(\mathbf{Q}')$] is weakly enlarged by the inter-orbital (3, 4) [(2, 4)] nesting, while $\chi_{x^2-y^2}^c(\mathbf{q})$ is relatively small and AF-like. Thus, the RPA cannot explain the structure transition that requires the divergence of $\chi_{x^2-y^2}^c(\mathbf{0})$.

Next, we study the role of VC due to the Maki-Thompson (MT) and Aslamazov-Larkin (AL) terms in Fig. 2 (a), which become important near the critical point [29, 30]. Here, $\hat{X}^{c(s)}(q) \equiv \hat{X}^{\uparrow,\uparrow}(q) + (-)\hat{X}^{\uparrow,\downarrow}(q)$, and wavy lines represent $\chi^{s,c}$. The AL term (AL1+AL2) for the charge sector, $X_{ll',mm'}^{\text{AL},c}(q)$, is given as

$$\begin{aligned} \frac{T}{2} \sum_k \sum_{a \sim h} \Lambda_{ll',ab,ef}(q; k) \{V_{ab,cd}^c(k+q)V_{ef,gh}^c(-k) \\ + 3V_{ab,cd}^s(k+q)V_{ef,gh}^s(-k)\} \Lambda'_{mm',cd,gh}(q; k), \end{aligned} \quad (5)$$

where $\hat{V}^{s,c}(q) \equiv \hat{\Gamma}^{s,c} + \hat{\Gamma}^{s,c}\hat{\chi}^{s,c}(q)\hat{\Gamma}^{s,c}$, $\hat{\Lambda}(q; k)$ is the three-point vertex made of three Green functions in Fig. 2 (a) [28], and $\Lambda'_{mm',cd,gh}(q; k) \equiv \Lambda_{ch,mg,dm'}(q; k) + \Lambda_{gd,mc,hm'}(q; -k-q)$. We include all U^2 -terms, which are important for reliable results. The expressions of other VCs will be published in future.

Both MT and AL terms correspond to the first-order mode-coupling corrections to the RPA susceptibility: The intra- (inter-) bubble correction gives the MT (AL) term [29]. In single-orbital models, the VC due to MT+AL terms had been studied by the self-consistent-renormalization (SCR) theory [29] or FLEX approximation with VC [30], and successful results had been obtained. In the former (latter) theory, the susceptibility is calculated in the self-consistent (self-inconsistent) way.

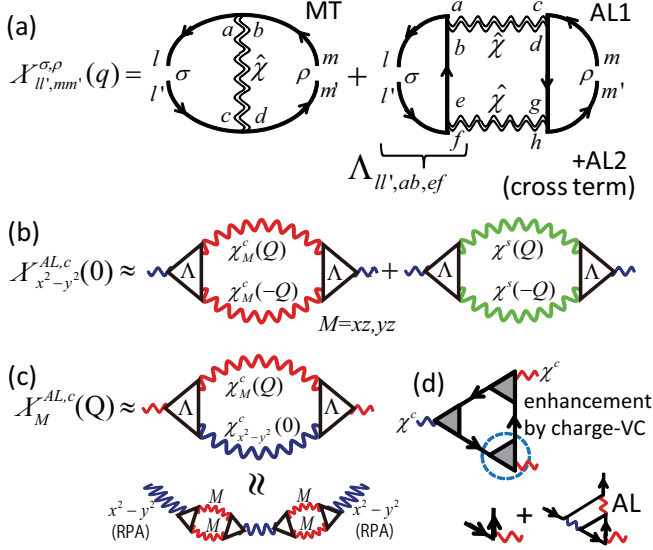


FIG. 2: (color online) (a) The MT and AL terms: The wavy and solid lines are susceptibilities and electron Green functions, respectively. $\Lambda_{ll',ab,ef}$ is the three-point vertex. (b) Dominant AL terms for $\chi_{x^2-y^2}^c(\mathbf{0})$; the first (second) term represents the two-orbion (two-magnon) process. (c) Dominant AL terms for $\chi_M^c(\mathbf{Q})$ ($M = xz, yz$); higher-order terms with bubbles made of $\chi_M^c(\pm\mathbf{Q})$ (= multi-fluctuation process) are relevant. (d) Enhancement of $\Lambda_{ll',ab,ef}$ due to charge VCs.

Here, we find a significant role of the AL term inherent in the multiorbital Hubbard model.

Now, we perform the SC-VC analysis, in the way to satisfy $\hat{\chi}^{c,s}(q)$ in the VC are equal to the total susceptibilities in Eq. (2). Then, $\hat{\chi}^c(q)$ is strongly enhanced by $X^{\text{AL},c}$ in Eq. (5), which is relevant when either $\hat{\chi}^c$ or $\hat{\chi}^s$ is large. On the other hand, we have verified numerically that $\hat{X}^s \sim T \sum \Lambda \cdot V^s V^c \cdot \Lambda$ is less important, although it could be relevant only when both $\hat{\chi}^c$ and $\hat{\chi}^s$ are large. Hereafter, we drop $\hat{X}^s(q)$ to simplify the argument. Figure 3 (a) show $\chi_{xz}^c(q)$ given by the SC-VC method for $n = 6.1$, $J/U = 0.088$ and $U = 1.53$, in which the Stoner factors are $\alpha_{\text{max}}^s = \alpha_0^s = 0.97$ and $\alpha_{\mathbf{Q}}^c = 0.86$. Compared to the RPA, both $\chi_{x^2-y^2}^c(q)$ and $\chi_{xz}^c(q)$ are strongly enhanced by the charge AL term, $\hat{X}^{\text{AL},c}$, since the results are essentially unchanged even if MT term is dropped. In the SC-VC method, the enhancements of other charge multipole susceptibilities are small. Especially, both the density and dipole susceptibilities, $\sum_{l,m} \hat{\chi}_{ll,mm}^c(q)$ and $\chi_\mu^c(q)$ ($\mu = x, y, z$) respectively, are suppressed.

Here, we discuss the importance of the AL term: At $q \approx \mathbf{0}$ or \mathbf{Q} , $\chi_\gamma^c(q)$ is enlarged by the diagonal vertex correction with respect to γ , $X_\gamma^{\text{AL},c}(q) \equiv \text{Tr}\{\hat{O}_\gamma \hat{X}^{\text{AL},c}(q) \hat{O}_\gamma\} / \text{Tr}\{\hat{O}_\gamma^2\}$, since the off-diagonal terms are absent or small [28]. The charge AL term in Eq. (5) is given by the products of two χ^c 's (two-orbion process) and two χ^s 's (two-magnon process), shown in Fig. 2 (b). The former process was

discussed in Ref. [28], and the latter has a similarity to the spin nematic theory in Ref. [16] based on a frustrated spin model. Now, we consider the orbital selection rule for the two-orbion process: Because of the relation $\text{Tr}\{\hat{O}_{x^2-y^2} \hat{O}_M^2\} \neq 0$ for $M = xz, yz$ and a rough relation $\Lambda_{ll',ab,cd} \sim \Lambda_{ll',l'b,bl} \delta_{l',a} \delta_{b,c} \delta_{d,l}$ [28], the two-orbion process for $\gamma = x^2 - y^2$ is mainly given by $\chi_M^c(\mathbf{Q})^2$. According to Eq. (5) and Ref. [28], $X_{x^2-y^2}^{\text{AL},c}(\mathbf{0}) \sim \Lambda^2 U^4 T \sum_q \{\chi(q)\}^2$ grows in proportion to $T \chi(\mathbf{Q}) [\log\{\chi(\mathbf{Q})\}]^2$ at high [low] temperatures. In the case of Fig. 3 (a), two-magnon process is more important for $\chi_{x^2-y^2}^c(\mathbf{0})$ because of the relation $\alpha_{\mathbf{Q}}^s > \alpha_{\mathbf{Q}}^c$. We checked that the two-magnon process is mainly caused by $\chi_{22,22}^s(\mathbf{Q})^2 - \chi_{22,33}^s(\mathbf{Q})^2 > 0$.

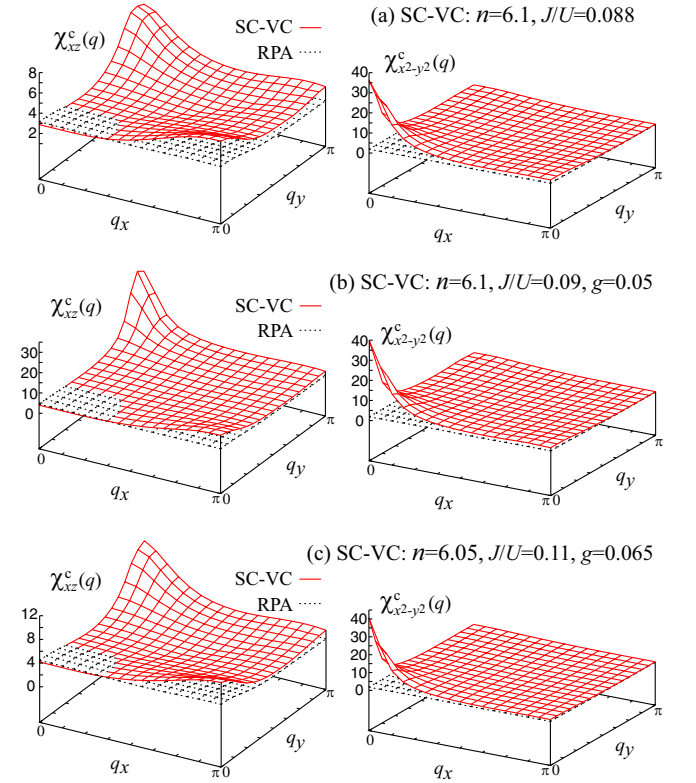


FIG. 3: (color online) $\chi_{xz}^c(q)$ and $\chi_{x^2-y^2}^c(q)$ given by the SC-VC method. The relation $\alpha_{\text{max}}^s = \alpha_0^s = 0.97$ is satisfied in all cases: (a) $n = 6.1$ and $J/U = 0.88$ ($\alpha_{\mathbf{Q}}^c = 0.86$), (b) $n = 6.1$, $J/U = 0.9$ and $g = 0.05$ ($\alpha_{\mathbf{Q}}^c = 0.96$), and (c) $n = 6.05$, $J/U = 0.11$ and $g = 0.065$ ($\alpha_{\mathbf{Q}}^c = 0.87$).

In the same way, $X_M^c(\mathbf{Q}) \sim \Lambda^2 U^4 T \sum_q \chi_M^c(q + \mathbf{Q}) \chi_{x^2-y^2}^c(q)$ is enlarged by the two-orbion process due to $\chi_M^c(\mathbf{Q})$ and $\chi_{x^2-y^2}^c(\mathbf{0})$, shown in Fig. 2 (c). (In this case, two-magnon process is less important since $\chi^s(\mathbf{0})$ is small.) The obtained $\chi_{xz}^c(q)$ has peaks at $q = \mathbf{Q}$ and \mathbf{Q}' since the inter-orbital scattering is emphasized by $X_{xz}^c(\mathbf{Q}) \propto \chi_{x^2-y^2}^c(\mathbf{0}) \gg 1$. Thus, both $\chi_{xz}^c(\mathbf{Q})$ and $\chi_{x^2-y^2}^c(\mathbf{0})$ are strongly enlarged in the SC-VC method, because of the “positive feedback” brought by these two

AL terms: Figure 2 (c) shows an example of the higher-order terms that are automatically generated in the SC-VC method. Such “multi-fluctuation processes” inherent in the self-consistent method magnify the RPA results.

Thus, strong ferro- and AF-orbital fluctuations are caused by AL terms. Both fluctuations work as the pairing interaction for the s_{++} -state, while the ferro-fluctuations are also favorable for the s_{\pm} -state. For $J/U < (J/U)_c \equiv 0.088$, the relation $\alpha_{\max}^s < \alpha_0^c = 0.97$ is realized and α_Q^c increases towards unity. In this case, orbital order occurs prior to the spin order as increasing U with J/U is fixed, since the VC (due to two-orbion process) can efficiently enlarge orbital susceptibilities because of large α_{\max}^c (RPA). This situation would be consistent with wider non-magnetic orthorhombic phase in Nd(Fe,Co)As and many 1111 compounds.

Since the present SC-VC method is very time-consuming, we applied some simplifications: We have verified in the self-inconsistent calculation that $\text{Tr}\{\hat{O}_\gamma \hat{X}(q) \hat{O}_{\gamma'}\}$ with $\gamma \neq \gamma'$ is zero or very small, especially at $\mathbf{q} = \mathbf{0}$ and \mathbf{Q} for the reason of symmetry. Since we are interested in the enhancement of $\chi_\gamma^c(\mathbf{q})$ at $\mathbf{q} = \mathbf{0}$ and \mathbf{Q} and the dominant interferences between $\gamma = xz, yz, x^2 - y^2$, we calculated $X_{l',mm'}(q)$ only for $\{(l, l'), (m, m')\} \in xz, yz, x^2 - y^2$. [$(l, l') \in \gamma$ means that $O_\gamma^{l,l'} \neq 0$.] That is, $\{(l, l'), (m, m')\} = \{(1, 2), (3, 4), (2, 5)\}, \{(1, 3), (2, 4), (3, 5)\}$, and $\{(1, 5), (2, 2), (3, 3)\}$.

We stress that both $(J/U)_c$ and AF-orbital fluctuations increase by considering following two factors: The first one is the charge VC at each point of the three-point vertex in Fig. 2 (d), as a consequence of the Ward identity between $\hat{\Lambda}$ and $\hat{\chi}^{\text{irr}}$. The enhancement factor at each point is estimated as $1 + X_\gamma^c/\chi_\gamma^0 = 1.3 \sim 2.5$ for $\gamma = xz$ and $x^2 - y^2$ in the present calculation near the critical point. This effect will increase $(J/U)_c$ sensitively. The second factor is the e -ph interaction: We introduce the quadrupole interaction in Eq. (1) due to Fe-ion oscillations [6, 14, 27]. As shown in Fig. 3 (b), very strong AF-orbital fluctuations are obtained for $J/U = 0.09$ and $g = 0.05$; $\alpha_{\max}^s = \alpha_0^c = 0.97$ and $\alpha_Q^c = 0.96$. The corresponding dimensionless coupling is just $\lambda = gN(0) \sim 0.035$ [6, 27]. We also study the case $n = 6.05$ and $g = 0.065$, and find that the relation $\alpha_{\max}^s = \alpha_{\max}^c = 0.97$ is realized at $(J/U)_c = 0.11$, as shown in Fig. 3 (c). For these reasons, strong ferro- and AF-orbital-fluctuations would be realized by the cooperation of the Coulomb and weak e -ph interactions.

Finally, we make some comments: The present multi-fluctuation mechanism is not described by the dynamical-mean-field theory (DMFT), since the irreducible VC is treated as *local*. Also, the local density approximation (LDA), in which the VC is neglected, does not reproduce the nonmagnetic orthorhombic phase. Although Yanagi *et al.* studied $U' > U$ model [7] based on the RPA, that was first studied in Ref. [31], $\chi_{3z^2-r^2}^c(\mathbf{0})$ develops

while $\chi_{x^2-y^2}^c(\mathbf{0})$ remains small, inconsistently with the structure transition. Our important future issue is to include the electron self-energy correction into the SC-VC method, which is important to discuss the filling and T -dependences of orbital and spin fluctuations, and to obtain more reliable $(J/U)_c$.

In summary, we developed the SC-VC method, and obtained the Coulomb-interaction-driven nematic and AF-orbital fluctuations due to the multimode (orbitons+magnons) interference effect [28] that is overlooked in the RPA. For $J/U \lesssim (J/U)_c$, the structure transition ($\alpha_0^c \approx 1$) occurs prior to the magnetic transition ($\alpha_Q^s \approx 1$), consistently with experiments. When $\alpha_{\max}^s \sim \alpha_{\max}^c$, both s_{++} - and s_{\pm} -states could be realized, depending on model parameters like the impurity concentration [3, 6]. In a sense of the renormalization group scheme, the quadrupole interaction in Eq. (1) is induced by the Coulomb interaction beyond the RPA. We expect that orbital-fluctuation-mediated superconductivity and structure transition are realized in many iron-based superconductors due to the cooperation of the Coulomb and e -ph interactions.

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