Nematicity and magnetism in FeSe and other families of Fe-based superconductors

Youichi Yamakawa\textsuperscript{1}, Seiichiro Onari\textsuperscript{2}, and Hiroshi Kontani\textsuperscript{1}

\textsuperscript{1} Department of Physics, Nagoya University, Furo-cho, Nagoya 464-8602, Japan.
\textsuperscript{2} Department of Physics, Okayama University, Okayama 700-8530, Japan.

(Dated: May 6, 2016)

Nematicity and magnetism are two key features in Fe-based superconductors, and their interplay is one of the most important unsolved problems. In FeSe, the magnetic order is absent below the structural transition temperature $T_{\text{str}} = 90$K, in stark contrast that the magnetism emerges slightly below $T_{\text{str}}$ in other families. To understand such amazing material dependence, we investigate the spin-fluctuation-mediated orbital order ($n_{xz} \neq n_{yz}$) by focusing on the orbital-spin interplay driven by the strong-coupling effect, called the vertex correction. This orbital-spin interplay is very strong in FeSe because of the small ratio between the Hund’s and Coulomb interactions ($J/\bar{U}$) and large $d_{xz}, d_{yz}$-orbitals weight at the Fermi level. For this reason, in the FeSe model, the orbital order is established irrespective of the small spin fluctuations. Thus, the orbital-spin interplay due to the vertex correction is the key ingredient in understanding the rich phase diagram with nematicity and magnetism in Fe-based superconductors in a unified way.

PACS numbers: 74.70.Xa, 75.25.Dk, 74.20.Pq

I. INTRODUCTION

In Fe-based superconductors, the origin of the electronic nematic state and its relation to the magnetism have been a central unsolved problem. Recently, the non-magnetic nematic state in FeSe has attracted increasing attention as a key to solve the origin of the nematicity. FeSe undergoes a structural and superconducting transitions at $T_{\text{str}} = 90$K and $T_c = 9$K, respectively, whereas the magnetic transition is absent down to 0 K [1]. The strength of the low-energy antiferro-magnetic (AFM) fluctuations is very weak above $T_{\text{str}}$, while it starts to increase below $T_{\text{str}}$. In stark contrast, the magnetic transition occurs at $T_{\text{mag}}$ slightly below $T_{\text{str}}$ in other undoped Fe-based superconductors. Since the relation $T_{\text{str}} > T_{\text{mag}}$ is unable to be explained by the random-phase-approximation (RPA), we should develop the microscopic theory beyond the mean-field-level approximations.

Up to now, two promising triggers for the structure transition have been discussed intensively: In the spin-nematic scenario [8–12], the trigger is the spin-nematic order. This spin-fluctuation induced spin-quadrupole order could emerge above $T_{\text{mag}}$ in highly magnetically frustrated systems. In the orbital order scenario [13–16], the trigger is the ferro-orbital (FO) order $n_{xz} \neq n_{yz}$. Above $T_{\text{str}}$, the strong orbital or spin-nematic fluctuations are observed by the measurements of shear modulus $C_{06}$ [2, 17, 18], Raman spectroscopy [19–22], and in-plane resistivity anisotropy [23, 24]. The nematic orbital fluctuations originate from the strong orbital-spin mode-coupling due to the strong-coupling effect, which is described by the Aslamazov-Larkin vertex correction (AL-VC). The electronic nematic state studied in single-orbital models [25] is more easily realized in multiorbital systems thanks to the AL-VC mechanism [16].

Except for the presence or absence of magnetism below $T_{\text{str}}$, FeSe and other Fe-based superconductors show common electronic properties. Below $T_{\text{str}}$, in both FeSe and BaFe$_2$As$_2$, large orbital polarization $\Delta E \equiv E_{yz} - E_{xz} \sim$ 50 meV [26–33] is observed. Such large $\Delta E$ originates from the electron-electron correlation since the lattice distortion $(a - b)/(a + b)$ is just 0.2 ~ 0.3%, as we discuss based on band calculation in Appendix A. Above $T_{\text{str}}$, the electronic nematic susceptibility is enhanced in both BaFe$_2$As$_2$ [17, 19, 23] and FeSe [2, 18], following the similar Curie-Weiss behavior. These facts indicate that the common microscopic mechanism drives the nematic order and fluctuations in all Fe-based superconductors, in spite of the presence or absence of the magnetism.

The realistic multiorbital Hubbard models for Fe-based superconductors, which are indispensable for the present study, were derived by using the first-principles method in Ref. [34]. To understand the absence of the magnetism below $T_{\text{str}}$ in FeSe, one significant hint is the smallness of the ratio between the Hund’s and Coulomb interactions, $J/\bar{U}$, since the Hund’s coupling enlarges (suppresses) the intra-site magnetic (orbital) polarization, which is verified by the functional renormalization-group (fRG) theory [35, 36]. Another significant hint is the absence of the $d_{xy}$-orbital hole-pocket in FeSe, which is favorable for the orbital-spin interplay on the $(d_{xz}, d_{yz})$-orbitals due to the AL-VC mechanism.

The goal of this paper is to explain the amazing variety of the electronic nematic states in Fe-based superconductors, especially the non-magnetic nematic state in FeSe, on the same footing microscopically. For this purpose, we study the spin-fluctuation-mediated orbital order by applying the self-consistent vertex-correction (SC-VC) method [16] to the first-principles models. In FeSe, the orbital-spin interplay is significant because of the smallness of $J/\bar{U}$ and the absence of $d_{xy}$-hole pocket. For
this reason, the orbital order is realized even when the spin fluctuations are substantially weak. The rich variety of the phase diagrams in Fe-based superconductors, such as the presence or absence of the magnetic order in the nematic phase, are well understood by analyzing the vertex correction seriously. The SC-VC theory had been successfully applied to explain the phase diagram in LaFeAs(O,H) [37], nematic CDW in cuprates [38, 39], and triplet superconductivity in Sr$_2$RuO$_4$ [36].

We comment that the localized spin models have been successfully applied to the nematic order, stripe magnetic order, and so on [40]. On the other hand, weak-coupling theories have also been applied to Fe-based superconductors satisfactorily [41]. In the present study, we study the mechanisms of the nematicity and magnetism in various Fe-based superconductors in terms of the itinerant picture, by taking the strong-coupling effect due to the AL-VC into account. The significant role of the AL-VC on the orbital fluctuations has been confirmed by the fRG theory [35, 36]. The AL-VC is important to reproduce the Kugel-Khomskii-type orbital-spin interaction [16].

II. MODEL HAMILTONIAN AND SC-VC THEORY

In the present study, we study the realistic $d$-$p$ Hubbard models

$$H_M(r) = H_0^M + rH_M^U$$

for $M$=LaFeAsO and FeSe by applying the SC-VC method [16]. In Eq. (1),

$$H_0^M = \sum_{k,l,m,\sigma} c_{k,l,\sigma}^\dagger h_{k,l,m,\sigma}(k)c_{k,l,\sigma}$$

is the 8-orbital $d$-$p$ tight-binding (TB) model in $k$-space, which is obtained by using the WIEN2k and WANNIER90 softwares; see Appendix A for detailed explanation. $\sigma$ is the spin index, and $l, m$ are the orbital indices: Hereafter, we denote the five $d$-orbitals as $d_{xz}, d_{yz}, d_{xy}, d_{x^2-y^2}$ as 1, 2, 3, 4, 5, and three $p$-orbitals as 6 ~ 8. The bandstructure and Fermi surfaces (FSs) in the LaFeAsO model are shown in Figs. 1 (a) and (b), respectively. Similar FSs with three hole-like FSs (h-FSs) and two electron-like FSs (e-FSs) exist in many Fe-based superconductors. In FeSe, however, h-FS3 composed of $d_{xy}$-orbital is absent, and the size of each FS is very small as clarified by the ARPES [27, 32, 33] and dHvA [42, 43] studies. To reproduce experimental bandstructure of FeSe, we introduce the additional intra-orbital hopping parameters into $H_0^M$ in order to shift the $d_{xy}$-orbital band [$d_{xz}/d_{yz}$-orbital band] at $(\Gamma, M, X)$ points by $(0, -0.25, +0.24) [(0.24, 0, +0.12)]$ in unit eV; see Appendix A. These energy shifts might be induced by the self-energy [44]. The constructed FSs in the FeSe model is shown in Fig. 1 (c). Since each Fermi pocket is very shallow, the superconductivity in FeSe could be close to a BCS-BEC crossover [45].

In Eq. (1), $H_M^U$ is the first-principles screened Coulomb potential for $d$-orbitals given by the “constrained-RPA method” [34] given as

$$H_M^U = \frac{1}{2} \sum_{i,l,\sigma,\sigma'} \{ U_{m,l} n_{i,l,\sigma} n_{i,m,\sigma'} (1 - \delta_{l,m} \delta_{\sigma,-\sigma'})$$

$$+ J_{m,l} c_{i,l,\sigma}^\dagger c_{i,m,\sigma'} (c_{i,l,\sigma'}^\dagger c_{i,\sigma,\sigma'} + c_{i,\sigma,\sigma'}^\dagger c_{i,\sigma,\sigma'}) \}$$

where $U_{m,l}$ and $J_{m,l}$ are orbital-dependent Coulomb and Hund’s interactions for $d$-electrons, respectively [34]. The averaged intra-orbital Coulomb interaction $\bar{U} = \frac{1}{5} \sum_{i,l} U_{i,l}$ and Hund’s interactions $\bar{J} = \frac{1}{10} \sum_{l,m} J_{i,m}$ are $(\bar{U}, \bar{J}) = (7.21, 0.681)$ for FeSe, and $(\bar{U}, \bar{J}) = (4.23, 0.568)$ for LaFeAsO in unit eV [34]. Thus, the ratio $\bar{J}/\bar{U} = 0.0945$ in FeSe is much smaller than the ratio $J/\bar{U} = 0.134$ in LaFeAsO. Such strong material dependence of $(\bar{U}, \bar{J})$ is understood as follows: $U_{i,l}$ is strongly screened by the screening bands (excluding the 8 bands in $H_0^M$) whereas the screening of $J_{i,m}$ is much weak, and the number of the screening bands is small in FeSe [46]. The factor $r(<1)$ in Eq. (1) is introduced to adjust the spin fluctuation strength. The ratio $J_{i,m}/U_{i,m}$ is unchanged by introducing the factor $r$ [47, 48].

FIG. 1: (color online) (a) Bandstructures of the eight-orbital TB models for LaFeAsO and FeSe. (b) FSs for the LaFeAsO TB model. (c) FSs for the FeSe TB model. The colors correspond to 2 (green), 3 (red), and 4 (blue), respectively.

The $8 \times 8$ Green function in the orbital basis is given as

$$\hat{G}(k) = (\hat{z}^{-1}\hat{\epsilon}_n + \mu - \hat{h}_{i,i}(k))^{-1},$$

(4)
where \( k = (k, \epsilon_n = (2n + 1)\pi T) \), \( \hat{h}_k^0(k) \) is the kinetic term in Eq. (2), and \( \hat{\Sigma}_k \rightarrow -\partial \hat{\Sigma}_k/\partial \epsilon_n \) represents the mass-enhancement due to the self-energy at the Fermi level. Here, we introduce the constant mass-enhancement factor for \( d \)-orbital \( 1/\tilde{z}_l (\geq 1) \). Then, Eq. (4) gives the coherent part of the Green function, which mainly determines the low-energy electronic properties. In the present study, \( r \) and \( z_l \) are the fitting parameters. In FeSe, the orbital order is obtained in the real first-principles Hamiltonian \( (r \approx 1) \) by taking the experimental mass-enhancement factors \( \tilde{z}_l \approx 4 \) into account, as shown later.

The \( d \)-orbital charge (spin) susceptibilities (per spin) is given in the following \( 5^2 \times 5^2 \) matrix form:

\[
\chi_c^{(sc)}(q) = \Phi_c^{(sc)}(q)(1 - \hat{\Gamma}_c^{(sc)}\Phi_c^{(sc)}(q))^{-1} \tag{5}
\]

where \( \Phi_c^{(sc)}(q) = \chi_0^{(sc)}(q) + \hat{\chi}_c^{(sc)}(q) \) is the irreducible susceptibility for the charge (spin) channel. In the SC-VC theory, we employ the AL-VC as \( \hat{\chi}_c^{(sc)}(q) \), and perform the self-consistent calculation with respect to the AL-VC and susceptibilities. Using the Green function in Eq. (4), the bare susceptibility is

\[
\chi_{l';m'}^{0} = -T \sum_k G_{l;m}(k + q)G_{m';l'}(k), \tag{6}
\]

where \( q = (q, \omega_n = 2l\pi T) \). Also, the AL-VC for the charge susceptibility is given as

\[
\chi_{l';m'}^{ALC}(q) = \frac{T}{2} \sum_p \sum_{\alpha = h} \Lambda_{l';m';\alpha,b,c,f}(q;p) \times \{3V_{\alpha,b,c,d}(p + q)V_{\alpha,f,g,h}(-p) + V_{\alpha,b,c,d}(p + q)V_{\alpha,c,g,h}(-p)\}
\]

\[
\times \Lambda_{m,m';c,d,g,h}(q;p), \tag{7}
\]

where \( p = (p, \omega_m) \), and \( \hat{\chi}_c^{(sc)}(q) = \hat{\chi}_c^{(sc)} + \hat{\chi}_c^{(sc)}\chi_c^{(sc)}(q)\hat{\chi}_c^{(sc)} \). The three-point vertex \( \Lambda(q;p) \), which gives the coupling between two-magnon and one-orbition, is given as

\[
\Lambda_{l',m';\alpha,b,c,f}(q;p) = -T \sum_{k'} G_{l,a}(k' + q)G_{f,l'}(k')G_{b,c}(k' - p), \tag{8}
\]

and \( \Lambda_{m,m';c,d,g,h}(q;p) \equiv \Lambda_{c,h,m,g;d,m'}(q;p) + \Lambda_{g,d,m;c,h,m'}(q;p - q) \). We stress that the strong temperature dependence of the three-point vertex is significant for realizing the orbital order. We include all \( U^2 \)-terms without the double counting in order to obtain quantitatively reliable results. Equation (7) means that the charge AL-VC becomes large in the presence of strong spin fluctuations. More detailed explanations are presented in the textbook [49].

In the present study, we neglected the spin-channel VCs since it is expected to be unimportant as discussed in Ref. [50]. In Appendix B, we verify validity of this simplification in the present model by performing a time-consuming self-consistent calculation with respect to both charge- and spin-channel Maki-Thompson (MT) and AL-VCs.

Hereafter, we mainly discuss the total spin susceptibility, \( \chi^s(q) \equiv \sum_m \chi^s_{l;m}(q) \), and the orbital susceptibilities for \( O_{xz-y^2} = n_{xz} - n_{y^2} \). \( \chi^s_{xz-y^2}(q) \equiv \chi_{2,2;2,2}^{c}(q) + \chi_{4,4;4,4}^{c}(q) \). The divergence of \( \chi^s_{xz-y^2}(q) \) at \( q = 0 \) gives rise to the FO order \( n_{xz} \neq n_{y^2} \). The charge (spin) Stoner factor \( \alpha_{C(S)} \) is given by the maximum eigenvalue of \( \hat{\Gamma}_c^{(sc)}\Phi_c^{(sc)}(q) \) in Eq. (5), and the charge (spin) susceptibility is enlarged in proportion to the charge (spin) Stoner enhancement factors \( S_{C(S)} = (1 - \alpha_{C(S)})^{-1} \).

As explained in Ref. [50], the development of \( \chi^s_{xz-y^2}(0) \) is mainly induced by the diagonal elements of \( \Phi_c^{(sc)} \) with respect to \( l = 2, 3 \). If we drop the off-diagonal elements of \( \Phi_c^{(sc)} \) approximately, \( \chi^s_{xz-y^2}(0) \) is given as

\[
\chi^s_{xz-y^2}(0) \approx 2\Phi_c^{(sc)}/(1 - (1 - 5J/U)\Phi_c^{(sc)}), \tag{9}
\]

where \( U = U_{2,2} = U_{3,3}, J = J_{2,3} \), \( \Phi_c^{(sc)} \equiv \chi_{0}^{c;l,l}(0) + \chi_{l,l}(0) \) \((l = 2, 3) \). Thus, the charge Stoner factor is

\[
\alpha_C = (1 - 5J/U)\Phi_c^{(sc)} = (1 - 5J/U)(1 + UX^c), \tag{10}
\]

considering the relation \( \chi^c_0(q) \approx 1/U \). Within the RPA (\( \Phi^c_0 = \chi^c_0 = 0 \)), only the spin fluctuations develop since the relation \( \alpha_S > \alpha_C \) is satisfied for \( J > 0 \). However, the opposite relation \( \alpha_C > \alpha_S \) is realized if the relation \( \Phi^c > \chi^c_0 \) is satisfied due to the charge-channel AL-VC [37].

### III. NUMERICAL RESULTS FOR LAFeAsO AND FeSe

First, we analyze the LaFeAsO model based on the SC-VC theory. For \( z = 1 \) for each \( l \), the obtained \( \chi^s(q) \) and \( \chi^s_{xz-y^2}(q) \) are shown in Fig. 2 (a) and (b), respectively, for \( r = 0.41 \) \((U = 1.74 \text{ eV}) \) at \( T = 50 \text{ meV} \). Here, the number of \( k \)-meshes is \( 32 \times 32 \), and the number of Matsubara frequencies is 256. Thus, both AFM and FO susceptibilities develop divergently, and the realized enhancement factors are \( S_C \approx 40 \) and \( S_S \approx 50 \). The \( r \)-dependences of the enhancement factors at \( T = 50 \text{ meV} \) are shown in the inset of Fig. 2 (c): Both \( S_C \) and \( S_S \) increase with \( r \), and they are equivalent at \( r^* = 0.41 \). The lower the temperature is, the smaller \( r^* \) is, whereas the value of \( S_S = S_C \) at \( r^* \) is approximately independent of \( T \). Similar result is obtained in BaFe2As2 model as shown in Appendix C.

In addition, other antiferro-orbital susceptibilities \( \chi_{4,4;4,4}(q) = 2[\chi_{4,4;4,4}(q) + \chi_{4,4;4,4}(q)] \) and \( \chi_{0,z}(q) = 2[\chi_{4,4;4,4}(q) + \chi_{4,4;4,4}(q)] \) develop secondary as reported in previous studies [16, 37, 49]. The obtained results are essentially similar to the results obtained in the five \( d \)-orbital Hubbard model for LaFeAsO explained in Ref. [16].

Figure 2 (c) shows the temperature dependences of the Stoner enhancement factors at \( r = 0.41 \). Both \( S_C \) and \( S_S \) approximately follow the Curie-Weiss behaviors with the charge and spin Weiss temperatures.
\( \theta_C = 48 \text{ meV} \) and \( \theta_S = 40 \text{ meV} \), respectively. The obtained relation \( \chi^x_{c_{zz} - z^2}(0) \propto (T - \theta_C)^{-1} \) is consistent with the Curie-Weiss behavior of the nematic susceptibilities in BaFe\(_2\)As\(_2\), derived from \( C_66 \) [17, 18], Raman spectroscopy [19, 20], and in-plane resistivity anisotropy [23]. Since \( \theta_C \sim \theta_S \), one could interpret that the orbital order in LaFeAsO is driven by the spin fluctuations.

The orbital-spin interplay due to the AL-VC is intuitively understood in terms of the strong-coupling picture \( U \gg W_{\text{band}} \) [37]: As shown in Fig. 2 (d), when the FO order \( n_{xz} \gg n_{yz} \) is realized, the nearest-neighbor exchange interaction has large anisotropy \( J^{(1)}_s \gg J^{(1)}_b \). Then, the stripe AFM order with \( Q = (\pi, 0) \) appears if \( J^{(2)} \) is not too small. Thus, the FO order/fluuctuations and AFM order/fluuctuations emerge cooperatively in the localized model, and such Kugel-Khomskii-type orbital-spin interplay is explained by the AL-VC in the weak-coupling picture.

Next, we analyze the FeSe model, in which the ratio \( J/\bar{U} \) is considerably small. In FeSe, the experimental mass-enhancement factor is \( \sim 10 \) for \( d_{yz} \)-orbital, and \( 3 \sim 4 \) for other \( d \)-orbitals according to the ARPES study [27]. Therefore, we put \( z_4^{-1} = z^{-1} \) for \( l \neq 4 \) and \( z_4^{-1} = 3z^{-1} \) in the present study. We find that the peak of \( \chi^x(q) \) moves from \( q = (\pi, \pi) \) to the experimental peak position \( q = (\pi, 0) \) [4-7] for \( z_4^{-1} = 1.5z^{-1} \), and the results of the SC-VC method are essentially unchanged for \( z_4^{-1} \geq 1.5z^{-1} \). Figures 3 (a) and (b) show the obtained \( \chi^x(q) \) and \( \chi^y_{c_{zz} - z^2}(q) \) for \( r = 0.25 (\bar{U} = 1.76 \text{ eV}) \) at \( T = 50 \text{ meV} \) in the case of \( z = 1 \). We see that only the FO susceptibility develop divergently \([S_C \approx 50]\), whereas the AFM susceptibility remains small \([S_S \approx 8]\), consistently with experiments for FeSe. The \( r \)-dependences of the Stoner enhancement factors at \( T = 50 \text{ meV} \) are shown in the inset of Fig. 3 (c): With increasing \( r \), \( S_C \) rapidly increases whereas \( S_S \) remains small.

Figure 3 (c) shows the temperature dependences of the enhancement factors at \( r = 0.25 \). We stress that \( S_C \) approximatively follow the Curie-Weiss behavior with the Weiss temperature \( \theta_C = 48 \text{ meV} \), which is consistent with the experimental Curie-Weiss behavior with positive \( \theta_C \) in FeSe [2]. Since the spin Weiss temperature takes a large negative value \( \theta_S \sim -20 \text{ meV} \), which is also consistent with experiments, one may consider that the orbital order in FeSe stems from causes other than spin fluctuations.

### IV. ORIGIN OF THE RELATION \( S_C \gg S_S \) IN FESE

In this section, we discuss why the relation \( S_C \gg S_S \) \((\theta_C > 0 \text{ and } \theta_S < 0) \) is realized in FeSe. First, we focus on the ratio between the Hund’s and Coulomb interactions \( J/\bar{U} \). It is intuitively obvious that the ratio \( J/\bar{U} \) is an important control parameter for the orbital nematicity. For larger \( J/\bar{U} \), the local configuration of the two-electrons in the \((d_{xz}, d_{yz})\)-orbitals is \( |d_{xz}, \uparrow \rangle \otimes |d_{yz}, \uparrow \rangle \), where the magnetic moment is \( s_z = 1 \) whereas the orbital polarization is \( n_{xz} - n_{yz} = 0 \). Thus, the smallness of \( J/\bar{U} \) in FeSe is favorable for the emergence of the orbital order without magnetization.

Microscopically, as we discuss in Sec. II, the charge St"oner factor for \( \chi^y_{c_{zz} - y^2}(0) \) is \( C_S \approx (1 - 5J/\bar{U})(1 + UX^c) \), where \( X^c \) is the charge AL-VC for orbital 2 or 3 at \( q = 0 \). Since \( \bar{U}/U \approx 0.0945 \) in FeSe, the orbital order is realized by relatively small AL-VC: \( X^c \sim 0.9\chi^0(0) \). In LaFeAsO, in contrast, large AL-VC of order \( 2\chi^0(0) \) is required to realize the orbital order. The obtained AL-VCs in both systems are shown in Fig. 9 (c) in Appendix D.

We discuss why the AL-VC is important in the FeSe model with \( \theta_S < 0 \): As we explain in Appendix D, the \( T \)-dependence of the AL-VC is given as \( X^c \sim \Lambda^2TS_S \), where \( \Lambda \) is the three-point vertex that represents the interference between two short-living magnons. We find the relation \( \Lambda^2 \propto T^{-a} \) with \( a \approx 1 \) at low temperatures due to the good nesting between h-FSs and e-FSs [20]. Thanks to the strong enhancement of \( \Lambda \) at low temperatures, the orbital order \((\alpha_C = 1) \) is realized even if \( \theta_S \) is negative. (Note that \( TS_S \) decreases as \( T \rightarrow 0 \) when \( \theta_S < 0 \).) Thus, serious diagrammatic analysis of the AL-VC is necessary to understand the rich normal-state phase diagrams in Fe-bases superconductors.
The enhancement of the nematic susceptibility due to the significant $T$-dependence of $\Lambda^2(\propto T^{-a})$ had been discussed in Refs. [20, 21, 51, 52]. However, the reported exponent $a$ is not universal, since it depends on the bandstructure and temperature range. In Appendix E, we show the $T$-dependence of the three-point vertex for LaFeAsO and FeSe models for wide temperature range. It is found that $a \approx 1$ for $T = 20z \sim 100z[\text{meV}]$, where $z \ll 1$ is the band-renormalization factor. Due to such large $T$-dependence of $a$, $\chi_{z}^{c_{x2-y^2}}(0)$ obtained by the present study follows the Curie-Weiss law only approximately.

Finally, we stress the importance of the orbital dependence of the spin fluctuation strength. Since the $d_{xy}$-orbital $h$-FS ($h$-FS3) is absent in FeSe, the relation $\chi_{z}^{c_{x2-y^2}}(q) \gg \chi_{z}^{s}(q)$ ($\chi_{z}^{s}(q) \equiv \chi_{z}^{s}(0,q)$) is realized. This condition is favorable for the development of $\chi_{z}^{c_{x2-y^2}}(0)$ since $X_{2(3)}^{c}$ is enlarged by the spin fluctuations on the $(d_{xz}, d_{yz})$-orbitals. More detailed analysis is given in Appendix D.

V. EFFECT OF THE MASS-ENHANCEMENT FACTOR

Here, we study the effect of the mass-enhancement factor: We study the FeSe model in the case of $z^{-1} = 4$ for $(d_{xz}, d_{yz})$-orbitals. The obtained $S_{C,S}$ as functions of $r$ are shown in the inset of Fig. 3 (d) at $T = 12.5$ meV. Here, $S_{S}$ remains small even for $r \sim 1$ since the bare susceptibility is suppressed by $z$. In contrast, $S_{C}$ is enlarged to 50 for $r \approx 0.97$, which is very close to the exact first-principles Hubbard model $H_{\text{FeSe}}(r = 1)$. The $T$-dependences of $S_{C,S}$ are shown in Fig. 3 (d): Beautiful Curie-Weiss behavior with $\theta_{C} = 12$ meV is obtained by putting $r = 0.97$.

To understand the similarity between the results in Fig. 3 (c) for $z = 1$ and the results in Fig. 3 (d) for $z^{-1} = 4$, we prove that both $\alpha_{C}$ and $\alpha_{S}$ are independent of $z$ under the rescaling $T \rightarrow zT$ and $(U, J) \rightarrow (U, J)/z$. Here, we assume that $z^{-1} = z^{-1}$ and neglect the $T$-dependence of $\mu$ for simplicity. Under the scaling $T \rightarrow zT$, the Green function $G(k, n)$ at Matsubara integer $n$ given in Eq. (4) is independent of $z$. For this reason, the bare susceptibility $\chi_{z}^{(q)}(q) = -T \sum_{k,n} G(k + q, n)G(k, n)$ is proportional to $z$. By following the same procedure, the three-point vertex $\Lambda$ is scaled by $z$, and therefore the AL-VC $X_{z}^{c}(0) \sim T U^{4} \sum_{q} \Lambda(0, q) \chi_{z}^{s}(q)^{2}$ is proportional to $z$ under the scaling $T \rightarrow zT$ and $(U, J) \rightarrow (U, J)/z$. Thus, both spin and charge irreducible susceptibilities are proportional to $z$, and both $\alpha_{S}$ and $\alpha_{C}$ are unchanged under the rescaling $T \rightarrow zT$ and $(U, J) \rightarrow (U, J)/z$. That is, the Weiss temperatures $\theta_{C,S}$ are scaled by $z$. The validity of these scaling relations are confirmed by the numerical study in Fig. 3.

It is possible to obtain $z^{-1}$ by calculating the self-energy $\Sigma(k)$ together with $\chi_{z}^{s-C}(q)$ self-consistently. In this case, fine tuning of $r$ will be unnecessary since the relation $\alpha_{S,C} < 1$ is assured if $\hat{\Sigma}(k)$ and $\hat{\chi}_{z}^{s-C}(q)$ are calculated self-consistently in two-dimensional systems (Mermin-Wagner theorem) [53]. This is our important future issue.

VI. DISCUSSIONS

A. Spin fluctuation strength and $k$-dependent orbital-polarization below $T_{\text{str}}$

Here, we study the electronic states in the FO order $n_{z} \neq n_{yz}$ established below the structure transition temperature $T_{\text{str}}$, at which the shear modulus $C_{66}$ reaches zero. According to the linear-response theory, $C_{66} \propto 1 - g\chi_{z}^{c_{x2-y^2}}(0)$, where $\chi_{z}^{c_{x2-y^2}}(0) \propto (T - \theta_{C})^{-1}$ is the electronic orbital susceptibility given by the SC-
VC theory, and $g$ is the phonon-mediated Jahn-Teller energy [54]. Therefore, $C_{66} \propto (T - T_{str})/(T - \theta_C)$, and $T_{str} = \theta_C + g$ is slightly higher than $\theta_C$ due to the weak electron-phonon coupling ($g \approx 10 \sim 50$ K) [2, 17, 18].

Figure 4 (a) shows the $T$-dependence of $S_S$ given by the RPA for LaFeAsO and FeSe for $z = 1$. Here, we introduce the orbital polarization $-\Delta E/2 (\Delta E/2)$ for the $a_{xz}(yz)$-level. We put $S_S = 20 (5)$ for LaFeAsO (FeSe) at $T_{str} = 30$ meV, and assume a mean-field-type $T$-dependence; $\Delta E = \Delta E_0 \tan h(1.74 \sqrt{T_{str}/T - 1})$ with $\Delta E_0 = 80$ meV. (For $z^{-1} = 4$, the renormalized orbital polarization $s\Delta E_0$ is just 20 meV.) In both LaFeAsO and FeSe, $S_S$ are enhanced by $\Delta E$, since $\alpha_S$ increases linearly with $\Delta E$ at $q = (\pi, 0)$ as discussed in Ref. [54]. In LaFeAsO, the magnetic order temperature $T_{mag}$ increases from $\theta_S$ to just below $T_{str}$ since $S_S$ is already large at $T_{str}$. In contrast, in FeSe, the enhancement of $\chi^s(\pi, 0)$ is much moderate [55].

We also perform the self-consistent analysis of the orbital-polarization $(\Delta E_{xz}(k), \Delta E_{yz}(k))$ and anisotropic $\chi^s(q)$, which is a natural extension of the SC-VC theory into the orbital ordered state [56]. The obtained $S_S$ and $k$-dependent orbital polarization are shown in Figs. 4 (a) (inset) and (b), respectively. The parameters are $r = 0.256$ and $1/z_4 = 1.6$. The difference $\Delta n = n_{xz} - n_{yz}$ is 0.2%. The hole-pocket around $\Gamma$-point becomes ellipsoidal along the $k_x$-axis due to the “sign-reversing orbital polarization”, in which $\Delta E_{xz}(0, 0) - \Delta E_{yz}(0, 0)$ shows the sign reversal as shown in Fig. 4 (c). Due to this sign reversal, $S_S$ in the inset of Fig. 4 (a) tends to saturate below 40 meV [33]. Also, two Dirac-cone FSs appear around X-point when $\Delta E_{yz}(\pi, 0) > 50$ meV. These results are essentially consistent with the recent ARPES studies reported in Refs. [27-33]. The obtained orbital-polarization $(\Delta E_{xz}(k), \Delta E_{yz}(k))$ belongs to $B_{1g}$ representation, and therefore it is consistent with the “$d$-wave orbital order” discovered in Ref. [31]. The $d$-wave orbital order is theoretically obtained by the mean-field approximation by introducing phenomenological long-range interaction [57], whose microscopic origin might be the ALVC studied in this paper.

In the present FeSe model with $z_4^{-1} = 3$, $\chi^s(q, 0)$ has the maximum at $q = (\pi, 0), (0, \pi)$ without orbital order at $T = 50$ meV, as shown in Fig. 4. (a) In the orbital ordered state, $\chi^s(q, 0)$ at $q = (\pi, 0)$ increases as shown in Fig. 4. (d). These results are consistent with the neutron scattering data for FeSe for both $T > T_{str}$ and $T < T_{str}$ [4-7]. Essentially similar results are obtained for $z_4^{-1} > 1.5$ at $T = 50$ meV. We verified that $\chi^s(q, 0)$ has clear maximum peak at $q = (\pi, 0)$ even for $z_4^{-1} = 1.1$ below $T = 10$ meV using $128 \times 128$ $k$-mashes. Experimentally, $z_{2,3}/z_4$ is about three [27], and the relation $z_{2,3}/z_4 \sim 1.3$ is predicted by the dynamical-mean-field-theory in Ref. [44].

According to Ref. [4], $\chi^s(q, \omega)$ shows the broad maximum at $q = (\pi, 0)$ at low-energies ($\omega \lesssim 10$ meV), and its strength is almost independent for $T > T_{str}$. The magnitude of the low-energy spin susceptibility in FeSe is one order of magnitude smaller than that in BaFe$_2$As$_2$ [58], whereas its magnitude would be comparable to that in LiFeAs [59]. This experimental report in FeSe will be consistent with the present theoretical result with the moderate $S_S \sim 10$ in Figs. 4 (c) and (d). Note that experimental dispersion relation in $\chi^s(q, \omega)$ for $\omega \lesssim 100$ meV is qualitatively understood based on the present FeSe model by considering the band-renormalization factor [7].

![Figure 4](image)

**FIG. 4:** (color online) (a) $T$-dependences of $S_S$ for both LaFeAsO and FeSe models ($z = 1$). The FO order is introduced below $T_{str} = 50$ meV. Inset: $T$-dependences of $S_S$ for the FeSe model obtained by calculating the $k$-dependent orbital polarization and $\chi^s(q)$ self-consistently. $S_S$ tends to saturate below 40 meV due to the sign-reversing orbital polarization. (b) Self-consistent solution of the orbital polarization $(\Delta E_{xz}(k), \Delta E_{yz}(k))$ in the orbital ordered state in the FeSe model at $T = 50$ meV. The shape of the $C_2$-symmetric FSs in (b) is consistent with the experimental reports [27-33]. We also show (c) the $\Delta E_{xz}(yz)(k)$ along the $k_x$-axis, and (d) the $C_2$-symmetric $\chi^s(q)$ in the orbital-ordered state.

**B. The ratio $\theta_S/\theta_C$ for FeSe, NaFeAs, BaFe$_2$As$_2$ and LaFeAsO as functions of $J/U$**

In Fig. 5 (a), we summarize the ratio $\theta_S/\theta_C$ obtained in FeSe, NaFeAs, BaFe$_2$As$_2$, and LaFeAsO as function
In both FeSe and LaFeAsO, strong orbital susceptibility \(\chi_{\alpha}^\omega(0)\) is positive with \(\theta_C\) for the strong orbital-spin interplay due to the strong-coupling effect, called the Aslamazov-Larkin vertex correction in the field theory. In the FeSe model, ferro-orbital order is established even when the spin Weiss temperature \(\theta_S\) is negative as shown in Fig. 3, since the three-point vertex \((=\text{the coupling between two-magnon and one-orbital})\) increases at low temperatures as \(\Delta \propto T^{-0.5}\). In contrast, the spin-nematic susceptibility driven by the spin susceptibility should be \(T\)-independent if \(\theta_S < 0\), as discussed in Ref. [7]. Therefore, we conclude that the nematicity in FeSe originates from the orbital order/fluctuations.

The nematic orbital fluctuations might play important roles in the pairing mechanism in Fe-based superconductors [64]. In FeSe, \(T_C\) increases from 9 K to 40 K under pressure, accompanied by the enhancement of spin fluctuations [1]. At the same time, the system approaches to the orbital critical point since \(T_{str}\) decreases to zero under pressure. These facts indicate the important role of the spin+orbital fluctuations in FeSe.
Acknowledgments

We are grateful to A. Chubukov, P.J. Hirschfeld, R. Fernandes, J. Schmalian, Y. Matsuda, T. Shibauchi and T. Shimojima for useful discussions. This study has been supported by Grants-in-Aid for Scientific Research from MEXT of Japan.

Appendix A: Eight-orbital models for FeSe and LaFeAsO

Here, we introduce the eight-orbital $d$-$p$ models for FeSe and LaFeAsO analyzed in the main text. We first derived the first principles tight-binding models using the WIEN2k and WANNIER90 codes. Crystal structure parameters of FeSe and LaFeAsO are given in Refs. [65] and [66], respectively. The obtained bandstructure and FSs in the LaFeAsO model are shown in Fig. 1 in the main text. In deriving the FeSe model, we introduce the $k$-dependent shifts for orbital $l$, $\delta E_l$, in order to obtain the experimentally observed FSs. In FeSe, we introduce the intra-orbital hopping parameters into $H^0_{\text{FeSe}}$ in order to shift the $d_{xy}$-orbital band [$d_{yz/yz}$-orbital band] at $(\Gamma, M, X)$ points by $(0, -0.25, +0.24)$ $([-0.24, 0, +0.12])$ in unit eV. Such level shifts are introduced by the additional intra-orbital hopping integrals: $\delta^{\text{on-site}} = \delta E_x/4 + \delta E_y/4 + \delta E_z/2$, $\delta^{\text{nn}} = \delta E_x/8 - \delta E_y/8$, and $\delta^{\text{nnn}} = \delta E_x/16 + \delta E_y/16 - \delta E_z/8$. The bandstructure is slightly violated: The splitting between the $d_{x^2}$- and $d_{yz}$-bands, $\Delta E_{\text{band}} \equiv E_{yz} - E_{x^2}$, is $16$ meV at X-point, and $\Delta E_{\text{band}} = 2$ meV at $\Gamma$-point.

Figure 6 (a) is the non-magnetic bandstructure in the orthorhombic LaFeAsO obtained by the WIEN2k software. The spin-orbit interaction is not taken into account. The crystal structure parameters in the orthorhombic phase is given in Ref. [66]. The orthorhombic structure deformation $(a - b)/(a + b)$ is $0.3\%$. Due to the electron-phonon interaction, the four-fold symmetry of the bandstructure is slightly violated: The splitting between the $d_{xz}$- and $d_{yz}$-bands, $\Delta E_{\text{band}} \equiv E_{yz} - E_{xz}$, is $16$ meV at X-point, and $\Delta E_{\text{band}} = 2$ meV at $\Gamma$-point.

Figure 6 (b) is the bandstructure in the orthorhombic FeSe. In the orthorhombic phase, the nearest Fe-Fe length is $a = 2.6716$Å and $b = 2.6610$Å, so $(a - b)/(a + b)$ is $0.2\%$ [65]. Here, the $k$-dependent orbital shift to fit the ARPES bandstructure introduced above is not taken into account. In FeSe, $\Delta E_{\text{band}} = 14$ meV at X-point, and $\Delta E_{\text{band}} = 3$ meV at $\Gamma$-point. Thus, the sign reversing orbital splitting observed in Ref. [33] cannot be explained by the band calculation.

The splitting is reduced by the renormalization factor $z$ due to the self-energy. Since $z \sim 1/3$ in FeSe and LaFeAsO, the renormalized splitting at X-point is $z\Delta E_{\text{band}} \sim 5$meV, which is one order of magnitude smaller than the experimental orbital splitting. Therefore, it is confirmed that the origin of the electronic nematic state in Fe-based superconductors is the electron-electron correlation.

FIG. 6: (color online) Bandstructure of (a) LaFeAsO and (b) FeSe in the experimental orthorhombic crystal structures obtained by the WIEN2k software.
Appendix B: Smallness of the VC for the spin susceptibility

In the original SC-VC theory, the spin and charge susceptibilities are calculated self-consistently, by including the MT-VC and AL-VC for the spin and charge susceptibilities [16, 49]. The strong orbital fluctuations are induced by the charge-channel AL-VC in Fe-based SCs, Ru-oxides and cuprate SCs [16, 37, 50]. In the main text, we studied the eight-orbital $d$-$p$ Hubbard models based on the SC-VC theory, by taking the charge-channel AL-VC into account self-consistently. The obtained susceptibilities are very similar to the RPA since the spin-channel VCs are dropped. It is easy to verify that the charge- and spin-channel MT-VCs are negligible in the present model. The charge- and spin-channel VCs self-consistently. The obtained results are quantitatively equivalent to Fig. 3 in the main text. This fact means that the VC for the spin channel is negligible. (c) $X_{\chi^{MT+s}}(q)$ and (d) $X_{\chi^{MT+AL+}}(q)$ obtained by the present self-consistent calculation.

Appendix C: Analysis of effective models of BaFe$_2$As$_2$ and NaFeAs

In the main text, we introduced the first principles models for LaFeAsO and FeSe, and analyzed these models by using the SC-VC method. Here, we also introduce the effective models for BaFe$_2$As$_2$ and NaFeAs, and analyze them using the SC-VC method.

In both BaFe$_2$As$_2$ and NaFeAs, the FSs have relatively large three-dimensional characters. In addition, the unfolding of the bandstructure in BaFe$_2$As$_2$ cannot be exactly performed because of its body-centered tetragonal crystal structure. Here, we introduce an simple effective BaFe$_2$As$_2$ TB model $H_{\text{BaFeAs}}^{0}$ by magnifying the size of the $d_{x^2-y^2}$ orbital hole-FS around $k = (\pi, \pi)$ in the LaFeAsO unfolded model, in order to reproduce the ARPES bandstructure in Ba122 compounds. Here, we shifted the $d_{xy}$-orbital band at M point by $+0.20$ eV. As for NaFeAs, we just use $H_{\text{NaFeAs}}^{0}$ as an effective NaFeAs TB model, $c.g., H_{\text{NaFeAs}}^{0} = H_{\text{LaFeAsO}}^{0}$, considering that the FSs in NaFeAs in the $k_z = 0$ plane are similar to the FSs in LaFeAsO. We use $H_{\text{NaFeAs}}^{0}$ in place of $H_{\text{LiFeAs}}^{0}$ given in Ref. [34].

The bandstructures and the FSs of the effective TB models for BaFe$_2$As$_2$ and NaFeAs are shown in Figs. 8 (a)-(c). Here, we perform the SC-VC analysis for the models $H_{M} = H_{M}^{0} + rH_{U}^{0}$ ($M$=BaFe$_2$As$_2$, NaFeAs), where $r < 1$ is the reduction parameter. We choose the parameter $r$ to satisfy the charge Stoner factor is $\alpha_C = 0.98$. The obtained $T$-dependences of the spin and charge Stoner enhancement factors, $S_S = (1 - \alpha_S)^{-1}$ and $S_C \equiv (1 - \alpha_C)^{-1}$ respectively, are shown in Fig. 8 (d) and (e). As for BaFe$_2$As$_2$, both spin and orbital fluctuations strongly develop at $T \approx 50$ meV in the case of $r = 0.36$. This result is consistent with experimental re-
T_{\text{mag}} \approx T_{\text{str}} \text{ in BaFe}_2\text{As}_2. \text{ As for NaFeAs, only orbital fluctuations strongly develop whereas spin fluctuations remain moderate at } T \sim 50 \text{ meV in the case of } r = 0.287. \text{ This result is consistent with experimental results in NaFeAs } [62], \text{ in which } T_{\text{mag}}(= 40 \text{ K}) \text{ is more than ten Kelvin smaller than } T_{\text{str}}(= 53 \text{ K}). \text{ Thus, normal-state phase diagrams in BaFe}_2\text{As}_2 \text{ and NaFeAs are well explained by analyzing their effective Hamiltonians using the SC-VC method.}

Appendix D: Why are strong orbital fluctuations induced by tiny spin fluctuations in FeSe?

In the main text, we studied the first-principles d-p Hubbard models for LaFeAsO and FeSe by applying the SC-VC theory. In both models, strong spin-fluctuation-driven orbital fluctuations are induced by AL-VC. In FeSe, we found that very small spin susceptibility \( \chi_{\text{max}} \) is sufficient to realize the orbital order, consistently with experimental results.

Here, we discuss why strong orbital fluctuations are induced by tiny spin fluctuations in FeSe. In Figs. 9 (a) and (b), we show the spin and orbital susceptibilities, \( \chi_{\text{max}}^s \equiv \chi^s(Q) \) and \( \chi_{\text{max}}^c(0) \equiv \chi_{2,2,2,2}(q) + \chi_{3,3,3,3}(q) - 2\chi_{2,2,2,2}(0) \), in the FeSe model and LaFeAsO model obtained by the SC-VC theory. Here, 32 \times 32 \mathbf{k}-meshes and 256 Matsubara frequencies are used. In both models, the charge Stoner factor is \( \alpha_C = 0.98 \) at \( T = 50 \) meV, and the obtained orbital susceptibilities show similar T-dependence. We set \( U = 1.76 \) (\( r = 0.25 \)) in FeSe, and \( U = 1.74 \) (\( r = 0.41 \)) in LaFeAsO, as we did in the main text. As for the spin susceptibility, in LaFeAsO, strong spin fluctuations develop at \( T = 50 \) meV (\( \alpha_S = 0.98 \)), consistently with previous theoretical studies [16, 37]. In FeSe, in contrast, \( \chi_{\text{max}}^s \) is almost constant till \( T = 50 \) meV (\( \alpha_S = 0.87 \)), consistently with experimental reports in FeSe.

Now, we discuss why the spin fluctuation strength required to realize \( \alpha_C \approx 1 \) is so different from LaFeAsO to FeSe. One reason is the difference in the ratio \( J/U \): Figure 9 (c) shows the T-dependence of the AL-VC on \( d_{xx,xy} \)-orbital, \( X_{2\text{AL,c}}(0) \equiv X_{2\text{AL,c}}^{\text{zero}}(0) \), obtained in the LaFeAsO and FeSe models. In both models, \( \alpha_C = 0.98 \) is satisfied at \( T = 50 \) meV. At \( T = 50 \) meV, the AL-VC for FeSe is about one-half of that in LaFeAsO. Thus, small AL-VC is enough to induce large orbital fluctuations in FeSe, since the charge Stoner factor is \( \alpha_C \approx (1 - 5J/U)\bar{\alpha}_2 \).

In Fig. 9 (d), we show that \( X_{2\text{AL,c}}(0) \equiv X_{2\text{AL,c}}^{\text{zero}}(0) \) is very small for both FeSe and LaFeAsO. Here, “zero” represents the zero-Matsubara term (classical contribution) in Eq. (7) in Sec. II. Thus, non-zero Matsubara terms in the AL-VC are negligible in the present calculation (by chance). Note that the \( U^2\)-term in AL-VC gives negative contribution.

Another reason for the relation \( \chi_{\text{max}}^s(\text{FeSe}) \ll \chi_{\text{max}}^s(\text{LaFeAsO}) \) at \( \alpha_C \approx 1 \) is the difference in the orbital dependence of the spin fluctuation strength: The AL-VC for the \( xz \)-orbital is approximately given as

\[
X_{2\text{AL,c}}(0) \approx 3TU^4 \sum_k |A_{2,2,2,2}(0;k)|^2 \chi_2^s(k)^2
\]

where we dropped the inter-orbital terms of \( \chi^s \) and \( \bar{\Lambda} \), and leave only the zero-Matsubara term in the Matsubara summation in Eq. (7) in Sec. II. If Eq. (D1) is justified, only the spin fluctuations on \( d_{xz,dyz} \)-orbitals are important for the FO fluctuations.

Figure 10 (a) shows \( \chi_2^c(Q) \) for FeSe and LaFeAsO for...
LaFeAsO. (c) and (c) orbital susceptibilities from Fig. 9 (a) and Fig. 10 (a), the ratio calculations in Fig. 9 (c).

\[ \chi^s(Q) \sim \frac{1}{2} \chi^c(Q) \]

\[ \chi^s(Q) \]

FIG. 9: (color online) (a) Spin susceptibility $\chi^s(Q)$ and (b) orbital susceptibilities $\chi^c_{x^2-y^2}(0)$ for FeSe and LaFeAsO as functions of $T$. We put $r = 0.25$ for FeSe, and $r = 0.41$ for LaFeAsO. (c) $X^c_{AL}(0)$ and (d) $X^c_{AL,\text{non-\text{z\text{z}}}0}(0)$ for FeSe and LaFeAsO.

\[ X^c_{AL}(0) \]

FIG. 10: (color online) (a) $\chi^s(Q)$, (b) $C_2 = \sum_q \chi^c(q)^2$, and (c) $|A_2|^2$ as functions of $T$ in FeSe and LaFeAsO. (d) The approximate AL-VC for $d_{xz}$-orbital $X^c_{AL}(0)$ is $C_2^x(T) = 3U^4 |A_2(0; (0, \pi))|^2TC_2$. In both FeSe and LaFeAsO, the obtained $X^c_{AL}(0)$ qualitatively agrees to the exact numerical calculations in Fig. 9 (c).

\[ C_2^x(T) \]

the same model parameters used in Fig. 9. As derived from Fig. 9 (a) and Fig. 10 (a), the ratio $\chi^c(Q)/\chi^s(Q)$ is just 0.22 in LaFeAsO, whereas the ratio increases to 0.53 in FeSe, since the relation $\chi^c(Q) \ll \chi^s(Q)$ ($\chi^s(Q) \sim \chi^c(Q)$) is satisfied in FeSe (LaFeAsO) because of the absence (presence) of h-FS3. This orbital dependence of the spin fluctuations in FeSe is favorable for realizing the FO fluctuations.

To understand the model-dependence of the AL-VC in more detail, we calculate $C_2 = \sum_q \chi^c(q)^2$ and show the result in Fig. 10 (b): The ratio $C_2^x/\chi^c_{\text{FeSe}}$ is just 1.35 since the width of the peak of $\chi^c(Q)$ around $Q = Q_0$ is much wider in FeSe. We also examine the scale of the three-point vertex for $d_{xz}$-orbital $A_2 = A_2(22222, q, k)$ at $q = 0$ and $k = Q$ in Fig. 10 (c). In both models, the relation $|A_2|^2 \propto T^a$ with $a \approx 1$ is satisfied for wide temperature range: Such strong $T$-dependence of the charge-spin coupling $A_2$ is essential for realizing the orbital fluctuations, so it should be taken into account in the numerical calculation. As results, we obtain a crude approximation for the AL-VC, $X^c_{AL}(Q) = 3U^4 |A_2(0; (0, \pi))|^2TC_2$, and show the result in Fig. 10 (d). This crude approximation qualitatively reproduces the exact numerical results for both FeSe and LaFeAsO given in Fig. 9 (c).

In summary, in both LaFeAsO and FeSe, strong orbital fluctuations are induced by AL-VC for the $d_{xz}$-orbital, $X^c_{AL}(0)$. In FeSe, very small spin susceptibility $\chi^s_{\text{max}}$ is sufficient to realize the spin-fluctuation-driven orbital order, because of both the smallness of $J/\xi$ and the largeness of $C_2$. Strong $T$-dependence of $A_2$ is essential for realizing the orbital fluctuations due to AL-VC.

Appendix E: Strong $T$-dependence of the three-point vertex

In this paper, we found that the strong orbital fluctuations in Fe-based superconductors originate from the AL-VC for the orbital susceptibility. The moderate increment of the AL-VC at low temperatures shown in Fig. 9 (c) gives rise to the Curie-Weiss behavior of $\chi^c_{x^2-y^2}(0)$. For the increment of the AL-VC, the strong $T$-dependence of the three-point vertex, shown in Fig. 10 (c), plays the significant role. Its strong $T$-dependence in Fe-based superconductors had been pointed out in Refs. [20, 21, 51, 52].

Here, we calculate the three-point vertex for LaFeAsO and FeSe models for wide temperature range with high numerical accuracy, using $512 \times 512 k$-mashes and the 2048 Matsubara frequencies. Figure 11 shows the square of the three-point vertex for $d_{xz}$-orbital $A_2(0; Q) \equiv A_2(22222, 0; Q)$ for $T \geq 10$ meV. In both LaFeAsO and FeSe models, the coefficient $a$ of $|A_2(0; Q)|^2 \propto T^a$ depends on the temperature range. In both models, $a \approx 1$ for $T = 20 \sim 100$ meV, so the numerical result in Fig. 10 (c) is confirmed by this accurate calculation. When the band renormalization due to $z < 1$ is considered, the relation $a \approx 1$ is realized for $T = 20z \sim 100z[\text{meV}]$.

For $T < 20z[\text{meV}]$, $|A_2(0; Q)|^2$ saturates since the
temperature is smaller than the energy scale of the nesting. For $T > 100 z$ [meV], the relation $\alpha \approx 2$ is realized as discussed in Ref. [51, 52]. Note that the chemical potential $\mu$ becomes higher than the of the hole-band at $\Gamma$ point when $T$ is higher than $100 z$ (300 z) [meV] in the FeSe (LaFeAsO) model.

In Ref. [20], we reported the relation $\chi_{2}^{AL,c}(0)/T \propto T^{-0.5}(1-\alpha_s)^{-1}$ based on approximate calculation for the five-orbital LaFeAsO TB model. The factor $T^{-0.5}$ originate from $|A_{2}|^2$. However, we performed more careful numerical analysis, and found that the approximate relation $|A_{2}|^2 \sim T^{-1} (a \approx 1)$ is realized for $T \sim 50 z$ [meV] in the five-orbital model.

Appendix F: Two definitions of the averaged Coulomb and Hund’s interactions

In the present study, the ratio between the intra-orbital Coulomb interaction and Hund’s interaction, $\tilde{J}/\tilde{U}$, is the essential control parameter. In this paper, we follow the Hubbard-Kanamori definition: $\tilde{U} \equiv \frac{1}{10} \sum_{l=1}^{5} U_{l,l}$ and $\tilde{J} \equiv \frac{1}{10} \sum_{l,m}^{2} J_{l,m}$. This definition is used in Ref. [34]. By using the Slater integrals [67], they are expressed as $\tilde{U} = U_0 + \frac{\tilde{U}}{\tilde{J}} (F^2 + F^4)$, $\tilde{U}' = U_0 - \frac{\tilde{U}}{\tilde{J}} (F^2 + F^4)$, and $\tilde{J} = \frac{5}{8} (F^2 + F^4)$ [67]. According to the first-principles cRPA method [34], the ratio $\tilde{J}/\tilde{U}$ is 0.0945 for FeSe, and the relation $\tilde{U}' = \tilde{U} - 2 \tilde{J}$ is approximately satisfied.

In the first principles studies, another definition of the averaged interaction is used frequently: $\tilde{U} = U_0$ and $\tilde{J} = \frac{\tilde{U}}{\tilde{J}} (F^2 + F^4)$. This definition is used in Refs. [70, 71]. They are equivalent to $\tilde{U} = \frac{1}{29} \left( \sum_{l=1}^{5} U_{l,l} + \sum_{l \neq m} U'_{l,m} \right)$ and $\tilde{J} = \tilde{U}_S - \frac{1}{10} \sum_{l \neq m} \left( U'_{l,m} - J_{l,m} \right)$ [68, 69].

Therefore, if we assume $\tilde{U}' = \tilde{U} - 2 \tilde{J}$, which is actually satisfied well in Ref. [34], we obtain the relations $\tilde{U} = \frac{1}{29} \tilde{J}$ and $\tilde{J} = \frac{1}{2} \tilde{J}$. Thus, $\tilde{J}/\tilde{U} = 0.224$ obtained by the first principles study for FeSe in Ref. [70] corresponds to $\tilde{J}/\tilde{U} = 0.127$. Also, $\tilde{J}/\tilde{U} = 0.294$ for LaFeAsO obtained in Refs. [70, 71] corresponds to $\tilde{J}/\tilde{U} = 0.157$.

As shown in Ref. 5 (b), the value of $\alpha_{ST}^S$ remains small ($\sim 0.9$) in the FeSe TB model with $H_{1}^{L,R} \propto K_{S} (\tilde{J}/\tilde{U} = 0.12)$ or with $H_{2}^{L,R} (\tilde{J}/\tilde{U} = 0.134)$. In each case, the obtained $T$-dependences of $S_S$ and $S_C$ are qualitatively similar to those shown in Fig. 3 (c). Therefore, the main results of the present study are unchanged even if $J/\tilde{U}$ in FeSe is slightly larger than 0.1.

[1] D. C. Johnston, *The puzzle of high temperature superconductivity in layered iron pnictides and chalcogenides*, Adv. Phys. 59, 803 (2010); Y. Mizuguchi and Y. Takano, *Review of Fe Chalcogenides as the Simplest Fe-Based Superconductor*, J. Phys. Soc. Jpn. 79, 102001 (2010).

[2] A. E. Böhm, T. Arai, F. Hardy, T. Hattori, T. Iye, T. Wolf, H. v. Lohneysen, K. Ishida, and C. Meingast, *Origin of the Tetragonal-to-Orthorhombic Phase Transition in FeSe: A Combined Thermodynamic and NMR Study of Nematicity*, Phys. Rev. Lett. 114, 027001 (2015).

[3] S.-H. Baek, D. V. Efremov, J. M. Ok, J. S. Kim, Jeroen van den Brink, and B. Böchner, *Orbital-driven nematicity in FeSe*, Nat. Mater. 14, 210 (2015).

[4] M. C. Rahn, R. A. Ewings, S. J. Sedlmaier, S. J. Clarke, and A. T. Boothroyd, *Strong $\pi$ spin fluctuations in $\beta$ – FeSe observed by neutron spectroscopy*, Phys. Rev. B 91, 180501(R) (2015).

[5] Q. Wang, Y. Shen, B. Pan, Y. Hao, M. Ma, F. Zhou, P. Steffens, K. Schmädl, T. R. Forrest, M. Abdel-Hafiez, X. Chen, D. A. Chareev, A. N. Vasiliev, P. Bourges, Y. Sidis, H. Cao, and J. Zhao, *Strong interplay between stripe spin fluctuations, nematicity and superconductivity in FeSe*, Nat. Mater. 15, 159 (2016).

[6] Q. Wang, Y. Shen, B. Pan, X. Zhang, K. Ikeuchi, K. Iida, A. D. Christianson, H. C. Walker, D. T. Adroja, M. Abdel-Hafiez, X. Chen, D. A. Chareev, A. N. Vasiliev, and J. Zhao, *Magnetic ground state of FeSe*, Nat. Comm. 8, 157003 (2016).

[7] S. Shamoto, K. Matsuoka, R. Kajimoto, M. Ishikado, Y. Yamakawa, T. Watashige, S. Kasahara, M. Nakamura, H. Kontani, T. Shibuchi, and Y. Matsuda, *Spin nematic susceptibility studied by inelastic neutron scattering in FeSe*, arXiv:1511.04267.

[8] R. M. Fernandes, L. H. VanBebber, S. Bhattacharya, P. Chandra, V. Keppens, D. Mandrus, M. A. McGuire, B. C. Sales, A. S. Sefat, and J. Schmalian, *Effects of Nematic Fluctuations on the Elastic Properties of Iron Arsenide Superconductors*, Phys. Rev. Lett. 105, 157003 (2010).

[9] F. Wang, S. A. Kivelson, and D.-H. Lee, *Nematicity and
quantum paramagnetism in FeSe, Nat. Phys. 11, 959 (2015).
[10] A. V. Chubukov, R. M. Fernandes, and J. Schmalian, Origin of nematic order in FeSe, Phys. Rev. B 91, 201105(R) (2015).
[11] J. K. Glashammer, I. I. Mazin, H. O. Jeschke, P. J. Hirschfeld, R. M. Fernandes, and R. Valenti, Effect of magnetic frustration on nematicity and superconductivity in iron chalcogenides, Nat. Phys. 11, 953 (2015).
[12] R. Yu and Q. Si, Antiferroquadrupolar and Ising-Nematic Orders of a Frustrated Bilinear-Biquadratic Heisenberg Model and Implications for the Magnetism of FeSe, Phys. Rev. Lett. 115, 116401 (2015).
[13] H. Kontani and Y. Yamakawa, Spin-orbital frustrations and anomalous metallic state in iron-pnictide superconductors, Phys. Rev. B 79, 054504 (2009).
[14] W. Lv, J. Wu, and P. Phillips, Orbital ordering induces structural phase transition and the resistivity anomaly in iron pnictides, Phys. Rev. B 80, 224506 (2009).
[15] C.-C. Lee, W.-G. Yin, and W. Ku, Ferro-Orbital Order and Strong Magnetic Anisotropy in the Parent Compounds of Iron-Pnictide Superconductors, Phys. Rev. Lett. 103, 267001 (2009).
[16] S. Onari and H. Kontani, Self-consistent Vertex Correlation Analysis for Iron-based Superconductors: Mechanism of Coulomb Interaction-Driven Orbital Fluctuations, Phys. Rev. Lett. 109, 137001 (2012).
[17] M. Yoshizawa, D. Kimura, T. Chiba, S. Simayi, Y. Nakamichi, K. Kihou, C.-H. Lee, A. Iyo, H. Eisaki, M. Nakajima, and S. Uchida, Structural Quantum Criticality and Superconductivity in Iron-based Superconductor Ba(Fe1−xCox)2As2, J. Phys. Soc. Jpn. 81, 024604 (2012).
[18] A. E. B¨ohmer, P. Burger, F. Hardy, T. Wolf, P. Schweiss, R. Fromknecht, M. Reinecker, W. Schranz, and C. Meingast, Nematic Susceptibility of Hole-Doped and Electron-Doped BaFe2As2 Iron-Based Superconductors from Shear Modulus Measurements, Phys. Rev. Lett. 112, 047001 (2014).
[19] Y. Gallais, R. M. Fernandes, I. Paul, L. Chauviere, Y.-X. Yang, M.-A. Mészáros, M. Cazayous, A. Sacuto, D. Colson, and A. Forget, Observation of Incipient Charge Nematicity in Ba(Fe1−xCox)2As2, Phys. Rev. Lett. 111, 267001 (2013).
[20] H. Kontani and Y. Yamakawa, Linear Response Theory for Shear Modulus C66 and Raman Quadrupole Susceptibility: Evidence for Nematic Orbital Fluctuations in Fe-based Superconductors, Phys. Rev. Lett. 113, 047001 (2014).
[21] M. Khodas and A. Levchenko, Raman scattering as a probe of nematic correlations, Phys. Rev. B 91, 235119 (2015).
[22] U. Karahasanovic, P. Kretzschmar, T. Böhm, R. Hackl, I. Paul, Y. Gallais, and J. Schmalian, Manifestation of nematic degrees of freedom in the Raman response function of iron pnictides, Phys. Rev. B 92, 075134 (2015).
[23] J.-H. Chu, H.-H. Kuo, J. G. Analytis, and I. R. Fisher, Divergent Nematic Susceptibility in an Iron Arsenide Superconductor, Science 337, 710 (2012).
[24] H.-H. Kuo, J.-H. Chu, S. A. Kivelson, and I. R. Fisher, Ubiquitous signatures of nematic quantum criticality in optimally doped Fe-based superconductors, arXiv:1503.00402.
[25] C. J. Halboth, W. Metzner, d-Wave Superconductivity and Pomeranchuk Instability in the Two-Dimensional Hubbard Model, Phys. Rev. Lett. 85, 5162 (2000); C. Honerkamp, M. Salmhofer, N. Furukawa, T. M. Rice, Breakdown of the Landau-Fermi liquid in two dimensions due to umklapp scattering, Phys. Rev. B 63, 035109 (2001); H. Yamase and H. Kohno, Instability toward Formation of Quasi-One-Dimensional Fermi Surface in Two-Dimensional t-J Model, J. Phys. Soc. Jpn. 69 (2000) 2151.
[26] M. Yi, D. Lu, J.-H. Chu, J. G. Analytis, A. P. Sorini, A. F. Kemper, B. Moritz, S.-K. Mo, R. G. Moore, M. Hashimoto, W.-S. Lee, Z. Hussain, T. P. Devereaux, I. R. Fisher, and Z.-X. Shen, Symmetry-breaking orbital anisotropy observed for detwinned Ba(Fe1−xCox)2As2 above the spin density wave transition, Proc. Natl. Acad. Sci. USA 108, 6878 (2011).
[27] J. Maletz, V. B. Zabolotnyy, D. V. Evtushinsky, S. Thirupathaiah, A. U. B. Wolter, L. Harnagea, A. N. Yaresko, A. N. Vasiliev, D. A. Chareev, A. E. Böhm, F. Hardy, T. Wolf, C. Meingast, E. D. L. Rienks, B. Büchner, and S. V. Borisenko, Unusual band renormalization in the simplest iron-based superconductor FeSe1−x, Phys. Rev. B 89, 220506(R) (2014).
[28] K. Nakayama, Y. Miyata, G. N. Phan, T. Sato, Y. Tanabe, T. Urate, K. Tanigaki, and T. Takahashi, Reconstruction of Band Structure Induced by Electronic Nematicity in an FeSe Superconductor, Phys. Rev. Lett. 113, 237001 (2014).
[29] M. D. Watson, T. K. Kim, A. A. Hagghighirad, N. R. Davies, A. McCollam, A. Narayanan, S. F. Blake, Y. L. Chen, S. Ghannadzadeh, A. J. Schofield, M. Hoesch, C. Meingast, T. Wolf, and A. I. Coldea, Emergence of the nematic electronic state in FeSe, Phys. Rev. B 91, 155105 (2015).
[30] T. Shimojima, Y. Suzuki, T. Sonobe, A. Nakamura, M. Sakano, J. Omachi, K. Yoshio, M. Kuwata-Gonokami, K. Ono, H. Kumigashira, A. E. Böhm, F. Hardy, T. Wolf, C. Meingast, H. v. Löhneysen, H. Ikeda, and K. Ishizaka, Lifting of zz/yz orbital degeneracy at the structural transition in detwinned FeSe, Phys. Rev. B 90, 121111(R) (2014).
[31] P. Zhang, T. Qian, P. Richard, X. P. Wang, H. Miao, B. Q. Lv, B. B. Fu, T. Wolf, C. Meingast, X. X. Wu, Z. Q. Wang, J. P. Hu, and H. Ding, Observation of two distinct dzz/dxz band splittings in FeSe, Phys. Rev. B 91, 214503 (2015).
[32] Y. Zhang, M. Yi, Z.-K. Liu, W. Li, J. J. Lee, R. G. Moore, M. Hashimoto, N. Masamichi, H. Eisaki, S.-K. Mo, Z. Hussain, T. P. Devereaux, Z.-X. Shen, and D. H. Lu, Distinctive momentum dependence of the band reconstruction in the nematic state of FeSe thin film, arXiv:1503.01556.
[33] Y. Suzuki, T. Shimojima, T. Sonobe, A. Nakamura, M. Sakano, H. Tsuji, J. Omachi, K. Yoshio, M. Kuwata-Gonokami, T. Watashige, R. Kobayashi, S. Kasahara, T. Shibauchi, Y. Matsuda, Y. Yamakawa, H. Kontani, and K. Ishizaka, Momentum-dependent sign-inversion of orbital polarization in superconducting FeSe, Phys. Rev. B 92, 205117 (2015).
[34] T. Miyake, K. Nakamura, R. Arita, and M. Imada, Comparison of Ab initio Low-Energy Models for LaFePO, LaFeAsO, BaFe2As2, LiFeAs, FeSe, and FeTe: Electron Correlation and Covalency, J. Phys. Soc. Jpn. 79, 044705
(2010).

[35] M. Tsuchiizu, Y. Ohno, S. Onari, and H. Kontani, Orbital Nematic Instability in the Two-Orbital Hubbard Model: Renormalization-Group + Constrained RPA Analysis, Phys. Rev. Lett. 111, 057003 (2013).

[36] M. Tsuchiizu, Y. Yamakawa, S. Onari, Y. Ohno, and H. Kontani, Spin-triplet superconductivity in Sr$_2$RuO$_4$ due to orbital and spin fluctuations: Analyses by two-dimensional renormalization group theory and self-consistent vertex-correction method, Phys. Rev. B 91, 155103 (2015).

[37] S. Onari, Y. Yamakawa, and H. Kontani, High-T$_c$ Superconductivity near the Anson Height Instability in Layered-Based Superconductors: Analysis of LaFeAsO$_{1-x}$F$_x$, Phys. Rev. Lett. 112, 187001 (2014).

[38] Y. Yamakawa and H. Kontani, Spin-Fluctuation-Driven Nematic Charge-Density Wave in Cuprate Superconductors: Impact of Aslamazov-Larkin Vertex Corrections, Phys. Rev. Lett. 114, 257001 (2015).

[39] M. Tsuchiizu, Y. Yamakawa, and H. Kontani, p-Orbital Density Wave with d Symmetry in High-T$_c$ Cuprate Superconductors, arXiv:1508.07218.

[40] C. Xu, M. Müller, and S. Sachdev, Ising and spin orders in the iron-based superconductors, Phys. Rev. B 78, 020501(R) (2008); C. Fang, H. Yao, W.-F. Tsai, J.P. Hu, and S. A. Kivelson, Theory of electronic nematic order in LaFeAsO, Phys. Rev. B 77 224509 (2008); E. Abrahams and Q. Si, Quantum criticality in the iron pnictides and chalcogenides, J. Phys.: Condens. Matter 23, 223201 (2011).

[41] A. V. Chubukov and P. J. Hirschfeld, Iron-based superconductors, seven years later, Physics Today 68, 46 (2015); H. Hosono and K. Kuroki, Iron-based superconductors: Current status of materials and pairing mechanism, Physica C 514, 399 (2015).

[42] T. Terashima, N. Kikugawa, E.-S. Choi, A. Audouard, F. Duc, L. Drigo, P. Toulemonde, S. Kasahara, T. Watashige, T. Hanaguri, Y. Kohsaka, T. Z. P. Yin, K. Haule, and G. Kotliar, A. V. Chubukov and P. J. Hirschfeld, C. Xu, M. Müller, and S. Sachdev, Ising and spin orders in the iron-based superconductors, Phys. Rev. B 78, 020501(R) (2008); C. Fang, H. Yao, W.-F. Tsai, J.P. Hu, and S. A. Kivelson, Theory of electronic nematic order in LaFeAsO, Phys. Rev. B 77 224509 (2008); E. Abrahams and Q. Si, Quantum criticality in the iron pnictides and chalcogenides, J. Phys.: Condens. Matter 23, 223201 (2011).

[43] A. V. Chubukov and P. J. Hirschfeld, Iron-based superconductors, seven years later, Physics Today 68, 46 (2015); H. Hosono and K. Kuroki, Iron-based superconductors: Current status of materials and pairing mechanism, Physica C 514, 399 (2015).

[44] T. Terashima, N. Kikugawa, A. Kiswandhi, Y. Kohsaka, T. Hanaguri, Y. von Löhneysen, T. Shibauchi, Y. Matsuda, T. Wolf, A. E. Böhm, F. Hardy, C. Meingast, H. v. Löhneysen, M.-T. Suzuki, R. Arita, F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H.-H. Wen, Contrastino Spin Dynamics between Underdoped and Overdoped BaFe$_{1-x}$Co$_{x}$As$_2$, Phys. Rev. Lett. 104, 037001 (2010).

[45] R. Arita, private communication.

[46] A. V. Chubukov and P. J. Hirschfeld, Iron-based superconductors, seven years later, Physics Today 68, 46 (2015); H. Hosono and K. Kuroki, Iron-based superconductors: Current status of materials and pairing mechanism, Physica C 514, 399 (2015).

[47] K. Suzuki, H. Usui, and K. Kuroki, Possible Three-Dimensional Nodes in the ± Superconducting Gap of BaFe$_2$(As$_{1-x}$P$_x$)$_2$, J. Phys. Soc. Jpn. 80, 013710 (2011).

[48] T. Misawa and M. Imada, Superconductivity and its mechanism in an ab initio model for electron-doped LaFeAsO, Nat. Commun. 5, 5738 (2014).

[49] S. Onari and H. Kontani, Iron-Based Superconductivity, (ed. P. D. Johnson, G. Xu, and W.-G. Yin, Springer-Verlag Berlin and Heidelberg GmbH & Co. K (2015).

[50] Y. Ohno, M. Tsuchiizu, S. Onari, and H. Kontani, Spin-Fluctuation-Driven Orbital Nematic Order in Ru-Oxides: Self-Consistent Vertex Correction Analysis for Two-Orbital Model, J. Phys. Soc. Jpn 82, 013707 (2013).

[51] A. Hinojosa, J. Cai, and A. V. Chubukov, Raman resonance in iron-based superconductors: The magnetic scenario, Phys. Rev. B 93, 075106 (2016).

[52] I. Paul, Nesting-induced large magnetoelectricity in the iron-arsenide systems, Phys. Rev. B 90, 115102 (2014).

[53] H. Kontani and M. Ohno, Effect of a nonmagnetic impurity in a nearly antiferromagnetic Fermi liquid: Magnetic correlations and transport phenomena, Phys. Rev. B 74, 014406 (2006).

[54] H. Kontani, T. Saito, and S. Onari, Origin of orthorhombic transition, magnetic transition, and shear-modulus softening in iron pnictide superconductors: Analysis based on the orbital fluctuations theory, Phys. Rev. B 84, 024528 (2011).

[55] S. Mukherjee, A. Kreisel, P. J. Hirschfeld, and B. M. Andersen, Model of Electronic Structure and Superconductivity in Orbitally Ordered FeSe, Phys. Rev. Lett. 115, 026402 (2015); A. Kreisel, S. Mukherjee, P. J. Hirschfeld, and B. M. Andersen, Spin excitations in a model of FeSe with orbital ordering, Phys. Rev. B 92, 224515 (2015).

[56] S. Onari, Y. Yamakawa, and H. Kontani, Sign-Reversing Orbital Polarization in the Nematic Phase of FeSe Driven by Aslamazov-Larkin Processes, arXiv:1509.01172.

[57] K. Jiang, J. Hu, H. Ding, and Z. Wang, Interatomic Coulomb interaction and electron nematic bond order in FeSe, Phys. Rev. B 93, 115138 (2016).

[58] 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, Normal-state spin dynamics and temperature-dependent spin-resonance energy in optimally doped BaFe$_{1.85}$Co$_{0.15}$As$_2$, Nature Physics 6, 178 (2010).

[59] N. Qureshi, P. Steffens, D. Lamago, Y. Sidis, O. Sobolev, R. A. Ewings, L. Harranaga, S. Wurmehl, B. Büncher, and M. Braden, Fine structure of the incommensurate antiferromagnetic fluctuations in single-crystaline LiFeAs studied by inelastic neutron scattering, Phys. Rev. B 90, 144503 (2014).

[60] Y. Nakai, S. Kitagawa, T. Iye, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, Enhanced anisotropic spin fluctuations below tetragonal-to-orthorhombic transition in LaFeAsO$_{1-x}$F$_x$ probed by $^{57}$As and $^{139}$La NMR, Phys. Rev. B 85, 134408 (2012).

[61] F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H.-H. Wen, Contrastino Spin Dynamics between Underdoped and Overdoped BaFe$_{1-x}$Co$_{x}$As$_2$, Phys. Rev. Lett. 104, 037001 (2010).

[62] L. Ma, G. F. Chen, D.-X. Yao, J. Zhang, S. Zhang, T.-L. Xia, and W. Yu, $^{23}$Na and $^{75}$As NMR study of antiferromagnetism and spin fluctuations in NaFeAs single
[63] A.V. Chubukov, M. Khodas, and R. M. Fernandes, *Magnetism, superconductivity, and spontaneous orbital order in iron-based superconductors: who comes first and why?*, arXiv:1602.05503.

[64] H. Kontani and S. Onari, *Orbital-Fluctuation-Mediated Superconductivity in Iron Pnictides: Analysis of the Five-Orbital Hubbard-Holstein Model*, Phys. Rev. Lett. **104**, 157001 (2010).

[65] A. E. Böhmer, F. Hardy, F. Eilers, D. Ernst, P. Adelmann, P. Schweiss, T. Wolf, and C. Meingast, *Lack of coupling between superconductivity and orthorhombic distortion in stoichiometric single-crystalline FeSe*, Phys. Rev. B **87**, 180505 (2013).

[66] T. Nomura, S. W. Kim, Y. Kamihara, M. Hirano, P. V. Sushko, K. Kato, M. Takata, A. L. Shluger, and H. Hosono, *Crystallographic phase transition and high-Tc superconductivity in LaFeAsO:F*, Supercond. Sci. Technol. **21**, 125028 (2008).

[67] J. C. Slater, *The Theory of Complex Spectra*, Phys. Rev. **34**, 1293 (1929).

[68] V. I. Anisimov, I. V. Solovyev, M. A. Korotin, M. T. Czyżyk, and G. A. Sawatzky, *Density-functional theory and NiO photoemission spectra*, Phys. Rev. B **48**, 16929 (1993).

[69] L. Vaugier, H. Jiang, and S. Biermann, *Hubbard U and Hund exchange J in transition metal oxides: Screening versus localization trends from constrained random phase approximation*, Phys. Rev. B **86**, 165105 (2012).

[70] M. Aichhorn, S. Biermann, T. Miyake, A. Georges, and M. Imada, *Theoretical evidence for strong correlations and incoherent metallic state in FeSe*, Phys. Rev. B **82**, 064504 (2010).

[71] M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, *Dynamical mean-field theory within an augmented plane-wave framework: Assessing electronic correlations in the iron pnictide LaFeAsO*, Phys. Rev. B **80**, 085101 (2009).