PHYSICAL REVIEW B **102**, 060504(R) (2020)

Rapid Communications

Suppression of superconductivity by spin fluctuations in iron-based superconductors

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(Received 20 November 2019; accepted 12 August 2020; published 27 August 2020)

We study the superconducting instability mediated by spin fluctuations in the Eliashberg theory for a minimal two-band model of iron-based superconductors. While antiferromagnetic spin fluctuations can drive superconductivity (SC) as is well established, we find that spin fluctuations necessarily contain a contribution to suppress SC even though SC can eventually occur at lower temperatures. This self-restraint effect stems from a general feature of the spin-fluctuation mechanism, namely, the repulsive pairing interaction, which leads to phase frustration of the pairing gap and consequently the suppression of SC.

DOI: 10.1103/PhysRevB.102.060504

Iron-based superconductors (FeSC) provide a platform to explore a mechanism of high-temperature (high-T_c) superconductivity (SC) [1]. Since SC is realized close to a spin-density-wave (SDW) phase, the importance of spin fluctuations is widely recognized as a possible mechanism of SC [2–4]. A close look at the phase diagram of FeSC reveals the presence of an electronic nematic phase, which is also close to the SC phase. While the origin of the nematic phase is still controversial [5], it was shown that orbital nematic fluctuations lead to strong-coupling SC with an onset temperature comparable to the observation [6,7]. The electronic structure of FeSC is characterized by multibands originating from five $3d$ orbitals of Fe ions [3]. Hence the orbital fluctuations are also explored as a possible mechanism of SC [8–10]. While electron-phonon coupling is present in real materials and is expected to lead to SC, the transition temperature (T_c) is believed to be too low compared to the observation [11].

The distinction between different SC mechanisms is a key issue of FeSC. Typically spin fluctuations lead to the so-called $s_{\pm}$-wave symmetry [2–4] whereas nematic [6,7,12] and orbital [8,9] fluctuations yield s_{++}-wave symmetry. Obviously this symmetry difference is crucial, but it is not easy to resolve the phase of SC order in experiments. Furthermore, an $s_{\pm}$-wave pairing gap was found to be stabilized even for nematic fluctuations when a partial contribution from spin fluctuations is considered in Ref. [13], suggesting that the gap symmetry itself cannot be decisive in identifying the SC mechanism.

The momentum dependence of the pairing gap is expected to depend on the underlying SC mechanism. However, it turned out [7] that nematic fluctuations lead to a pairing gap similar to that from spin fluctuations, except for the sign of the pairing gap. Considering simplifications involved in many theoretical studies, it is not easy to extract a robust and key difference of the gap structure, which can distinguish between the different SC mechanisms.

Irrespective of the underlying SC mechanism in FeSC, it is tacitly assumed that spin, orbital, and nematic fluctuations work positively on driving SC. However, in this Rapid Communication, we find that spin fluctuations tend to suppress the SC instability even though spin fluctuations can eventually lead to SC at lower temperatures. This self-restraint effect is a general feature originating from a repulsive pairing interaction, which yields a sign change of the pairing gap on the Fermi surfaces (FSs) connected by a momentum transfer of the spin fluctuations.

A minimal model for the band structure of FeSC may read as [14,15]

$$H_0 = \sum_{\mathbf{k},\sigma,\alpha,\beta} \epsilon_{\mathbf{k}}^{\alpha\beta} c_{\mathbf{k}\alpha\sigma}^{\dagger} c_{\mathbf{k}\beta\sigma} \tag{1}$$

on a square lattice, where the unit cell contains one iron and $\alpha = 1$ and 2 refer to the d_{xz} and d_{yz} orbitals, respectively; $c_{\mathbf{k}\alpha\sigma}^{\dagger}$ and $c_{\mathbf{k}\alpha\sigma}$ are the creation and annihilation operators for electrons with momentum $\mathbf{k}$, orbital α, and spin orientation σ; intraorbital dispersions are given by $\epsilon_{\mathbf{k}}^{11} = -2t_1 \cos k_x - 2t_2 \cos k_y - 4t_3 \cos k_x \cos k_y - \mu$ and $\epsilon_{\mathbf{k}}^{22} = -2t_2 \cos k_x - 2t_1 \cos k_y - 4t_3 \cos k_x \cos k_y - \mu$, whereas the interorbital dispersion is $\epsilon_{\mathbf{k}}^{12} = -4t_4 \sin k_x \sin k_y$; μ is the chemical potential. The typical FSs observed in FeSC are well captured by choosing the parameters as [15] $t = -t_1$, $t_2/t = 1.5$, $t_3/t = -1.2$, $t_4/t = -0.95$, and $\mu/t = 0.6$. In the following, we measure all quantities with the dimension of energy in units of t.

As shown in Fig. 1(a), the Hamiltonian (1) yields two hole FSs around $\mathbf{k} = (0, 0)$ and (π, π), and two electron FSs around $\mathbf{k} = (\pi, 0)$ and $(0, \pi)$, which we refer to as FS1, FS2, FS3, and FS4, respectively. FS1 and FS2 originate from both d_{xz} and d_{yz} orbitals whereas FS3 consists of the d_{yz} orbital and FS4 d_{xz} orbital. These FSs capture the orbital components obtained in a more realistic five-band model [16].

To clarify the effect of spin fluctuations on SC, we consider a general SU(2) symmetric two-particle interaction

$$H_I = \frac{1}{8N} \sum_{\mathbf{q},\mathbf{k},\mathbf{k}'} \sum_{\alpha,\beta,\sigma_j} V(\mathbf{k},\mathbf{k}',\mathbf{q}) \times \boldsymbol{\sigma}_{\sigma_1\sigma_2} \cdot \boldsymbol{\sigma}_{\sigma_3\sigma_4} c_{\mathbf{k}\alpha\sigma_1}^{\dagger} c_{\mathbf{k}+\mathbf{q}\alpha\sigma_2} c_{\mathbf{k}'+\mathbf{q}\beta\sigma_3}^{\dagger} c_{\mathbf{k}'\beta\sigma_4}, \tag{2}$$

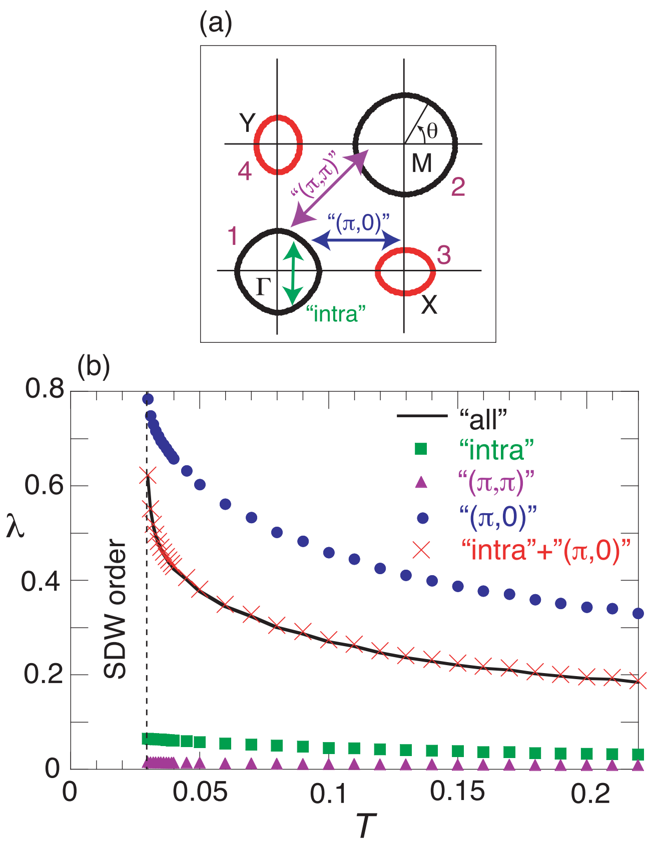

FIG. 1. (a) Hole Fermi pockets (1 and 2) around Γ and M points and electron pockets (3 and 4) around X and Y in the normal state. "intra," "$(\pi,0)$," and "(π,π)" denote scattering processes inside each pocket, between the hole and electron pockets, and between the two hole (or electron) pockets, respectively. (b) Temperature dependence of the eigenvalues λ (solid line). The eigenvalues are also computed by focusing on particular scattering processes as denoted by "intra," "(π,π)," and "$(\pi,0)$." Below $T = 0.030$, SDW order occurs before SC instability.

where j runs from 1 to 4, $\boldsymbol{\sigma}$ are Pauli matrices, and N is the total number of lattice sites. This interaction is the effective one close to a SDW phase. It should not be associated with the Heisenberg-type spin interaction in the strong-coupling physics, because our model is defined in the usual Hilbert space where the double occupancy of electrons is allowed at any site. Microscopically the interaction (2) is obtained as a low-energy effective magnetic interaction generated by, for example, the repulsive Hubbard interaction by decreasing the energy scale in a functional renormalization group scheme [17,18]. The form of $V(\mathbf{k},\mathbf{k}',\mathbf{q})$ depends on the details of the high-energy fluctuations. To keep a connection with FeSC, we approximate $V(\mathbf{k},\mathbf{k}',\mathbf{q}) \approx V(\mathbf{q})$, so that a conventional SDW order can be stabilized. $V(\mathbf{q})$ should exhibit a peak at $\mathbf{q} = (\pm\pi, 0)$ and $(0, \pm\pi)$ with a negative sign to capture the stripe-type antiferromagnetic order typically observed in FeSC [19]. We consider $V(\mathbf{q}) = 2V_1(\cos q_x + \cos q_y) + 4V_2 \cos q_x \cos q_y$ with $V_2 > V_1/2 > 0$; we put $V_1 = 1$ for simplicity. In this case, the sizable interaction extends up to the second nearest-neighbor sites in real space. One may consider a different form of $V(\mathbf{q})$, but our major conclusions do not change [20].

For the interaction described by Eq. (2), the spin fluctuation propagator is computed from a bubble summation, namely,

$$\tilde{V}(\mathbf{q}, iq_m) = V(\mathbf{q}) - \frac{V(\mathbf{q})\chi_0(\mathbf{q}, iq_m)V(\mathbf{q})}{1 + V(\mathbf{q})\chi_0(\mathbf{q}, iq_m)} \tag{3}$$

and $\chi_0(\mathbf{q}, iq_m) = -\frac{T}{2N}\sum_{\mathbf{k},\sigma,n} \mathrm{Tr}\mathcal{G}_0(\mathbf{k}, ik_n)\mathcal{G}_0(\mathbf{k}+\mathbf{q}, ik_n + iq_m)$. Here $\mathcal{G}_0$ is a 2×2 matrix of the noninteracting Green's function defined for Eq. (1), ik_n (iq_m) fermionic (bosonic) Matsubara frequency, and T temperature. The first term in Eq. (3) does not depend on frequency and describes the instantaneous effect, whereas the second term accounts for the retardation effect on the pairing. A role of the instantaneous part for SC would be analyzed appropriately by including the Coulomb repulsion [21]. As a result, the superconducting tendency from the instantaneous part would be significantly suppressed. Even in this case, as we shall show below, the self-restraint effect itself is general and can occur also for the instantaneous part as long as it provides the repulsive pairing interaction. However, we believe that the dynamical effect is more important than the instantaneous effect as widely discussed for FeSC. To make the new mechanism of the self-restraint effect transparent as much as possible, we focus on dynamical spin fluctuations described by the second term in Eq. (3).

The Eliashberg gap equations involve two coupled nonlinear equations for the pairing gap $\Delta(\mathbf{k}, ik_n)$ and the renormalization function $Z(\mathbf{k}, ik_n)$. In many interesting cases, it is highly demanding to solve the Eliashberg equations numerically. Hence $Z(\mathbf{k}, ik_n)$ would be set to unity and yet computation would be limited to a temperature region much higher than T_c. To overcome these technological issues, we recall that SC instability is a phenomenon close to the FS and project the momentum on the FSs. We divide the FSs into many patches and define the Fermi momentum $\mathbf{k}_F$ on each patch. Thus $\mathbf{k}_F$ is a discrete quantity in this work. This idea allows us to achieve stable computations down to very low temperature with including the renormalization function [6] as well as a fine momentum resolution [7].

After linearizing the Eliashberg equations with respect to $\Delta(\mathbf{k}, ik_n)$, we obtain

$$\begin{aligned}\Delta(\mathbf{k}_F, ik_n)Z(\mathbf{k}_F, ik_n) \\ = -\pi T \sum_{\mathbf{k}'_F, n'} N_{\mathbf{k}'_F} \frac{\Gamma_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n)}{|k'_n|}\Delta(\mathbf{k}'_F, ik'_n),\end{aligned} \tag{4}$$

$$Z(\mathbf{k}_F, ik_n) = 1 - \pi T \sum_{\mathbf{k}'_F, n'} N_{\mathbf{k}'_F} \frac{k'_n}{k_n}\frac{\Gamma^Z_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n)}{|k'_n|}. \tag{5}$$

Here $N_{\mathbf{k}_F}$ is a momentum-resolved density of states on each FS patch and $\Gamma_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n)$ is the averaged pairing interaction over the FS patches specified by $\mathbf{k}_F$ and $\mathbf{k}'_F$:

$$\begin{aligned}\Gamma_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n) \\ = -\tfrac{1}{4}\langle W_{ab}(\mathbf{k},\mathbf{k}')^2[\tilde{V}(\mathbf{k}-\mathbf{k}', ik_n - ik'_n) \\ + 2\tilde{V}(\mathbf{k}+\mathbf{k}', ik_n + ik'_n)]\rangle_{\mathbf{k}_F\mathbf{k}'_F},\end{aligned} \tag{6}$$

where $\tilde{V}(\mathbf{k}-\mathbf{k}', ik_n - ik'_n)$ comes from longitudinal spin fluctuations and $\tilde{V}(\mathbf{k}+\mathbf{k}', ik_n + ik'_n)$ from transverse ones. The vertex part $W_{ab}(\mathbf{k},\mathbf{k}') = [U^\dagger(\mathbf{k})U(\mathbf{k}')]_{ab}$ comes

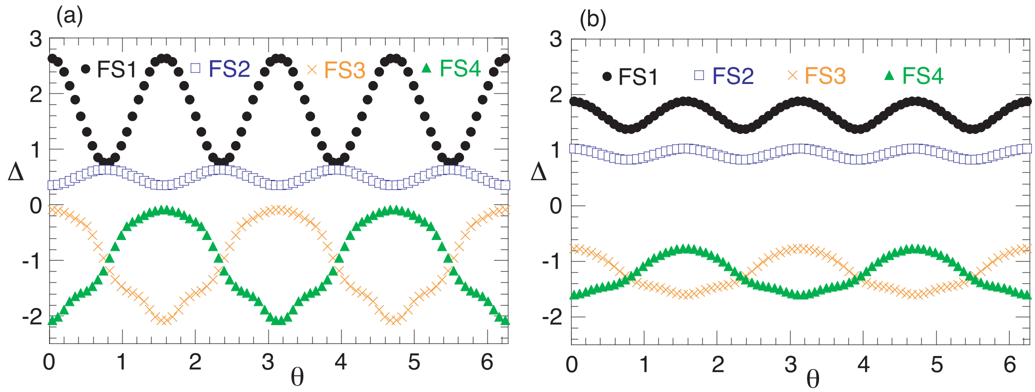

FIG. 2. Momentum dependence of the pairing gap Δ on each Fermi pocket at the lowest temperature $T = 0.03$ from "all" scattering processes (a) and "$(\pi, 0)$" scattering processes alone (b). The polar angle θ is measured from the horizontal axis on each pocket as shown in Fig. 1(a).

from the 2×2 unitary matrix diagonalizing the kinetic term Eq. (1), and a and b denote band indices. Since each band forms FSs, the indices a and b can be absorbed into the FS indices $\mathbf{k}_F$ and $\mathbf{k}'_F$. Similarly, we can compute $\Gamma^Z_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n)$ in Eq. (5) as

$$\Gamma^Z_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n) = \frac{1}{4}\langle W_{ab}(\mathbf{k}, \mathbf{k}')^2[3\tilde{V}(\mathbf{k}-\mathbf{k}', ik_n - ik'_n) - 2V(\mathbf{k}-\mathbf{k}')]\rangle_{\mathbf{k}_F\mathbf{k}'_F}. \quad (7)$$

$Z(\mathbf{k}_F, ik_n)$ is directly obtained from Eq. (5). It is then straightforward to solve the eigenvalue equation, Eq. (4), numerically. When the eigenvalue λ exceeds unity, SC instability occurs.

Since the SC instability is expected near the antiferromagnetic phase, we choose $V_2 = 1.7$, for which the stripe-type SDW order occurs below $T = 0.030$. The value of V_2 is a control parameter to tune the SDW phase in our low-energy effective model and our conclusion of the self-restraint effect does not depend on a choice of V_2.

The solid line in Fig. 1(b) shows the temperature dependence of the eigenvalue of Eq. (4). With decreasing temperature, the eigenvalue is enhanced and reaches as large as 0.6 at $T \approx 0.03$. If the temperature is decreased further, SDW instability would preempt SC instability. While the SC instability therefore does not occur in a strict sense, the eigenvalue less than unity is frequently obtained in many theoretical studies for FeSC and consistent with the literature [22–24]. Note that the eigenvalue can exceed unity if we neglect the self-energy effect (see Fig. 3).

For the FSs typical to FeSC, there are three different low-energy scattering processes, "intra," "$(\pi, 0)$," and "(π, π)," as shown in Fig. 1(a). To identify the dominant scattering process leading to the SC, we also compute the eigenvalue of the Eliashberg equation, Eq. (4), by choosing particular scattering processes. Since spin fluctuations are characterized by momenta $(\pi, 0)$ and $(0, \pi)$, it is reasonable that the eigenvalue for "$(\pi, 0)$" scattering processes becomes much larger than the other two. Our finding here is the substantial suppression of the eigenvalue from "$(\pi, 0)$" by including the intrapocket scattering processes [see the line of "intra + $(\pi,0)$" in Fig. 1(b)]. Intrapocket scattering processes are characterized by small momentum transfers and correspond to a tail of spin fluctuations with a peak around $(\pi, 0)$ and $(0, \pi)$. In fact, "intra" scattering processes alone yield the eigenvalue less than 0.1. Therefore the contribution from "intra" scattering processes seems irrelevant to SC, but Fig. 1(b) reveals that it plays a vital role to suppress the SC tendency, which is the major finding of this work.

This self-restraint effect can be understood in terms of phase frustration of the pairing gap. As is well known [25], spin fluctuations give rise to a repulsive pairing interaction and in fact $\Gamma_{\mathbf{k}_F\mathbf{k}'_F}(ik_n, ik'_n)$ in Eq. (6) is positive. In this case, the pairing gap tends to have the opposite sign between the hole and electron pockets connected by "$(\pi, 0)$" scattering processes. The resulting gap has the same sign inside each pocket. On the other hand, spin fluctuations necessarily contain "intra" scattering processes as a tail of the major antiferromagnetic fluctuations. These processes also yield a repulsive pairing interaction and thus tend to drive the sign change of the pairing gap *inside* each pocket. Therefore there occurs frustration of the phase of the pairing gap from "$(\pi, 0)$" and "intra" scattering processes. Figure 1(b) implies that this phase frustration effect is crucially important to the suppression of the eigenvalue of the Eliashberg equations even

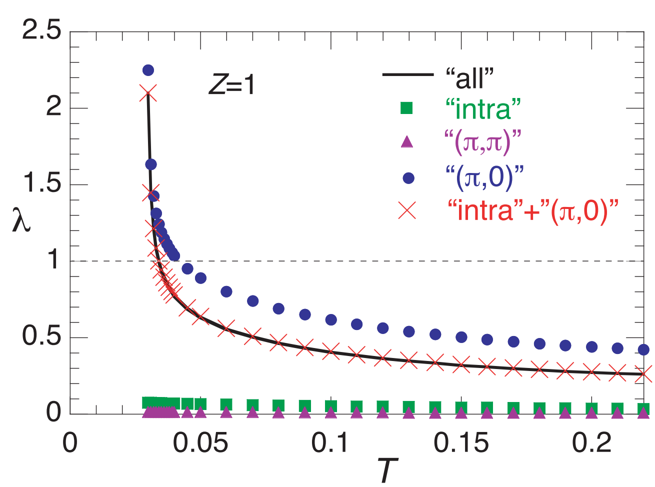

FIG. 3. Temperature dependence of the eigenvalues λ (solid line denoted by "all"). The eigenvalues are also computed by focusing on particular scattering processes as denoted by "intra," "(π, π)," and "$(\pi, 0)$." The self-energy effect is discarded by assuming $Z = 1$.

though the “intra” scattering processes alone are not effective to the SC instability itself. This self-restraint effect can be a general feature because the phase frustration is necessarily involved in the spin-fluctuation mechanism as long as it yields a repulsive pairing interaction.

While “intra” scattering processes are the major source of the self-restraint effect, “(π, π)” scattering processes also lead to the phase frustration of the SC gap. This is because they wish to have the *opposite* sign between the hole (electron) pockets whereas the major “$(\pi, 0)$” scattering processes eventually lead to the *same* sign between the hole (electron) pockets. Quantitatively, however, such a phase frustration effect is not effective compared to the “intra” processes as shown in Fig. 1(b). In fact, the eigenvalue of the Eliashberg equations is almost reproduced by considering only “intra” and “$(\pi, 0)$” scattering processes. That is, “intra” processes are much more destructive to the SC than “(π, π)” ones. For a different interaction $V(\mathbf{q})$, the contribution from “(π, π)” scattering processes can suppress SC more than Fig. 1(b), but still “intra” scattering processes play a major role of the self-restraint effect [20].

The “intra” scattering processes should not be confused with ferromagnetic fluctuations. The self-restraint effect cannot be understood in terms of the competition of, for example, singlet and triplet pairings. In fact, the static magnetic susceptibility does not show any peak around (0,0). Moreover, we checked that the eigenvector obtained from the “intra” pocket scattering processes alone is not triplet pairing.

To see how the self-restraint effect affects the momentum dependence of the pairing gap, we plot $\mathbf{k}_F$ dependence of the pairing gap in Fig. 2 (Ref. [26]). The pairing gap has the same sign in each pocket and the opposite sign between the hole (FS1 and FS2) and electron pockets (FS3 and FS4). The so-called $s_\pm$-wave symmetry is realized as expected [2,3]. The pairing gap exhibits a large $\mathbf{k}_F$ dependence on FS1, FS3, and FS4. While the gap has a fourfold symmetry on FS1 and FS2, it has a twofold symmetry on FS3 and FS3, because the FS has a twofold symmetry around $\mathbf{k} = (\pi, 0)$ and $(0, \pi)$, respectively. All these features are consistent with the literature [27]. The point here is that those gaps suffer from the self-restraint effect. The pairing gap without the self-restraint effect is obtained by considering “$(\pi, 0)$” scattering processes alone and the obtained results are shown in Fig. 2(b). A comparison with Fig. 2(a) demonstrates that the self-restraint effect causes the large $\mathbf{k}_F$ dependence of the pairing gap on FS1, FS3, and FS4 to minimize the phase frustration effect of the pairing gap although the $s_\pm$ symmetry does not change.

The self-restraint effect is different from the self-energy effect. We compute the eigenvalue of the Eliashberg equations by neglecting the self-energy effect, namely, by putting $Z = 1$. The result is shown in Fig. 3 in the same fashion as Fig. 1(b) and essentially the same results are obtained except for the absolute value of λ. The “$(\pi, 0)$” scattering processes yield the SC instability at $T = 0.042$, which is then reduced to $T = 0.034$ by adding “intra” scattering processes; the resulting eigenvalue then reproduces the eigenvalue for “all” scattering processes. The self-restraint effect reduces T_c by $(0.042 - 0.034)/0.042 = 19\%$. At $T = 0.042$, we have obtained $\lambda = 0.65$ in Fig. 1(b) for “$(\pi, 0)$” scattering processes. Hence the self-energy effect suppresses the SC tendency by $(1 - 0.65)/1 = 35\%$. That is, the suppression of the SC instability due to the self-restraint effect is comparable to that due to the self-energy effect.

Antiferromagnetic spin fluctuations are widely discussed as a possible high-T_c mechanism. While there is no doubt that spin fluctuations can drive the SC, this mechanism needs to overcome the self-restraint effect to achieve high T_c. In this sense, a favorable condition is required to realize high T_c from spin fluctuations. To reduce the self-restraint effect substantially, we would invoke an interaction term $V(\mathbf{q})$, whose magnitude becomes very small for a small momentum transfer so that the contribution from “intra” scattering processes is substantially weakened.

On the other hand, orbital fluctuations with a large momentum transfer [8,9] and nematic fluctuations [6,7] are also proposed as a possible high-T_c mechanism in FeSC. These fluctuations yield an attractive pairing interaction and thus tend to have the same sign of the pairing gap on all FSs as far as we neglect the effect of spin fluctuations [13,28]. Hence the self-restraint effect does not occur and all “intra,” “$(\pi, 0)$,” and “(π, π)” scattering processes work positively for the SC instability. In this sense, it seems easier to achieve high T_c if those fluctuations are dominant. While the electron-phonon coupling is believed to be too small to explain the T_c of FeSC [11], it is also free from the self-restraint effect as long as it yields an attractive pairing interaction.

In summary, it is tacitly assumed that antiferromagnetic spin fluctuations work positively for a SC instability. However, the present work finds that spin fluctuations have a contribution to *suppress* the SC tendency. This self-restraint effect comes from scattering processes inside the Fermi pockets with a small momentum transfer, which corresponds to a *tail* of the major antiferromagnetic spin fluctuations. We have shown that such a seemingly negligible contribution plays a remarkably important role to suppress the SC instability (Figs. 1 and 3). This effect is comparable to the suppression of SC by the self-energy effect. The self-restraint effect can be understood in terms of phase frustration of the pairing gap caused by a repulsive pairing interaction inherent in antiferromagnetic spin fluctuations. To compromise with the frustration, the system tends to have a larger $\mathbf{k}_F$ dependence of the pairing gap (Fig. 2). The self-restraint effect is general and thus expected also in other models of SC mediated by antiferromagnetic fluctuations [25].

The authors thank W. Metzner for a critical reading of the manuscript and T. Hotta, A. Katanin, and T. Kita for fruitful comments. This work was supported by JSPS KAKENHI Grants No. JP15K05189, No. JP18K18744, and No. JP20H01856.

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