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LETTER TO THE EDITOR

Spin-fluctuation-induced superconductivity controlled by orbital fluctuation

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Abstract
A microscopic Hamiltonian reflecting the correct symmetry of f orbitals is proposed for discussing superconductivity in heavy-fermion systems. In the orbitally degenerate region in which not only spin fluctuations (SF) but also orbital fluctuations (OF) show considerable development, cancellation between SF and OF destabilizes the $d_{x^2-y^2}$-wave superconductivity. On entering the non-degenerate region by increasing the crystalline electric field, $d_{x^2-y^2}$-wave superconductivity mediated by antiferromagnetic SF emerges from the suppression of OF. We argue that the present scenario can be applied to the recently discovered superconductors $CeTIn_5$ (T = Ir, Rh, and Co).

Unconventional superconductivity has been one of central issues in the research field of strongly correlated electron systems. Especially since the discovery of high-temperature superconductivity in cuprates, much effort has been focused on elucidating the mechanism of unconventional superconductivity, clarifying that a crucial role is played by antiferromagnetic (AF) 'spin fluctuations' (SF) [1]. The importance of AFSF is widely recognized, for instance, in 4f- and 5f-electron superconducting materials such as $CeCu_2Si_2$ [2] and UPd_2Al_3 [3] as well as in organic superconductors such as κ-(BEDT-TTF) [4]. Thus, it is widely believed that a broad class of unconventional superconductors originates from SF.

In d- and f-electron systems, however, the potential importance of *orbital* degrees of freedom has recently been discussed intensively. In fact, 'orbital ordering' is found to be a key issue for understanding microscopic aspects of the charge-ordered phase in colossal-magnetoresistive manganites [5]. This orbital ordering is primarily relevant to the insulating phase, while 'orbital fluctuations' (OF) should be significant in the metallic phase. Recently, the effects of OF have attracted attention, since it is hoped that a new scenario for superconductivity can be provided [6]. Especially from a conceptual viewpoint, it is important to clarify how superconductivity emerges when both SF and OF play active roles.

As a typical material for use in investigating superconductivity in a system with both SF and OF, let us introduce the recently discovered heavy-fermion superconductors $CeTIn_5$ (T = Ir, Rh, and Co) [7] with the $HoCoGa_5$-type tetragonal crystal structure. Due to this structure and to strong correlation effects, the similarity with cuprates has been emphasized. In particular, AFSF also play an essential role in the Ce-115 system, since it exhibits quasi-two-dimensional Fermi surfaces [8] and the AF phase exists next to the superconducting state in the phase diagram of $CeRh_{1-x}Ir_xIn_5$ [9]. In fact, a line node in the gap function has been observed in $CeTIn_5$ by various experimental techniques [10]. It may be true that the superconductivity itself is due to AFSF, but an important role for the OF has been overlooked in spite of the fact that both SF and OF are originally included in the ground-state multiplet of the Ce^{3+} ion. In actual materials, superconductivity occurs in a situation where OF are suppressed, since orbital degeneracy is lifted by the effect of the crystalline electric field (CEF). Thus, we envision a scenario where OF control the stability of the superconductivity even though they originate from AFSF.

In this letter, we investigate superconductivity based on the orbitally degenerate Hubbard model constructed by the tight-binding method [11]. Solving the gap equation with the pairing interaction evaluated using the random phase approximation (RPA), we obtain several superconducting phases around the spin- and orbital-ordered phases. In the orbitally degenerate region where both SF and OF are developed, it is found that singlet superconductivity is suppressed due to the competition between them, while triplet superconductivity is favoured since they are cooperative in this case. When a level splitting is included to lift the orbital degeneracy, $d_{x^2-y^2}$-wave superconductivity due to AFSF is stabilized in the vicinity of the AF phase. Thus, we claim that AFSF-induced superconductivity in Ce-115 systems is substantiated in consequence of the suppression of OF.

In order to construct the microscopic model for f-electron systems, let us start our discussion from a local basis of the Ce^{3+} ion. Among the 14-fold-degenerate 4f-electron states, due to the effect of strong spin–orbit coupling, only the $j = 5/2$ sextuplet effectively contributes to the low-energy excitations (j is total angular momentum). This sextuplet is further split into a Γ_7 doublet and a Γ_8 quadruplet due to the effect of cubic CEF, where the eigenstates are given by $|\Gamma_{7\pm}\rangle = \sqrt{1/6}|\pm5/2\rangle - \sqrt{5/6}|\mp3/2\rangle$, $|\Gamma^{(1)}_{8\pm}\rangle = \sqrt{5/6}|\pm5/2\rangle + \sqrt{1/6}|\mp3/2\rangle$, and $|\Gamma^{(2)}_{8\pm}\rangle = |\pm1/2\rangle$. Here + and − in the subscripts denote up and down 'pseudo-spins', respectively, within each Kramers doublet.

Now we discuss the relative positions of the energy levels of Γ_7 and $\Gamma_8^{(\tau)}$ while taking account of certain features of $CeTIn_5$. Since $CeTIn_5$ has a tetragonal crystal structure and a quasi-two-dimensional Fermi surface [8], it is natural to consider a two-dimensional square lattice composed of Ce ions. Due to the effect of anions surrounding the Ce ion, it is deduced that the energy level of Γ_7 becomes higher than those of the Γ_8s. Thus, in the following, the Γ_7 orbital is neglected for simplicity. Note also that we need to include the effect of the tetragonal CEF, which lifts the degeneracy of the $\Gamma_8^{(\tau)}$. Although a mixing between Γ_7 and $\Gamma_8^{(1)}$ generally occurs under a tetragonal CEF, such a mixing is expected to be small since the Γ_7 orbital has higher energy. Thus, in this situation, the effect of a tetragonal CEF can be included in terms of a level splitting ε between the $\Gamma_8^{(1)}$ and $\Gamma_8^{(2)}$ orbitals. In order to explain the magnetic anisotropy observed in experiments, $\Gamma_8^{(1)}$ must be lower than $\Gamma_8^{(2)}$, i.e., $\varepsilon > 0$ in $CeTIn_5$. The magnitude of ε for each $CeTIn_5$ compound will be discussed later. It should be noted that the above level scheme is consistent with the following two facts:

(i) Experimental results for $CeTIn_5$ exhibit a larger uniform susceptibility for magnetic field perpendicular to the $CeIn_3$ plane than for the parallel case [7, 12]. This significant anisotropy is well explained under the assumption that Γ_7 is *not* the lowest-energy state and ε is *positive*.

(ii) The band-structure calculation results suggest that the almost flat band corresponding to Γ_7 appears above the Fermi level [11].

In order to include the itinerant features of the 4f electrons, a simple way to proceed is to include nearest-neighbour hopping for the f electrons by the tight-binding method [11, 13]. Although the hybridization with the In 5p electronic states may be important, here such an effect is considered as renormalization of the effective hopping amplitude of f quasiparticles. Further, on adding the on-site Coulomb interaction terms among the f electrons, the Hamiltonian becomes

$$H = \sum_{ia\tau\tau'\sigma} t^{a}_{\tau\tau'} f^{\dagger}_{i\tau\sigma} f_{i+a\tau'\sigma} - \varepsilon \sum_{i} (n_{i1\sigma} - n_{i2\sigma})/2 + U \sum_{i\tau} n_{i\tau\uparrow} n_{i\tau\downarrow} + U' \sum_{i\sigma\sigma'} n_{i1\sigma} n_{i2\sigma'}, \quad (1)$$

where $f_{i\tau\sigma}$ is the annihilation operator for an f electron with pseudo-spin σ in the orbital $\Gamma_8^{(\tau)}$ at site $\boldsymbol{i}$, $\boldsymbol{a}$ is the vector connecting nearest-neighbour sites, and $n_{i\tau\sigma} = f^{\dagger}_{i\tau\sigma} f_{i\tau\sigma}$. The first term represents the nearest-neighbour hopping of f electrons, with the amplitude $t^{a}_{\tau\tau'}$, between $\Gamma_8^{(\tau)}$ and $\Gamma_8^{(\tau')}$ along the $\boldsymbol{a}$-direction, given by $t^{x}_{11} = -\sqrt{3}t^{x}_{12} = -\sqrt{3}t^{x}_{21} = 3t^{x}_{22} = 1$ for $\boldsymbol{a} = \boldsymbol{x}$ and $t^{y}_{11} = \sqrt{3}t^{y}_{12} = \sqrt{3}t^{y}_{21} = 3t^{y}_{22} = 1$ for $\boldsymbol{a} = \boldsymbol{y}$, respectively, in energy units where $t^{x}_{11} = 1$. Note the positive sign of the first term in H, since the M point, not the Γ point, is at the bottom of the bands forming the Fermi surfaces [11]. Note also that the present $t^{a}_{\tau\tau'}$ is just the same as that of the e_g electrons [5], but this point will be discussed elsewhere. The second term denotes the tetragonal CEF. In the third and fourth terms, U and U' are the intra- and inter-orbital Coulomb interactions, respectively. In reality, $U = U'$, since they originate from the same Coulomb interactions among f orbitals in the $j = 5/2$ multiplet, but in this letter, we also treat the case where $U \neq U'$ in order to analyse the roles of SF and OF. Since we consider quarter-filling (one f electron per site), the model in the limit of $\varepsilon = \infty$ reduces to the half-filled, single-orbital Hubbard model.

Now, in order to investigate superconductivity around the spin- and/or orbital-ordered phases, we calculate spin and orbital susceptibilities, $\hat{\chi}^{s}(\boldsymbol{q})$ and $\hat{\chi}^{o}(\boldsymbol{q})$, respectively. Within the RPA, these are given in matrix form as

$$\hat{\chi}^{s}(\boldsymbol{q}) = [\hat{1} - \hat{U}^{s}\hat{\chi}(\boldsymbol{q})]^{-1}\hat{\chi}(\boldsymbol{q}), \quad (2)$$

$$\hat{\chi}^{o}(\boldsymbol{q}) = [\hat{1} + \hat{U}^{o}\hat{\chi}(\boldsymbol{q})]^{-1}\hat{\chi}(\boldsymbol{q}), \quad (3)$$

where labels of rows and columns in the matrix appear in the order 11, 22, 12, and 21, these being pairs of orbital indices 1 and 2. Note that $\hat{1}$ is the 4×4 unit matrix. $\hat{U}^{s}$ is given by $U^{s}_{11,11} = U^{s}_{22,22} = U$, $U^{s}_{12,12} = U^{s}_{21,21} = U'$, otherwise zero, while $\hat{U}^{o}$ is expressed as $U^{o}_{11,11} = U^{o}_{22,22} = U$, $U^{o}_{11,22} = U^{o}_{22,11} = 2U'$, $U^{o}_{12,12} = U^{o}_{21,21} = -U'$, otherwise zero. The matrix elements of $\hat{\chi}(\boldsymbol{q})$ are defined by $\chi_{\mu\nu,\alpha\beta}(\boldsymbol{q}) = -T\sum_{\boldsymbol{k},n} G^{(0)}_{\alpha\mu}(\boldsymbol{k}+\boldsymbol{q}, \mathrm{i}\omega_n) G^{(0)}_{\nu\beta}(\boldsymbol{k}, \mathrm{i}\omega_n)$, where T is temperature, $G^{(0)}_{\mu\nu}(\boldsymbol{k}, \mathrm{i}\omega_n)$ is the non-interacting Green function for f electrons with momentum $\boldsymbol{k}$ propagating between μ- and ν-orbitals, and $\omega_n = \pi T(2n+1)$ with integer n. The instabilities for the spin- and orbital-ordered phases are determined by the conditions $\det[\hat{1} - \hat{U}^{s}\hat{\chi}(\boldsymbol{q})] = 0$ and $\det[\hat{1} + \hat{U}^{o}\hat{\chi}(\boldsymbol{q})] = 0$, respectively.

By using $\hat{\chi}^{s}(\boldsymbol{q})$ and $\hat{\chi}^{o}(\boldsymbol{q})$, the superconducting gap equation is given as

$$\boldsymbol{\Delta}^{\xi}(\boldsymbol{k}) = \sum_{\boldsymbol{k}'} \hat{V}^{\xi}(\boldsymbol{k}-\boldsymbol{k}')\hat{\phi}(\boldsymbol{k}')\boldsymbol{\Delta}^{\xi}(\boldsymbol{k}'), \quad (4)$$

where $\boldsymbol{\Delta}^{\xi}(\boldsymbol{k}) = [\Delta^{\xi}_{11}(\boldsymbol{k}), \Delta^{\xi}_{22}(\boldsymbol{k}), \Delta^{\xi}_{12}(\boldsymbol{k}), \Delta^{\xi}_{21}(\boldsymbol{k})]^{t}$ is the gap function in the vector representation for a singlet ($\xi = \mathrm{S}$) or triplet ($\xi = \mathrm{T}$) pairing state, and the matrix elements of the singlet and triplet pairing potentials, respectively, are given by

$$V^{\mathrm{S}}_{\alpha\beta,\mu\nu}(\boldsymbol{q}) = [(-3/2)\hat{W}^{s}(\boldsymbol{q}) + (1/2)\hat{W}^{o}(\boldsymbol{q}) + \hat{U}^{s}]_{\alpha\mu,\nu\beta}, \quad (5)$$

$$V^{\mathrm{T}}_{\alpha\beta,\mu\nu}(\boldsymbol{q}) = [(1/2)\hat{W}^{s}(\boldsymbol{q}) + (1/2)\hat{W}^{o}(\boldsymbol{q}) - \hat{U}^{s}]_{\alpha\mu,\nu\beta}. \quad (6)$$

Here, the spin and orbital susceptibilities are included as $\hat{W}^{s}(q) = \hat{U}^{s} + \hat{U}^{s}\hat{\chi}^{s}(q)\hat{U}^{s}$ and $\hat{W}^{o}(q) = -\hat{U}^{o} + \hat{U}^{o}\hat{\chi}^{o}(q)\hat{U}^{o}$. The elements of the pair correlation function $\hat{\phi}(k)$ are given by $\phi_{\alpha\beta,\mu\nu}(k) = T\sum_n G^{(0)}_{\alpha\mu}(k, -i\omega_n)G^{(0)}_{\nu\beta}(-k, i\omega_n)$. The superconducting transition is obtained for each respective irreducible representation by solving equation (4), where the maximum eigenvalue becomes unity.

Here we mention the essential symmetries of the Cooper pairs in multi-orbital systems. The pairing states are classified into four types owing to spin $SU(2)$ symmetry and space inversion symmetry:

(1) spin singlet and orbitally symmetric with even parity,
(2) spin triplet and orbitally symmetric with odd parity,
(3) spin singlet and orbitally antisymmetric with odd parity, and
(4) spin triplet and orbitally antisymmetric with even parity.

Note that $SU(2)$ symmetry does not exist in orbital space, since the lattice and the local wavefunctions rotate simultaneously. For (1) and (2), f electrons in the same band form the Cooper pair, while for (3) and (4), such a pair is formed only between different bands. Except for the special case in which two Fermi surfaces connect with each other, orbitally antisymmetric pairing is unstable due to depairing effects destroying inter-band pairs. In fact, even after careful calculations, we do not find any region for (3) and (4) in the parameter space considered here. Thus, in the following, we discuss only the orbitally symmetric pair.

Let us discuss first the effect of ε on the correlation between SF and OF. Since we are considering a realistic situation corresponding to $CeTIn_5$, we restrict ourselves to the case of $U = U'$ for a while. In figure 1, the principal components of $\hat{\chi}^{s}(q)$ and $\hat{\chi}^{o}(q)$ are shown in q-space for $U = U' = 0.9U_m$, where U_m denotes spin instability determined from $\det[\hat{1} - \hat{U}^{s}\hat{\chi}(q)] = 0$. In the orbitally degenerate case with $\varepsilon = 0$, as shown in figures 1(a) and (b), the overall magnitude of OF is comparable with that of SF. As for the q-dependence, common structures are found in $\chi^{s}(q)$ and $\chi^{o}(q)$. Namely, there are two peaks around $(\pi/2, \pi/2)$ and $(\pi/2, 0)$ owing to the nesting properties of the Fermi surface (see the inset). Thus, the orbitally degenerate region is characterized by competition between SF and OF. With increasing ε, the Fermi surface approaches a shape having the nesting vector (π, π) (see the inset). For $\varepsilon = 4$, as shown in figures 1(c) and (d), OF are almost completely suppressed, and SF around (π, π) become dominant. Note here that the increase of ε makes the lower energy state favourable and suppresses excitations to the upper energy state, indicating the suppression of OF. Thus, even at this stage, it is understood that the suppression of OF leads to the development of SF for the paramagnetic system which is wavering between spin and orbital ordering.

Next, we consider how a superconducting phase emerges when ε is increased. In figure 2(a), the maximum eigenvalues for several irreducible representations are depicted as a function of ε for $U = U' = 2.5$. The calculations are carried out for a fixed $T = 0.02$, and the first Brillouin zone is divided into 128×128 meshes. In the orbitally degenerate region, several eigenvalues are very close to each other, owing to multi-peak structures in $\hat{\chi}^{s}(q)$ and $\hat{\chi}^{o}(q)$. With increasing ε, as is easily understood from the growth of AFSF mentioned above, the eigenvalue for B_{1g} symmetry becomes dominant and finally, at $\varepsilon \approx 3$, the B_{1g} superconducting phase is stabilized. On further increasing ε, the AF instability eventually occurs, since the system asymptotically approaches the half-filled single-orbital Hubbard model. We emphasize that the increase of ε brings about transitions successively, in the order of PM, B_{1g} superconducting, and AF phases.

On the basis of the above calculated results, let us try to explain the differences among three Ce-115 compounds, $CeRhIn_5$ (Néel temperature $T_N = 3.8$ K), $CeIrIn_5$ (superconducting

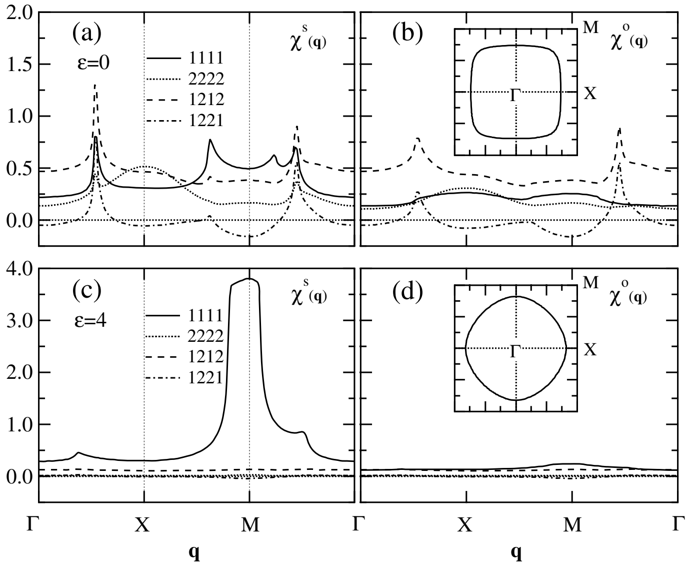

Figure 1. (a) Spin and (b) orbital susceptibilities in $\boldsymbol{q}$-space for $\varepsilon = 0$ and $U = U' = 0.9U_{\rm m}$. (c) and (d) are for $\varepsilon = 4$. Insets indicate the Fermi-surface lines.

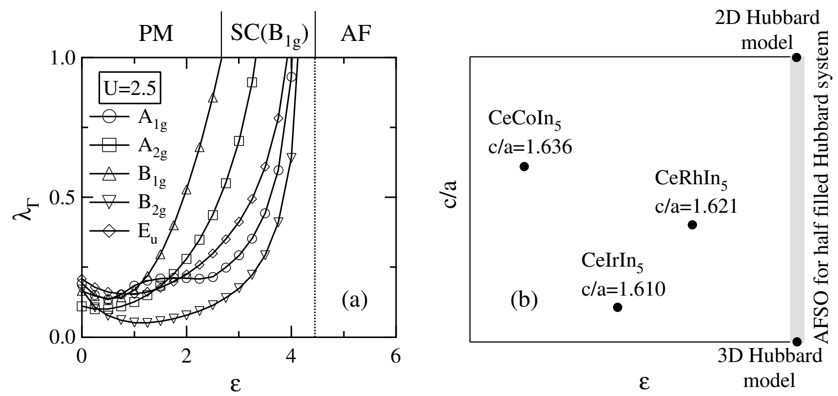

Figure 2. (a) Maximum eigenvalue versus ε for the respective irreducible representation at $U = U' = 2.5$. (b) A schematic plot of c/a versus ε to illustrate the comparison between our theory and actual $CeTIn_5$ compounds.

transition temperature $T_c = 0.4$ K), and $CeCoIn_5$ ($T_c = 2.3$ K) [7]. One property which distinguishes these compounds is the two dimensionality, as expressed by the ratio c/a between lattice constants. Two dimensionality is naturally expected to become stronger in the order $CeIrIn_5$, $CeRhIn_5$, and $CeCoIn_5$ [7]. Since the magnetic state should be stabilized with increasing three dimensionality, $CeIrIn_5$ should be the most favourable to the occurrence of antiferromagnetism among three compounds, but this is obviously inconsistent with experimental results. This inconsistency is resolved by introducing another important ingredient, ε. One can show that the increase of magnetic anisotropy just corresponds to the increase of ε. Analysis of experimental results for the anisotropy of magnetic susceptibilities [7, 12] leads to the conclusion that ε becomes larger in the order $CeCoIn_5$, $CeIrIn_5$, and $CeRhIn_5$. Taking into account the effect of ε in addition to dimensionality,

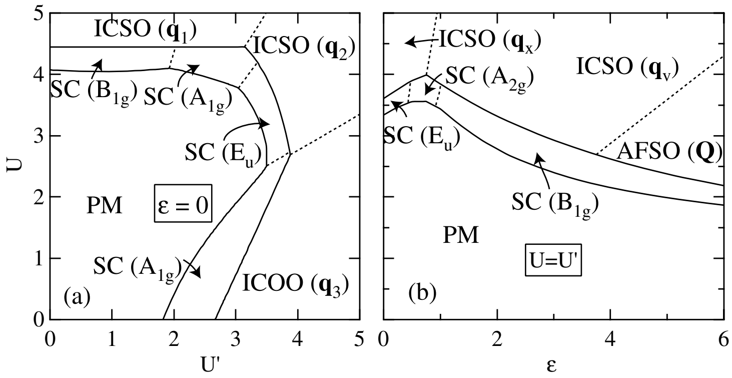

Figure 3. Phase diagrams (a) in the U–U' plane for $\varepsilon = 0$ and (b) in the U–ε plane for $U = U'$. The meanings of the abbreviations used here are as follows: SC(Γ) denotes the superconducting phase with Γ symmetry, ICSO(q) indicates the incommensurate spin-ordered phase with wavevector q, ICOO(q) is the incommensurate orbital-ordered phase with wavevector q, and AFSO(Q) denotes the AF spin-ordered phase. Several wavevectors are defined: $q_1 = (0.56\pi, 0.56\pi)$, $q_2 = (0.53\pi, 0)$, $q_3 = (\pi, 0)$, $q_x = (q_x, 0)$, $q_y = (\pi, q_y)$, and $Q = (\pi, \pi)$. Solid curves show the phase boundaries determined by actual calculations, while dotted lines are schematic phase boundary guides for the eye.

we arrive at the picture shown schematically in figure 2(b). Due to the enhancement of AFSF induced by increasing ε, as shown in figure 1, it may be understood that $CeRhIn_5$ rather than $CeIrIn_5$ is more favourable to antiferromagnetism. That is, it is considered that $CeRhIn_5$ with the largest ε is antiferromagnetic, while $CeCoIn_5$ and $CeIrIn_5$ with smaller ε than $CeRhIn_5$ exhibit superconductivity. The difference in T_c between $CeCoIn_5$ and $CeIrIn_5$ may be attributed to the extent of the two dimensionality. Concerning the superconductivity and antiferromagnetism of $CeTIn_5$ compounds, the combination of two dimensionality and the crystalline-field splitting is necessary to reach a consistent picture.

The orbitally degenerate model, equation (1), shows new, rich superconducting properties both for singlet and for triplet pairings. In figure 3(a) and (b), we show the phase diagrams in the U–U' plane for $\varepsilon = 0$ and in the U–ε plane for $U = U'$, respectively. In these figures, four characteristic superconducting phases are observed:

(1) a SF-mediated spin-singlet superconducting phase with B_{1g} symmetry in the vicinity of a spin-ordered phase;
(2) a g-wave superconducting phase with A_{2g} symmetry;
(3) a spin-triplet superconducting phase with E_u symmetry due to the cooperation between SF and OF around $U = U'$ and $\varepsilon = 0$; and
(4) an OF-mediated spin-singlet superconducting phase with A_{1g} symmetry for $U \ll U'$ around an orbital-ordered phase.

As regards (1), it is thought that B_{1g} superconductivity is induced by SF around $q = Q = (\pi, \pi)$. Concerning the triplet superconductivity (3), note that for this region all superconducting instabilities are quite close to each other because of the multi-peak structures in $\hat{\chi}^s(q)$ and $\hat{\chi}^o(q)$, as shown in figure 1. We also note that the factors in front of the SF term in $\hat{W}^s(q)$ are $-3/2$ for singlet and $+1/2$ for triplet pairing, while the factor for the OF term in $\hat{W}^o(q)$ is $+1/2$ for both pairings. That is, OF are cooperative with SF for spin-triplet pairing, while SF and OF compete with each other for spin-singlet pairing [6]. Thus, the spin-triplet pairing phase appears in the region with $\varepsilon \approx 0$ and $U \approx U'$. The mechanism of the

OF-mediated singlet superconductivity (4) is interesting. In this state, two quasi-particles with different pseudo-spins form an on-site intra-orbital singlet pair since $U' \gg U$, leading to A_{1g} superconductivity. In short, singlet superconductivity with B_{1g} or A_{1g} symmetry is stabilized when either SF or OF are dominant, while triplet superconductivity is favoured when there is cooperation between SF and OF.

In summary, we have studied superconductivity in the Ce-115 systems on the basis of the orbitally degenerate Hubbard model, and found that on increasing the tetragonal CEF splitting, $d_{x^2-y^2}$-wave superconductivity due to AFSF emerges out of the suppression of OF in the vicinity of the AF phase. The concept of AFSF-induced superconductivity controlled by OF qualitatively explains the differences among Ce-115 materials.

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