Magnetic fluctuations and superconducting pairing in $\varepsilon$-iron

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We study Coulomb correlation effects and their role in superconductivity of $\varepsilon$-iron under pressure from 12 to 33 GPa by using a combination of density functional and dynamical mean-field theory. Our results indicate a persistence of the Fermi-liquid behavior below the temperature $\sim 1000$ K. The Coulomb correlations are found to substantially renormalize the density of states, reducing the distance from the peak to the Fermi level to 0.4 eV compared to 0.75 eV obtained in DFT calculations. We find significant antiferromagnetic correlations, which are accompanied by the formation of short-lived local magnetic moments. We use the obtained results as a starting point for construction of the multi-band Bethe-Salpeter equation, which eigenvalues indicate that antiferromagnetic spin fluctuations may result in the superconducting pairing in $\varepsilon$-Fe. Moreover, the tendency to superconducting instability becomes weaker with increase of pressure, that may explain the disappearance of superconductivity at $\sim 30$ GPa.

I. INTRODUCTION

Since the discovery of the hexagonal close-packed (hcp) iron ($\varepsilon$ phase) in 1964 [1], its puzzling properties have attracted significant interest. This phase appears at pressures above 12 GPa and, as shown by static compression experiments, is stable up to pressures and temperatures corresponding to Earth’s core conditions [2]. It makes the investigation of $\varepsilon$-Fe particularly relevant for geophysics and Earth’s magnetism [2–4].

One of the actively debated characteristics of $\varepsilon$-Fe is its magnetic behavior. The Mössbauer effect measurements found no hyperfine magnetic field at temperatures down to 30 mK [5–8], implying that hcp iron is non-magnetic. The static magnetic order was not also detected by x-ray magnetic circular dichroism (XMCD) [9] and neutron powder diffraction [10]. The anomalous mode splitting in the Raman spectra [11], interpreted initially as a hint of the presence of magnetic order, was later shown to be inconsistent with it [12]. At the same time, the x-ray emission spectroscopy (XES) consistently detected remnant magnetic, decreasing with pressure up to $\sim 35$ GPa [10, 13, 14].

The presence of magnetic correlations in $\varepsilon$-Fe is supported by density functional theory (DFT) studies. Most of early calculations predicted the antiferromagnetic type-II (AFM-II) ground state, which is stable up to $\sim 50$ GPa [15–18], or the spin-spiral ground state [19, 20]. It is worth noting that AFM-II structure leads to a better agreement of structural and elastic properties with experiments, as compared to non-magnetic calculations [15, 16]. Nevertheless, recent DFT study of Lebert et al. [21] revealed three intensity-modulated phases with a lower energy than AFM-II state [10]. In this study, the ground state at pressures 15–35 GPa was found to be formed by alternating AFM and non-magnetic bilayers.

At the same time, the paramagnetic state of $\varepsilon$-Fe was studied by a combination of DFT and dynamical mean-field theory (DFT+DMFT) [21]. These studies showed the importance of Coulomb correlations for description of the electronic [22, 23] and structural [24] properties at moderate pressures, as well as phase stability [25, 26], transport [27] and magnetic [23, 27] properties at Earth’s core conditions. In addition, a jump of resistivity at the $\alpha \rightarrow \varepsilon$ transition was qualitatively captured by Pourovskii et al. [23], while quantitative difference was attributed to non-local correlation effects (for a review, see [28]).

The properties of $\varepsilon$-Fe have become even more intriguing with the discovery of superconductivity in the pressure range from 15 to 30 GPa, with the largest critical temperature $T_c = 2$ K at about 21 GPa [29]. The superconducting properties are also unusual. In particular, the superconductivity seems to occur in paramagnetic phase; the large slope of the critical field curve at $T_c$ demonstrates that heavy particles are involved in the pairing (see [30] and review in Ref. [31]). Moreover, the temperature dependence of resistivity shows a $T^{5/3}$ power law behavior below 30 K over the entire superconducting pressure region [30, 32, 33]. This non-Fermi-liquid behavior (in contrast with the $T^2$ dependence for Fermi liquid) was attributed to ferromagnetic (FM) fluctuations according to Moriya’s theory of weak itinerant magnetism [34]. These experimental results point to an unconventional superconductivity; the presence of FM fluctuations may be a signature of triplet pairing symmetry. However, it is at odds with the DFT calculations, predicting a proximity to antiferromagnetism, which would likely yield singlet superconducting state.

A narrow pressure range of superconductivity also suggests that it has a spin-fluctuation origin. This was confirmed by calculations of the electron-phonon coupling within DFT, which showed that the phonon mechanism can explain the appearance of superconductivity in $\varepsilon$-Fe, but not its rapid disappearance with pressure [38, 39]. In particular, the calculated range of superconductivity was found to extend well above 100 GPa. To consider the magnetic pairing mechanism, Jarlborg estimated the coupling constant within DFT and concluded that superconductivity mediated by spin fluctuations is more likely than that owing to electron-phonon interaction [10].
Above discussed observations stress the importance of studying correlation effects in \(\varepsilon\)-iron. In this paper we study the electronic and magnetic properties of paramagnetic \(\varepsilon\)-Fe within superconducting pressure range by employing DFT+DMFT approach. We find considerable local correlations, leading to formation of short-lived local magnetic moments, as well as pronounced incommensurate spin fluctuations. To clarify whether these fluctuations can mediate the electron pairing, we derive and solve the multi-band Bethe-Salpeter equation (BSE) in the particle-particle channel. The obtained results indicate that the spin fluctuations in \(\varepsilon\)-Fe may lead to superconducting instability in the experimental pressure range.

The plan of the paper is the following. In Sect. II we list the computational details. In Sect. III we present the results on electronic, magnetic and superconducting properties, and finally in Sect. IV the conclusions are given. In Appendix we derive the gap equation and pairing interaction.

II. COMPUTATIONAL DETAILS

We have performed DFT calculations of \(\varepsilon\)-iron using the full-potential linearized augmented-plane wave method implemented in the ELK code supplemented by the Wannier function projection procedure. The Perdew-Burke-Ernzerhof form of generalized gradient approximation (GGA) was considered. The calculations were carried out with the experimental lattice constants at the corresponding pressures \[11\]. The convergence threshold for total energy was set to \(10^{-6}\) Ry. The integration in the reciprocal space was performed using \(22 \times 22 \times 12\) \(k\)-point mesh in all calculations except those of the Fermi surface and momentum-dependent susceptibility, where \(42 \times 42 \times 22\) mesh was employed. From converged DFT results we have constructed effective Hamiltonians in the basis of Wannier functions, which were built as a projection of the original Kohn-Sham states to site-centered localized functions as described in Ref. \[42\] considering \(3d\), \(4s\) and \(4p\) states.

In DMFT calculations we use for \(d\)-states the Hubbard parameter \(U \equiv F^0 = 4.3\) eV and Hund’s rule coupling \(J = 1.0\) eV, which were used in previous study of \(\varepsilon\)-Fe and resulted in accurate descriptions of its structural properties at the same pressures as in current work \[23\]. We note that these values are in overall agreement with estimates for \(\alpha\)-iron \[14\]. Our calculations have been performed using the AMULET code \[11\]. The double-counting correction has been taken in the around mean-field form. We also verified that the fully localized double-counting correction leads to similar results, although providing a slightly larger (\(\sim 0.1\)) filling of \(d\) states. The impurity problem has been solved by the hybridization expansion continuous-time quantum Monte Carlo method \[15\] with the density-density form of Coulomb interaction.

![FIG. 1: (a) Imaginary part of self-energy for degenerate \(xy\) and \(x^2-y^2\) states as a function of imaginary frequency \(i\nu\) obtained by DFT+DMFT method at \(\beta = 100\) eV\(^{-1}\). Inset: temperature dependence of the ratio of quasiparticle damping \(\Gamma\) to temperature \(T\). (b) Imaginary part of the above mentioned self-energy at the first Matsubara frequency as a function of temperature. Inset: the same for \(U = 6\) eV. The straight lines depict the fit to linear dependence.

We checked that the full charge self-consistency in DFT+DMFT scheme does not qualitatively affect the results, and neglected it in our calculations.

III. RESULTS AND DISCUSSION

A. Electronic properties

We find the filling of various \(d\)-orbitals in the range \(n = 1.30 \pm 1.36\), which is close to the values in \(\alpha\)- and \(\gamma\)-iron \[10\[16\], but the peak of the density of states is sufficiently far from the Fermi level (see below). We first analyze possible deviations from the Fermi-liquid (FL) regime by using several criteria employed earlier at Earth’s core conditions \[8\[24\]. For brevity, we present the results only for degenerate \(xy\) and \(x^2-y^2\) states, while similar results were obtained for \(z^2\) and degenerate \(xz\) and \(yz\) states.

In a FL state, the imaginary part of self-energy depends linearly on imaginary frequency \(i\nu\) at small \(|\nu\) as \(\text{Im } \Sigma(i\nu) \approx -\Gamma - (Z^{-1} - 1)|\nu|\), where \(\Gamma\) is the quasiparticle damping (i.e., inverse quasi-particle lifetime), \(Z\) is the quasiparticle residue. In Fig. 1(a) we show the obtained
imaginary part of $\Sigma(i\nu_n)$, which scales almost linearly for all considered pressures. To obtain a more quantitative estimate, we compute the quasiparticle damping $\Gamma$, which in a FL depends quadratically on temperature, implying $\Gamma/T \propto T$. To this end, we perform the analytical continuation of self-energies by using the Padé approximants [49]. The obtained temperature dependence of $\Gamma/T$, shown in the inset of Fig. 1(a), also corresponds to the coherent FL regime.

To obtain an accurate estimate of the temperature $T^*$, below which the crossover to the FL regime occurs, avoiding use of Padé approximants, we consider the “first Matsubara frequency” rule [50], according to which in the local approximation $\text{Im} \Sigma$ at first Matsubara frequency $\nu_1 = \pi T$ does not contain any terms beyond $O(T)$ in the FL regime. As shown in Fig. 1(b), $\text{Im} \Sigma(i\pi T)$ is proportional to temperature below $T^*$ which is $\sim 1500$ K at 12 GPa. A gradual increase of $T^*$ with pressure reflects the reduction of the Coulomb correlation strength due to larger bandwidth. These results suggest a possibility of suppression of coherence temperature $T^*$ for larger Hubbard $U$. However, our calculations with $U = 6$ eV lead to only a weak change of $T^*$, which is $\sim 1000$ K at pressure of 12 GPa (see inset in Fig. 1(b)). The value of $T^*$ can be also affected by using the full rotationally-invariant Coulomb interaction, while the density-density approximation was shown to underestimate the magnitude of scattering $|\text{Im} \Sigma(i\nu_n)|$ in $\varepsilon$-Fe [28]. The FL behavior of $\varepsilon$-Fe was also predicted by Pourovskii et al. at Earth’s core conditions [24, 29], even at the relatively high temperature of $\sim 6000$ K, while a contrary conclusion was obtained by Zhang et al. [20].

The Coulomb correlations in $\varepsilon$-Fe result in substantial mass enhancement $m^*/m = Z^{-1}$, which, averaged over all $d$ orbitals, is equal to 1.8 at 12 GPa and gradually decreases to 1.55 at 33 GPa in agreement with earlier studies [28]. The enhancement of the quasiparticle mass is accompanied by the renormalization of the density of states (DOS) near the Fermi level as displayed in Fig. 2. As a result, the distance from the peak lying above the Fermi level is decreased to 0.4 eV compared to 0.75 eV obtained in DFT calculations at 12 GPa. The DOS at the Fermi level decreases with pressure, but not rapidly enough to explain the disappearance of superconductivity, as was also noted previously [38].

### B. Magnetic properties

In Fig. 3(a) we present the local spin-spin correlation functions $\langle S_z(\tau)S_z(0) \rangle$ at $T = 290$ K, which have a significant instantaneous average $\langle S_z^2 \rangle \simeq 1$, and decay rapidly with imaginary time $\tau$. This $\tau$-dependence corresponds to fast quantum fluctuations of instantaneous magnetic moments on lattice sites. To estimate the degree of spin localization, we Fourier transform the spin correlator to imaginary bosonic frequencies, $\chi_{\text{loc}}(i\omega_n) = \int_0^\beta \exp(i\omega_n \tau) \langle S_z(\tau)S_z(0) \rangle$, and then analytically continue it to real frequency $\omega$. The real part of the obtained $\chi_{\text{loc}}(\omega)$ is shown in Fig. 3(b) in comparison with that of $\alpha$-iron [51] (see also Refs. [46, 50]), being a system with well-defined local moments. The half width of the peak in $\text{Re} \chi_{\text{loc}}(\omega)$ at half of its height yields approximately inverse lifetime of local magnetic moments [47].

For $\varepsilon$-iron we therefore obtain the inverse lifetime in energy units ranging from 0.3 eV at $p = 12$ GPa to 0.5 eV at

![FIG. 2: Density of states of $\varepsilon$-Fe obtained within DFT and DFT+DMFT at $T = 290$ K for different pressures. The Fermi level is at zero energy.](image)

![FIG. 3: Local spin-spin correlation functions in the imaginary-time (a) and real energy (b) domains calculated by DFT+DMFT method at $\beta = 40$ eV$^{-1}$. In the bottom panel, the obtained correlation functions are compared with that for $\alpha$-Fe [51]. Inset: instantaneous average $\langle S_z^2 \rangle$ for $\varepsilon$-Fe as a function of pressure.](image)
at $p = 33$ GPa, which corresponds to the lifetime of local moments ranging from 14 fs to 8 fs, respectively. This is comparable to the lifetime of local moments, discussed previously in pnictides [52]. Thus, the obtained results show a presence of short-lived local magnetic moments, which lifetime decreases with increasing pressure in agreement with XES measurements [10, 13, 14].

To get further insight into the formation of local moments, we calculate the uniform magnetic susceptibility as a response to a small external magnetic field introduced in the DMFT part (see Fig. 3(a)). In particular, we have used the magnetic field corresponding to splitting of the single-electron energies by 0.02 eV, which was checked to provide a linear response. The calculated uniform susceptibility, shown in Fig. 4(a), increases almost linearly with temperature at low temperatures and decreases above a certain temperature $\sim 3000$ K, determined by the distance from the peak of the density of states to the Fermi level, similarly to other systems with a peak of the density of states shifted off the Fermi level [47, 53].

Increasing value of the density of states shifted off the Fermi level, similarly to other systems with a peak of the density of states, which lifetime decreases with increasing pressure [54], the density of states shifted off the Fermi level [47, 53].

The static local spin susceptibility $\chi_{\text{loc}} = 4\mu_B^2\chi_{\text{loc}}(0)$ shows a Pauli-like behavior below the temperature $T^*$ of a crossover to the Fermi liquid regime and a Curie-Weiss behavior $\chi_{\text{loc}} \propto (T - T_{\text{loc}})^{-1}$ above $T^*$ due to presence of the unscreened local moments (see Fig. 3(b)). The “Weiss” temperature $T_{\text{loc}}$ is negative and determines the Kondo temperature $T_K \sim -T_{\text{loc}}$ below which the local moments are screened by conduction electrons, similarly to the single-impurity Kondo model [54], for which $\chi_{\text{loc}} \propto |T_{\text{loc}}|$. In particular, we use the magnetic field corresponding to splitting of the single-electron energies by 0.02 eV, which was checked to provide a linear response. The calculated uniform susceptibility $\chi_{\text{loc}} = 4\mu_B^2\chi_{\text{loc}}(0)$ shows a Pauli-like behavior below the temperature $T^*$ of a crossover to the Fermi liquid regime and a Curie-Weiss behavior $\chi_{\text{loc}} \propto (T - T_{\text{loc}})^{-1}$ above $T^*$ due to presence of the unscreened local moments (see Fig. 3(b)).

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equation

$$\lambda_{s,t}^{\alpha} f_{k,\alpha}^{s,t} = - \sum_{k',i\nu_n,\beta} V_{kk',\alpha\beta}^{s,t} f_{k',\beta}^{s,t} G_{k\beta}(i\nu_n) G_{k\beta}(-i\nu_n)$$  \hspace{1cm} (2)$$

where $G_{k\alpha}(i\nu_n)$ is the Green function for the band $\alpha$, obtained by the diagonalization of the functions $G_{k}(i\nu_n)$ with respect to orbital indexes. We parameterize the gap functions $f_{k,\alpha}$ for momenta lying on the Fermi surface and assume them to be independent on the radial direction of $k$, since the summation in Eq. (2) is anyhow dominated by the vicinity of the Fermi surface. Among many sheets of the Fermi surface (see Fig. 6) we choose the two most important (Figs. 6(a,b)), which parts are connected by nesting vectors. We parameterize points of these Fermi surface sheets by the polar and azimuthal angles $(\theta, \phi)$, where $\phi = 0$ corresponds to point M of the Brillouin zone, $\theta = 0, \pi$ to the “North” and “South” poles.

Calculated leading eigenvalues of singlet and triplet pairing are shown in Fig. 7. For the considered range of pressures, singlet and triplet eigenvalues are close to each other. Interestingly, we obtain crossing of eigenvalues of singlet and triplet pairing near the pressure $p_c = 22$ GPa, close to that, at which maximum value of superconducting $T_c$ is obtained. Yet, both eigenvalues increase with decreasing pressure, so that the obtained pressure dependence agrees with the experimental data for $p > p_c$. For $p < p_c$ the considered theory does not describe the suppression of superconductivity with decreasing pressure. The increase of the eigenvalues with decreasing pressure, however, goes along with the increase of the non-uniform susceptibility, see Fig. 5 and shows the spin-fluctuation origin of the obtained superconducting order parameters. We note that the obtained close competition of singlet and triplet pairing is similar to that discussed previously for Sr$_2$RuO$_4$ (see, e.g., Refs. [57, 60]) and may be lifted by going beyond second-order perturbation theory, including nearest-neighbor Coulomb repulsion, spin-orbit coupling etc.

Obtained momentum dependence of the gap functions for singlet order parameters (Fig. 8(a,b)) take the values $\approx 0, \pm \Delta$ at the subsequent vertical faces of the considered Fermi surface sheets. This momentum dependence allows one to achieve a change of the sign of gap function on the opposite sides of the Fermi surface, connected by the wave vector of preferred incommensurate fluctuations in the K-M direction. On the contrary, the triplet pairing gap is almost $\phi$-independent and changes sign at $\theta = \pi/2$, having a single line of the nodes.

**IV. CONCLUSIONS**

We have studied electronic and magnetic properties of $\varepsilon$-Fe at pressures up to 33 GPa by employing the DFT+DMFT approach and using its results as a starting point for multi-band Bethe-Salpeter equation. Our main results are as follows.

(i) We have not found a significant deviation from the
FIG. 8: Eigenfunction of the Bethe-Salpeter equation for singlet (a,b) and triplet (c,d) pairing as a function of polar $\theta$ and azimuthal $\phi$ angles calculated at pressure of 21 GPa and $\beta = 100$ eV$^{-1}$. Panels (a,c) and (b,d) correspond to sheets of Fermi surface depicted in Figs. 6(a) and 6(b), respectively. Angle $\phi = 0$ corresponds to point M of the Brillouin zone.

FL behavior below 1000 K even with relatively large $U = 6$ eV. This points to the importance of non-local correlations effects, neglected in DMFT.

(ii) At the same time, the Coulomb correlations lead to substantial electron mass enhancement up to $m^*/m \sim 1.8$, which is accompanied by renormalization of the density of states. As a result, the distance from the peak lying above the Fermi level is decreased to 0.4 eV compared to 0.75 eV obtained in DFT calculations.

(iii) We have found short-lived magnetic moments with the lifetime $\sim 10$ fs, comparable to that estimated previously in pnictides. This may explain the discrepancy of XES experiments with the Mössbauer spectroscopy because of the longer time scales of the latter. Namely, the lifetime of local magnetic moments or spin fluctuations can be small enough to be undetected by the Mössbauer measurements.

(iv) We find that the antiferromagnetic spin fluctuations are the dominant ones. At the same time, we do not find substantial ferromagnetic correlations responsible for the $T^{5/3}$ dependence of the resistivity. This indicates that the non-local correlation effects beyond DMFT may play a significant role in $\varepsilon$-Fe. In particular, non-local correlation effects corresponding to virtual transitions into a peak lying near the Fermi level may give a contribution. For this contribution the peak needs not to lie directly at $E_F$; moreover, its distance from $E_F$ needs not to be too small.

(v) The antiferromagnetic spin fluctuations in $\varepsilon$-Fe can result in the superconducting instability. The obtained results allow us to explain the appearance of superconducting instability in the considered temperature range and its disappearance with pressure, but not a gradual grow of the superconducting critical temperature $T_c$ from 15 to 21 GPa. The growth of $T_c$ with pressure in the low-pressure range may be explained by earlier suggested presence of magnetic clusters of $\alpha$-phase within $\varepsilon$-phase, which both are metastable in a wide pressure range. These magnetic clusters, more common at low pressures, or, alternatively, other lattice defects can affect the electron pairing, leading to suppression of the critical temperature. The latter explanation is supported by the experimental fact that $\alpha \rightarrow \varepsilon$ martensitic transition is sluggish, and pressures higher than 20 GPa are needed for a complete transformation.

As it follows from the above discussion, further study of the non-local correlations in $\varepsilon$-iron seems to be important. This would allow studying in more detail mechanism of superconductivity and its pressure dependence, and may allow to explain observed $T^{5/3}$ behavior of resistivity, accompanied by a quantum phase transition at the pressure $p = 21$ GPa. On the other hand, further experimental studies are required to clarify the magnetic and superconducting properties of $\varepsilon$-Fe, that may also have significant implications for geophysics and Earth’s core magnetism.

Acknowledgments

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where usually we separate singlet and triplet terms:

Due to Pauli principle, the interaction vertex $\Gamma$ is antisymmetric:

where for $SU(2)$ symmetric interaction the vertices $V$ do not depend on spin indexes.

To discuss superconducting pairing, we perform the mean field decoupling

We suppose that the quadratic part $\hat{H}_0$ of the Hamiltonian is diagonalized by the transformation

where $\epsilon_{k,\alpha\sigma}$ are the fermionic operators for the band $\alpha$ and spin projection $\sigma$, such that

where $E_{k\alpha} = \sum_{mm'} \varepsilon_k^{mm'} (U_k^\alpha)^* U_k^{m'}$ and $\sum_{m'm''m'''} \varepsilon_k^{mm'} (U_k^\alpha)^* U_k^{m''} = 0$ ($\alpha \neq \beta$). Then we obtain

where

As usual, we separate singlet and triplet terms:

$$(\Delta_{s,\alpha,\sigma})_s = (\Delta_{s,\alpha,\sigma})_s + (\Delta_{s,\alpha,\sigma})_t,$$

$$(\Delta_{s,\alpha,\sigma})_t = (\Delta_{s,\alpha,\sigma})_t + i (d_{k,\alpha})_s (\sigma^y)^* {d_{k,\alpha}^*}.$$
where $\sigma$ are the Pauli matrices. From these relations, we find

$$
(\Delta_{k,\alpha}^{\sigma,\sigma'}(s(t)) = \frac{1}{2} \left( \Delta_{k,\alpha}^{\sigma,\sigma'} + \Delta_{k,\alpha}^{\sigma',\sigma} \right) = -\frac{1}{2} \sum_{\beta,\sigma''\sigma'''} \Gamma_{s(t),k,p}^{\sigma,\sigma'';\sigma''',\beta} \langle \epsilon_{p,\beta,\sigma,\beta'} - \epsilon_{p,\beta,\sigma} \rangle
$$

where

$$
\Gamma_{s(t),k,p}^{\sigma,\sigma'';\sigma''',\beta} = \frac{1}{2} \left( \Gamma_{k,p}^{\sigma,\sigma'';\sigma''',\beta} + \Gamma_{k,p}^{\sigma'',\sigma''';\sigma,\beta} \right),
$$

and

$$
V_{k,p}^{\sigma,\sigma'';\sigma'''}(s(t)) = \frac{1}{2} \sum_{\alpha,\gamma,\delta} \left( V_{mm',m''m'''}(s(t)) \mp V_{m''m'''m'''}(s(t)) \right) \left( U_{k}^{\alpha} \right)^{*} \left( U_{p}^{\gamma} \right)^{*} U_{p}^{\gamma'} U_{p}^{\gamma''}.
$$

From Eq. (A10) we obtain gap equations:

$$
\psi_{k,\alpha} = - \sum_{k',i'\nu'} V_{kk',\alpha}\mathcal{F}_{k'(i'\nu')} \tag{A13a}
$$

$$
d_{k,\alpha} = - \sum_{k',i'\nu'} V_{kk',\alpha}\mathcal{F}_{k'(i'\nu')} \tag{A13b}
$$

where $\mathcal{F}_{k\alpha}(i\nu_n) = (i\sigma_{\alpha}^0 / 2)(\epsilon_{k\alpha}(\tau)\epsilon_{k\alpha}(0))_{i\nu_n}$ and $\mathcal{F}_{k\alpha}(i\nu_n) = (i/2)(\sigma_{\alpha}\tau^0\epsilon_{k\alpha}(\tau)\epsilon_{k\alpha}(0))_{i\nu_n}$ are the anomalous Green functions of the Hamiltonian $H_0 + H_{\text{BCS}}$, the index $i\nu_n$ denotes the Fourier transform. The corresponding Bethe-Salpeter equations are obtained by linearizing the gap equations and considering the eigenvalues and eigenfunctions of the operators in the right-hand-side of Eq. (A13), see Eq. (2) of the main text.

2. Bare and second-order vertex

To consider the vertex $V$, we use the second-order perturbation theory. To preserve spin-rotational symmetry, we consider the following orbital-dependent interactions

$$
H_{\text{int}} = U \sum_{im} n_{im\uparrow} n_{im\downarrow} + \sum_{i,m>\sigma} U_{1}^{mm'} n_{im\sigma} n_{im'\sigma} + \sum_{i,m>\sigma} U_{2}^{mm'} n_{im\sigma} n_{im'\sigma}\delta_{mm'}
$$

$$
+ \sum_{i,m>\sigma} (U_{2}^{mm'} - U_{1}^{mm'}) c_{im\sigma}^{i} c_{im'\sigma}\delta_{im}\delta_{im'},\delta_{mm'} + \sum_{i,m>\sigma} J_{mm'} \delta_{im}\delta_{im'}\delta_{mm'}
$$

(for the standard Kanamori parameterization with Hund exchange $I$, interorbital exchange $J$, and pair hopping $J$ we have $U_{2}^{mm'} = U, U_{1}^{mm'} = U' - I, J_{mm'} = J$). For the bare local vertex $V_{0}^{mm',m''m'''}$ defined by putting the Hamiltonian (A14) into the form of Eqs. (A2) and (A3a) we have

$$
V_{0}^{mm',m''m'''} = U \delta_{mm'} \delta_{mm''} + U_{2}^{mm'} \delta_{mm''} (1 - \delta_{mm'})
$$

$$
+ \left( U_{2}^{mm'} + U_{1}^{mm'} \right) \delta_{mm''}(1 - \delta_{mm'}) + J_{mm'} \delta_{mm''} (1 - \delta_{mm'}).
$$

In the second order in $V_0$ the effective interaction is given by $V_{0}^{mm',m''m'''} + V_{2,k,-k,p,p}$ where

$$
V_{2,k,-k,p,p}^{mm',m''m'''} = \frac{1}{2} \left[ -2V_{0}^{mm',m''m'''}(k - k) + V_{0}^{mm',m''m'''}(k + p) \right] \left( \tilde{\mathcal{F}}_{mm',m''m'''}(k - k) \right)
$$

$$
+ \left( \tilde{\mathcal{F}}_{mm',m''m'''}(k + p) \right) V_{0}^{mm',m''m'''}(k - k) + \left( \tilde{\mathcal{F}}_{mm',m''m'''}(k + p) \right) V_{0}^{mm',m''m'''}(k + p),
$$

where we assume summation over repeating orbital indices and

$$
L_{ph}^{mm',m''m'''} = -T \sum_{k,i\nu_n} c_{k\alpha}^{mm'}(i\nu_n) G_{k+q}^{m''m'''}(i\nu_n)
$$

is the particle-hole bubble.

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