Variational cluster approach to \(s\)-wave pairing in heavy-fermion superconductors

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We study \(s\)-wave Cooper pairing in heavy-fermion systems. We analyze the periodic Anderson model by means of the variational cluster approach (VCA) focusing on the interorbital Cooper pairing between a conduction electron (\(c\) electron) and an \(f\) electron, called the “\(c-f\) pairing.” It is shown that the \(s\)-wave superconductivity appears coexisting with long-range antiferromagnetic order when electrons or holes are doped into the system at half filling. The antiferromagnetic order vanishes when the doping concentration exceeds a certain critical value, leading to a pure \(s\)-wave superconducting state. Moreover, the comparative study with different reference systems used in the VCA shows that the interorbital \(c-f\) pairing is essential for the appearance of the \(s\)-wave superconductivity.

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I. INTRODUCTION

Heavy-fermion systems have provided opportunities to study various types of unconventional superconductivity. For example, the Ce-based compound CeCoIn\(_5\) has two kinds of superconducting states, one of which observed in higher magnetic fields is a strong candidate for the Fulde-Ferrell-Larkin-Ovchinnikov state with finite center-of-mass momentum of the Cooper pairs.\(^1\) In superconductors without inversion symmetry, such as CePt\(_3\)Si and CeRhSi\(_3\), the exotic parity mixing between spin-singlet and spin-triplet states is expected to occur due to the existence of the antisymmetric spin-orbit interaction.\(^2\) The coexistence of superconductivity and long-range magnetic order has been observed in several ferromagnets (UGe\(_2\), URhGe, etc.) as well as in several antiferromagnets (UPd\(_2\)Al\(_3\), UNi\(_2\)Al\(_3\), etc.).\(^3\) A variety of experimental and theoretical efforts have been devoted to understand those exotic states.

Superconductivity with simple \(s\)-wave pairing symmetry is another unconventional phenomenon in heavy-fermion systems. Usually, heavy-fermion superconductors favor the nodal pairing states, such as the \(d\)-wave and \(p\)-wave states, rather than the \(s\)-wave state. This is because the strong Coulomb repulsion in those systems is incompatible with intrasite Cooper pairing, which gives the nodal \(d\)-wave and \(p\)-wave states. In fact, nuclear resonance (NMR and NQR) experiments have demonstrated that many of heavy-fermion superconductors possess the nodal superconducting gaps.\(^4\) On the other hand, some heavy-fermion compounds, such as CeRu\(_2\)\(_2\)\(_3\)\(_2\)\(_2\) and recently reinvestigated CeCu\(_2\)Si\(_2\)\(_2\)\(_2\) are known to exhibit \(s\)-wave superconductivity. In the BCS theory, such \(s\)-wave superconductivity is explained as a result of the electron-phonon attraction between electrons. However, as mentioned above, heavy-fermion compounds have the strong Coulomb repulsion, which is considered as the dominant interaction between electrons. Thus, the \(s\)-wave superconductivity in those compounds may come from another mechanism.

The multiorbital nature is one of the characteristic features of heavy-fermion systems, which are composed of itinerant electrons in the conduction orbitals (\(c\) electrons) and localized electrons in the \(f\) orbitals (\(f\) electrons). The correlation between \(c\) and \(f\) electrons leads to various intriguing phenomena, such as the Kondo effect,\(^4\)\(^1\)\(^4\) quantum critical behavior,\(^2\)\(^5\)\(^1\)\(^7\) and magnetic orderings due to the RKKY interaction.\(^1\)\(^4\)\(^8\) Recently, the importance of such orbital degrees of freedom has also been recognized in the studies of superconductivity in the other strongly correlated electron systems. For example, the material dependence in the critical temperature of cuprates has been explained by using the multiorbital Hubbard models.\(^9\) Moreover, the multiorbital nature is considered to be the key for understanding the high-\(T_c\) superconducting properties in iron pnictides.\(^1\)\(^0\) Previous studies\(^2\)\(^1\)\(^\)\(^2\)\(^1\) suggested that the multiorbital nature can be a source of \(s\)-wave superconductivity in heavy-fermion systems. Hanzawa and Yosida\(^2\)\(^1\)\(^\)\(^2\)\(^3\) and Spaepen\(^2\)\(^4\) discussed the interorbital Cooper pairing between \(c\) and \(f\) electrons, which we call the “\(c-f\) pairing,” as a possible mechanism for \(s\)-wave superconductivity. They estimated the order of the critical temperature in the periodic Anderson model with infinitely large Coulomb repulsion. More recently, the present authors\(^1\)\(^9\) also studied the \(c-f\) pairing for finite Coulomb repulsion, and presented a mean-field phase diagram of \(s\)-wave superconducting state. Note, however, that the mean-field approximation cannot properly describe local charge, spin, and orbital fluctuation effects, which are crucial in heavy-fermion systems. Thus, more sophisticated treatment is required to achieve a deeper understanding of the nature of the interorbital pairing.

In this paper, we use the variational cluster approach (VCA)\(^2\)\(^4\) to study \(s\)-wave superconductivity in heavy-fermion systems. The VCA can properly take into account the local Coulomb repulsion\(^2\)\(^5\)\(^2\)\(^6\) and allows us to deal with various long-range orders, such as charge-density-wave\(^2\)\(^7\) \(d\)-wave superconducting\(^2\)\(^8\)\(^2\)\(^9\) and antiferromagnetic\(^3\)\(^0\)\(^3\)\(^1\) orders. Here, we apply the VCA...
to the standard periodic Anderson model considering all three types of s-wave Cooper pairings, i.e., between c electrons (c-c pairing), between f electrons (f-f pairing), and between c and f electrons (c-f pairing). We also consider possible antiferromagnetic order, which has been shown to emerge when the Coulomb repulsion is sufficiently strong. We calculate those order parameters and find five different phases depending on the parameters. At half filling, the system undergoes a second-order phase transition from nonmagnetic Kondo insulator to antiferromagnetic state when we increase the Coulomb repulsion. In the VCA, the self-energy matrix of the reference system by using the off-diagonal components. We can also calculate the self-energy matrix of the reference system by using the diagonal terms allow for describing the cluster chemical potential $\mu'$ is also treated as a variational parameter. The corresponding Weiss fields, $h'_{cc}$, $h'_{ff}$, $h'_{cf}$, and $h'_{AF}$, are determined by the variational conditions as mentioned below. To keep the thermodynamic consistency, the cluster Hamiltonian $H'_g$ includes four types of Weiss-field terms, $H'_cc$, $H'_ff$, $H'_cf$, and $H'_AF$. The first three terms allow for describing the c-c, f-f, and c-f pairing orders, respectively. The last term gives long-range antiferromagnetic order. The corresponding Weiss fields, $h'_{cc}$, $h'_{ff}$, $h'_{cf}$, and $h'_{AF}$, are determined by the variational conditions as mentioned below. To keep the thermodynamic consistency, the cluster chemical potential $\mu'$ is also treated as a variational parameter. We denote the set of these variational parameters as $\mathbf{t}' = (h'_{cc}, h'_{ff}, h'_{cf}, h'_{AF}, \mu')$. We assume that the Weiss-field $h'_{AF}$ acts only on $f$ electrons, which is justified by the fact that the antiferromagnetic order in this system is mainly due to the Coulomb repulsion between $f$ electrons.

We introduce the following Nambu spinor defined on each cluster:

$$\Psi = (c_{i\uparrow}, c_{i\downarrow}, f_{i\uparrow}, f_{i\downarrow}, c_{i\uparrow}^\dagger, c_{i\downarrow}^\dagger, f_{i\uparrow}^\dagger, f_{i\downarrow}^\dagger)^T,$$

where the two sites on the cluster $\Gamma$ are labeled 1 and 2. By diagonalizing the two-site Hamiltonian $H'_g$, we can easily obtain the Green’s-function matrix $G' = \langle \Psi | \Gamma | \Psi \rangle$ and the grand potential $\Omega'$ of the reference system $H'$. Note that $G'$ includes the anomalous Green’s functions regarding the c-c, f-f, and c-f pairings as the off-diagonal components. We can also calculate the self-energy matrix $\Sigma'$ of the reference system by using

$$\mathbf{H}' = \mathbf{H}_{PAM}' + H'_cc + H'_ff + H'_cf + H'_AF,$$

where

$$H'_{PAM} = -t \sum_{\langle ij \rangle} \sum_{\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) + \epsilon_f \sum_{i\sigma} n_{i\sigma}^f
- V \sum_{i\sigma} (f_{i\sigma}^\dagger c_{i\sigma} + \text{H.c.}) + U \sum_{i} n_{i\uparrow}^f n_{i\downarrow}^f
- \mu \sum_{i\sigma} (n_{i\sigma}^c + n_{i\sigma}^f),$$

for the emergence of the “Weiss fields” in the reference system. We compare two different reference systems with and without the Weiss field that acts as a pair potential for the c-f pairing, and conclude that the formation of Cooper pairs between $c$ and $f$ electrons is indeed an essential mechanism to stabilize the s-wave superconducting states in heavy-fermion systems.
\[ \Sigma'(t') = G_0^{-1} - G'^{-1}, \] where \( G_0 \) is the free Green’s function of the reference system obtained by setting \( U = 0 \) in Eq. (2).

According to the SFT, the grand potential of the original system can be written as
\[
\Omega(t') = \Omega' - \frac{N}{2} \text{Tr} \ln [-G'] + \sum_k \text{Tr} \ln [-G_{VCA}(k)] - 2N(\mu - \mu'),
\]
where \( N \) is the total number of lattice sites. In the VCA, the self-energy of the original system is approximated by that of the reference system as \( G_{VCA}(k) \equiv (G_0(k)^{-1} - \Sigma')^{-1} \). Here, \( G_0(k) \) is the free Green’s function of the original system with \( k \) being the wave vector in the Brillouin zone of the reference system. The last term on the right-hand side of Eq. (7), \(-2N(\mu - \mu')\), arises from the anticommutation relation of electron operators when we rewrite the Hamiltonians, Eqs. (1) and (2), using the Nambu spinor. Practical details of the evaluation of Eq. (9) are given in the Appendix. We determine the optimal values of the variational parameters \( \Omega' \) by solving the variational problems \( \Omega' = \Omega(\Omega') \) for \( \mu' = \mu'_{\text{opt}} \), \( \bar{\mu} = \bar{\mu}_{\text{opt}} \), and \( \psi = \psi_{\text{opt}} \).

Using the same Green’s function \( G_{VCA}(k) \mid t' = t'_{\text{opt}} \), we evaluate the following quantities:
\[
\Delta_{cc} = \frac{1}{N} \sum_i \langle c_{i\uparrow} c_{i\uparrow} \rangle, \quad \Delta_{ff} = \frac{1}{N} \sum_i \langle f_{i\uparrow} f_{i\uparrow} \rangle, \quad \Delta_{cf} = -\frac{1}{2N} \sum_i \langle c_{i\uparrow} f_{i\downarrow} - c_{i\downarrow} f_{i\uparrow} \rangle,
\]
where \( \Delta_{cc} \), \( \Delta_{ff} \), and \( \Delta_{cf} \) represent the \( s \)-wave superconducting order parameters for the \( c-c \), \( f-f \), and \( c-f \) pairings, respectively. The quantity \( m_c \) (\( m_f \)) is the staggered magnetization in the \( c \) (\( f \)) orbital. Throughout this work, we fix the value of \( \epsilon_f \) to \(-U/2\), considering the situation where the Fermi level is located near the center of the upper and lower Hubbard bands of \( f \) electrons. Under the symmetric condition \( \epsilon_f = -U/2 \), the models for electron-doped \((n = 2.0 + \delta)\) and hole-doped \((n = 2.0 - \delta)\) systems are symmetric with each other about half filling \((n = 2.0)\). Thus, we discuss only the electron-doped case hereafter. We set the hybridization \( V = t \) and the temperature \( T = 0 \) in the present study.

### III. RESULTS

At half filling, the system exhibits the Kondo insulating state, which changes into an antiferromagnetic state when the Coulomb repulsion exceeds a critical value \( U/t = 2.3 \). Our VCA analysis gives \( U_c/t \approx 2.31 \). In the following, we focus on the case of \( U > U_c \). Figure 1 shows the phase diagram in the \((n, U/t)\) plane. To explain each phase in the phase diagram, we show in Fig. 2a) the \( n \) dependences of the order parameters at \( U/t = 2.6 \), which is marked by the horizontal dashed line in Fig. 1. We also show the corresponding behavior of the Weiss fields in Fig. 2b. Away from half filling, only the superconducting order parameters \( \Delta_{ff}, \Delta_{cf} \), and \( \Delta_{cc} \) have finite values, which means that the system is in the pure \( s \)-wave superconducting (SC) phase. The values of the order parameters satisfy the inequality \( \Delta_{ff} > \Delta_{cf} > \Delta_{cc} \). When we decrease the density \( n \), the staggered magnetizations \( m_f \) and \( m_c \) appear at the critical density \( n_c \approx 2.12 \), below which the \( s \)-wave superconductivity coexists with the antiferromagnetic (AF) order. When \( n \) is decreased further, the system exhibits phase separation (PS). Since the difference of the grand potentials at \( \mu = \mu_A \) and \( \mu = \mu_B \) was given by \( \Delta \Omega = -N \int_{n_A}^{n_B} n(\mu) d\mu \), we determined the boundaries \( n_1 \) and \( n_2 \) of the PS region from the Maxwell construction in the \((n, \mu/t)\) plane, as shown in Fig. 3. This type of phase separation was also found in the previous VCA studies that discussed the coexistence of \( d \)-wave superconductivity and antiferromagnetic order in the Hubbard model. One of these studies has predicted that the PS region becomes narrower as the cluster size increases and may vanish in the limit of large cluster size. This may also be the case for the PS region.

FIG. 1. The phase diagrams in the \((n, U/t)\) plane. The horizontal dashed line indicates the line of \( U/t = 2.6 \).
FIG. 2. The $n$ dependences of (a) the order parameters and (b) the Weiss fields for $U/t = 2.6$.

FIG. 3. The $n$ dependence of the chemical potential for $U/t = 2.6$.

in our results. Finally, near half filling ($2 < n < n_1$), the system exhibits the coexistence phase again.

We also note that the $f-f$ pairing order parameter $\Delta_{ff}$ is larger than the other superconducting order parameters $\Delta_{cc}$ and $\Delta_{cf}$, as shown in Fig. 2(a). This behavior cannot be readily understood since the Coulomb repulsion $U$ in the $f$ orbital suppresses the on-site $s$-wave Cooper pairing between $f$ electrons. We recall that the density of states (DOS) for $f$ electrons is much larger than that for $c$ electrons near the Fermi level in the periodic Anderson model. Thus, although the effective pairing attraction between $f$ electrons is weakened by the Coulomb repulsion, $\Delta_{ff}$ is still large due to the large DOS for $f$ electrons.

We argue that the $c-f$ pairing is a key for the appearance of $s$-wave superconductivity in the periodic Anderson model, which has also been pointed out in our previous mean-field study. To confirm this within the VCA, we carry out additional analyses using the following cluster Hamiltonians instead of Eq. (2): (i) $H_{\Gamma,1} = H_{\text{PAM}} + H_{cc} + H_{ff}$ (i.e., the $c-f$ pairing field is not considered); (ii) $H_{\Gamma,2} = H_{\text{PAM}} + H_{cf}$ (i.e., only the $c-f$ pairing field is considered). In the first case (i), we found only a trivial solution $h_{cc} = h_{ff} = 0$, namely, no superconducting solution is obtained ($\Delta_{cc} = \Delta_{ff} = \Delta_{cf} = 0$). This indicates that the occurrence of the $s$-wave superconductivity requires the $c-f$ pairing field $h_{cf}$, i.e., the $c-f$ pairing plays a crucial role in the mechanism for the $s$-wave superconductivity. Indeed, in the second case (ii), we find a solution with $h_{cc} \neq 0$ and $\Delta_{cf} \neq 0$. Note that the other order parameters $\Delta_{cc}$ and $\Delta_{ff}$ also have finite values even though the corresponding Weiss fields $h_{cc}$ and $h_{ff}$ are not taken into account. This stems from the hybridization $V$ between $c$ and $f$ states. Due to the existence of $h_{cf}'$ and $V$, the self-energy $\Sigma'(t')$ has the off-diagonal components $\Sigma_{cc}'(t')$ for the $c-c$ pairing and $\Sigma_{ff}'(t')$ for the $f-f$ pairing as well as $\Sigma_{cf}'(t')$ for the $c-f$ pairing, through the diagonalization of $H_{\Gamma,2}$. Thus, all the superconducting order parameters, $\Delta_{cc}$, $\Delta_{ff}$, and $\Delta_{cf}$, have finite values although $H_{\Gamma,2}$ does not include $h_{cc}'$ and $h_{ff}'$. This comparative study indicates that the pair potential for the $c-f$ pairing is essential for the occurrence of the $s$-wave superconductivity.

Finally, we consider the reason why the $s$-wave superconductivity can coexist with the long-range antiferromagnetic order. In usual single-orbital systems, $s$-wave superconductivity and antiferromagnetism compete with

(a) single-orbital system

antiferromagnetic order

$s$-wave Cooper pair

site $i$ site $i+1$

antiferromagnetic state $s$-wave superconducting state

(b) multi-orbital system

coexistence state of the $s$-wave $c/f$ pairing and antiferromagnetic orders

FIG. 4. Schematic pictures on the relationship between $s$-wave superconductivity and antiferromagnetism. The panels (a) and (b) correspond to the cases of single-orbital and multi-orbital systems, respectively.
each other. As seen in Fig. 4a, the antiferromagnetic state has local spin polarization, which is incompatible with the formation of local spin singlets in the $s$-wave superconducting state. On the other hand, these two conditions can be simultaneously satisfied for interorbital pairing in multiorbital systems, as seen in Fig. 4b).

**IV. CONCLUSION**

We have investigated $s$-wave superconductivity in heavy-fermion systems in terms of the variational cluster approach (VCA) to the periodic Anderson model. In the VCA, we have taken into account all the three types of $s$-wave Cooper pairings: the intraorbital pairings between $c$ electrons and between $f$ electrons, and the interorbital pairing between $c$ and $f$ electrons. We have shown that $s$-wave superconducting states appear when electrons or holes are doped to the system at half filling. In a region close to half filling, the $s$-wave superconductivity coexists with long-range antiferromagnetic order. The VCA comparative analysis with different reference systems indicated that the $c$-$f$ pairing plays a dominant role in the formation of the $s$-wave superconducting state. These results might advance the understanding of the systems indicated that the $s$-wave superconducting state.

The superconducting state. On the other hand, these two $s$-wave superconducting states appear when electrons are assumed to be all-f wave superconductivity in heavy-fermion superconducting state. We have investigated $s$-wave superconductivity in heavy-fermion systems and between $f$ electrons. We have shown that $s$-wave superconducting states appear when electrons or holes are doped to the system at half filling. In a region close to half filling, the $s$-wave superconductivity coexists with long-range antiferromagnetic order. The VCA comparative analysis with different reference systems indicated that the $c$-$f$ pairing plays a dominant role in the formation of the $s$-wave superconducting state. These results might advance the understanding of the systems indicated that the $s$-wave superconducting state.

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**Appendix A: Evaluation of the grand potential**

We evaluate the grand potential given by Eq. (4) at $T = 0$. We first introduce the matrices $Q^{(e)}$ and $Q^{(h)}$ whose elements are given by:

$$Q_{m,n}^{(e)} = \langle 0 | \Psi_m | n \rangle, \quad Q_{m,n}^{(h)} = \langle n | \Psi_m | 0 \rangle, \quad (A1)$$

with

$$H_0^{(e)} | 0 \rangle = E_0 | 0 \rangle, \quad H_0^{(h)} | n \rangle = E_n | n \rangle. \quad (A2)$$

Here, $| 0 \rangle (| n \rangle)$ is the ground ($n$-th excited) state of the cluster Hamiltonian $H_0^{(e)}$ and $\Psi_m$ is the $m$-th component of the Nambu spinor $\Psi$. Note that the excited states with even (odd) numbers of electrons can be ignored when the ground state $| 0 \rangle$ consists of odd (even) numbers of electrons. Therefore, $Q^{(e)}$ and $Q^{(h)}$ are $8 \times 8$ (4/2) matrices in the present case. Using $Q^{(e)}$ and $Q^{(h)}$, we define the $8 \times 2N_e$ $Q$-matrix $Q$ which has following elements:

$$Q_{m,l} = \begin{cases} Q_{m,l}^{(e)} (1 \leq l \leq N_e) \\ Q_{m,l-N_e}^{(h)} (N_e + 1 \leq l \leq 2N_e). \end{cases} \quad (A3)$$

We also introduce the $2N_e \times 2N_e$ diagonal matrix $\Lambda$ whose diagonal elements are given by

$$\Lambda_{l,l} = \begin{cases} E_l - E_0 & (1 \leq l \leq N_e) \\ -E_{l-N_e} + E_0 & (N_e + 1 \leq l \leq 2N_e). \end{cases} \quad (A4)$$

In the Lehmann representation, the cluster Green’s function $G'(\omega)$ can be written as:

$$G'(\omega) = Qg(\omega)Q^1, \quad (A5)$$

where $g(\omega) = (\omega - A)^{-1}$.

We can rewrite the second term on the right-hand side of Eq. (9) as follows:

$$- \frac{N}{2} \text{Tr} \ln [-G'(\omega)] = - \frac{N}{2} \sum_{l=1}^{2N_e} \omega_l \Theta(-\omega_l) + R, \quad (A6)$$

where $\omega_l$ is the pole of the cluster Green’s function. In the present work, the matrix $\Theta(\omega)$ is Heaviside step function defined by $\Theta(\omega) = 1$ for $\omega \geq 0$ and $\Theta(\omega) = 0$ for $\omega < 0$. The last term $R$ represents the contribution from the poles of the self-energy $\Sigma'$. We note that since $\omega_l$ is given by the diagonal elements of $\Lambda$, the first term of Eq. (10) is simplified as

$$- \frac{N}{2} \sum_{l=1}^{2N_e} \omega_l \Theta(-\omega_l) = - \frac{N}{2} \sum_{l=1}^{N_s} (E_0 - E_l). \quad (A7)$$

In a similar way, the third term on the right-hand side of Eq. (9) is rewritten as follows:

$$\sum_{\mathbf{k}} \text{Tr} \ln [-G_{\text{VCA}}(\mathbf{k})] = \sum_{\mathbf{k}} \sum_{l=1}^{2N_\mathbf{k}} \omega_l(\mathbf{k}) \Theta(-\omega_l(\mathbf{k})) - R, \quad (A8)$$

where $\omega_l(\mathbf{k})$ is the pole of the VCA Green’s function $G_{\text{VCA}}(\mathbf{k})$ with $\mathbf{k}$ being the wave vector in the Brillouin
zone of the reference system. The details of the numerical method to find \(\omega(\mathbf{k})\) will be given in the next paragraph. With the help of Eqs. (A11)-(A13), we obtain the following expression for the grand potential per site:

\[
\frac{\Omega}{N} = \frac{E_0}{2} - \frac{1}{2} \sum_{l=1}^{N_s} (E_0 - E_l) + \frac{1}{N} \sum_{\mathbf{k}} \sum_{l=1}^{2N_s} \omega_l(\mathbf{k}) (\omega_l(\mathbf{k}) - 2(\mu - \mu')). \quad (A9)
\]

Here, the summation \(\frac{1}{N} \sum_{\mathbf{k}}\) is replaced by the integration \(\frac{1}{(2\pi)^d} \int_{-\pi/2}^{\pi/2} d\tilde{x} \int_{-\pi/2}^{\pi/2} d\tilde{y}\) in thermodynamic limit \(N \to \infty\).

We finally present the numerical method to find the poles of the VCA Green’s function \(G_{\text{VCA}}(\mathbf{k})\). The VCA Green’s function \(G_{\text{VCA}}(\mathbf{k})\) is given by\(^{36,37}\)

\[
G_{\text{VCA}}(\mathbf{k}) = \frac{1}{G_0(\mathbf{k})^{-1} - \Sigma'} = \frac{1}{(\omega - \mathbf{T}(\mathbf{k})) - (\omega - \mathbf{T}' - G'^{-1})} = \frac{1}{G'^{-1} - V(\mathbf{k})}, \quad (A10)
\]

where the matrices \(\mathbf{T}(\mathbf{k})\) and \(\mathbf{T}'\) are

\[
\mathbf{T}(\mathbf{k}) = \begin{pmatrix} A(\mathbf{k}) & 0 \\ 0 & -A(\mathbf{k}) \end{pmatrix}, \quad \mathbf{T}' = \begin{pmatrix} B & C \\ C & D \end{pmatrix}, \quad (A11)
\]

with

\[
A(\mathbf{k}) = \begin{pmatrix} -\mu & \epsilon(\mathbf{k}) & -V & 0 \\ -\mu & 0 & -V & 0 \\ -V & 0 & -\epsilon_f - \mu & 0 \\ 0 & -V & 0 & -\epsilon_f - \mu \end{pmatrix}, \quad (A12)
\]

\[
\epsilon(\tilde{k}) = -t(1 + e^{-2t\tilde{k}_x} + e^{-t(\tilde{k}_x - \tilde{k}_y)} + e^{-t(\tilde{k}_x + \tilde{k}_y)}), \quad (A13)
\]

\[
\mathbf{B} = \begin{pmatrix} -\mu' & t & -V & 0 \\ -t & -\mu' & 0 & -V \\ -V & 0 & \epsilon_f - \mu' - h'_{\text{AF}} & 0 \\ 0 & -V & 0 & \epsilon_f - \mu' + h'_{\text{AF}} \end{pmatrix}, \quad (A14)
\]

\[
\mathbf{C} = \begin{pmatrix} -h'_{cc} & 0 & h'_{cf} & 0 \\ 0 & -h'_{cc} & 0 & h'_{cf} \\ h'_{cf} & 0 & h'_{ff} & 0 \\ 0 & h'_{cf} & 0 & h'_{ff} \end{pmatrix}, \quad (A15)
\]

and

\[
\mathbf{D} = \begin{pmatrix} \mu' & t & V & 0 \\ t & \mu' & 0 & -V \\ V & 0 & -\epsilon_f + \mu' - h'_{\text{AF}} & 0 \\ 0 & V & 0 & -\epsilon_f + \mu' + h'_{\text{AF}} \end{pmatrix}. \quad (A16)
\]

The matrix \(\mathbf{V}(\mathbf{k}) = \mathbf{T}(\mathbf{k}) - \mathbf{T}'\) denotes the intercluster hopping. By substituting Eq. (A5) into Eq. (A10), we obtain

\[
G_{\text{VCA}}(\tilde{k}) = \frac{1}{g^{-1} - Q^\dagger \mathbf{V}(\mathbf{k}) Q}. \quad (A17)
\]

This expression shows that the poles of the VCA Green’s function are given as the eigenvalues of the matrix \(\mathbf{L}(\tilde{k}) = \mathbf{A} + Q^\dagger \mathbf{V}(\mathbf{k}) \mathbf{Q}\).
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