Double-Exchange Ferromagnetism and Orbital-Fluctuation-Induced Superconductivity in Cubic Uranium Compounds

Takashi Hotta

Department of Physics, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo 192-0397, Japan

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A double-exchange mechanism for the emergence of ferromagnetism in cubic uranium compounds is proposed on the basis of a $j$-$j$ coupling scheme. The idea is orbital-dependent duality of 5$f$ electrons concerning itinerant $\Gamma_7^-$ and localized $\Gamma_8^-$ states in the cubic structure. Since orbital degree of freedom is still active in the ferromagnetic phase, orbital-related quantum critical phenomenon is expected to appear. In fact, odd-parity $p$-wave pairing compatible with ferromagnetism is found in the vicinity of an orbital ordered phase. Furthermore, even-parity $d$-wave pairing with significant odd-frequency components is obtained. A possibility to observe such exotic superconductivity in manganites is also discussed briefly.

KEYWORDS: Ferromagnetism, Orbital, Superconductivity, Uranium Compounds, Manganites

In the Bardeen-Cooper-Schrieffer theory for superconductivity,\textsuperscript{1} it was simply considered that magnetism suppresses superconductivity, since the singlet $s$-wave electron pair mediated by phonon-induced attraction is easily destroyed by an applied magnetic field. However, since the pioneering discovery of superconductivity in Ce-based heavy-fermion materials\textsuperscript{2} and some uranium compounds,\textsuperscript{3-7} it has been gradually recognized that anisotropic superconducting pair mediated by magnetic fluctuations generally appears in strongly correlated electron materials. In particular, due to successive discoveries of superconductivity near an antiferromagnetic phase both in $d$- and $f$-electron systems, nowadays it is confirmed that “magnetism is a good friend to superconductivity”.\textsuperscript{8}

When we turn our attention to the relation between ferromagnetism and superconductivity, it was discussed that critical magnetic fluctuations can mediate triplet Cooper pair.\textsuperscript{9} In fact, superconductivity has been observed in a ferromagnetic phase of uranium compounds such as UGe\textsubscript{2},\textsuperscript{10} URhGe,\textsuperscript{11} UIr,\textsuperscript{12} and UCoGe.\textsuperscript{13} However, for $f$-electron systems, a microscopic theory for superconductivity has not been satisfactorily developed so far, mainly due to the difficulty in multi-orbital nature and strong spin-orbit coupling of $f$ electrons.

A way to overcome such a situation is to exploit a $j$-$j$ coupling scheme. Along this research direction, the present author has developed microscopic $f$-electron theories on the basis of the $j$-$j$ coupling scheme.\textsuperscript{14-16} In the model, one $f$-electron state is characterized by an appropriate linear combination of the $z$ component of total angular momentum $j$. Usually it is convenient to use the basis which diagonalizes the crystalline electric field (CEF) potential. In any case, we accommodate plural numbers of $f$ electrons in such one-$f$-electron states due to the effect of Hund’s rule interaction.

For the case of cubic CEF potential, it is well known that the $j=5/2$ sextet is split into $\Gamma_7^-$ doublet and $\Gamma_8^-$ quartet. Since the $\Gamma_7^-$ orbital has nodes along the cubic axes, it has strong localized nature, while $\Gamma_8^-$ states have itinerant nature in comparison with $\Gamma_7^-$ electrons. This orbital-dependent duality of $f$ electrons seems to be a key issue of rich phenomena in $f$-electron materials. Since electrons in localized $\Gamma_7^-$ and itinerant $\Gamma_8^-$ orbitals are coupled with the Hund’s rule interaction, we envisage a situation similar to double-exchange manganites with mobile $e_g$ and localized $t_{2g}$ electrons.

In this Letter, a double-exchange scenario for the emergence of ferromagnetism in cubic uranium compounds is proposed on the basis of the orbital-dependent duality nature of $5f$ electrons. We also propose some experiments to confirm the double-exchange ferromagnetism in cubic uranium materials. In the ferromagnetic phase, we obtain the reduced Hamiltonian with active orbital degree of freedom. By analyzing the model within a random phase approximation (RPA), we find both odd-parity $p$-wave and even-parity $d$-wave pairing states in the vicinity of an orbital ordered state, suggesting orbital-related quantum critical phenomena. Finally, we briefly discuss a possibility of superconductivity in manganites, which is well described by the double-exchange model.

First we briefly explain the $j$-$j$ coupling scheme. We include the spin-orbit coupling so as to define the state labelled by the total angular momentum $j$, given by $j=s+\ell$, where $s$ and $\ell$ are spin and angular momenta, respectively. For $f$-orbitals with $\ell=3$, we immediately obtain an octet with $j=7/2$ and a sextet with $j=5/2$, which are well separated by the spin-orbit interaction. Since the octet level is higher than the sextet one, it is enough to consider $j=5/2$ sextet when local $f$-electron number is less than six.

Next we define the one $f$-electron state in the cubic crystal structure. It is well known that under the cubic CEF potential, the sextet of $j=5/2$ is split into $\Gamma_7^-$ doublet and $\Gamma_8^-$ quartet. Note, however, that the ground state depends on the crystal structure. For instance, in the AuCu$_3$-type cubic structure, the energy level for $\Gamma_7^-$ doublet is lower than that for $\Gamma_8^-$, while for CaF$_2$-type cubic structure, $\Gamma_8^-$ quartet becomes the ground state. In this paper, we assume the case with $\Gamma_7^-$ ground state.

Since we consider the metallic uranium compounds, the valance of uranium ion takes the value between three and four, corresponding to the local $f$-electron number between three and two. When we accommodate two or three electrons in $\Gamma_7^-$ and $\Gamma_8^-$ levels, we find two possibilities of low- and high-spin states, if we borrow the terminology of $d$-electron systems, depending on the balance between the Hund’s rule interaction and the CEF splitting between $\Gamma_7^-$ and $\Gamma_8^-$ levels.

Readers may consider that the high-spin state is always stabilized in $f$-electron ions, but we should note that the effective Hund’s rule interaction $J_{\text{eff}}$ in the $j$-$j$ coupling scheme is reduced from the original Hund’s rule coupling among $f$-
orbitals \( J_H \) as \( J_{\text{eff}}=J_H(g_f-1)^2/J_H/49,14 \) where \( g_J \) is the Landé’s g-factor and \( g_f=6/7 \) for \( J=5/2 \). In fact, we have proposed the low-spin state for actinide ions to understand spin and orbital structure of AnTGA\(_5\) (An=U and Np; T=Ni, Pt, Fe and Co)\(^{17,18}\) and multipole order in NpO\(_2\).\(^{19,20}\) In this paper, on the other hand, we attempt to find new possibility of high-spin state concerning ferromagnetism and superconductivity.

Now we discuss the \( f \)-electron kinetic term in a tight-binding approximation. When we evaluate \( f \)-electron hopping amplitude \( t_{\alpha\tau}^{\ast} \), for nearest-neighbor hopping via the \( \sigma \) bond between adjacent \( f \) orbitals, it is given by \( t_{\alpha b}^{\ast}=-\sqrt{3}t_{ab} \), \( t_{\alpha a}^{\ast}=\sqrt{3}t_{ab} \), and \( t_{bb}^{\ast}=3t/4, \), where indices \( a \) and \( b \) distinguish two \( \Gamma_8 \) states (see Fig. 1) and \( t \) is given for \( t=3(f f \sigma)/7 \) with the use of Slater-Koster integral \((f f \sigma)\).\(^{21,22}\) Note that \( \Gamma_7 \) orbital is localized, since the corresponding wavefunction has nodes along the axis directions, as shown in Fig. 1. On the other hand, \( \Gamma_8 \) orbitals are itinerant and their hopping amplitudes are just the same as those of \( e_g \) orbitals of \( 3d \) electrons,\(^{15,23}\) since \( \Gamma_8 \) is isomorphic to \( \Gamma_3 \times \Gamma_6 \), where \( \Gamma_3 \) indicates \( E \) representation for the orbital part and \( \Gamma_6 \) denotes the spin part.

As mentioned above, we assume the high-spin state in this paper. Namely, the Hund’s rule interaction works among \( \Gamma_7 \) and \( \Gamma_8 \) orbitals. Note that Coulomb interaction in \( \Gamma_7 \) states is larger than those for \( \Gamma_8 \) ones in the order of \( J_{\text{eff}} \). The difference of the magnitude of Coulomb interaction between itinerant and localized orbitals is not significant in comparison with \( d \)-electron systems, but in the combination with the orbital dependent duality nature, we arrive at the double-exchange model, which is used as a canonical model for manganites.\(^{15,23}\) In this model, in order to gain the kinetic energy, the ferromagnetic phase appears, which is called the double-exchange ferromagnetism. This is established in the qualitative understanding of ferromagnetism in manganites.

In order to confirm the emergence of double-exchange ferromagnetism in uranium compounds, we propose a couple of experiments in analogy with manganites. One is the observation of large negative magnetoresistance phenomenon. Since electrons can move smoothly in the ferromagnetic phase in comparison with the paramagnetic one, the resistivity is drastically decreased, when we apply a magnetic field on double-exchange materials. The large negative magnetoresistance in cubic uranium compounds may be an evidence for the double-exchange mechanism. For instance, in \( \beta \)-US\(_2\), large magnetoresistance phenomenon has been observed,\(^{24}\) although this material does not have cubic structure.

Another is more direct evidence for the relation between the Curie temperature \( T_C \) and the kinetic energy. In manganites with relatively wide bandwidth for conduction electrons, it has been observed that \( T_C \) is increased with the hole doping.\(^{23}\) Since the double-exchange ferromagnetism occurs so as to gain the kinetic energy, the ferromagnetic transition occurs more easily when electrons can move smoothly. Thus, we propose the appearance of the ferromagnetic metallic phase due to the application of hydrostatic pressure or the hole doping on insulating and/or antiferromagnetic states of cubic uranium compounds. In the case of uranium compounds, hole doping can be done by thorium substitution. It is a drastic phenomenon that ferromagnetism appears due to thorium doping into antiferromagnetic uranium compounds.

When \( \Gamma_8 \) is lower than \( \Gamma_7 \) with large Hund’s rule coupling, the double-exchange ferromagnetism occurs for itinerant \( \Gamma_7 \) and localized \( \Gamma_8 \). Such a situation is realized in Nd-based filled skutterudite compounds, in which ferromagnetism is frequently observed. It may be interesting to seek for evidence of double-exchange ferromagnetism in such materials. Note also that the present mechanism cannot be directly applied to the tetragonal Uranium material, but it works even in the tetragonal system, when the CEF level splitting among Kramers doublets is less than \( J_{\text{eff}} \) and the lower level has localized nature.

Let us discuss the superconductivity in the ferromagnetic phase. For the purpose, we consider the spinless \( f \)-electron model with active orbital degree of freedom as

\[
H = \sum_{i,a,r,r'} v_{\alpha}^a \hat{c}_{i,r'}^\dagger \hat{c}_{i,a+r'} + U \sum_{i} n_{i\alpha} n_{i\beta},
\]

(1)

where \( f_{i\tau} \) is the annihilation operator for an \( f \)-electron in the \( \tau \)-orbital of \( \Gamma_8 \) at site \( i \), \( \sum_{\tau} f_{i\tau}=f_{i\tau}^\dagger f_{i\tau} \), and \( U \) is the inter-orbital Coulomb interaction. Throughout this paper, we set \( U=4t \), which is less than the bandwidth \( 6t \).

Note that if \( t_{\alpha\tau}^{\ast}=\delta_{\alpha\tau} \), \( H \) is equivalent to the well-known Hubbard model and we simply deduce that \( d \)-wave superconductivity appears near the antiferro orbital-ordered phase. However, in actuality, electrons hop among different adjacent orbitals. The type of superconductivity in such a realistic multiorbital system has been discussed actively,\(^{25,27}\) and quite recently, it has attracted much attention due to the discovery of Fe-based superconductors.\(^{28}\)

The non-interacting Green’s function \( \hat{G} \) is given by

\[
\hat{G}^{-1}(k) = \begin{pmatrix}
    i\omega_n + \mu - \varepsilon_{kaa} & \varepsilon_{kab} \\
    -\varepsilon_{kba} & i\omega_n + \mu - \varepsilon_{kbb}
\end{pmatrix},
\]

(2)

where we introduce the abbreviation \( k=(k_x, i\omega_n) \), \( k \) is the momentum, \( \omega_n=(2n+1)\pi T \) is the fermion Matsubara frequency with an integer \( n \) and a temperature \( T \), \( \varepsilon_{kaa}=3t(\cos k_x + \cos k_y)/2 \), \( \varepsilon_{kbb}=t(\cos k_x + \cos k_y + 4\cos k_z)/2 \), \( \varepsilon_{kab}=\varepsilon_{kba}=-\sqrt{3}t(\cos k_x - \cos k_y)/2 \), and a chemical potential \( \mu \) controls the \( \Gamma_8 \) electron number \( \langle n \rangle \). Since \( \Gamma_7 \) electron is assumed to be localized, the cases of \( \langle n \rangle=1 \) and 2 correspond to \( U^+ \) and \( U^+ \) ions, respectively.

In order to discuss superconductivity, we solve the linearized gap equation for anomalous self-energy \( \phi \), given by

\[
\phi_{\tau_1\tau_2}(k) = -T \sum_{n'} \sum_{k',\tau'_1,\tau'_2} K_{\tau_1\tau_2,\tau'_1\tau'_2}(k,k') \phi_{\tau'_1\tau'_2}(k'),
\]

(3)

where \( K(k,k')=\hat{V}(k,k')\hat{G}(k')\hat{G}(k') \) and \( \hat{V} \) is given by

\[
\hat{V}(k,k') = \hat{J} + \hat{J}\hat{\chi}(k-k')\hat{I} - \hat{J}\hat{\chi}(k-k')^{-1}\hat{J} + \hat{L} - \hat{L}\hat{\chi}(k+k')\hat{I} + \hat{\lambda}(k+k')\hat{J} - \hat{\lambda}(k+k')^{-1}\hat{L},
\]

(4)

Fig. 1. Charge distribution of (a) \( \Gamma_8 \), (b) \( \Gamma_{-8} \), and (c) \( \Gamma_7 \) states.
In the calculation, we use a 32 lattice and 1024 Matsubara frequencies. The ordering vector is \( q \). The boundary curve is determined from the divergence in \( \chi(q) \) at \( \mu/\tau \approx 1.76 \), while \( \mu/\tau \) is relatively smaller than those of the even-frequency ones.

Fig. 2. (Color online) Phase diagram for \( U=4t \) near the quantum critical region. Inset shows the whole phase diagram.

In Fig. 2, we show the phase diagram in the \( (\mu, T) \) plane. The boundary curve is determined from the divergence in the RPA susceptibility. The inset shows the whole phase diagram: In the region I \((0 < \mu/\tau < 0.85)\), the orbital ordered state appears. The ordering vector is \( Q=(\pi, \pi, \pi) \) at \( \mu=0 \), but it is changed as \( (\pi, \pi, \delta) \), where \( \delta \) is monotonically decreased with the decrease of \( \mu \) and eventually becomes zero for \( \mu/\tau > 0.7 \). In the narrow region II \((0.85 < \mu/\tau < 1.05)\), we find \( Q=(\delta, \delta, \pi) \) with \( \delta=11\pi/16 \). In the region III \((1.05 < \mu/\tau < 1.76)\), \( Q=(\delta, \delta, \delta) \), where \( \delta=\pi \) for \( T/\tau > 0.1 \), while \( \delta < \pi \) for \( T/\tau < 0.1 \).

On the analogy of anisotropic superconductivity near an antiferromagnetic critical point, we expect the appearance of superconductivity when the orbital order is suppressed. In the present case, as shown in Fig. 2, there appears superconducting pairing state due to orbital fluctuations with \( (\delta, \delta, \delta) \) around at a quantum critical point \( \mu/\tau \approx 1.76 \). Note that orbital is not the conserved quantity, since there exists nonzero hopping amplitude between different orbitals. Thus, it is meaningless to define orbital singlet and triplet by analogy with spin singlet and triplet in the standard single-orbital Hubbard model. Here the superconducting pair is classified only by parity. In fact, we find that the superconducting state is labelled by even- and odd-parity, as shown in Fig. 2.

We remark that even- and odd-frequency components are mixed in the present case. In order to understand this point, it is convenient to redefine the anomalous self-energy as \( \phi_1(k)=\phi_{ab}(k) \), \( \phi_2(k)=\phi_{bb}(k) \), \( \phi_3(k)=\phi_{ab}(k)+\phi_{ba}(k) \), and \( \phi_4(k)=\phi_{ab}(k)-\phi_{ba}(k) \). First we note that the relation \( \phi_3(k)=-\phi_2(-k) \) always holds for \( j=1\sim 4 \), since it is due to the fermion property. The odd-parity solutions are characterized by \( \phi_i(k, i\omega_n)=-\phi_i(-k, i\omega_n) \) for \( i=1\sim 3 \) and \( \phi_4(k, i\omega_n)=-\phi_4(-k, i\omega_n) \). Note that \( \phi_3(k) \) has odd-frequency property. On the other hand, the even-parity solutions are characterized by \( \phi_i(k, i\omega_n)=\phi_i(-k, i\omega_n) \) for \( i=1\sim 3 \) and \( \phi_4(k, i\omega_n)=\phi_4(-k, i\omega_n) \). Note that \( \phi_4(k) \) with \( i=1\sim 3 \) have odd-frequency properties.

Let us first examine the odd-parity solution in the low-temperature region. In Fig. 3(a), we plot \( \phi_i(k) \) vs. \( \omega_n \) at \( \mu=\pi/3 \). As mentioned above, \( \phi_1(k) \) vs. \( \omega_n \) are even-frequency functions, while \( \phi_4 \) is odd-frequency one. The absolute value of \( \phi_4 \) is relatively smaller than those of the even-frequency ones. Since \( \phi_4(k) \) is exactly equal to the amplitude for the antisymmetric pair of electrons on different Fermi surfaces, the contribution of \( \phi_4 \) is suppressed.

Here \( J_{ab,ba}=J_{ba,ab}=L_{aa,ba}=L_{ab,aa}=U, \hat{I} \) denotes unit matrix, and \( G_{\tau_1,\tau_2}(q)=-\sum_k G_{\tau_1,\tau_2}(k+q)G_{\tau_2,\tau_1}(k) \). Here \( q=\left(q, \nu_n \right) \), \( q \) is the momentum, and \( \nu_n=2n\pi T \) is the boson Matsubara frequency. In the calculation, we use a \( 32 \times 32 \times 32 \) lattice and 1024 Matsubara frequencies.

In Fig. 3, we show the phase diagram in the \( (\mu, T) \) plane. Red and blue colors denote plus and minus signs, respectively. (c) Sign of \( \phi_3(k, i\pi T) \) with odd-parity on the Fermi surfaces.

![Fig. 3](image-url)

![Fig. 4](image-url)

![Fig. 4](image-url)
of Fermi surfaces in the first Brillouin zone. From the results, the pairing symmetry is found to be \( p \)-wave. We do not show the results for \( \phi_2 \) and \( \phi_4 \), but \( \phi_2 \) is similar to \( \phi_1 \) and the magnitude of \( \phi_4 \) is small in comparison with other components. The node positions of \( \phi_3 \) are different from those of \( \phi_1 \), but it is due to the difference in local symmetry of \( \Gamma_8^{-a} \) and \( \Gamma_8^{-b} \). In fact, we find \( \phi_3 \sim \phi_1 (\cos k_x - \cos k_y) \).

In Fig. 4, we show the results for even-parity solution in the high-temperature region. The \( n \) dependence is depicted in Fig. 4(a). We find that \( \phi_1 \)'s for \( i=1\sim3 \) are odd-frequency functions, while \( \phi_4 \) is even-frequency one. Also in this case, the contribution of \( \phi_4 \) is relatively small in comparison with other components. Thus, the even-parity solution is characterized by the odd-frequency components, leading to a way to observe peculiar odd-frequency pairing. From Fig. 4(b), the gap function is found to be characterized by \( d \)-wave. As observed in Fig. 4(c), \( \phi_3 \) seems to be \( s \)-wave, due to the relation of \( \phi_3 \sim \phi_1 (\cos k_x - \cos k_y) \).

Note that \( \mu \) is related to the valence of uranium ion. The critical point of \( \mu/(\approx 1.76) \) corresponds to \( \langle n \rangle \approx 1.6 \), i.e., \( U^{3.1+} \). The width of the superconducting region for the valence of uranium ion is the order of 0.01. Namely, the region is limited, but the value in the middle of \( U^{3+1} \) and \( U^{4+} \) is realistic for actual uranium metallic compounds. Thus, we believe that the superconductivity induced by orbital fluctuations could appear in ferromagnetic cubic uranium compounds.

Here we mention a possibility to apply the theory to manganites, which are well described by the double-exchange model. The superconducting region corresponds to \( \langle n \rangle \approx 1.6 \), which denotes \( \epsilon_g \) electron number for manganites. The situation indicates 0.4 electrons per manganese ion form the particle-hole symmetry. Thus, the situation is close to the half-doped manganites with orbital ordering. In cubic manganites with relatively wide bandwidth, the metallic ferromagnetic phase is known to appear near the orbital ordering. The pattern of orbital ordering is different from the present one, but it is expected to observe superconductivity in manganites with high quality near half-doping. We note that the superconducting transition temperature \( T_C \) in Fig. 2 seems to be higher than that of the single-band Hubbard model within the same RPA. The stabilization of the even-parity solution due to significant odd-frequency components seems to be relevant to the increase \( T_C \). Since this point may open a new route to high-\( T_C \) materials, further investigations will be required in future.

Five comments are in order. (1) We have discussed orbital ordering and superconductivity in the ferromagnetic phase, but in order to confirm that the Curie temperature is higher than the orbital-ordering temperature and \( T_C \), it is necessary to estimate the magnitude of Coulomb interaction among \( d \) orbitals. This point is out of the scope of this paper, but it is one of future problems. (2) We have pointed out that \( \Gamma_7^- \) becomes localized orbital when we take into account only \( \sigma \) bond for \( f \) electron hopping. In general, hopping amplitudes through \( \pi \) and \( \phi \) bonds appear and effective hoppings through ligand anions exist. Thus, \( \Gamma_7^- \) is not perfectly localized in actual systems. However, we still believe that orbital dependent duality has an important starting point for the discussion on ferromagnetism and superconductivity. (3) We have ignored normal self-energy effects, but it is possible to include them, for instance, in the fluctuation-exchange (FLEX) approximation. Without considering the vertex corrections, it overestimates the normal self-energy effect such as damping of quasi-particle, but in future, we can perform the FLEX calculation in the combination with dynamical mean-field approximation. (4) We have discussed superconductivity in the ferromagnetic phase from a microscopic viewpoint, but in actuality, it is necessary to consider how magnetic flux penetrates the system. If the magnetic flux forms some pattern such as the Abrikosov lattice, it indicates the ordering of localized \( \Gamma_7^- \) electrons carrying magnetic moments. This point may lead to an interesting possibility of the coupling between flux-lattice formation and spin-orbital order. (5) We have proposed the spinless model, but from a realistic viewpoint, we should include both majority and minority spin bands. However, the minority spin band is virtually ignored, when minority spin density is so small that the intra-orbital Coulomb repulsion is effectively reduced in comparison with inter-orbital Coulomb interaction, indicating that orbital fluctuations dominate spin ones.

In summary, we have proposed the double-exchange scenario for the emergence of ferromagnetism in cubic uranium compounds. We have found orbital-related quantum critical phenomena such as odd-parity \( p \)-wave and even-parity \( d \)-wave superconducting states in the vicinity of orbital-ordered phase. This orbital-fluctuation-induced superconductivity is expected to be found in ferromagnetic cubic uranium compounds and cubic perovskite manganites near the half-doping.

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