Anisotropic Superconductivity Emerging from the Orbital Degrees of Freedom in a $\Gamma_3$ Non-Kramers Doublet System

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We study superconductivity in a three-orbital model for $f^2$ ions with the $\Gamma_3$ crystalline electric field (CEF) ground state. An antiferromagnetic interaction between the $\Gamma_7$ and $\Gamma_8$ orbitals is introduced to stabilize the $\Gamma_3$ CEF state. This interaction also works as an on-site attractive interaction for spin-singlet pairing between electrons in these orbitals. The interorbital pairing state composed of the $\Gamma_7$ and $\Gamma_8$ orbitals on the same site has the $E_g$ symmetry. Indeed, by applying the random phase approximation, we find that the $E_g$ spin-singlet superconducting state is realized over a wide parameter range.

Unconventional superconductivity, such as in cuprate high-temperature superconductors and heavy-fermion materials, has been widely discussed. In particular, the magnetic-fluctuation-mediated superconducting mechanism has been widely discussed.

In addition, the orbital degrees of freedom may play an important role in superconductivity in orbitally degenerate systems. Indeed, there have been suggestions that orbital fluctuations are important in Fe-based superconductors. However, the presence of both orbital and spin degrees of freedom has made it difficult to discern the specific role of the orbital degrees of freedom.

On the other hand, in f-electron systems, an ion with an even number of f electrons has the $\Gamma_3$ non-Kramers doublet level under a cubic crystalline electric field (CEF). The $\Gamma_3$ state has the same symmetry as the spinless $e_g$ electron, and possesses quadrupole and octupole moments but no dipole moment. Thus, the $\Gamma_3$ system can be regarded as an ideal system for investigating orbital physics, and may provide a route to unconventional superconductivity other than the spin-fluctuation mechanism.

Superconductivity was observed in PrT$_2$X$_{20}$ (where T denotes a transition metal and X is Zn or Al) in which the CEF ground state of the $f^2$ electronic configuration in a Pr$^{3+}$ ion is the $\Gamma_3$ doublet. (Strictly, in PrRh$_2$Zn$_{20}$, the CEF ground state is the $\Gamma_{23}$ doublet owing to symmetry lowering at the Pr site that is induced by a structural transition.) In PrIr$_2$Zn$_{20}$, superconductivity is observed below the antiferroquadrupole ordering temperature. The order parameter for antiferroquadrupole ordering in PrIr$_2$Zn$_{20}$ was determined to be $O^2 = x^2 - y^2$. In PrTi$_2$Al$_{20}$, superconductivity appears below the ferroquadrupole ordering temperature of $O^2 = 3z^2 - r^2$. In PrRh$_2$Zn$_{20}$, superconductivity occurs simultaneously with antiferroquadrupole ordering. In any case, superconductivity is realized inside the quadrupole ordered phase. The relation between superconductivity and the quadrupole degrees of freedom has therefore attracted much attention.

To discuss superconductivity with quadrupole or orbital degrees of freedom, it may be useful to consult two-orbital models. In two-orbital models, there is an interesting possibility to realize anisotropic superconductivity originating from the orbital anisotropy. For example, we obtained d-wave spin-triplet superconductivity in a model for $e_g$ orbitals on a square lattice. However, it was shown that a two-orbital model is inadequate for describing the multipole degrees of freedom in the $\Gamma_3$ CEF state. For example, the intermediate $f^3$ state is always the $\Gamma_8$ state in the two-orbital model for the $\Gamma_3$ doublet, but for a realistic parameter set to realize the $f^2$-$\Gamma_3$ state in a local model considering all the f-electron orbitals, the $f^3$ ground state is the $\Gamma_6$ state. It is therefore necessary to look beyond the two-orbital model. Indeed, we have found that a three-orbital model can remedy the above shortcomings.

The present study considers the three-orbital model for the $\Gamma_3$ CEF state by applying the random-phase approximation (RPA) and clarifies the characteristics of superconductivity in the $\Gamma_3$ systems. In the present model, the $\Gamma_3$ doublet is composed of the two singlets between the $\Gamma_7$ and $\Gamma_8$ orbitals (Fig. 1). The interaction that stabilizes the $\Gamma_3$ doublet also works as an on-site attractive interaction for the spin-singlet pairing between the electrons in these orbitals. The orbital symmetry can be rewritten as $\Gamma_7 = \Gamma_2 \times \Gamma_6$ and $\Gamma_8 = \Gamma_3 \times \Gamma_6$, where $\Gamma_6$ describes the Kramers or spin degeneracy. Thus, the interorbital spin-singlet pairing state, composed of the $\Gamma_7$ and $\Gamma_8$ orbitals on the same site has the $E_g$ ($= \Gamma_3 = \Gamma_2 \times \Gamma_3$) symmetry. It is therefore natural to expect d-wave superconductivity in this model.

This study considers the f-electron states with total angular momentum $j = 5/2$ as one-electron states. These states split into the $\Gamma_7$ and $\Gamma_8$ states in a cubic CEF. The $\Gamma_7$ states at site $r$ are given by $c_{r71}^c|0\rangle = (1/\sqrt{6})(a_{r5/2}^x + \sqrt{5}a_{r3/2}^y)|0\rangle$ and $c_{r71}^s|0\rangle = (1/\sqrt{6})(a_{r5/2}^y - \sqrt{5}a_{r3/2}^x)|0\rangle$, where $a_{rj}^\alpha$ is the...
creation operator for the electron with \( j \) as the \( z \)-component of the total momentum at \( r \), and \( |0 \rangle \) denotes the vacuum state. The \( \Gamma_6 \) states are given by \( c_{\alpha l}^\dagger |0\rangle = (1/\sqrt{6})(\sqrt{3}a_{r-3/2} + a_{\tau-3/2}^\dagger) |0\rangle \), \( c_{\alpha l}^\dagger |0\rangle = (1/\sqrt{6})(\sqrt{3}a_{r-3/2} + a_{\tau-3/2}^\dagger) |0\rangle \), \( c_{\sigma l}^\dagger |0\rangle = a_{\tau-1/2}^\dagger |0\rangle \), and \( c_{\sigma l}^\dagger |0\rangle = a_{\tau-1/2}^\dagger |0\rangle \). In these states, \( \sigma = \uparrow \) or \( \downarrow \) denotes the Kramers degeneracy of the one-electron states. Although this is not a real spin, owing to spin-orbit coupling, we may nonetheless henceforth call it spin for simplicity.

To realize the \( \Gamma_3 \) state as the ground state of an \( f^2 \) ion, we consider an antiferromagnetic interaction \( J \) between electrons in the \( \Gamma_7 \) and \( \Gamma_8 \) orbitals (Fig. 1). While it may be possible to derive \( J \) by taking account of the effects of the higher-energy \( j = 7/2 \) states,\(^{29}\) we introduce it here phenomenologically to realize the \( \Gamma_3 \) state. This interaction favors spin singlets between these orbitals. These singlets are the main components of the \( \Gamma_3 \) doublet in realistic situations.\(^{27,29}\) When \( J \) is large enough, the ground state among the \( f^2 \) states is the \( \Gamma_6 \) doublet.\(^{27}\) The drawback of the two-orbital model discussed above is thus eliminated.

The model Hamiltonian is given by
\[
H = \sum_{\tau \rho, \gamma \gamma'} \left( \frac{\beta}{2} \right) c_{\tau \rho}^\dagger c_{\gamma \gamma'} + J \sum_{\rho} s_{\tau \rho} s_{\gamma \rho},
\]
where \( \beta_{\gamma \gamma'} \) is the hopping integral, with the vector \( \mu \) connecting nearest-neighbor sites and \( \gamma = (\tau, \sigma') \) with \( \tau = \alpha, \beta \), or \( 7 \). \( s_{\tau \rho} \) is \((1/2) \sum_{\sigma'} c_{\tau \rho}^\dagger \sigma \sigma' c_{\sigma' \rho}^\dagger\) and \( s_{\gamma \rho} \) is \((1/2) \sum_{\sigma'} c_{\tau \rho}^\dagger \sigma \sigma' c_{\sigma' \rho}^\dagger\) with \( \nu = \alpha \) or \( \beta \). \( \sigma \) are the Pauli matrices. Note that \( \beta_{\gamma \gamma'} \equiv \beta_{\gamma'} \gamma \), since \( H \) is Hermitian.

With regard to the kinetic energy term, as one of the simplest possible models, we consider only \( f \)-electron hopping through \( \sigma \) bonding \((ff\sigma)\) on a simple cubic lattice. In this case, the hopping integrals are nonzero only between the \( \Gamma_8 \) orbitals and they can be expressed as \( 4 \times 4 \) matrices.\(^{31,32}\) The hopping integrals are given by \( d_{\gamma \gamma'}^{(1,0,0)} = (1 - \tilde{\eta}) \gamma \), \( d_{\gamma \gamma'}^{(0,1,0)} = (1 - \tilde{\eta}) \gamma \), \( d_{\gamma \gamma'}^{(0,0,1)} = (1 - \tilde{\eta}) \gamma \), and \( d_{\gamma \gamma'}^{(0,0,1)} = (1 - \tilde{\eta}) \gamma \), where \( \tilde{\eta} = \delta_{\gamma \gamma'} \delta_{\gamma \gamma'} \), \( \tilde{\eta} = \delta_{\gamma \gamma'} \delta_{\gamma \gamma'} \), and \( \tilde{\eta} = (\pm \sqrt{3} k^2 - \bar{k})/2 \). We have set the lattice constant to unity and \( t = 3(\beta/4)\). The bandwidth is \( W = 12t \).

When strong electron correlations are properly included for the \( f^2 \) case (that is, with two electrons per site in this model), the electron number in each of the \( \Gamma_7 \) and \( \Gamma_8 \) levels should be nearly equal to one: \( n_7 = \left\langle \sum_{\tau \rho} c_{\tau \rho}^\dagger c_{\tau \rho} \right\rangle \approx 1 \) and \( n_8 = \left\langle \sum_{\tau \rho} c_{\tau \rho}^\dagger c_{\tau \rho} \right\rangle \approx 1 \), where \((\cdots)\) denotes the expectation value. Such strong correlation effects can be included partly within the RPA by fixing \( n_7 \) and \( n_8 \), via the independent tuning of the chemical potentials for these orbitals.

The gap equation is expressed as\(^{30,33}\)
\[
\lambda \Delta_{\gamma \gamma'}(k) = -\frac{1}{N} \sum_{k' \tau \gamma'} V_{\gamma \gamma'}^f (k - k') \phi_{\gamma \gamma'}(k') \Delta_{\gamma \gamma'}(k'),
\]
where \( N \) is the number of lattice sites and \( \Delta^f(k) [\Delta^f(k)] \) is the gap function for spin-singlet \([\text{-triplet}] \) pairing. The eigenvalue \( \lambda \) reaches unity at the superconducting transition temperature \( T_c \). The pair-correlation function \( \phi(k) \) is given by
\[
\phi_{\gamma \gamma'}(k) = T \sum_{i_\alpha} G_{\gamma \gamma'}(k, i_\alpha) G_{\gamma \gamma'}(-k, -i_\alpha),
\]
where \( T \) is the temperature and \( G_{\gamma \gamma'}(k, i_\alpha) \) is the non-interacting Green’s function. The pairing interactions are written as
\[
V_{\gamma \gamma'}(q) = \left[ U_{\gamma}^x \chi^x(q) U_{\gamma}^x + \frac{U_{\gamma}^z}{2} \right] - \frac{1}{2} \left[ U_{\gamma}^x \chi^x(q) U_{\gamma}^z - \frac{U_{\gamma}^z}{2} \right],
\]
\[
V_{\gamma \gamma'}(q) = \left[ U_{\gamma}^x \chi^x(q) U_{\gamma}^x + \frac{U_{\gamma}^z}{2} \right] - \frac{1}{2} \left[ U_{\gamma}^x \chi^x(q) U_{\gamma}^z - \frac{U_{\gamma}^z}{2} \right],
\]
where the spin and charge susceptibilities are
\[
\chi^{\alpha \beta}(q) = \left[ \chi^\alpha(q) \left[ 1 + \frac{U^2}{U^x \chi^x(q)} U^z \right] \right]^{-1},
\]
with
\[
\chi_{\gamma \gamma'}(k, q) = -\frac{1}{N} \sum_{i_\alpha} G_{\gamma \gamma'}(k, i_\alpha) G_{\gamma \gamma'}(-k, -i_\alpha).
\]

The matrices \( U^x \) and \( U^z \) are defined as \( U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = 2J_T \tau_q \), \( U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = 2J_T \tau_q \), \( U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = 2J_T \tau_q \), and \( U_{\gamma \gamma'}^{\alpha \beta} = U_{\gamma \gamma'}^{\alpha \beta} = 2J_T \tau_q \), and \( U^z = 2J_T \tau_q \), and \( U^z = 2J_T \tau_q \), and \( U^z = 2J_T \tau_q \), and \( U^z = 2J_T \tau_q \). In the evaluation of Eqs. (3) and (7), the summation over the fermion Matsubara frequency \( \epsilon_n \) can be executed analytically.

The multipole operators can be expressed in the form
\[
O_{\gamma \gamma'}(k, q) = \sum_{\gamma' \gamma''} c_{\gamma' \gamma''} O_{\gamma' \gamma''} c_{\gamma' \gamma''}.
\]

We normalize \( \bar{O} \) so that \( \text{Tr} \bar{O}^2 = 1 \), where \( \text{Tr} \) denotes the trace of the matrix. Using linear-response theory, we can calculate the multipole susceptibility.\(^{34}\) The susceptibility for a magnetic [electric] multipole moment can be expressed by using \( \chi^x(q) [\chi^{x}(q)] \). If we define the multipole ordering temperature from the divergence of the corresponding susceptibility \( X \), we will always find superconducting instability before multipole ordering takes place, owing to the enhancement of the pairing interaction. The root of this unrealistic consequence is the ignorance of the self-energy within RPA.\(^{35}\) In this study, we identify the multipole ordering temperature as the temperature where \( X \) reaches a particular threshold.

In the following, we show the results for a simple cubic lat-
transition temperature among the multipole-ordering and superconducting states, we found that only the conducting transitions. Having considered all the superconducting states and the other superconducting states and $T_{\text{SDW}}$ for this parameter.

Figure 2(a) shows the temperature dependences of the eigenvalue $\lambda$ for the $E_g$ spin-singlet pairing and of the susceptibility $\chi$ for $T^\alpha$ at $q_{\text{max}}$ for $n_\gamma = n_\delta = 1$ and $J = 5t$. (b) Wave vector dependences of the susceptibilities for $n_\gamma = n_\delta = 1$, $J = 5t$, and $T = 1.1t$.

Of size $N = 32 \times 32 \times 32$. We also performed calculations for a $16 \times 16 \times 16$ lattice and found the size dependence to be negligible except for the dilute cases $n_\delta \approx 0$. Concerning multipole ordering, we define the transition temperature by the condition $\chi = 10/t$. Then, we determine the highest transition temperature among the multipole-ordering and superconducting transitions. Having considered all the superconducting states, we found that only the $E_g$ spin-singlet pairing state displays the highest transition temperature. For multipole ordering, we consider the charge, dipole, quadrupole, and octupole moments. Among them, only the $\Gamma_4$ octupole ($T^4$) state is realized. It has the same symmetry as the dipole moment, and should appear simultaneously with the dipole moment in the ordered phase. We found that the susceptibilities for $T^\alpha$ and for the dipole moment diverge at the same temperature, if we ignore the superconducting instability. Thus, we determined the spin-density-wave (SDW) transition temperature $T_{\text{SDW}}$ by using the susceptibility for $T^\alpha$.

Figure 2(a) shows the temperature dependences of the eigenvalue $\lambda$ for the $E_g$ spin-singlet superconductivity and of the susceptibility $\chi$ for $T^\alpha$ at the wave vector $q_{\text{max}}$, where the susceptibility has the maximum value for $n_\gamma = n_\delta = 1$ and $J = 5t$. The value of $\lambda$ reaches unity at $T_c \approx 1.06n$. The susceptibility $\chi$ is not enhanced for this parameter set. Thus, the superconductivity is not mediated by such multipole fluctuations with a particular wave vector. The fluctuations just above the transition temperature depend weakly on the wave vector $q$.

![Fig. 2.](image)

![Fig. 3.](image)

Fig. 2. (Color online) (a) Temperature dependences of the eigenvalue $\lambda$ for the $E_g$ spin-singlet pairing and of the susceptibility $\chi$ for $T^\alpha$ at $q_{\text{max}}$ for $n_\gamma = n_\delta = 1$ and $J = 5t$. (b) Wave vector dependences of the susceptibilities for $n_\gamma = n_\delta = 1$, $J = 5t$, and $T = 1.1t$.

Fig. 3. (Color online) (a) $T_c$ for the $E_g$ spin-singlet pairing and $T_{\text{SDW}}$ as functions of $J$ for $n_\gamma = n_\delta = 1$. The open squares represent $T_c$ evaluated without $\chi(q)$ and $\chi(q)$ in Eq. (4). (b) $T_c$ for the $E_g$ spin-singlet pairing as a function of $n_\delta$ for $n_\gamma = 1$ and $J = 5t$. [Fig. 2(b)], that is, the local fluctuations may still be important for the emergence of superconductivity.

Figure 3(a) shows $T_c$ for the $E_g$ spin-singlet pairing and $T_{\text{SDW}}$ as functions of the strength of the antiferromagnetic interaction $J$. In the cases of weak interactions ($J \lesssim t$), the SDW state is realized but its transition temperature $T_{\text{SDW}}$ is very low. For stronger interactions ($J \gtrsim t$), the $E_g$ spin-singlet superconducting state is realized. The transition temperature $T_c$ increases with $J$. In a wide parameter region $J \lesssim W = 12t$, where we can apply weak-coupling theory, the $E_g$ spin-singlet superconducting state is realized. Note that, for a larger $J$, local spin-singlet formation (which is beyond the scope of the present weak-coupling treatment) should become strong, leading to the suppression of superconductivity. In Fig. 3(a), we also show $T_c$ evaluated without $\chi(q)$ and $\chi(q)$ in Eq. (4). In this case, $T_c$ becomes much lower. We therefore recognize that the local fluctuations $\sim \sum_q \chi(q)$ are still important for the realization of superconductivity and that we cannot ignore $\chi(q)$, even though $\chi(q_{\text{max}})$ is not large.

To examine the stability of the $E_g$ spin-singlet superconducting state, we varied the electron number $n_\delta$ in the $\Gamma_4$ level from unity. For example, we show the $n_\delta$ dependence of $T_c$ for $J = 5t$ in Fig. 3(b). We find that $T_c$ is always higher than for the other superconducting states and $T_{\text{SDW}}$ for this parameter. Thus, the $E_g$ spin-singlet state is not restricted to $n_\delta = 1$.

By determining the highest transition temperature for each parameter set, we constructed the phase diagram shown in
Fig. 4. (Color online) Phase diagram for \( n_7 = 1 \).

Fig. 4. The change in \( J \) corresponds to the external hydrostatic pressure and the change in \( n_8 \) is regarded as carrier doping. In this phase diagram, the \( E_g \) spin-singlet superconductivity is realized over a wide range of parameters. This superconductivity would therefore be robust against such perturbations. Notably, this superconducting phase extends far from the SDW phase and does not require particular fluctuations. Even in an unrealistic two-orbital model for the \( \Gamma_3 \) CEF state, \( E_g \) superconductivity was obtained,\(^{20}\) indicating that \( E_g \) superconductivity is relatively stable in \( \Gamma_3 \) systems.

In summary, we have investigated superconductivity in the \( \Gamma_3 \) non-Kramers doublet system. The \( E_g \) spin-singlet pairing state originating from the orbital degrees of freedom is realized over a wide parameter range. This superconductivity is not mediated by fluctuations with a particular wave vector and would therefore be stable against perturbations.

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