Multipole Fluctuations in Filled Skutterudites

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In order to clarify exotic multipole properties of filled skutterudites, we evaluate multipole susceptibility for n=1~5, where n is the local \( f \)-electron number, on the basis of a multiorbital Anderson model constructed using the \( j-j \) coupling scheme. For \( n=1 \), magnetic fluctuations dominate over low-temperature electronic properties, while for \( n=2 \) and 4, electronic states are dominated by both magnetic and quadrupole fluctuations. For \( n=3 \) and 5, octupole fluctuations are found to be significant, depending on the crystalline electric field potential. We discuss possible relevance of the results to actual materials.

KEYWORDS: Multipole susceptibility, Filled skutterudites, Multiorbital Anderson model, \( j-j \) coupling scheme, Numerical renormalization group method

Recent discovery of heavy-fermion superconductivity in PrOs\(_4\)Sb\(_{12}\) has triggered a rapid increase of research activities on filled skutterudite compounds.\(^1,2\) This material is believed to belong to the group of strongly correlated electron systems, but the superconductivity cannot be simply understood in terms of \( d \)-wave pairing mediated by antiferromagnetic spin fluctuations, since the inverse of the nuclear spin-lattice relaxation time for PrOs\(_4\)Sb\(_{12}\) does not show the \( T^3 \) behavior at low temperature \( T \) characteristic of a line-node gap.\(^3\) In addition, PrOs\(_4\)Sb\(_{12}\) exhibits several exotic superconducting features such as multiple superconducting phases,\(^4\) point-node behavior in the gap function,\(^4,5\) and the breaking of time-reversal symmetry as detected by \( \mu \)SR experiments.\(^6\)

It is premature to settle on the mechanism of superconductivity in PrOs\(_4\)Sb\(_{12}\), but recently, the possibility of exotic pairing due to quadrupole fluctuations has been suggested experimentally\(^7\) and theoretically.\(^8\) In fact, neutron scattering experiments have revealed that a quadrupolar ordered state is induced by a magnetic field in the vicinity of the superconducting phase of PrOs\(_4\)Sb\(_{12}\),\(^9\) implying a potential role for quadrupole fluctuations in superconducting pair formation. Moreover, PrFe\(_4\)P\(_{12}\) exhibits a second-order phase transition at 6.5K,\(^10\) but this transition is considered to be due to antiferro-quadrupolar ordering from neutron scattering experiments\(^11\) and theoretical efforts.\(^12,13\) It has been gradually recognized that the quadrupolar degree of freedom plays a crucial role in filled skutterudite compounds.

From the theoretical viewpoint, multipole phenomena in \( f \)-electron systems have been investigated phenomenologically on the basis of the \( LS \) coupling scheme. This Heisenberg-like model for relevant multipole moments could successfully explain some of the experimental results for filled skutterudites. In particular, the phase diagram for PrOs\(_4\)Sb\(_{12}\) has been reproduced\(^14\) in a quasi-quartet system with \( \Gamma_1 \) ground and \( \Gamma_4^{(2)} \) excited states with a very small excitation energy,\(^15\) for a crystalline electric field (CEF) potential with \( T_1 \) symmetry.\(^16\) However, the microscopic approach to electronic properties of filled skutterudites has been very limited mainly due to the complexity of multipole degrees of freedom.

In order to overcome such difficulties, it has been proposed to construct a microscopic model for \( f \)-electron systems based on the \( j-j \) coupling scheme.\(^17\) With this approach, for instance, the microscopic origin of octupole ordering in NpO\(_2\) has recently been established.\(^18\) Furthermore, the magnetism and superconductivity of filled skutterudite materials have been studied from a microscopic viewpoint on the basis of a multiorbital Anderson model.\(^19,20\) However, the effect of multipole fluctuations has not been investigated satisfactorily at the microscopic level, even though their potential role has been emphasized in research on filled skutterudites. In the \( j-j \) coupling scheme, we can microscopically evaluate physical quantities related to multipoles by treating them as combined spin and orbital degrees of freedom. Such a study is believed to open a door which leads to a new stage for multipole physics of \( f \)-electron systems.

In this Letter, we attempt to clarify what kind of multipole fluctuations are dominant in filled skutterudites, by analyzing a multiorbital Anderson model based on the \( j-j \) coupling scheme for \( n=1\sim5 \), where \( n \) denotes local \( f \)-electron number. For this purpose, we evaluate susceptibilities for fifteen kinds of dipole, quadrupole, and octupole moments up to rank 3 using a numerical technique. In this way, we find that for \( n=1 \), magnetic fluctuations are dominant at low temperatures, while for \( n=2 \) and 4, electronic states are dominated by magnetic and quadrupole fluctuations at low temperatures. For \( n=3 \) and 5, octupole fluctuations are found to appear for some ranges of CEF parameters. We provide some explanations and predictions on filled skutterudites from the present results.

In general, in the \( j-j \) coupling scheme, a large spin-orbit interaction is first assumed, and then, only the \( j=5/2 \) sextet is taken into account. Concerning the applicability of the \( j-j \) coupling scheme to rare-earth compounds, readers can consult Ref. 20. Note that the main conduction band of filled skutterudites is \( a_\text{u} \) with \( xyz \) symmetry,\(^21\) where the subscript “\( u \)” means ungerade, i.e., odd parity for space inversion. Then, the Anderson Hamiltonian is written as

\[
H = \sum_{k\sigma} \varepsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k\sigma} (V_{c_{f\sigma}} f_{\sigma} + \text{h.c.}) + H_{\text{loc}},
\]

where \( \varepsilon_k \) is the dispersion of \( a_\text{u} \) conduction electrons with \( \Gamma_7 \) symmetry in terms of the \( j-j \) coupling scheme, \( c_{k\sigma} \) is the annihilation operator for conduction electrons with momentum \( k \) and spin \( \sigma \), \( f_{\gamma\sigma} \) is the annihilation operator of \( f \) electrons on the impurity site with pseudospin \( \sigma \) and orbital \( \gamma \), \( V \) is the hybridization between conduction and \( f \) electrons with \( \Gamma_7 \) symmetry, and \( H_{\text{loc}} \) is the local \( f \)-electron interaction term.
The orbital index $\gamma$ is introduced to distinguish three kinds of the Kramer doublets, two $\Gamma_8$ and one $\Gamma_7$. Here “a” and “b” denote the two $\Gamma_8$’s and “c” indicates the $\Gamma_7$. Throughout this paper, we set $V=0.05$, where the energy unit is taken as $D$, half of the bandwidth of the conduction band. From band-structure calculations, the bandwidth is estimated as 2.7 eV in PrRu$_4$P$_{12}$, indicating that $D=1.35$ eV.

The local $f$-electron term $H_{loc}$ is given by\(^17\)

$$H_{loc} = \sum_{\gamma} B_{\gamma} f_{\gamma 1}^d f_{\gamma 1}^{\dagger} + (1/2) \sum_{\gamma_1 \neq \gamma_2} \sum_{\sigma_1, \sigma_2} U_{\gamma_1 \gamma_2}^{\sigma_1 \sigma_2} f_{\gamma_1 \gamma_2}^{\sigma_1} f_{\gamma_2 \gamma_1}^{\sigma_2} (1 - \delta_{\sigma_1 \sigma_2}) + \sum_{\gamma} \delta f_{\gamma 1},$$

where $B_{\gamma}$ is the CEF potential, which is already diagonalized. It is convenient to introduce a level splitting $\Delta$ between $\Gamma_7$ and $\Gamma_8$ as $\Delta = B_{\gamma}^8 - B_{\gamma}^7$. The Coulomb integral $U$ in the $j$-$j$ coupling scheme is expressed in terms of three Racah parameters, $E_0$, $E_1$, and $E_2$.\(^17\) To set the local $f$-electron number, we adjust the $f$-electron chemical potential for each $n$.

In order to reproduce the CEF scheme for Pr-based filled skutterudites, we prefer positive $\Delta$. In particular, the quasiquartet situation with singlet ground and triplet excited states is reproduced by taking a small positive $\Delta$. However, since the difference between $O_h$ and $T_h$ is not fully included in the $j$-$j$ coupling scheme,\(^20\) the excited state for $n=2$ is always $\Gamma_4$ for $\Delta>0$, although $\Gamma_4^{(2)}$ (a mixture of $\Gamma_4$ and $\Gamma_6$) is the ground state in actual material. The extent of the mixture explains the difference between PrOs$_4$Sb$_{12}$ and PrFe$_4$P$_{12}$\(^23\) but we simply ignore such effect. It is one of future problems to include the effect of $T_h$ in the $j$-$j$ coupling scheme.

In order to discuss multipole properties, we evaluate multipole susceptibilities for $f$ electrons, given by

$$\chi_X^{\gamma} = \frac{1}{Z} \sum_n e^{-E_n/T} \langle m | X_{\Gamma\gamma} | n \rangle^2,$$

where $X_{\Gamma\gamma}$ is the multipole operator, $X$ denoting the multipole symbol, $\Gamma$ is the irreducible representation with $\gamma$ to distinguish degenerate representations, $E_n$ is the energy eigenvalue for the $n$-th eigenstate $|n\rangle$, and $Z$ is the partition function given by $Z=\sum_n e^{-E_n/T}$. For $j=5/2$, we can define multipole operators up to rank 5 in general, but we are primarily interested in multipole properties from the $\Gamma_8$ quartet. Thus, we concentrate on multipole moments up to rank 3.\(^24\) Note that in the $j$-$j$ coupling scheme, physical quantities for multi-$f$-electron systems can be evaluated from one-particle operators on the basis of quantum-field theory, as we have performed for d-electron systems. We have the clear advantage that it is not necessary to redefine $X_{\Gamma\gamma}$ depending on the ground-state multiplet, when we change local $f$-electron number.

Now let us show explicit forms for multipole operators following Ref. 24. As for dipole moments with $\Gamma_4$ symmetry, we express the operators as

$$J_{aux} = J_x, \quad J_{auy} = J_y, \quad J_{azu} = J_z,$$

where $J_x$, $J_y$, and $J_z$ are three angular momentum operators for $j=5/2$, respectively. Concerning quadrupole moments, they are classified into $\Gamma_{3g}$ and $\Gamma_{5g}$, where $g$ denotes gerade. We express the $\Gamma_{3g}$ quadrupole operators as

$$O_{3gu} = (2J_x^2 - J_z^2 - J_y^2)/2, \quad O_{3gy} = \sqrt{3}(J_x^2 - J_y^2)/2,$$

For the $\Gamma_{5g}$ quadrupole, we have the three operators

$$O_{5g0} = \sqrt{3}(J_x^2 + J_y^2)/2, \quad O_{5g1} = \sqrt{3}(J_x^2 - J_y^2)/2, \quad O_{5g2} = \sqrt{3}(J_y^2 - J_x^2)/2,$$

where the bar denotes the operation of taking all possible permutations in terms of cartesian components.

Finally, regarding octupole moments, there are three types as $\Gamma_{2u}$, $\Gamma_{4u}$, and $\Gamma_{5u}$. We express the $\Gamma_{2u}$ octupole as

$$T_{2u} = \gamma J_x J_y J_z/6.$$

For the $\Gamma_{4u}$ octupole, we express the operators as

$$T_{4ux} = (2J_x^3 - J_x J_y^2 - J_x J_z^2)/2, \quad T_{4uy} = (2J_y^3 - J_x J_z^2 - J_y J_z^2)/2, \quad T_{4uz} = (2J_z^3 - J_x J_y^2 - J_y J_z^2)/2,$$

while the $\Gamma_{5u}$ octupole operators are given by

$$T_{5ux} = \sqrt{15}(J_x^2 J_y^2 - J_x J_y J_z)/6, \quad T_{5uy} = \sqrt{15}(J_y^2 J_z^2 - J_y J_z J_x)/6, \quad T_{5uz} = \sqrt{15}(J_z^2 J_x^2 - J_z J_x J_y)/6.$$

For the evaluation of multipole susceptibility, we resort to the numerical renormalization group (NRG) method,\(^25\) in which momentum space is logarithmically discretized to include efficiently the conduction electrons near the Fermi energy. In actual calculations, we introduce a cut-off $\Lambda$ for the logarithmic discretization of the conduction band. Due to the limitation of computer resources, we keep $m$ low-energy states. In this paper, we set $\Lambda=5$ and $m=3000$. Note that the temperature $T$ is defined as $T=\Lambda^{-(N-1)/2}$ in the NRG calculation, where $N$ is the number of the renormalization step.

In Figs. 1(a)-(e), we show the temperature dependence of $T^{\Gamma_X}$ for $E_0=5$, $E_1=2$, and $E_2=0.5$ with $\Delta=10^{-5}$. We note that such a small value of $\Delta$ is taken to reproduce the CEF level scheme for Pr-based filled skutterudites.\(^19,20\) Note also that we show one of $\chi^{\Gamma_X}$’s in each irreducible representation, since susceptibilities in the same irreducible representation take the same values owing to cubic symmetry. First of all, in the degenerate region with very small $\Delta$, electron-hole symmetry approximately holds within the $j=5/2$ sextet. Namely, the results for $n=1$ and $n=2$ are quite similar to those for $n=5$ and $n=4$, respectively, as observed in Figs. 1(a)-(e). For $n=1$ and 5, at low temperatures, we observe that $\Gamma_{4u}$ magnetic octupole fluctuations are predominant. Note that the fluctuations of the $\Gamma_{4u}$ dipole also survive, since it belongs to the same symmetry as the $\Gamma_{4u}$ octupole. Also, for $n=3$, there remain magnetic fluctuations of the $\Gamma_{4u}$ dipole and octupole.

For $n=2$ and 4, on the other hand, we find significant fluctuations for the $\Gamma_{5g}$ and $\Gamma_{3g}$ quadrupoles, in addition to those of the $\Gamma_{4u}$ dipole and octupole. Let us explain the results for the case of $n=2$. As shown in Fig. 1(f), for $\Delta>0$, the local $f$-electron ground state is a $\Gamma_1$ singlet composed of two $\Gamma_7$ electrons. The first excited state is a $\Gamma_4$ triplet formed by $\Gamma_7$ and $\Gamma_8$ electrons, while the second excited state is a $\Gamma_5$ including a couple of $\Gamma_8$ electrons. Even if the $\Gamma_1$ singlet is the ground state, there exists a $\Gamma_4$ triplet state with very small excitation energy. This $\Gamma_4$ triplet state carries a quadrupole moment. Thus, significant quadrupole fluctuations remain for $n=2$. The same explanation works also for $n=4$ due to the electron-hole symmetry for very small $\Delta$. 
Now we consider the case with larger $\Delta$, since the CEF potential is easily changed due to the substitution of rare-earth ion R, transition metal ion T, and pnictogen X for the filled skutterudite $RTX_{12}$. In Figs. 2 (a)-(e), we show the temperature dependence of $T_X\chi^f$ for $\Delta=0.1$. For $n=1$, the low-temperature behavior of multipole susceptibilities is almost unchanged from those for $\Delta=10^{-5}$. Since one $f$ electron is always located in a $\Gamma_7$ orbital, $\Gamma_5$ is irrelevant for the case of $n=1$. Since the Kondo temperature is very low for $V=0.05$, the local moment of the $\Gamma_7$ electron still persists in the present temperature range, but it should be screened eventually for extremely low $T$. For $n=2$, the $\Gamma_1$ singlet ground state is well separated from the $\Gamma_4$ triplet excited state and thus, we do not observe any fluctuations at low temperatures.

For $n=3$ and 5, we note another electron-hole relation in the $\Gamma_8$ quartet for very large $\Delta$, as is easily understood from Fig. 2(f). In fact, the main results in Figs. 3(c) and (e) look similar to each other. In particular, we find that $\Gamma_{2u}$ octupolar fluctuations are predominant at low temperatures for $n=3$ and 5. Note that $\Gamma_{4u}$ and $\Gamma_{5u}$ octupolar fluctuations also become significant for $n=5$. Enhancement of octupolar fluctuations is a characteristic point for $n=3$ and 5 in the region of large $\Delta$. Finally, for $n=4$ with large $\Delta$, since the $\Gamma_7$ levels are fully occupied, a couple of electrons in the $\Gamma_8$ quartet form a local $\Gamma_5$ triplet, leading to dominant magnetic fluctuations.

In order to see the change of multipole fluctuations, it is convenient to show the Curie constant for multipole susceptibility, defined as $C_{X}^{\gamma} = T_X \chi_{X}^{\gamma} / T = 10^{-10}$. In Figs. 3(a)-(e), we plot $C_{X}^{\gamma}$ vs. $\Delta$ for each $n$. In Fig. 3(f), we summarize the multipole triples for $n=1$. As mentioned above, the multipole properties do not depend on $\Delta$. Note again that for $T < 10^{-10}$, the local moment of the $\Gamma_7$ electron should be screened eventually. For $n=2$, in the region of $\Delta < 5 \times 10^{-5}$, both magnetic and quadrupole fluctuations are dominant, while for $\Delta > 5 \times 10^{-5}$, multipole fluctuations are all “dead”. For $n=3$, in the region of $\Delta < 0.02$, $\Gamma_{4u}$ magnetic octupole and dipole fluctuations are dominant, even though the absolute values are small. On the other hand, for $\Delta > 0.02$, $\Gamma_{2u}$ octupolar fluctuations gradually die out as $\Delta$ increases. Finally, for $n=5$, in the region of $\Delta < 2 \times 10^{-4}$, $\Gamma_{4u}$ magnetic octupole fluctuations are predominant, while for $\Delta > 2 \times 10^{-4}$, fluctuations of the $\Gamma_{2u}$ octupole are dominant and those of $\Gamma_{4u}$ and $\Gamma_{5u}$ are subordinate, similar to the results for $n=3$ with very large $\Delta$, due to electron-hole symmetry in the $\Gamma_8$ subspace.

Next we discuss relevance of our results to $RT_{4}X_{12}$. Although the actual system is not exactly described by the impurity model, the multipole properties are deduced from the dominant fluctuations. The case of $n=1$ would seem to correspond to the case of R=Ce, but in filled skutterudites, the cerium ion is considered to be tetravalent, rather than trivalent. Thus, it may be difficult to claim a direct relation between the $n=1$ results and Ce$_{T}X_{12}$. For $n=2$, on the other hand, we expect a clear correspondence to the case of R=Pr, in which we confirm significant magnetic and quadrupole fluctuations at low temperatures. The range of $\Delta$ with magnetic and quadrupole fluctuations is very narrow, which may be related to the experimental fact that the appearance of exotic superconductivity is very limited in Pr-based filled skutterudites.

The cases of $n=3$ and 5 correspond to R=Nd and Sm, respectively. For both cases, in the region of small $\Delta$, the predominance of magnetic fluctuations suggests that magnetic ordering occurs in actual systems, although we have not determined whether it is ferro or antiferro. If it is possible to increase $\Delta$ experimentally for Nd- and Sm-based filled skutterudites, we expect exotic electronic properties to result from octupolar fluctuations, including $\Gamma_{2u}$ and $\Gamma_{5u}$. In fact,
for SmRu₄P₁₂, signs of octupole ordering have been suggested by elastic constant measurements and μSR experiments. When we turn our attention to actinides, quite recently, NpFe₄P₁₂ has been successfully synthesized. Since the neptunium ion is considered to be tetravalent, Np-based filled skutterudites correspond to \( n=3 \). Thus, it may be natural to consider that the low-temperature properties of Np-based filled skutterudites are dominated by magnetic fluctuations.

It is difficult to find actual filled skutterudite materials corresponding to \( n=4 \), but Pu-based filled skutterudites may be good candidates if they can be synthesized, since actinide ions are tetravalent in the filled skutterudite structure. For \( n=4 \) with relatively small \( \Delta \), based on an approximate electron-hole relation in the \( j-j \) coupling scheme, one expects to find electronic properties similar to those of Pr-based filled skutterudites. In particular, exotic superconductivity may also occur in Pu-based filled skutterudites with relatively high transition temperatures, since hybridization is large in actinide compounds owing to the difference in the nature of \( 4f \) and \( 5f \) electrons. This point should be carefully discussed, when Pu-based filled skutterudites are synthesized at some future point.

Two comments are in order. (i) In this paper, we have simply ignored the effect of the rattling motion of rare-earth ion, but it is interesting to see whether magnetic and quadrupolar fluctuations are enhanced or not due to the effect of rattling in Pr-based filled skutterudites. This point can be investigated based on an extended model incorporating further electron-phonon interaction terms into \( H_{\text{loc}} \). This is among possible future developments. (ii) We have suggested \( \Gamma_{2n} \) for the dominant type of octupole moment for \( n=5 \), but in the \( j-j \) coupling scheme, the effect of \( T_1 \) symmetry is not fully included. Since \( \Gamma_{4u} \) and \( \Gamma_{5u} \) octupole moments should also occur in actual filled skutterudites, we cannot conclude which is the winner among octupole moments for \( R=\text{Sm} \) from the present results. To resolve this point, it is necessary to analyze a model including seven \( f \) orbitals, as has been done in Ref. 20. This is another future problem.

In summary, we have analyzed the multiorbital Anderson model to evaluate multipolar susceptibilities. It has been found that for \( n=1 \), magnetic fluctuations are always significant at low temperatures. For \( n=2 \), electronic states are dominated by magnetic and quadrupolar fluctuations at low temperatures, while multipolar fluctuations are dead for large \( \Delta \). For \( n=3 \) and 5, at small \( \Delta \) magnetic fluctuations are dominant, while for large \( \Delta, \Gamma_{2n} \) octupolar fluctuations are found to occur. For \( n=4 \) at small \( \Delta \), the situation is quite similar to the case of \( n=2 \), but for increasing \( \Delta \), quadrupolar fluctuations gradually die out.

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Fig. 3. Curie constant for each multipole susceptibility as a function of \( \Delta \) for (a) \( n=1 \), (b) \( n=2 \), (c) \( n=3 \), (d) \( n=4 \), and (e) \( n=5 \). (f) Summary for the types of relevant multipole. Here “mag.” indicates magnetic, while “quad.” indicates quadrupole. The boundary between small and large \( \Delta \) depends on \( n \), but we simply ignore such a quantitative point in this table.