Dear Editor,

We resubmit our manuscript retitled *Unconventional Superconductivity arising from Multipolar Kondo Interactions* to SciPost with appropriate revisions addressing all comments raised by the referees. Details of our responses to the referee comments are provided below, as well as the changes made in the manuscript (in blue font in the revised manuscript).

Sincerely yours,
Adarsh S. Patri
Yong Baek Kim

RESPONSE TO THE REFEREE 1

In the report, the referee discusses the importance of our study to understanding the superconducting nature of strongly correlated non-Kramers systems. We address the interesting questions and comments below.

(1) In Sec. 3 and Appendix C, the authors describe the electron-electron interactions from multipolar Kondo effects. However, the assumed condition to obtain the attractive force for the superconductivity is unclear. The authors should clarify such condition.

We thank the referee for raising this important point. Equations (C7)–(C10) describe the various potential terms contributing to the electron-electron interaction of Eq. (C6). Indeed, the potential is composed of three pieces: (i) an explicit momentum dependence $\mathbf{q}$, (ii) matrix structure from the Kondo interaction (the $\Gamma^{x,y,z}$ matrices), and (iii) phenomenological order parameters $m_{\mathcal{Q}}$, $m_{\mathcal{O}}$, $a_{0,1,2}$. As such depending on the location in momentum space, different sections of the interaction potential become attractive ($\mathcal{V}_{\alpha\beta\gamma\delta} > 0$) or repulsive ($\mathcal{V}_{\alpha\beta\gamma\delta} < 0$). The values of phenomenological Landau values (given at end of Appendix H) were chosen so as to connect with the experimental setting. In particular, the quadrupolar mass term ($m_{\mathcal{Q}} \sim (T - T_{\mathcal{Q}})$) was taken to be smaller than the octupolar mass ($m_{\mathcal{O}} \sim (T - T_{\mathcal{O}})$) to reflect the fact the proposed octupolar ordering temperature $T_{\mathcal{O}}$ is lower than the quadrupolar ordering temperature $T_{\mathcal{Q}}$; here T is the temperature, which is taken to be above $T_{\mathcal{Q},\mathcal{O}}$ in the paramagnetic phase (Ref. 65). The choices of the other phenomenological values $a_{0,1,2} > 0$ were chosen to reflect the stiffness associated with spatial fluctuations of the order parameters; mathematically, these choices also ensured that the Gaussian multipolar action was non-singular. With these physically motivated choices, we were able to uncover a large number of non-trivial superconducting states, with their own characteristic quasiparticle excitation features. One may indeed be able to select other numerical choices for these parameters, as well as modify the conduction electron band structure. Such changes may result in stabilizing some of the other pairing functions that were not found with our choice of parameters; for instance, the $A_2^{J=3}$ superconducting state may be realized by changes to the Landau parameters. Ultimately, the determination of which order parameter may describe the superconducting ground state would require additional microscopic information (such as the conduction electron density of states that mediate the multipolar fluctuations) to connect with ongoing/recent experiments.

We highlight the physical reasoning behind the choices for the phenomenological parameters in Appendix H.

(2) In Fig. 4, the authors show gapless nodes for the odd-parity order parameters. However, it is still difficult for readers to visualize. The authors should revise it, for example, by showing the Bogoliubov Fermi surface. Besides, the labels for the subfigures (a), (b) etc. should be added.

The Bogoliubov Fermi *surfaces* depicted in Figure 4 are in fact point nodes indicated by the blue dots, while the orange sphere is the itinerant electron Fermi surface. The indicated dashed lines were used to guide the eye along the various cubic axes. We have added such information to the figure caption as well as clearly indicated the subfigure labels to aid the reader.

(3) In Sec. 6, the authors describe the properties of the relevant superconducting state found in this research. In my view, it is helpful for readers if you can include some description about the topological feature and whether time reversal symmetry is preserved or not for the relevant superconducting states.

We thank the referee for this interesting question. The time-reversal properties of the superconducting state can be examined by considering the complex nature of the non-vanishing order parameters. To be specific, a pairing $|\mathbf{\Delta}\rangle$ can be written as a linear combination over the "real" angular momentum basis states,

$$|\mathbf{\Delta}\rangle = \sum_u \Delta_u \, |O_h; u\rangle \,, \tag{1}$$

where $|O_h; u\rangle$ are the time-reversal invariant cubic irrep basis function given in Appendix E, and u sums over the cubic irreps, for example $|E^{J=2}\rangle$ etc. In this basis, time-reversal symmetry even order parameters are thus given by order parameters Δ_u that are equal to their complex conjugate (up to a global phase); we have appropriately added in the global factor of i into the definition of the cubic irreps and the Cooper pair operators in the manuscript to make this explicitly apparent.

Thus, for the non-trivial pairing states arising from the two-channel Kondo interaction (given in Table 1), the even J states are time-reversal invariant, while the odd J break time-reversal. More specifically, $A_1^{J=0}$ and $E^{J=2}$ are time-reversal invariant, while $T_2^{J=3}$, $T_\pm^{J=1,3}$, $T_2^{J=1,3}$ break time-reversal symmetry. These correspond to the non-vanishing order parameters realized in Fig. 1. For the novel Kondo interaction pairing states, we have two possibilities (Figs. 2, 3). Focussing on Fig. 2, these states also follow the same prescription: even (odd) J states preserve (break) time-reversal symmetry. For the pairing states formed from one electron from $j = 1/2$ and $j = 3/2$ (Fig. 3), interestingly both $E_-^{J=2}$ and $T_2^{J=2}$, $T_-^{J=1}$ break time-reversal symmetry in the mean-field theory solutions. Indeed, a thorough topological classification of the pairing states (realized in the paramagnetic phase discussed in this work, as well as the superconducting states coexisting with multipolar ordering as in ongoing experiments, Ref. 75) is an important and intriguing direction that we suggest for future theoretical studies.

We update our manuscript to describe the time-reversal properties of the corresponding pairing states.

(4) The authors use the point-group irrep to classify the pairing functions. I understand this is theoretically rigorous for the spin-orbit coupled system, but I think such notation is not accessible for experimentalists and non-experts. I would recommend the authors to provide the relation of the point-group irrep to the conventional notation such as p-wave or d-wave with some figures illustrating the order parameters in k-space?

The point group irrep classification is used to categorize the total angular momentum J nature of the Cooper pair. Indeed, this is distinct from the momentum $\mathbf{k}$-space distribution of the pairing function typically used to describe conventional superconductors. As one may see in Eqs. (C7)–(C10), the pairing potentials (and subsequently the pairing functions) have a complicated momentum space distribution. For example, the $E^{J=2}$ irrep from the two-channel Kondo interaction in Eq. F2 has $\mathbf{k}$-space distribution that includes both an isotropic contribution from f_1 as well as $d_{x^2-y^2}$ and $d_{2z^2-x^2-y^2}$ contributions from $f_{2\nu}$ and $f_{2\mu}$, respectively. As well, the "mixing" between the two components of the aforementioned $E^{J=2}$ irrep further complicates the momentum space distribution. We depict the real and imaginary components of the realized order parameters in newly constructed Appendix I. As well, to clarify any possible source of confusion, we emphasize the distinction of the $\mathbf{k}$-space distribution acquired from the pairing potentials and the point group classification in the main text.

(5) There are some errors in the reference list; Ref. [12] and [13] are duplicated, and there are stylistic errors in the publication titles (for example, URu_2Si_2, UPt_3 , UNi_2Al_3, Knight shift, etc. are not capitalized). Authors should check carefully and revise them.

We have corrected the stylistic errors for the relevant references in the revised manuscript.

RESPONSE TO THE REFEREE 2

We thank the referee for highlighting the rigorous nature of our work and for the applicability of our study to understanding novel forms of superconductivity arising from multipolar fluctuations. We address the insightful comments raised by the referee below.

(1) While I think that the authors calculations are sound, the interpretation seems incorrect, or at least poorly explained. The symmetry aspects here are not clear. The authors claim that superconducting order parameters (OPs) in different irreducible representations coexist, but it is really not clear what they mean. Do they mean something like $s + id$ pairing, where the OPs couple quadratically? They discuss the Cooper pairs scattering off of one another, which suggests a quadratic coupling. However, the language in the main paper, and the appendices (particularly F and G) suggest a linear coupling. If that is the case, these are not different irreps at all, and the effect of the different symmetrizing/antisymmetrizing, spin-orbit coupling terms has just not been properly accounted for. I would like to see a Ginzburg-Landau theory for the superconducting order parameters, with the final irrep of the order parameter given. Given that the high temperature state is paraquadrupolar, there should be no linear couplings between distinct irreps allowed.

The variety of possible superconducting states examined in this work are characterized by the total angular momentum J of the Cooper pair. Due to cubic nature of the system, these angular momentum states are further classified in terms of irreducible representations of the O_h point group. Here, the O_h point group is the symmetry group of the high-temperature paramagnetic phase. Moreover, these Cooper pair operators have been appropriately anti-symmetrized to satisfy Fermi-Dirac statistics as described in Sec. 4, and as we have explicitly verified with the Cooper pair operators presented in Appendix E. Now, as seen in Figures 1,2,3 and the complete Hamiltonian in Appendices F and G, there exist "mixing" or "cross" terms between the various Cooper pair irreps. The reason for this "mixing" is due to the momentum-space dependent pairing potentials f_0, f_1 (that contain isotropic in linear-momentum $\mathbf{q}$ dependency), as well as $f_{2\nu}$, $f_{2\mu}$ (that possess $d_{x^2-y^2}$ and $d_{2z^2-x^2-y^2}$ $\mathbf{q}$ space dependency). Indeed, if the pairing potential was momentum independent and a mere constant, such linear mixing terms would be forbidden by symmetry. It is the explicit non-trivial momentum dependence that permits such "mixing" terms in the Hamiltonian.

To illustrate this point more concretely, we can consider the interaction terms in Eq. 99, where we have mixing between an A_1 irrep Cooper pair and a E irrep Cooper pair,

$$H_{\mathrm{int}} = \sum_{\mathbf{k},\mathbf{k}'} \left[\sqrt{2} f_{2\nu} J_2^2\right]_{\mathbf{k},\mathbf{k}'} \left(A^\dagger_{1;j=\frac{1}{2};\mathbf{k}} E^{J=2}_{2;\mathbf{k}'} + \mathrm{h.c.}\right) + \sum_{\mathbf{k},\mathbf{k}'} \left[\sqrt{2} f_{2\mu} J_2^2\right]_{\mathbf{k},\mathbf{k}'} \left(A^\dagger_{1;j=\frac{1}{2};\mathbf{k}} E^{J=2}_{1;\mathbf{k}'} + \mathrm{h.c.}\right). \tag{2}$$

The A_1 irrep transforms as the identity, while the components of the E irrep Cooper pair transform as (in angular momentum space) the basis functions $x^2 - y^2$ and $3z^2 - r^2$. Such a coupling between these different irreps is allowed due to the momentum space basis functions $f_{2\nu}$, $f_{2\mu}$ that transform as $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ in $\mathbf{q}$ space. In effect, the pairing potential and the E irreps form an identity irrep (i.e. group theoretically, $E \otimes E = A_1$) and thus such a coupling is allowed from symmetry. The other mixing terms in the superconducting Hamiltonian can similarly be understood. Though we can understand the permitted terms from such group theoretic considerations, we would like to emphasize that the interactions terms we found were generated by integrating out the multipolar fluctuations (as described in Sec. 3 in the manuscript).

(2) Given the authors are examining these higher order J pairings and their parity, the time-reversal properties should also be discussed.

We thank the referee for this interesting question. Indeed, this question was also asked by Referee #1; we restate our answer to the question here for convenience.

A pairing $|\boldsymbol{\Delta}\rangle$ can be written as a linear combination over the "real" angular momentum basis states,

$$|\boldsymbol{\Delta}\rangle = \sum_u \Delta_u \, |O_h; u\rangle \,, \tag{3}$$

where $|O_h; u\rangle$ are the time-reversal invariant cubic irrep basis function given in Appendix E, and u sums over the cubic irreps; for example $|E_1^{J=2}\rangle$ etc. In this basis, time-reversal symmetry even order parameters are thus given by order parameters Δ_u that are equal to their complex conjugate (up to a global phase). We note that we have appropriately added in the global factor of i into the definition of the cubic irreps and the Cooper pair operators in the manuscript to make this explicitly apparent.

Examining our mean-field theory order parameter solutions, we thus discover that the non-trivial pairing states arising from the two-channel Kondo model (given in Table 1), the even J states are time-reversal invariant, while the odd J break time-reversal. More specifically, $A_1^{J=0}$ and $E^{J=2}$ are time-reversal invariant, while $T_2^{J=3}$, $T_\pm^{J=1,3}$, $T_2^{J=1,3}$ break time-reversal symmetry. These correspond to the non-vanishing order parameters realized in Fig. 1. For the novel Kondo interaction pairing states we have two possibilities (Figs. 2, 3). Focussing on Fig. 2, these states also follow the same prescription: even (odd) J states preserve (break) time-reversal symmetry. For Fig. 3, where the pairing states are formed from one electron from $j = 1/2$ and $j = 3/2$, both $E_-^{J=2}$ and $T_2^{J=2}$, $T_-^{J=1}$ break time-reversal symmetry. This is a surprisingly interesting feature of Cooper pairs formed from electrons belonging to distinct j sectors, and is reminiscent of the feature that even-J and odd-J Cooper pairs may be both parity even and odd (regardless of the even-ess of J) for the pairing states arising from the novel Kondo interaction.

We have correspondingly updated our manuscript to include this discussion.

(3) Fig. 3 is the most confusing, as they claim not only to be mixing irreps, but also to be mixing time-reversal odd and even order parameters (odd and even J). Again, I don't see how these can mix if the high temperature state is not breaking time-reversal, and particularly given that the OPs have a fixed parity. Inversion symmetry is not broken, and all the order parameters appear to be even-frequency. This behavior needs to be explained in detail, ideally with a Ginzburg-Landau theory.

To help to understand the nature of the pairing between the $T_2^{J=2}$ and $T_-^{J=1}$ Cooper pairs (the case highlighted by the referee), we consider their interaction Hamiltonian (Eq. 100),

$$H_{\text{int (1)}} = -\sum_{\mathbf{k},\mathbf{k}'} \left[f_0\beta^2 + \frac{1}{2}f_{2\nu}\beta^2 - \frac{\sqrt{3}}{2}f_{2\mu}\beta^2\right]_{\mathbf{k},\mathbf{k}'} T^\dagger_{2+^{(1)}\mathbf{k}} T_{2+^{(1)}\mathbf{k}'} - \sum_{\mathbf{k},\mathbf{k}'} \left[f_0\beta^2 - \frac{1}{2}f_{2\nu}\beta^2 + \frac{\sqrt{3}}{2}f_{2\mu}\beta^2\right]_{\mathbf{k},\mathbf{k}'} T^\dagger_{1-^{(1)}\mathbf{k}} T_{1-^{(1)}\mathbf{k}'}$$
$$+ \sum_{\mathbf{k},\mathbf{k}'} \left[-\frac{\sqrt{3}}{2}f_{2\nu}\beta^2 - \frac{1}{2}f_{2\mu}\beta^2\right]_{\mathbf{k},\mathbf{k}'} \left(-iT^\dagger_{2+^{(1)}\mathbf{k}} T_{1-^{(1)}\mathbf{k}'} + \text{h.c.}\right), \tag{4}$$

where we present the (1) component for illustration purposes. Importantly, it is the $\mathbf{k}$-space distribution of the interaction potential that permits the "mixing" between the two irreps in question. In question #1, we provided the group-theoretic means to understand a similar mixing term between $A_1^{J=0}$ and $E^{J=2}$ irreps, which was simpler to understand as the A_1 irrep transforms as an identity under the O_h point group. In this case, we recall the group-theoretic product $T_2 \otimes T_1 = A_2 \oplus E \oplus T_1 \oplus T_2$. Since the pairing potential is composed of E irrep basis functions (and since $E \otimes E = A_1$) one can notice that the product of the pairing potential with the Cooper pairs of interest is $E \otimes T_2 \otimes T_1 \supset A_1$. Thus, from symmetry-considerations such terms are indeed permitted. We do note that if the pairing potential was a constant (and independent of $\mathbf{k}$-space), such a term would indeed not be allowed. It is the momentum space distribution (acquired from the Ginzburg-Landau theory of multipolar fluctuations) that allows such non-trivial interaction Hamiltonians. Indeed, as we described above, both these order parameters break time-reversal and coexist together in the mean-field theory solutions. That is, the pairing states pointed out by the referee in fact break time-reversal i.e. the order parameters corresponding to $T_2^{J=2}$ and $T_-^{J=1}$ are not equal to their complex conjugate (up to a global phase).

(4) The role of symmetrization or antisymmetrization needs to be discussed carefully. It appears to be playing the role of another degree of freedom that comes into the overall antisymmetric nature of the electron wavefunction.

The Cooper pair operators presented in Appendix E are appropriately anti-symmetrized such that under the exchange of the angular momentum j and momenta $\mathbf{k}$ the operator picks up a minus sign; this minus sign is indicative of the fermionic nature of the electrons forming the Cooper pair. As we describe in Sec. 4, the Cooper pair operators are constructed using angular momentum via Clebsch-Gordan (CG) coefficients. Importantly, the CG coefficients take into account the j angular momentum exchange via the phase factor identity $\langle j_1, m_1; j_2, m_2 | j_1, j_2; J, M\rangle = (-1)^{J-j_1-j_2}\langle j_2, m_2; j_1, m_1 | j_2, j_1; J, M\rangle$. The subsequent parity of the Cooper pair operator (i.e. $\mathbf{k} \to -\mathbf{k}$) is such that the overall operator satisfies Fermi-Dirac statistics. An interesting and unusual feature is when we consider Cooper pair operators formed by one electron from $j = 1/2$ and the other from $j = 3/2$. This permits us to define a Cooper pair operator $b^\dagger_{J,M,\mathbf{k}}$ ($\tilde{b}^\dagger_{J,M,\mathbf{k}}$) with $j = \frac{3}{2}$ ($j = \frac{1}{2}$) fermion at $\mathbf{k}$ and $j = \frac{1}{2}$ ($j = \frac{3}{2}$) fermion at $-\mathbf{k}$, where the two Cooper pair operators are related to each other by the CG phase factor under exchange of j_1 and j_2. Due to the interaction being parity symmetric ($V_\mathbf{q} = V_{-\mathbf{q}}$), this allows us to then consider parity even and odd Cooper pair operators, $b^\dagger_{J,M;\mathbf{k};\pm}$, which are linear combinations of $b^\dagger_{J,M,\mathbf{k}}$ and $\tilde{b}^\dagger_{J,M,\mathbf{k}}$. These operators are appropriately antisymmetrized and possess definite parity. In the manuscript, the aforementioned linear combinations were referred to as “symmetric and anti-symmetric linear combinations”, which may have created a source of confusion. To avoid this possible source of confusion, we revise the manuscript to describe these operators as linear combinations of $b^\dagger_{J,M,\mathbf{k}}$ and $\tilde{b}^\dagger_{J,M,\mathbf{k}}$.