

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I recommend publication of this manuscript in Communications Physics. The manuscript presents an experimental (inelastic neutron scattering) and theoretical study of a magnetic spin resonance mode in the material CeCoIn₅, and comes to the conclusion that the origin of this mode is not an spin-excitonic bound state due to a sign change in the superconducting order parameter, but rather, most probably, due to a reduction of damping to a magnon-like mode in the superconducting state. This finding is of considerable interest in the light of the unclear origin of superconductivity in the material CeCoIn₅, and its proximity to a quantum critical point. Of particular interest is the finding that the mode splits in an (in-plane) applied magnetic field into a doublet rather than into a triplet. All together the conclusions are threefold: first, the resonance mode cannot be employed as evidence for a sign change in the superconducting order parameter; second, the normal state has a hidden overdamped magnetic excitation that is uncovered in the superconducting state by the removal of the damping mechanism; and third, the resonance mode splits into a doublet (rather than a triplet) under an in-plane applied field and becomes overdamped similar as in the normal state. The findings are of strong interest for the superconductivity and the correlated electron systems communities, have implications for possible pairing mechanisms as well as give insight into the role of magnetism in the material CeCoIn₅.

Reviewer #2 (Remarks to the Author):

This manuscript addresses the origin of the spin resonance observed by inelastic neutron scattering in the superconducting phase of CeCoIn₅. To do so, the authors performed new inelastic neutron scattering measurements in both zero and applied fields. The zero-field dispersion of the resonance mode was characterized and shown to have an upward curvature, while a magnetic field was shown to split the resonance mode into two distinct excitations. Theoretically, a spin-exciton bound state should have a downward curvature and a magnetic field should split the resonance into three distinct modes. Thus, the authors concluded the spin resonance in CeCoIn₅ is not a spin-exciton bound state, which would be a sign of its underlying superconducting d-wave order parameter. Instead, the authors propose the resonance mode in CeCoIn₅ is a "spin wave-like" excitation and the resonance appears below T_c due to a reduction of the damping within the superconducting phase.

The results of Y. Song et al. are interesting for researchers working in the field of unconventional superconductivity where a great deal of attention is devoted to characterizing the symmetry of the superconducting wave function. However, it should be known that both the zero-field upward dispersion of the resonance mode and its splitting in an applied field have both been observed in previous works (examples: ref.[43] and [47]). Furthermore, the conclusions reached by the authors have also been proposed earlier (example: ref.[15]). Nonetheless, the authors show better quality data than previously published results and their manuscript is worth publication. My concerns/comments are given below and should be addressed.

(1) Looking at Fig. 1(f), it looks like a downward curvature could be observed below ~ 0.45 meV. It seems that an "hour-glass" dispersion could be visible with very low spectral weight associated with part of the dispersion that would be downward. Can the authors comment on this?

(2) CeCoIn₅ being a d-wave superconductor, wouldn't the scattering associated with the spin resonance mode necessarily exist? In the case of CeCoIn₅, the spin resonance mode could be buried/hidden underneath a brighter excitation corresponding to the spin-wave-like excitation discussed by the authors? It seems one cannot rule out this possibility.

(3) It would be important to know how the authors accounted for resolution function effects in

their determination of the zero-field dispersion reported in Fig.1 (f)?

(4) I have a lot of difficulties to understand how the resonance mode in CeCoIn5 can be associated with spin-wave excitations when there is no sign of magnetic correlations building up in the quasi-elastic channel? Strictly speaking, a spin-wave excitation is a coherent deformation of a static spin configuration. In the case of CeCoIn5, no static correlations are observed (not even short-range correlations?). Could there be magnetic order that is not visible by neutron such as a multipolar phase?

(5) Why the authors claim the dispersion of the spin resonance mode in CeCoIn5 is shifted by $(\delta, \delta, 0)$ away from $(0.5, 0.5, 0.5)$, where $\delta = 0.034$? The dispersion drawn for example in Fig.1(f) is centered at $(0.5, 0.5, 0.5)$. Can the authors claim the data collected at $E=0.375$ meV (Fig.2(a)) support two peaks instead of a single one centered at $(0.5, 0.5, 0.5)$? Note the authors use a single gaussian peak to fit that particular cut.

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The authors present a comprehensive inelastic neutron study of the spin resonance mode (SRM) in the dx^2+y^2 -wave heavy fermion superconductor CeCoIn5. By analyzing both the momentum and magnetic field dependence of the SRM, the authors concluded that the SRM is not consistent with predictions of a spin-exciton origin, thus its presence is not experimentally indicative – although cannot necessarily discount – of the presence a sign-changing superconducting order parameter.

The authors emphasize that the conclusion of the SRM not being spin-exitonic in origin for CeCoIn5, a heavy fermion superconductor where spin-fluctuation-mediated pairing is believed to be at play, demonstrates the importance of testing the origins of the SRM when utilizing its presence as proof for sign-changing unconventional superconductivity.

The clear demonstration that the presence of the SRM does not imply a sign-changing superconducting order parameter is both of great interest in the broader unconventional superconductor community and serves an important pedagogical tool in the identification and testing of spin-fluctuation-mediated pairing. The manuscript is very well-written with the reader being introduced to the motivation and overall importance for such a study, followed by inelastic neutron scattering studies performed in the absence and presence of an external magnetic field. The analysis is detailed and is presented in a way that it is easy for the reader to follow. In my opinion, the manuscript is of broad interest and deserves to be published in Communication Physics, although I have a few minor remarks that I would like the authors to consider:

1. In Figs.1(e) and (f), the authors performed two inelastic neutron studies, one on PANDA, the other on MACS, that clearly demonstrated an upward dispersion. May the authors choose different symbols/colors to help the reader appreciate the fact that this conclusion is a result from two independent measurements. In its current format, it is very difficult to distinguish the two.

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Conclusion: The manuscript deserves to be published in Communication Physics with minor revisions.

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We thank the reviewer for reviewing our work, accurately summarizing our findings and recommending it for publication.

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published results and their manuscript is worth publication. My concerns/comments are given below and should be addressed."

We thank the reviewer for reviewing our work, accurately summarizing our findings and recommending it for publication.

The reviewer is correct that some of our observations have been previously reported. The comprehensive measurements in this work further allowed us to observe for the first time: (1) the detailed behavior of the SRM for $E < 0.5 \text{ meV}$, ruling out any significant downward dispersing branch; (2) the evolution of the dispersive SRM under applied field and showing that doublet splitting occurs not only at a single momentum $Q = (0.5, 0.5, 0.5)$ but for the entire upward-dispersing branch with increased damping. We respond to the reviewer's concerns/comments in detail below.

"(1) Looking at Fig. 1(f), it looks like a downward curvature could be observed below $\sim 0.45 \text{ meV}$. It seems that an "hour-glass" dispersion could be visible with very low spectral weight associated with part of the dispersion that would be downward. Can the authors comment on this?"

For the lowest 3 energies in Fig. 1e-f, the fit values of δ are the same with error considered. Therefore, we believe our data do not suggest a downward dispersion. In the revised manuscript, we have made this point clear and added fit values and uncertainties of δ in Fig 2.

"(2) CeCoIn₅ being a d-wave superconductor, wouldn't the scattering associated with the spin resonance mode necessarily exist? In the case of CeCoIn₅, the spin resonance mode could be buried/hidden underneath a brighter excitation corresponding to the spin-wave-like excitation discussed by the authors? It seems one cannot rule out this possibility."

We agree that this cannot be ruled out, given the pairing symmetry of CeCoIn₅ is rather well-established. Our work shows the dominant magnetic spectral weight is not spin-excitonic, however a small spin-excitonic contribution can NEVER be ruled out, because any experiment that fails to observe it can only lower the upper limit of its contribution to the total spectral weight. In the revised manuscript, we have made this point more clear.

This does not affect the conclusion of our manuscript, which is that the detectable SRM in CeCoIn₅ is not spin-excitonic. This means that for unconventional superconductors whose pairing symmetry is not yet as well-established compared to CeCoIn₅, more stringent tests should be needed when using the observation of a SRM as evidence for sign-changing superconductivity.

"(3) It would be important to know how the authors accounted for resolution function effects in their determination of the zero-field dispersion reported in Fig.1 (f)?"

To accurately account for effects of the resolution function one needs a parametrized model of excitations in full 4D E-Q space, but because we only carried out scans along $(h, h, 0)$ centered at

(0.5,0.5,0.5), to do so we will need to make assumptions that also need to be justified. We therefore carried out our analysis without correction for resolution effects, and have described our experimental configuration in the Methods section so that it can be fully reproduced.

Qualitatively, because the two dispersive branches at $\pm\delta$ are observed to have similar intensities, the resolution ellipsoid in the scattering plane is small relatively to the broadness of the excitations. Out-of-plane resolution can in principle affect the observed splitting of the two peaks at $(0.5\pm\delta, 0.5\pm\delta, 0.5)$, due to contributions from signals at $(0.5\pm\delta, 0.5\mp\delta, 0.5)$. To gauge to what extent this may affect our observations, it is instructive to examine Supplementary Figure 3, where we compare our data (vertically focused analyzer and $k_f=1.3\text{\AA}^{-1}$) with published data (flat analyzers and $k_f=1.15\text{\AA}^{-1}$) measured using a configuration that has better resolution. The overall agreement between data, as well as the agreement between extracted values of δ (relative to error bars), both suggest the effects due to resolution should be small. To summarize, although resolution can in principle affect the observed value of δ , our data suggest its effect to be small; more importantly, the qualitative upward-dispersing nature of the SRM is robust to resolution effects.

“(4) I have a lot of difficulties to understand how the resonance mode in CeCoIn5 can be associated with spin-wave excitations when there is no sign of magnetic correlations building up in the quasi-elastic channel? Strictly speaking, a spin-wave excitation is a coherent deformation of a static spin configuration. In the case of CeCoIn5, no static correlations are observed (not even short-range correlations?). Could there be magnetic order that is not visible by neutron such as a multipolar phase?”

We thank the reviewer for raising this important point. We are referring to magnon-like excitations observed in the paramagnetic state that resemble spin waves in the ordered state (paramagnons). These are commonly seen in cuprate and iron-based superconductors in the absence of static order, e.g. near optimal doping.

“(5) Why the authors claim the dispersion of the spin resonance mode in CeCoIn5 is shifted by $(\delta, \delta, 0)$ away from $(0.5, 0.5, 0.5)$, where $\delta = 0.034$? The dispersion drawn for example in Fig.1(f) is centered at $(0.5, 0.5, 0.5)$. Can the authors claim the data collected at $E=0.375$ meV (Fig.2(a)) support two peaks instead of a single one centered at $(0.5, 0.5, 0.5)$? Note the authors use a single gaussian peak to fit that particular cut.”

We did not plot a point for $E=0.375$ meV in Fig. 1f, the lowest energy point is for $E=0.4$ meV. For $E\geq 0.4$ meV, we extracted δ by fitting 2 peaks. As noted in our manuscript, we used data from Fig. 2b-f (but not Fig. 2a) for making Fig. 1e-f.

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We have revised these figures based on the reviewer’s suggestion. In addition, we have revised Supplementary Fig. 2 for consistency.

“2. It would be perhaps be instructive to include the predicted downward dispersion from other isostructural unconventional superconductors (e.g. that shown in Fig. 4 in PRL 114, 247005, a paper already referenced by the authors as [34], where the main mode below the multimagnon continuum shows such behavior. This would emphasize to the reader the predicted behavior that was shown in the Fig. 1(d)).”

PRL 114, 247005 reports a neutron scattering study of antiferromagnetically ordered CeRhIn₅ (non-superconducting), which exhibits upward-dispersing spin waves. In Fig. 1(f), we compare the dispersion of spin waves in CeRhIn₅ extracted from two previous works (including PRL 114, 247005) with the our observed dispersion in CeCoIn₅. As far as we know, there are no neutron scattering studies on CeIrIn₅, a superconducting analog of CeCoIn₅.

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In the revised manuscript, we have expanded our discussions on this point. It was found in CeRhIn₅ that field along (1-10) reduces magnetic anisotropy energy for the (110) direction, and if the same happens then it is possible for CeCoIn₅ to evolve from a (001) easy axis to a (001)x(110) easy plane when the anisotropy energies for these two axes become similar.

“4. Is there a citation for the indium self-flux method used for crystal growth?”

We have added a reference for the self-flux method used to synthesize Ce115 materials.

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We thank the referee for carefully reviewing our work and finding these corrections. For the second suggested correction, we intended to write CeRhIn₅ rather than CeCoIn₅. We have revised to reference ‘magnetically ordered CeRhIn₅’ to make this clearer.

“6. In reviewing the potential pitfalls of spin-exciton models of SRMs in unconventional superconductors, the authors should consider discussing other, earlier breakdowns in other systems, such as the electron-doped cuprates (Phys. Rev. B 76, 094506 (2007)) to name one example.”

We thank the reviewer for reminding us of this work, and have added it to the references. We have also added a reference on Bi2212 that argued against a quasiparticle approach in accounting for spin dynamics (Nat. Phys. 5, 642-646).

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