nature portfolio

Peer Review File

Spin excitations in metallic kagome lattice FeSn and CoSn

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In my previous review I asked:
" I find that the current version is mixed up and will be very confusing for a general audience. It is therefore not suitable for publication in the present form. It is also not clear whether the remaining verified results are of sufficient interest to warrant publication in Nature Communications. I would suggest that the authors rewrite the paper along the following lines so an accurate judgement can be made:
1) Discuss the correct results i.e. the low-energy spin waves and the model.
2) Remove most of the discussion about the Stoner band, since it is not observed.
3) Make clear that the bands of scattering are 170meV and 350 meV are from the glue (e.g. label them as such in the figures) from the start.
4) Discuss the glue in the methods section or in one place do not mix it in with the rest of the results and introduction. Scientific papers do not need the history of the experiment in this way. A reference to the discussion about the glue can be made in the abstract, and when discussing the experimental data."

The authors have not taken these remarks fully onboard although they have addressed specific points. As a understand it the current situation is that they have observed (i) some magnetic excitations, they have also observed (ii) some spurious background. The authors need to go through the paper and make changes in my view including: (1) separate aspects (i) and (ii); (2) remove the "history"; (3) make the discussion of the spurious scattering more scientific so that other scientists can make use of what they have found and understand why it is determined to be due to the CYTOP-M and not hydrocarbons. Below I give some suggestions for changes, I would expect the authors to make additional changes too to address these points.
[1] Abstract " Although our initial measurements on FeSn indeed reveal well-defined spin waves extending well above 140 meV coexisting with a flat excitation at 170 meV, subsequent experiments on CoSn indicate that the flat mode actually arises mostly from hydrocarbon scattering of the CYTOP-M commonly used to glue the samples to aluminum holder. Therefore, our results established the evolution of spin excitations in FeSn and CoSn, and identified an anomalous flat mode that has been overlooked by the neutron scattering community for the past 20 years."
=>"In addition to the spin excitations we observed non-dispersive excitations (at 150-180 meV and 350-380 meV) of the CYTOP-M used to glue the samples to aluminum holder."

[2] " In this paper, we report inelastic neutron scattering (INS) studies of spin excitations in 2D metallic Kagome lattice antiferromagnetic FeSn35 and paramagnetic CoSn36. For FeSn, our initial measurements reveal well-defined spin waves extending well above 140 meV and a narrow ~24 meV wide band of excitations that cannot be described by a simple spin-wave model. While these data suggest the presence of electron-hole-pair Stoner excitations from the majority-spin flat band below the Fermi level to minority-spin bands near or above the Fermi level in FeSn, subsequent experiments on paramagnetic CoSn also have the same flat mode coexisting with expected paramagnetic spin excitations. Through carefully analysis INS spectra under different conditions, we conclude that the observed flat mode actually arises mostly from hydrocarbon scattering of the CYTOP-M commonly used to glue the samples to aluminum holder37. Therefore, our results established the evolution of spin excitations in FeSn and CoSn, and revealed that the observed flat electronic band below the Fermi level does not induce a narrow band Stoner continuum. In addition,

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[3] "By shifting the incident beam neutron away from the sample, the hydrocarbon contamination is still present with the similar intensity ratio between 170 meV and 360 meV modes (Table 1). These results suggest that the scattering can only come from C-H bending mode instead C-F bending mode. " Is the 360 meV mode not a bond stretching?
[4] Please give approximate amounts (grams) of glue and sample.
[5] The authors say that the 360 meV is also observed in the absence of CYTOP-M. What fraction of the scattering is actually due to the CYTOP-M?
[6] Figure 3 caption "Flat band like excitations in FeSn" This caption is misleading because the a lot of the scattering described it due to CYTOP-m e.g. the flat band, please read and correct.
[7] Figure 4 caption. Again the excitations shown include those from CYTOP.
[8] Please check the supplementary information captions to make sure they say what materials are being measured.

If the authors make changes along the line above then manuscript can be published.

Replies to comments by the referee:

We would like to express our sincere appreciation to the referee for his/her comments and recommendations on the paper. We address them below, and discuss changes made to the original manuscript. With these changes, we hope that the paper should now be suitable for publication.

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We appreciate these comments and feel that they are an accurate description of the situation.

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=>"In addition to the spin excitations we observed non-dispersive excitations (at 150-180 meV and 350-380 meV) of the CYTOP-M used to glue the samples to aluminum holder."”

We have revised the abstract following the referee's suggestion mentioned above.

"[2] " In this paper, we report inelastic neutron scattering (INS) studies of spin excitations in 2D metallic Kagome lattice antiferromagnetic FeSn35 and paramagnetic CoSn36. For FeSn, our initial measurements reveal well-defined spin waves extending well above 140 meV and a narrow ~24 meV wide band of excitations that cannot be described by a simple spin-wave model. While these data suggest the presence of electron-hole-pair Stoner excitations from the majority-spin flat band below the Fermi level to minority-spin bands near or above the Fermi level in FeSn, subsequent experiments on paramagnetic CoSn also have the same flat mode coexisting with expected paramagnetic spin excitations. Through carefully analysis INS spectra under different conditions, we conclude that the observed flat mode actually arises mostly from hydrocarbon scattering of the CYTOP-M commonly used to glue the samples to aluminum holder37. Therefore, our results established the evolution of spin excitations in FeSn and CoSn, and revealed that the observed flat electronic band below the Fermi level does not induce a narrow band Stoner continuum. In addition, we identified an anomalous flat mode that has been overlooked by the neutron scattering community for the past 20 years."
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Following the referee's suggestions, we have revised the paragraph and eliminated discussion about the history of the process for discovering the CYTOP-M mode. We have also added a reference where we reanalyze the data of high-energy spin excitations of iron pnictides glued using CYTOP-M, and found the presence of a weak flat mode at 170 meV. Fortunately, the intensity of this mode is significantly less than spin excitations and therefore does not affect the conclusions of the original paper.

"[3] "By shifting the incident beam neutron away from the sample, the hydrocarbon contamination is still present with the similar intensity ratio between 170 meV and 360 meV modes (Table 1). These results suggest that the scattering can only come from C-H bending mode instead C-F bending mode. " Is the 360 meV mode not a bond stretching?"

We appreciate very much that the referee caught this problem. What we meant to say is that since ratio between 170 meV and 360 meV mode are the same with and without sample being in the same, this provide direct evidence that the 170 meV mode see in FeSn does not have a magnetic component. We have revised the sentence in question.

"[4] Please give approximate amounts (grams) of glue and sample."

We have kept the CYTOP-M glued plates, and measured it to be approximately 0.34 grams. We noted this in the revised draft. The weight of FeSn and CoSn has already been stated in the paper.

"[5] The authors say that the 360 meV is also observed in the absence of CYTOP-M. What fraction of the scattering is actually due to the CYTOP-M?"

The amount of hydrocarbon contamination from pump oil in time of flight measurements is difficult to estimate, as the signal is highly dependent on how long the cryostat is operational in a particular experiment. For this reason, it is rather difficult to estimate the signal ratio of hydrocarbons with and without CYTOP-M on the same sample. Nevertheless, we can compare the intensity of the 170 meV obtained at ISIS (Fig. 3a, 3b), which has CYTOP-M, with those obtained without CYTOP-M (Figs. 5e and 5f), which has pure FeSn but without CYTOP-M since both measurements were normalized to absolute units. We find that the ratio to be (18.4+-0.3)/(1.25+-0.16), so majority of the signal at 170 meV arises from CYTOP-M. We added a sentence to make this clear in caption of Fig. 5 so a reader can have this information.

"[6] Figure 3 caption "Flat band like excitations in FeSn" This caption is misleading because the a lot of the scattering described it due to CYTOP-m e.g. the flat band, please read and correct."

Thanks a lot. This is corrected to be "Dispersionless excitations from FeSn+CYTOP-M".

"[7] Figure 4 caption. Again the excitations shown include those from CYTOP."

This is corrected as well to be "Measurement of excitations in CoSn+CYTOP-M"

"[8] Please check the supplementary information captions to make sure they say what materials are being measured."

Thanks, we have checked and updated the captions when necessary.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed the points I raised and I am happy for the manuscript to manuscript to be published.