Authors' Response to Reviews of

μSR study of unconventional pairing symmetry in the quasi-1D $Na_2Cr_3As_3$ superconductor

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Magnetochemistry, Manuscript ID: magnetochemistry-2199042

RC: *Reviewers' Comment*, AR: Authors' Response, ☐ Manuscript Text

To
The Editor
Magnetochemistry

Dear Editor,

We thank you very much for the review of our manuscript and for conveying the referees' reports. We appreciate the time and work you and the reviewers put into providing us with insightful comments on our article which have indeed helped us to improve the quality of our manuscript. Please find attached an updated version of our manuscript which we would like to resubmit for publication in Magnetochemistry.

We have answered the referees' all comments and questions in point by point details. We provided a list of changes made in the revised manuscript in the authors' response file and highlighted the revisions in the main manuscript in blue color. Having dealt with the referees comments comprehensively, we believe that our manuscript will now be suitable for publication in Magnetochemistry.

We hope to hear a positive response to our manuscript soon.

Yours sincerely,
A. Bhattacharyya
(On behalf of all the authors)

1. Reviewer #1

RC: *Comments and Suggestions for Authors*
The presented research is very interesting and I recommend publishing the article after a serious improvement.

AR: We thank the referee for taking the time to critically read and review our work and providing valuable comments to improve the manuscript. We appreciate the referee for identifying the importance of our work and his/her recommendation for publication in Magnetochemistry after some corrections. Having dealt with the referee's comments comprehensively, we believe that our manuscript will now be suitable for publication.

RC: *Introduction section should be improved. The use of muSR method in the paper must be explained, specifying the importance of the results that can be obtained/ as well as the use of the electronic structure calculations.*

AR: We thank the referee for suggesting to discuss the importance of muon spin spectroscopy and electronic structure. We have modified the introduction section in the modified manuscript. We have included the following line

"In order to investigate the pairing mechanism and time reversal symmetry breaking in the ground state of novel superconductors, we have performed a systematic muon spin rotation and relaxation (μSR) study. Muon, a spin 1/2 subatomic particle, probed into the sample, precise around the local magnetic field, is a powerful tool to investigate superconducting materials [20]. Furthermore, our results are also supported by the electronic structure calculation."

[20] Hillier, A.D.; Blundell, S.J.; McKenzie, I.; Umegaki, I.; Shu, L.; Wright, J.A.; Prokscha, T.; Bert, F.; Shimomura, K.; Berlie, A.; et al. Muon spin spectroscopy. Nature Reviews Methods Primers 2022, 2.

RC: *Usually, when we talk about the crystal structure in an experimental paper, an XRD spectrum is provided to confirm the quality of the studied sample. Likewise with magnetization. The custom is that specific experimental curves must be given. Instead, the authors describe these properties by reffering to ref. 29. Is the mSR experiment performed by the authors on the same sample studied in ref. 29 or is it a different sample with the same chemical formula? These details must be specified and/or completed as appropriate very clearly.*

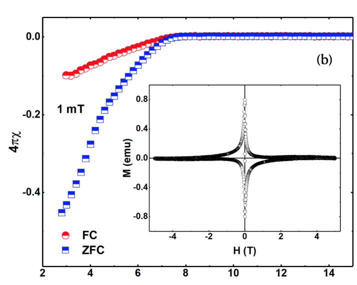

Figure 1: Temperature dependence of magnetization measurements. This clearly reveals the signature of a superconducting phase transition.

AR: Thank you for raising such an important question. We agree with the referee on his/her statements. It was challenging to perform the XRD experiment because the samples are highly air sensitive. By measuring magnetization [**Figure 1**] and MuSR, we have examined the sample quality. Both experiments yield comparable Tc results to those found in Reference 29. The most significant finding of our zero field MuSR data is that there is no evidence of a spontaneous magnetic field in the superconducting ground state. That confirms the existence of any magnetic impurity. Therefore, the primary physics we investigated for this research came exclusively from the Na2Cr3As3 sample.

RC: *The authors write: "Previous theoretical and experimental studies confirm the presence of nodal gap in A2Cr3As3 (A=K, Rb, Cs) systems [11,16]." What experimental studies?*

AR: We appreciate the referee for pointing out the statement. We have corrected the statement in the modified manuscript.

"Previous theoretical and experimental studies of magnetic penetration depth using tunnel-diode oscillator, μSR and nuclear quadrupole resonance measurements confirm the presence of nodal gap in $A_2Cr_3As_3$ (A = K, Rb, Cs) systems [13–16,18]."

[13] Wu, X.; Yang, F.; Le, C.; Fan, H.; Hu, J. Triplet p z-wave pairing in quasi-one-dimensional A2Cr3As3 superconductors (A= K, Rb, Cs). Physical Review B 2015, 92, 104511.

[14] Pang, G.; Smidman, M.; Jiang, W.; Bao, J.; Weng, Z.; Wang, Y.; Jiao, L.; Zhang, J.; Cao, G.; Yuan, H. Evidence for nodal superconductivity in quasi-one-dimensional K2Cr3As3. Physical Review B 2015, 91, 220502.

[15] Adroja, D.; Bhattacharyya, A.; Telling, M.; Feng, Y.; Smidman, M.; Pan, B.; Zhao, J.; Hillier, A.; Pratt, F.; Strydom, A. Superconducting ground state of quasi-one-dimensional K2Cr3As3 investigated using μSR measurements. Physical Review B 2015, 92, 134505.

[16] Adroja, D.; Bhattacharyya, A.; Smidman, M.; Hillier, A.; Feng, Y.; Pan, B.; Zhao, J.; Lees, M.R.; Strydom, A.; Biswas, P.K. Nodal superconducting gap structure in the quasi-one-dimensional $Cs_2Cr_3As_3$ investigated using μSR measurements. Journal of the Physical Society of Japan 2017, 86, 044710.

[18] Luo, J.; Yang, J.; Zhou, R.; Mu, Q.; Liu, T.; Ren, Z.a.; Yi, C.; Shi, Y.; Zheng, G.q. Tuning the Distance to a Possible Ferromagnetic Quantum Critical Point in $A_2Cr_3As_3$. Physical review letters 2019, 123, 047001.

RC: *Further the authors write: "To understand the enigmatic superconducting gap structure of Na2Cr3As3, we have carried out the TF-mSR measurements." TF-mSR measurements are also experimental. And here, again, the logical question arises, why mSR? Nowhere in the text, in the introduction or in the mRR section, is this explained.*

AR: We thank the referee for the remark. In the modified introduction part of the manuscript we have discussed the importance of μSR in this context.

2. Reviewer #2

RC: *Lines 9 and 247 : chiral superconductivity should be explained*

AR: We thank the referee for carefully reading the manuscript and for suggestions on improvements to the manuscript. As suggested by the referee we have discussed the chiral superconductivity in the introduction

part of the manuscript
"Sr_2RuO_4 and UPt_3 are two promising candidates of chiral superconductors with plausible triplet p-wave and triplet f -wave pairing [10,11]. In a chiral superconductor, an angular momentum spontaneously develops and lowers its free energy by eliminating nodes in the gap."

[10] Kallin, C. Chiral p-wave order in Sr_2RuO_4. Reports on Progress in Physics 2012, 75, 042501.

[11] Campos, P.G. Visualization of chiral superconductivity in UPt_3. PhD thesis, Université Grenoble Alpes [2020-....], 2021.

RC: *Lines 94-95 Eq (1) should be corrected : some symbols are not clear*

AR: We have corrected this in our modified manuscript. Thank you.

RC: *Line 97 : $\gamma^{-}/2\pi$ = 135.5 MHz T-1 the index should be rewritten and does not be rewritten in line 107*

AR: Thank you for the suggestion. We have corrected this in our modified manuscript.

RC: *Line 107 : where $\gamma_\mu/2\pi$ = 135.5 MHz/T is the muon gyromagnetic ratio and. . . .. This parameter has been already defined in line 97*

AR: We are sorry for the mistake, however, in the modified manuscript we have taken care of it.

RC: *The vertical legend in Figure 2 c should be reoriented upwards*

AR: As recommended by the referee we have reoriented the vertical legend in Figure 2 c.

RC: *Line 155 : $H^{-} = \sigma/\gamma^{-}$) : The index is not visible*

AR: Thank you for pointing out the mistake. We have corrected it in the modified manuscript.

RC: *Therefore, the authors are expected to address the following questions:*

The authors worked on a powdered sample. Didn't they have the possibility to prepare a single crystal?

AR: Yes, It is possible to synthesize the single crystal of $Na_2Cr_3As_3$. The sample prepared by Q. G. Mu *et al.* [Phys. Rev. Materials 2018, 2, 034803] is a single crystal. However, for the μSR measurements minimum of 1 gm sample is needed. 1 gm single crystal preparation is difficult. So, we have performed the μSR measurements in the high quality powder sample which has similar T_C as of the single crystal.

RC: *It is known that Cr2+ (or 3+) has a 3/2 (or 1) spin and is susceptible to antiferromagnetic coupling. The authors have focused here only on ferromagnetism? What can be stated about the antiferromagnetism of Cr ions?*

AR: Thank you very much for suggesting to discuss the antiferromagnetic nature of the Chromium (Cr) atom. Initially, CrAs was found to be a pressure-induced superconductor near an antiferromagnetic instability [1-3]. However, the Cr-based arsenides $A_2Cr_3As_3$ (A = Na, K, Rb, and Cs) are of great interest in terms of low dimensionality, strong ferromagnetic (FM) fluctuations, and noncentrosymmetric superconductivity. Due to the presence of an alkaline atom (A), a strong spin–orbit coupling (SOC) has remarkable effect in $A_2Cr_3As_3$ which suppresses the antiferromagnetic nature of Chromium [4,5].

[1] W. Wu, J. Cheng, K. Matsubayashi, P. Kong, F. Lin, C. Jin, N. Wang, Y. Uwatoko, and J. Luo, Nat. Commun. 2014, 5, 5508.

[2] H. Kotegawa, S. Nakahara, H. Tou, and H. Sugawara, J. Phys. Soc. Jpn. 2014, 83, 093702.

[3] Y. Shen, Q. Wang, Y. Hao, B. Pan, Y. Feng, Q. Huang, L. W. Harriger, J. B. Leao, Y. Zhao, R. M. Chisnell, J. W. Lynn, H. Cao, J. Hu, and J. Zhao, Phys.Rev.B 2016, 93, 060503(R).

[4] C. Xu, N. Wu, G. X. Zhi, B.H. Lei, X. Duan, F. Ning, C. Cao and Q. Chen npj Computational Materials 2020, 6, 30.

[5] C.-C. Liu, C. Lu, L. D. Zhang, X. Wu, C. Fang, F. Yang Phys. Rev. Research 2020, 2, 033050.