

Peer Review File

Static magnetic order with strong quantum fluctuations in spin-1/2 honeycomb magnet Na₂Co₂TeO₆



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7th May 24

Dear Professor Ma,

Thank you for submitting your manuscript, "Static magnetic order with strong quantum fluctuations in spin-1/2 honeycomb magnet Na₂Co₂TeO₆", to Communications Materials. It has now been seen by 2 referees, whose comments are appended below. You will see that while they find your work of potential interest, they have raised substantial concerns on experimental measurements and data interpretation. In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that convincingly addresses these serious concerns.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data and analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

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Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Aldo

Dr Aldo Isidori

Senior Editor

Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

This work is focused on the honeycomb cobaltate $\text{Na}_2\text{Co}_2\text{TeO}_6$ (NCTO). The authors' central claim is that through a combination of muon spin relaxation and neutron scattering measurements, they find support for not only frustrated Kitaev interactions but also field-induced quantum fluctuations, suggesting that NCTO may be near the onset of a spin-liquid state. Because the ground state magnetic order of NCTO is complex, the authors attempted to take advantage of the magnetic field

dependence of the magnetic Bragg reflections. The main conclusions of the work are that (1) the magnetic order in NCTO is multi-domain zigzag order rather than a multi-k structure and (2) the presented measurements evidence strong quantum fluctuations and highlight the presence of the Kitaev interaction.

There are a number of issues with this work. My main impression is that evidence for quantum fluctuations is being overstated based on the results. I would appreciate it if the authors could clarify the following questions:

- 1) A central claim of the neutron diffraction work is a bump in the order parameter in Fig. 1(c) around 15 K. This bump does not appear to be statistically significant, or at least not nearly enough to disprove triple-Q order. The fits in the range of 12 K to 17 K should be shown clearly in the SI.
- 2) The discussion of the anisotropic evolution of the peak intensities at the M_{-1} and M points is confusing. Neutron scattering is only sensitive to the perpendicular component of the moment due to the polarization factor, meaning that as the field aligns the moments along the $(-1\ 1\ 0)$ direction one would expect the scattering intensity to decrease as a function of field. Instead, Fig. 1(d) reports the opposite. Was any type of magnetic structure refinement performed as a function of field accounting for domain populations? Some kind of quantitative model to support the domain picture at all would be helpful.
- 3) A central claim of the work from the inelastic scattering is that broadening as a function of field at the K-point is evidence for quantum fluctuations. This broadening should be present not only in the SIKA measurement in Fig. 2(c), but also in the SEQ measurement in Fig. 2(b). Why are the SEQ data not included in Fig. 2(c)? From the color plots, it looks like the SEQ data shows no evidence of broadening. The resolution of the SIKA measurement is also insufficient to determine if this is broadening or if there are two modes.
- 4) Why does broadening necessarily imply quantum fluctuations? This is an overall imprecise statement as it comes with no meaningful discussion about why quantum fluctuations would coexist with a fully ordered moment, as suggested by the muon data in Fig. 3(c). Broadening could originate from enhanced magnon-magnon interactions, domain effects, or some other means, but to jump directly to the most exotic explanation does not sit well with me.
- 5) The measurement of T^* at 8 K in the muon data in Fig. 4(d) is not statistically significant and should not be used as evidence of redistribution of magnetic domains, especially since it is not visible at all in the neutron diffraction. The asymmetry analysis is more convincing.

6) Were any attempts made to use linear spin-wave theory to model your inelastic neutron scattering spectra?

In its current state I cannot recommend this work for publication, but I do hope that the authors can address these questions.

Reviewer #2 (Remarks to the Author):

This manuscript reports the neutron scattering and muon spin rotation/relaxation (μ SR) results in single crystals of the honeycomb magnet $\text{Na}_2\text{Co}_2\text{TeO}_6$, a candidate material for Kitaev magnet. I understand that there are two main arguments in the manuscript. The first one is that the magnetic ordering state is the multi-domain state, which is revealed by their neutron scattering experiments. Since the existence of the multiple- q -ordered state has been discussed in the community, the result is particularly important for the research field. The second one is that they found a substantial residual magnetic fluctuation even in the magnetic ordered state from their μ SR results. This is also valuable information. However, I have several questions and comments about the μ SR data analysis in the manuscript as shown below. Thus, I could not recommend the manuscript for publication in Nature in the present form. I would be grateful if I received enough elaboration from the authors.

Q1. Although I agree that the residual magnetic fluctuation appears in the magnetic ordered state because I can find some slow relaxation in ZF- μ SR spectra at 2 K in Fig. S3, the estimated $\lambda_L = 0.45 \text{ us}^{-1}$ seems to be overestimated. Generally, estimation of relaxation rate λ using an equation, $Ae^{-\lambda t}$, strongly depends on the value of the initial asymmetry A . λ tends to be large when A is small, whereas the λ may be small when A is large. Therefore, the λ_L is considered to depend on the value of A_1 and $(1-\alpha)$ in Eq. (2), which is the equation the authors used for the analysis of ZF- μ SR spectra. I would like to ask the authors if the A_1 and $(1-\alpha)$ are compressed and the λ_L is overestimated. The Eq. (2) has another non-oscillating term $A_2 e^{-\lambda_{\text{tail}} t} G_{\text{KT}}$ from the second muon site. Since the second muon site is not affected by spontaneous internal field from the ordered magnetic moment, the muon site contributes to the rotating asymmetry in wTF- μ SR data, A_{NM} , even in the magnetic ordered state. The A_{NM} is estimated to be 0.02--0.03 for wTF- μ SR analysis as shown in Fig. 3(b). The A_2 in Eq. (2) should be compatible with the A_{NM} value. However, the A_2 is estimated to be 0.05--0.06 for ZF- μ SR analysis as shown in Fig. S4. I am afraid I consider that the A_2 value for the ZF- μ SR analysis is too large and the A_1 may be compressed. Then, the

λ_L becomes large. I would like to suggest a reanalysis of the ZF-muSR data with some restrictions for A_2 .

Q2. The authors pointed out the similarity of the temperature dependence of muon spin relaxation rate between the NCTO and a quantum spin liquid candidate YbMgGaO_4 . However, one for the YbMgGaO_4 showed no sign of magnetic ordering and the λ increased with decreasing temperature before the following saturating behavior near the lowest temperature as shown in Ref. 59. I consider that the present result is more like the muSR result for $\text{FeTe}_2\text{O}_5\text{Br}$ (M. Pregelj et al., PRL, 109, 227202 (2012).). The authors should refer to the previous muSR results showing persistent spin dynamics in the magnetic ordered state.

Q3. The lack of muSR data at longitudinal-field (LF) conditions, which is an external field applied along the initial muon spin polarization, gives some ambiguity for the interpretation of the present muSR data. I agree that the slow relaxation in the ZF-muSR spectra may represent the microscopic dynamical magnetic properties of the sample. However, no-decoupling behavior should be checked by applying LF to confirm that the slow relaxation certainly originated from the magnetic fluctuation. If the authors have extra muSR data, especially at LF, these should be added to the manuscript.

Reply to comments:

Referee 1:

Q1: A central claim of the neutron diffraction work is a bump in the order parameter in Fig. 1(c) around 15 K. This bump does not appear to be statistically significant, or at least not nearly enough to disprove triple-Q order. The fits in the range of 12 K to 17 K should be shown clearly in the SI.

Response: We are grateful for the thorough review and valuable feedback from the reviewer. In response to the concern regarding the anomaly observed around 15K at point M1 in Figure 1(c), we have included a detailed fitting for the 12K-17K range in Fig. E1 (S1) (b) of the Supplementary Materials. With the data of M1 from 12K to 17K, the bump of M1 is confirmed. As the temperature-dependences of M and M1 are different, the ground state of NCTO should not be triple-Q. The extra sentence “Each integrated intensity was calculated by fitting the raw data with a Gaussian function, as illustrated in Fig. S1 of the Supplementary materials.” has been added to the paragraph on Page 4.

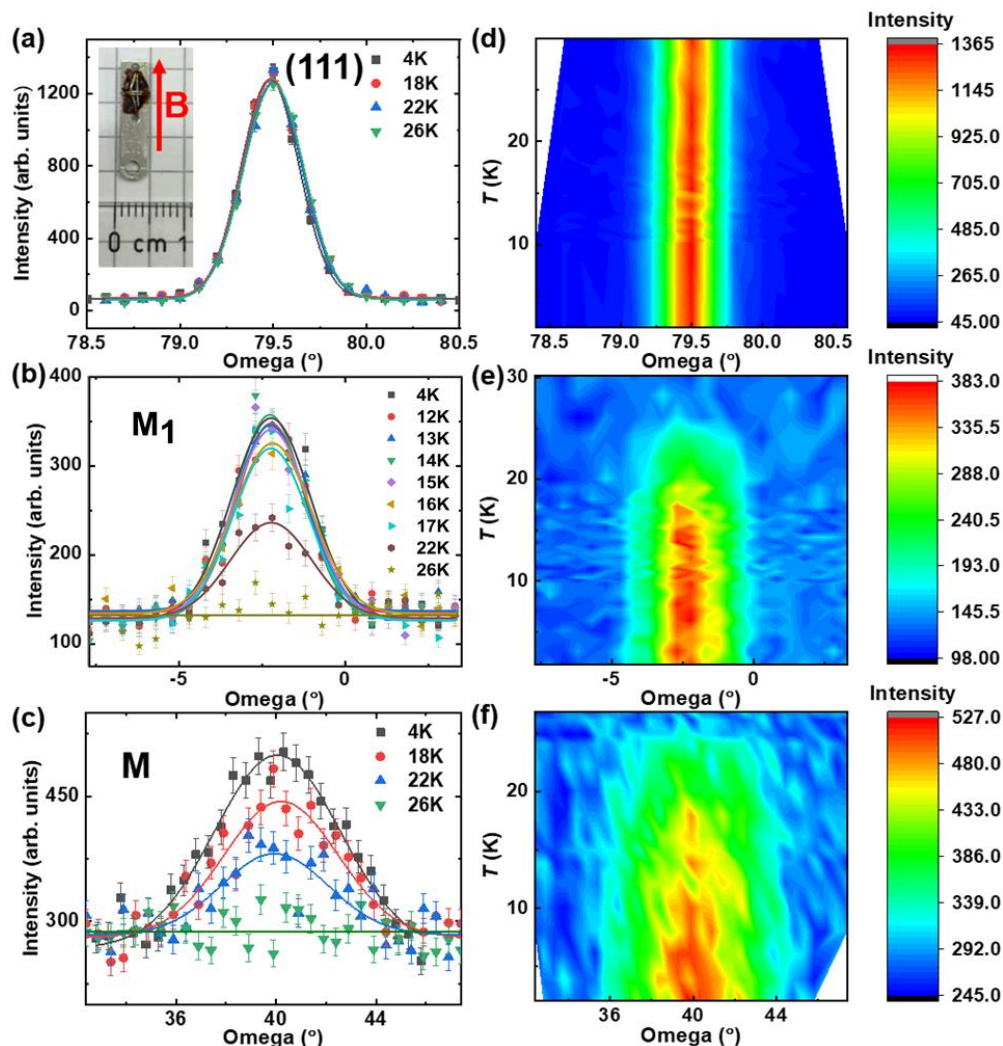


Fig. E1 (S1). Single-crystal neutron diffraction of NCTO. (a), (b), and (c) Zero-field temperature dependence of nuclear peak (111), magnetic peak M₁ (-0.5, 0.5, 1) and M (0.5, 0, 1) at selected temperatures. The data collected at each temperature was fitted by a Gaussian function (the solid lines) to obtain an integrated intensity plotted in Figure 1. Inset: the sample used for the single crystal neutron

diffraction experiment with the magnetic field applying along $[-1, 1, 0]$ direction. (d), (e), and (f) The contour maps of (111), $(-0.5, 0.5, 1)$, and $(0.5, 0, 1)$ peaks. Apparently, there is no any lattice distortion or ferromagnetic contribution with temperature evolution from (d).

Q2: The discussion of the anisotropic evolution of the peak intensities at the M_1 and M points is confusing. Neutron scattering is only sensitive to the perpendicular component of the moment due to the polarization factor, meaning that as the field aligns the moments along the $(-1\ 1\ 0)$ direction one would expect the scattering intensity to decrease as a function of field. Instead, Fig. 1(d) reports the opposite. Was any type of magnetic structure refinement performed as a function of field accounting for domain populations? Some kind of quantitative model to support the domain picture at all would be helpful.

Response: We sincerely appreciate the insightful feedback provided by the reviewer. i) The representation of $M_1(-0.5, 0.5, 1)$ point in the Fig. E2(a) corresponds to the $(-H, H, L)$ plane. As the domain populations increase with the increasing magnetic field $H \parallel (-0.5, 0.5, 0)$, the perpendicular component also increases, and a gradual enhancement of the intensity of the peak is observed. ii) for the intensity of $(0.5, 0, 1)$, as the domain populations decrease with the increase of the magnetic field H , the spin magnetic moment also decreases and the peak intensity decreases gradually. Therefore, the scattering intensity of M_1 reported in Fig. E2(c) (Fig. 1(d) in main text) increases as a function of the field, whereas the intensity at M exhibits the opposite behavior. This phenomenon aligns with the multi-domain scenario. However, due to limited beamtime, we were unable to collect sufficient magnetic reflections for the refinement with a function of the field. Therefore, we only discussed the magnetic structure qualitatively and cannot provide a quantitative magnetic structure model with the magnetic fields.

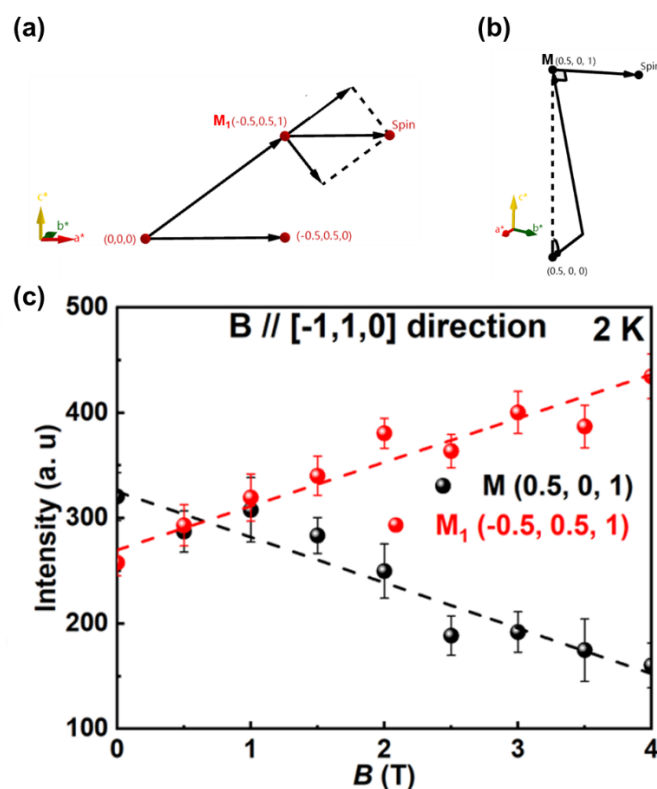


Fig. E2. The schematic of the spin and magnetic-field directions at $(-0.5, 0.5, 1)$ (a) and $(0.5, 0, 1)$ (b) points in the reciprocal space. (c) (Fig. 1(d) in main text) Field-dependent intensity of magnetic

reflections at M and M_1 points, collected at $T = 2$ K and with the magnetic field applied along $[-1, 1, 0]$ direction. Dashed lines are guides to the eyes.

Q3: A central claim of the work from the inelastic scattering is that broadening as a function of field at the K-point is evidence for quantum fluctuations. This broadening should be present not only in the SIKA measurement in Fig. 2(c), but also in the SEQ measurement in Fig. 2(b). Why are the SEQ data not included in Fig. 2(c)? From the color plots, it looks like the SEQ data shows no evidence of broadening. The resolution of the SIKA measurement is also insufficient to determine if this is broadening or if there are two modes.

Response: We agree with the referee's view that the broadening of the excitation spectrum with increasing magnetic field is not readily apparent in the SEQ data. This can be explained by several factors.

1) the dispersion diagram from Time-of-Flight (TOF) data was measured by a high flux/intermediate resolution instrument. Due to the technique limitation, the instrument resolution at the excitation (~ 3 meV) can be obtained accurately and we only could estimate the resolution from the elastic peak line for $E_i = 18$ meV, around 0.41 meV. We attempted a constant-Q cut for the SEQ data at the K point, as shown in the attached figure. Although the exact values of the Full Width at Half Maximum (FWHM) are not as same as the ones from SIKA, the FWHM increases with the field.

2) the FWHMs in the manuscript were measured from a high-resolution cold triple-axis instrument, SIKA, which allows us to perform precise energy scans at specific momentum points. At a fixed final neutron energy of 5 meV, SIKA could achieve a resolution as good as 0.13 meV. This higher resolution is expected to clearly reveal any splits in the excitation spectrum during constant-Q scans. Obviously, the SIKA data shown the broadening of the K-point excitation spectrum with increasing magnetic field, which can be attributed to the quantum fluctuations induced by field effects. In order to make statement clear, we add the resolution information of SIKA in the "Methods" part.

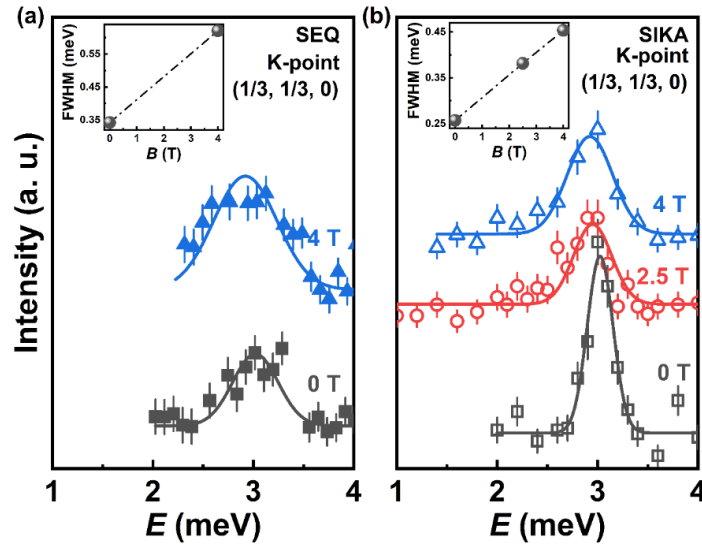


Fig. E3. (a) The constant-Q cut of Time-of-Flight (TOF) data of the K point collected at SEQ by applying magnetic fields up to 4 T along the $[-1,1,0]$ direction. The solid lines represent the fits using a Gaussian function. The inset shows the field dependence of the FWHM of the excitation gap. (b) (Fig. 2(c) in main text) The constant-Q scans of the K-point $(1/3, 1/3, 0)$ collected at SIKA by applying magnetic fields up

to 4 T along the $[-1,1,0]$ direction. The solid lines represent the fits using a Gaussian function. The inset shows the field dependence of the FWHM of the excitation gap.

Q4: Why does broadening necessarily imply quantum fluctuations? This is an overall imprecise statement as it comes with no meaningful discussion about why quantum fluctuations would coexist with a fully ordered moment, as suggested by the muon data in Fig. 3(c). Broadening could originate from enhanced magnon-magnon interactions, domain effects, or some other means, but to jump directly to the most exotic explanation does not sit well with me.

Response: We agree with the referee's view that the conclusion of strong quantum fluctuations in the material cannot be drawn based solely on the broadening in INS results. In fact, this conclusion on strong quantum fluctuations was derived with the μ SR data. The neutron scattering and μ SR respond to different ranges of spin fluctuation rate: μ SR is sensitive to fluctuations in the range of 10^4 to 10^{12} Hz, while neutron scattering is sensitive to fluctuations in the range of 10^8 to 10^{13} Hz. In Fig. 4(e) of main text, the spins of NCTO still exhibit strong fluctuations at a frequency of $0.42 \mu\text{s}^{-1}$, which is 2.2×10^6 Hz (μ SR range) and coexists with the stable internal magnetic field caused by long-range magnetic order, as characterized in Fig. 4(c) in main text. As the μ SR range, we use both complementary methods. To make this conclusion more straightforward, we have added the description "and such strong quantum fluctuations are more intuitively reflected in our μ SR data" at the end of Page 6.

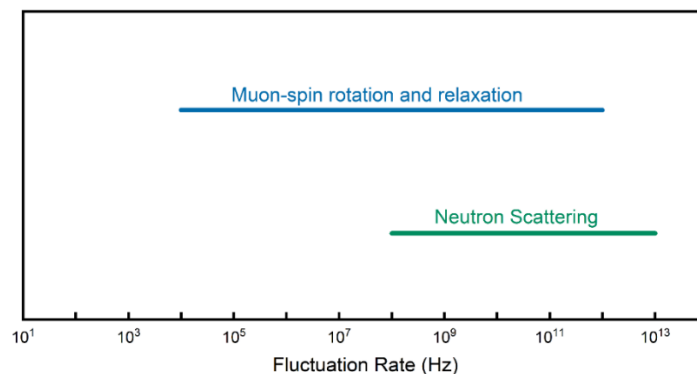


Fig. E4. Detectable spin fluctuation rates for Muon-spin rotation and relaxation and Neutron Scattering methods.

On the other hand, as suggested by the referee, the presence of such strong spin fluctuations in a fully ordered magnet is different from classical fluctuations. These fluctuations originate from the quantum mechanical uncertainty principle and zero-point energy, leading to quantum fluctuations. At finite temperatures, thermal energy disrupts the perfect alignment of spins, making these quantum fluctuations dominant. In fact, the coexistence of strong spin fluctuations and long-range magnetic order has been observed in many materials using neutron scattering and μ SR techniques, such as $\text{Gd}_2\text{Ti}_2\text{O}_7$ [1], $\text{Tb}_2\text{Sn}_2\text{O}_7$ [2], and $\text{FeTe}_2\text{O}_5\text{Br}$ [3]. To make our description more precise, we have added the statement "The presence of such strong spin fluctuations in this nearly fully ordered magnet clearly differs from classical fluctuations and instead arises from quantum fluctuations. Similar behavior has been reported in $\text{FeTe}_2\text{O}_5\text{Br}$ " to the paragraph on Page 9. One more reference has been added as Ref. [61].

Q5: The measurement of T^* at 8 K in the muon data in Fig. 4(d) is not statistically significant and should not be used as evidence of redistribution of magnetic domains, especially since it is not

visible at all in the neutron diffraction. The asymmetry analysis is more convincing.

Response: We agree that it is not statistically significant and should not be used as evidence for the redistribution of magnetic domains, particularly since it is not observed in the neutron diffraction data. Consequently, we have revised the manuscript to remove this interpretation about T^* and instead focus on the more convincing asymmetry analysis. To make our description more precise, we have added the statement " Additionally, anomalies around 8K were observed with $S_\mu // c$, which are consistent with previous magnetic susceptibility results." to the paragraph on Page 10. We believe these revisions will help to eliminate the confusion for readers.

Q6: Were any attempts made to use linear spin-wave theory to model your inelastic neutron scattering spectra?

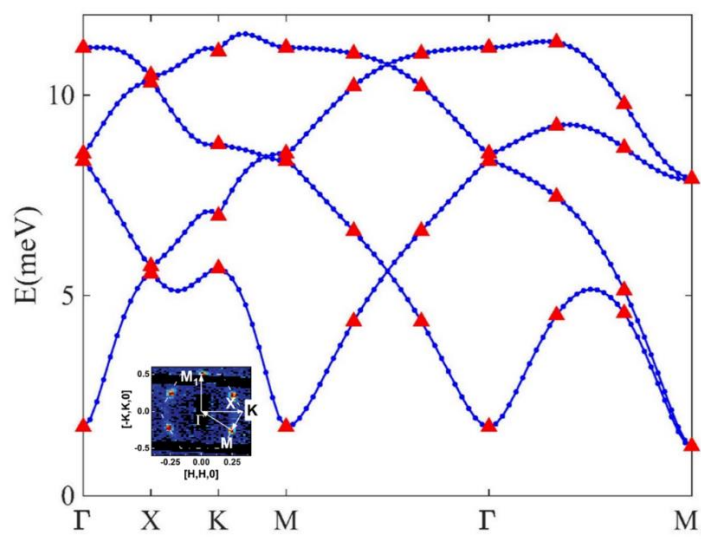


Fig. E5. Calculated zero-field magnon excitation spectrum using linear spin-wave theory, with inset highlighting high-symmetry points in the Brillouin zone.

Response: We are grateful for the reviewer's suggestion. We have indeed performed linear spin-wave theory calculations, as shown in the attached figure. The high-symmetry points Γ , X , K , and M in the figure correspond to those used in our experiments. However, these results have already been reported in another publication of ours (<https://doi.org/10.21203/rs.3.rs-2034295/v1>). To avoid redundancy, we decided not to include this information in the current manuscript and cite it as a reference, Ref. [19]. We believe this approach maintains the originality and focus of our present work. One more sentence has been added "... such as the nearest-neighbor (NN) Heisenberg coupling J_1 and the third NN Heisenberg coupling J_3 beyond the linear spin-wave theory [19]."

Referee 2:

Q1: Although I agree that the residual magnetic fluctuation appears in the magnetic ordered state because I can find some slow relaxation in ZF-muSR spectra at 2 K in Fig. S3, the estimated $\lambda_L = 0.45 \text{ us}^{-1}$ seems to be overestimated. Generally, estimation of relaxation rate λ using an equation, $Ae^{-\lambda t}$, strongly depends on the value of the initial asymmetry A . λ tends to be large when A is small, whereas the λ may be small when A is large. Therefore, the λ_L is considered to depend on the value of A_1 and $(1-\alpha)$ in Eq. (2), which is the equation the authors used for the analysis of ZF-muSR spectra. I would like to ask the authors if the A_1 and $(1-\alpha)$ are compressed and the λ_L is overestimated. The Eq. (2) has another non-oscillating term $A_2 e^{-\lambda_{\text{tail}} t} G_{\text{KT}}$ from the second muon site. Since the second muon site is not affected by spontaneous internal field from the ordered magnetic moment, the muon site contributes to the rotating asymmetry in wTF-muSR data, A_{NM} , even in the magnetic ordered state. The A_{NM} is estimated to be 0.02--0.03 for wTF-muSR analysis as shown in Fig. 3(b). The A_2 in Eq. (2) should be compatible with the A_{NM} value. However, the A_2 is estimated to be 0.05--0.06 for ZF-muSR analysis as shown in Fig. S4. I am afraid I consider that the A_2 value for the ZF-muSR analysis is too large and the A_1 may be compressed. Then, the λ_L becomes large. I would like to suggest a reanalysis of the ZF-muSR data with some restrictions for A_2 .

Response: We are grateful for the reviewer's detailed analysis and comments. We agree with the reviewer: imposing restrictions on A_2 indeed affects other fitting parameters. Following the suggestion, we re-analyzed the ZF-muSR data with A_2 constrained to 0.03. As a result, the reanalyzed data showed a reduction in λ_L . As shown in Fig. E6(c)-(f), the black data points represent the previous fitting results, while the red data points represent the fitting results with A_2 restricted to 0.03. These fitting results were derived from the $S_\mu // a$ data. Due to the reduction of A_2 , λ_L was compressed, and the fitting results were poor as shown in the attached figure from our revised analysis.

$$\mathbf{A}_{\text{ZF}}(t) = A_1 \cdot \left[\alpha \cos(\gamma_\mu B_{\text{int}} t + \varphi) \cdot e^{-\lambda_T t} + (1 - \alpha) \cdot e^{-\lambda_L t} \right] + A_2 e^{-\lambda_{\text{tail}} t} \cdot G_{\text{KT}} . \quad (1)$$

It is worth noting that the fitting parameters for zero-field (ZF) and weak transverse-field (wTF) conditions should be distinguished, considering that A_{NM} and A_2 respond differently under these conditions. We wish to clarify that the A_{NM} data were obtained under a magnetic field of 0.01T, whereas the A_2 data were collected under zero-field conditions. Taking these factors into account, along with the observed deterioration in fit results after modifications, we opted not to enforce such constraints and presented the results as outlined in the main text. This approach enabled us to derive a more precise depiction of the zero-field data.

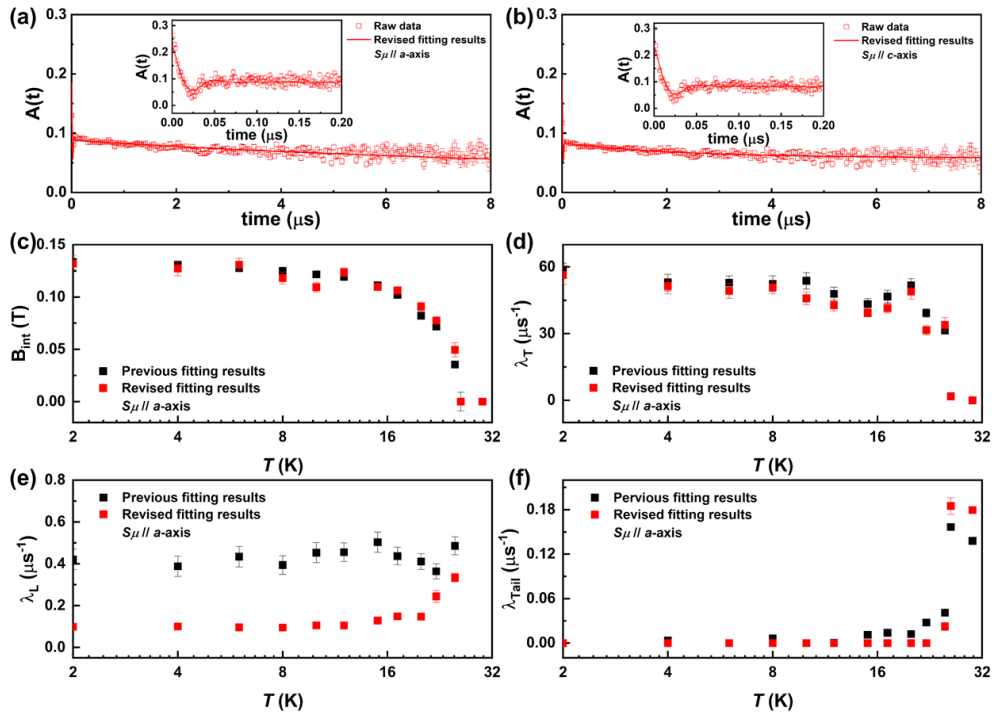


Fig.E6. (a) and (b) Zero-field (ZF) μ SR time spectra measured at $T=2$ K with the muon-spin direction S_μ // a and c -axis, respectively. The inset shows the results of the 0.2 μ s measurement. The solid curves represent fits to Eq. (1) after constraining A_2 . (c)-(f) Temperature dependence of the B_{int} , λ_T , λ_L , and λ_{tail} , respectively, as derived from ZF- μ SR analysis. The black data points are from the original submission, while the red data points are the revised fitting results after constraining A_2 .

Q2: The authors pointed out the similarity of the temperature dependence of muon spin relaxation rate between the NCTO and a quantum spin liquid candidate YbMgGaO₄. However, one for the YbMgGaO₄ showed no sign of magnetic ordering and the λ increased with decreasing temperature before the following saturating behavior near the lowest temperature as shown in Ref. 59. I consider that the present result is more like the μ SR result for FeTe₂O₅Br (M. Pregelj et al., PRL, 109, 227202 (2012)). The authors should refer to the previous μ SR results showing persistent spin dynamics in the magnetic ordered state.

Response: We are grateful for the reviewer's comment regarding the comparison of the temperature dependence of the muon spin relaxation rate. We agree that the present result is more comparable to the μ SR results for FeTe₂O₅Br (M. Pregelj et al., PRL, 109, 227202 (2012)). Accordingly, we have cited this reference as Ref. [61] and discussed the similarities between NCTO and FeTe₂O₅Br in our revised manuscript. To make our description more precise, we have added the statement "where strong spin fluctuations coexist with a fully magnetically ordered state" to the paragraph on Page 11.

Q3: The lack of μ SR data at longitudinal-field (LF) conditions, which is an external field applied along the initial muon spin polarization, gives some ambiguity for the interpretation of the present μ SR data. I agree that the slow relaxation in the ZF- μ SR spectra may represent the microscopic dynamical magnetic properties of the sample. However, no-decoupling behavior should be checked by applying LF to confirm that the slow relaxation certainly originated from the magnetic

fluctuation. If the authors have extra μ SR data, especially at LF, these should be added to the manuscript.

Response: We are grateful for the reviewer's thorough review of our manuscript and the valuable feedback provided. Regarding the absence of μ SR data under longitudinal-field (LF) conditions and the ambiguity it may introduce in interpreting our current μ SR data, we acknowledge and respect your concerns. The slow relaxation observed in the zero-field (ZF) μ SR spectra could indeed reflect the microscopic dynamical magnetic properties of the sample. However, the confirmation of non-decoupling behavior through LF application would be essential to ascertain that this slow relaxation originates from magnetic fluctuations.

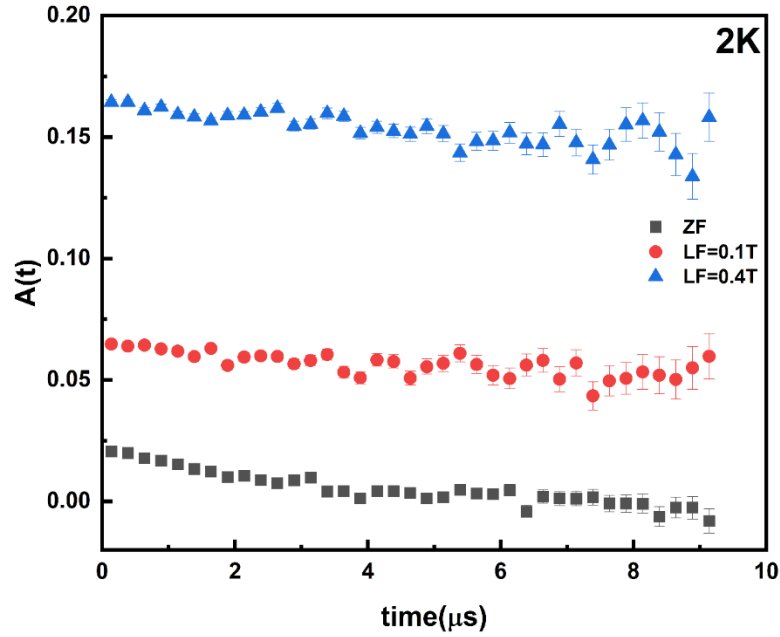


Fig.E7. The μ SR time spectra measured at $T = 2$ K by applying magnetic fields up to 0.4 T with the muon-spin parallel to the c-axis.

In consideration of the suggestion regarding LF data, due to limited beamtime, we only conducted LF experiments at base temperature. Specifically, given our sample's internal field of 0.1 T, we theoretically expected muon spins to decouple at temperatures below 2 K under a field greater than 0.1 T. However, the data have exhibited distinct relaxation, suggesting incomplete decoupling likely due to fluctuations. Therefore, we selected this magnetic field to investigate temperature dependence (Fig.S4 in Supplementary Materials). Although the LF data is not included in the main text, it is already clear that the slow relaxation certainly originated from the magnetic fluctuation.

References:

- [1] A. Yaouanc, P. Dalmas de Re'otier, V. Glazkov, C. Marin, P. Bonville, J. A. Hodges, P. C. M. Gubbens, S. Sakarya, and C. Baines. PRL 95, 047203 (2005).
- [2] P. Dalmas de Re'otier, A. Yaouanc, L. Keller, A. Cervellino, B. Roessli, C. Baines, A. Forget, C. Vaju, P. C. M. Gubbens, A. Amato, and P. J. C. King. PRL 96, 127202 (2006).
- [3] M. Pregelj, A. Zorko, O. Zaharko, D. Arc'on, M. Komelj, A. D. Hillier, and H. Berger. PRL 109, 227202 (2012).

26th Jun 24

Dear Professor Ma,

Thank you for submitting your revised manuscript, "Static magnetic order with strong quantum fluctuations in spin-1/2 honeycomb magnet Na₂Co₂TeO₆", to Communications Materials. It has now been seen again by our 2 referees, whose comments are appended below. You will see that while they find your work improved, there are still important points raised by Reviewer #2, as it appears that some of your claims regarding the λ_T behaviour are not fully supported by the current data.

While we are still interested in the possibility of publishing your study in Communications Materials, we would like to consider your response to these concerns and ask you to further revise your manuscript, by adding more convincing data and/or toning down some of the claims, before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

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We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Dr Aldo Isidori

Senior Editor

Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I believe that the authors have satisfied the comments and that there is sufficient evidence for the claims that are in the revised version of the manuscript - I would also like to thank them for addressing each of my questions in detail. If they have time, a follow-up study with detailed neutron diffraction measurements confirming their domain picture would be helpful, but I believe that the current version is ready for publication.

Reviewer #2 (Remarks to the Author):

I appreciate the author's efforts to answer my questions. I received enough elaboration for my previous three questions from the authors. However, I have a new question for the revised manuscript which is shown below.

The authors suggest that anomalies are observed at T_F and 8 K in the T dependence of λ_T , which is shown in Fig. 4(d), in the revised manuscript. However, the number of data points in Fig. 4(d) seems not enough to conclude the existence of anomalies at T_F and 8 K in the T dependence of λ_T .

Furthermore, I am concerned about discussing something based on the small change of λ_T data. Generally, λ_T may be contributed by both static internal field distribution and spin dynamics, and it shows complex T-dependence. λ_T often makes small convex and concave in T dependence λ_T even when there is no phase transition. Interpretation of λ_T data would be difficult.

Even if the small convex downwards in Fig. 4(d) are true, it sounds difficult to understand such λ_T behavior is originating from the domain wall redistribution. Since the μ SR is a microscopic local probe, which detects the internal field from the localized magnetic moments located within a radius of 50-200 angstrom around the muon, it is natural not to detect the domain redistribution if the domain size is large enough. I am not sure about the domain size of this material, but the conventional magnetic domain size is known as 10^{-4} to 10^{-6} m.

I would like to recommend the authors not conclude domain redistribution from the present λ_T .

Comment:

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Response:

We want to thank the reviewer for the valuable feedback and for appreciating our efforts addressing previous questions. We acknowledge the new concern regarding the revised manuscript and would like to address them in detail.

Firstly, we agree with the reviewer's comment on the limited number of data points in Fig.E1(c)(Fig.4(d)): not being sufficient to conclusively identify anomalies at T_F and 8 K in the temperature dependence of λ_T . Furthermore, we agree that λ_T can be influenced by both static internal field distribution and spin dynamics, leading to a complex temperature dependence. This complexity makes the interpretation of λ_T data not straightforward. To provide a more comprehensive understanding of magnetic behavior, we combined multiple experimental techniques. Our previous magnetic susceptibility measurements have identified an anomaly at T_F in Fig.E1(b)(Fig.3(b)). Additionally, temperature-dependent neutron diffraction experiments, as shown in Fig.E1(a)(Fig.1(c)), have also revealed anomalies at T_F . Different magnetic domains, indicated by M and M_1 magnetic peaks, exhibit distinct behaviors that suggest domain redistribution. Therefore, while μ SR data alone may not provide conclusive evidence, it is consistent with the findings from these other techniques.

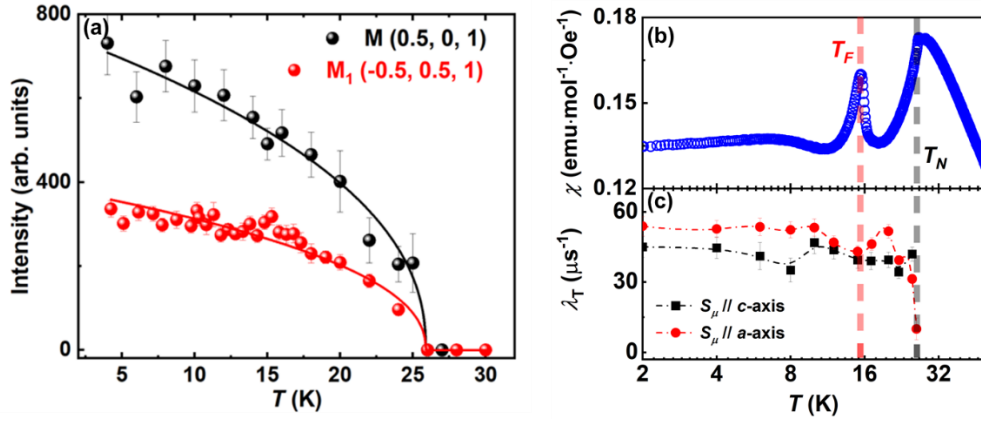


Fig. E1. (a)(Fig.1c) The temperature dependence of the zero-field intensity of two magnetic reflections at $M(0.5, 0, 1)$ and $M_1(-0.5, 0.5, 1)$. The solid lines represent the fits to the phenomenological model (see details in the text). (b)(Fig.3b) The temperature-dependent zero-field cooling (ZFC) magnetic susceptibility curve $\chi(T)$ measured at $B = 0.01$ T with $B \parallel a^*$ -axis. The magnetic susceptibility data were taken from Ref. [19]. (c)(Fig.4d) Temperature dependence of the λ_T , as derived from ZF- μ SR analysis.

Secondly, we understand that depending solely on μ SR data might not be sufficient for drawing firm conclusions about domain redistribution. In response to your constructive suggestion, we have revised our manuscript to soften the discussion regarding the anomaly in λ_T at T_F and its interpretation. We have reduced the focus on the role of μ SR data in concluding domain redistribution and have instead focused on the wider perspective provided by both neutron scattering and μ SR data, presenting a more balanced discussion of the results. The sentence “These results suggest that the anomaly observed at T_F should be attributed to a redistribution of the magnetic domains originating from three different k -vectors of the zigzag AFM order.” has been changed into “The observed anomaly at T_F is consistent with previous magnetic susceptibility and neutron scattering results, and this might suggest a possibility of redistribution of the magnetic domains originating from three different k -vectors of the zigzag AFM order.” on page 10. Moreover, we have revised the statement from “At the same time, the $\lambda_T(T)$ curves suggest that a domain reorientation occurs at T_F , coinciding with the multi-domain scenario.” to “At the same time, the potential transition trend observed in the $\lambda_T(T)$ curves is consistent with previous measurement results and might indicate a domain reorientation occurring at T_F , coinciding with the multi-domain scenario.” on page 10.

8th Jul 24

Dear Professor Ma,

Your manuscript titled "Static magnetic order with strong quantum fluctuations in spin-1/2 honeycomb magnet Na₂Co₂TeO₆" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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We hope to hear from you within two weeks; please let us know if the process may take longer.

Best regards,

Dr Aldo Isidori

Senior Editor

Communications Materials

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

I would like to appreciate the author's effort for the revision. I think that the revised manuscript well addressed my question and it discusses the domain redistribution more carefully than the previous version. I want to recommend the manuscript published in the journal.