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Peer Review File

Anomalous continuum scattering and higher-order van Hove singularity in the strongly anisotropic S = 1/2 triangular lattice antiferromagnet

Reviewers' Comments:

Reviewer #1:

Remarks to the Author:

In this manuscript, the authors experimentally examined magnetic excitation of a polycrystalline sample of spin-1/2 triangular lattice antiferromagnet Ba2La2Co2Te2O12 (BLCTO) by using inelastic neutron scattering (INS). The INS spectra above the Neel temperature (T_N) were fitted by using the Landau-Lifshitz dynamical simulations for a Heisenberg-type classical spin model with XXZ exchange anisotropy on a triangular lattice. The authors found that the model with anisotropy parameter Delta=0.56 well reproduces the INS data. Using the same parameters, dynamical spin structure factor was calculated at zero temperature by employing nonlinear spin-wave theory (NLSWT) and Schwinger boson (SB) approach. The INS spectra below T_N was well explained by the SB calculations, where continuum excitations higher in energy than single-magnon branch are clearly seen in contrast to spin-wave based theories. Based on these findings, the authors concluded that BLCTO is a novel quantum spin system with strong exchange anisotropy, which is different from other triangular antiferromagnets.

Experimental and theoretical data are well summarized and analyzed systematically. Therefore, this paper provides a new view of frustrated quantum spin systems in terms of exchange anisotropy. In particular, it is interesting and impressive that the Heisenberg model with strong anisotropy very well explains high-energy continuum excitation as well as momentum-independent flat spectral distributions at and above the single-magnon excitation energy, the latter of which was assigned as higher-order van Hove singularity. This will stimulate further theoretical investigations on spin dynamics in such exchange anisotropic models.

Concerning momentum-independent flat spectral distributions, the NLSWT (as well as LSWT) also shows such flat profile almost independent of Delta as shown in Supplementary Figs. 8 and 9. Therefore, the flat profile itself does not necessary exclude NLSWT. Even for the SB results, there are flat profiles for large Delta. Therefore, a careful discussion is necessary to conclude that higher-order van Hove singularity is the origin of the flat profile. One suggestion would be to claim that Delta=0.6 is flatter in momentum space than Delta=0.8 based on Supplementary Fig. 10. Note that Supplementary Figs. 7, 8, 9, 10, and 16 were not referred in any place. ("Supplementary Fig. 16" in the second paragraph of page 7 in Supplementary Information (SI) should be " Supplementary Fig. 17".)

Though the main text is well organized and connivingly described, there remains statements in the abstract, which are incoherent with the main text. In the abstract, it was mentioned that "a spinon confinement length that markedly exceeds the lattice spacing". However, there is no statement and information on the confinement length not only in the main text abut also in SI. Similarly, the word "a quantum melting point" in the abstract does not appear in the main text (only appears in SI). These terms in the abstract are a kind of key words of this manuscript. Therefore, they should be adequately explained in the main text.

In Fig. 2e, a vertical broken line indicating BLCTO is located at delta/lambda_SCO=2. However, it mentioned that delta/lambda_SCO=1.854 in Supplementary Notes. They should be consistent each other.

Reviewer #2:

Remarks to the Author:

This paper treats the magnetic excitations of $Ba_2La_2CoTe_2O_{12}$, abbreviated BLCTO. This compound is magnetically described as a spin-1/2 XXZ model on a triangular lattice. I read this manuscript with great interest. The authors investigated the excitation spectra of BLCTO powder by inelastic neutron scattering. They first evaluated the exchange anisotropy as D=0.56, applying LLD simulations to the excitation data above TN. Their result shows that the LLD simulation is a powerful tool for estimating the exchange anisotropy of the powder sample. Next, the authors analyzed the excitation spectrum below TN, using the Schwinger boson approach for spinons. The theoretical spectrum appears to capture the characteristics of the experimental spectrum. Their result shows that the spinon is an elementary excitation in the spin-1/2 triangular antiferromagnet. Since the spinon is the elementary excitation characteristic of quantum spin liquid, their result implies that the ground state has a significant spin liquid-like component. This manuscript is interesting and instructive. Thus, I recommend publication. However, I have some questions and comments as follows.

1. In Supplementary Information, the authors described the anisotropy of the g-factors. However, there is little description of the g-factor's anisotropy in the text, and hence, readers may misunderstand that the authors assume the isotropic g-factor.

2. As shown in Fig. 4, the best fit using the Schwinger boson calculation is given by J_1=1.77 meV, D=0.56, and J_2=0. Are these parameters consistent with the saturation fields

measured in Ref. [29]? Any statement on the issue is needed because the saturation fields are another constraint for the exchange parameters.

3. Some readers will be interested in how the excitation spectrum varies with decreasing D. Some explanations for this point are desired.

Reviewer #3:
Remarks to the Author:
The well-written paper shows the large extent of analysis possible from powder ToF INS data when using multiple incident wavelengths and temperature regimes. Indeed, examples of real TLAF with various types of anisotropy are missing to validate the proposed phase diagram. The presented paper is very valuable in this regard. My comments below are aimed at making the paper more consistent and on making the data analysis process more transparent.

• Often the authors are quick to pose the hypothesis of the significant role of quantum fluctuations without augmenting. No other hypotheses are considered for explaining the models/data disagreement (see eg. pg.7, but more places in the text).
• Often the authors argue the agreement with the model is excellent (eg. page 6) when it only describes a narrow region of the S(E). It is not clear how good it is at describing S(Q).
• What is the relation between the J1 obtained in the paramagnetic regime (2.6meV) and the one in the ordered phase (1.55-1.7meV). Why are they different, why is the paramagnetic larger? Would other exchange interactions increase in magnitude below TN? Would they contribute to LRO? They should discuss the relation between these two different results.
• Below TN, a continuum is seen above the magnons, it can partially be explained with Schwinger boson theory (the best option in the paper) but the contribution of two(N)-magnon continuum is not considered in the model. It is however discussed in the paper (see page 8). The authors should at least mention the energy range the two-magnon continuum should occupy perhaps by adding a sign/line in fig. supplementary 10e-g.
• Considering the Schwinger boson result on the magnitude of the magnetic exchange interactions. How can the authors re-interpret heat capacity and susceptibility data? Would they be able to describe the field dependency of the heat capacity data?
• Considering the models, can the authors interpret the two critical magnetic fields (~9 and 16T) found in Kojima's magnetization data?
• Can the authors comment on what are they doing to integrate the ToF data at low wavevectors extending in energy all the way to the max. transfer energy? What are they doing for the points outside the ToF E-Q window?
• The elastic line is cut off the data (see eg. fig.3), are they just not plotting the elastic line or has this been subtracted by using higher temperature data? This can drastically change fig 3d below 1meV, the most crucial comparison cut. Why not do the S(E) comparison using all three available data sets (Ei=6,10, 1.5meV -last from supplementary?)
• The main paper should show the q-dependency of the intensity for the data below TN for all the main features (arrows in fig 4 e), and show how these compare to the three models. In general, q-dependencies are missing in the whole data analysis.
• Can the authors include in the discussion an explanation (for the experimentalist) of the origin of the continuum in the Schwinger boson theory?
• Finally, can the authors comment on the advantages/disadvantages of studying an anisotropic system using only powder samples?

First Report of Referee 1

General Comment: In this manuscript, the authors experimentally examined magnetic excitation of a polycrystalline sample of spin-1/2 triangular lattice antiferromagnet Ba2La2Co2Te2O12 (BLCTO) by using inelastic neutron scattering (INS). The INS spectra above the Neel temperature (T_N) were fitted by using the Landau-Lifshitz dynamical simulations for a Heisenberg-type classical spin model with XXZ exchange anisotropy on a triangular lattice. The authors found that the model with anisotropy parameter Delta=0.56 well reproduces the INS data. Using the same parameters, dynamical spin structure factor was calculated at zero temperature by employing nonlinear spin-wave theory (NLSWT) and Schwinger boson (SB) approach. The INS spectra below T_N was well explained by the SB calculations, where continuum excitations higher in energy than single-magnon branch are clearly seen in contrast to spin-wave based theories. Based on these findings, the authors concluded that BLCTO is a novel quantum spin system with strong exchange anisotropy, which is different from other triangular antiferromagnets.

Experimental and theoretical data are well summarized and analyzed systematically. Therefore, this paper provides a new view of frustrated quantum spin systems in terms of exchange anisotropy. In particular, it is interesting and impressive that the Heisenberg model with strong anisotropy very well explains high-energy continuum excitation as well as momentum-independent flat spectral distributions at and above the single-magnon excitation energy, the latter of which was assigned as higher-order van Hove singularity. This will stimulate further theoretical investigations on spin dynamics in such exchange anisotropic models.

Reply: We appreciate the referee's positive view on the impact of our work and their supportive comment.

Comment #1: Concerning momentum-independent flat spectral distributions, the NLSWT (as well as LSWT) also shows such flat profile almost independent of Delta as shown in Supplementary Figs. 8 and 9. Therefore, the flat profile itself does not necessary exclude NLSWT. Even for the SB results, there are flat profiles for large Delta. Therefore, a careful discussion is necessary to conclude that higher-order van Hove singularity is the origin of the flat profile. One suggestion would be to claim that Delta=0.6 is flatter in momentum space than Delta=0.8 based on Supplementary Fig. 10.

Reply: We thank the referee for the thoughtful suggestion. Before going further into details regarding the referee's comment, we note that we have revised relevant sections in the main text and have modified a relevant figure in the Supplementary Information to improve our explanations in this regard (see Fig. R1 below). As noted by the Referee, all three calculations exhibit some flat spectral weight distribution at the top of the one-magnon bands independent of Δ, which is around 2.3 meV for LSWT, and 1.5~1.8 meV for NLSWT and Schwinger Boson (SB) calculations. However, when looking into the region below the top energy, the one-magnon dispersion becomes noticeably "flatter" if Δ is significantly smaller than 1, as the Referee already mentioned. For instance, the LSWT or NLSWT calculations with $0.7 < \Delta < 0.9$ (Supplementary Figs. 10–11) still possess dispersive branches from approximately 1 meV to the top of the one-magnon bands, unlike our experimental observation. On the other hand, decreasing Δ further makes them "flatter" and eventually merges them into the flat spectral weight at the top position, which better aligns with our observation. The higher-order van Hove singularity we refer to is more relevant to this "flatter" spectrum found in a larger portion of the one-magnon dispersion, instead of the usual flatness at the top of one-magnon bands that always appears regardless of the model. In other words, the Δ value that reaches a maximum flatness corresponds to the formation of a higher-order van Hove singularity at the Γ and K points.

A key point is that a maximal flatness characterized by the higher-order van Hove singularity is predicted to occur in the magnetic spectrum for appropriate values of $\Delta < 1$ for all three of the calculation methods discussed in this work (LSWT, NLSWT, and SB). It must be captured by any calculation due to the generality of the symmetry argument underpinning its existence, as derived in the main text using a Taylor expansion analysis (see Eq. (2) of the main text). For clarity, we have added the following sentence in the main text (page 10):

"The above arguments derived from Eq. (2) are independent of the models for elementary excitations, i.e., they hold for LSWT, NLSWT, and Schwinger Boson calculations alike."

In LSWT and NLSWT, however, the higher order van Hove singularity is predicted for $\Delta \approx 0.25$ and $\Delta \approx 0.35$, respectively, and the corresponding magnetic spectra in Supplementary Figures 10 and 11 thus exhibit the flattest features around $\Delta = 0.3$, much below the experimental anisotropy $\Delta \approx 0.56$. Indeed, the NLSWT calculation with $\Delta = 0.56$ (Fig. 4g & g) still yields a spectrum with noticeable dispersion between 1.3 and 1.6 meV, unlike our observation. In contrast, the Schwinger boson approach correctly predicts the higher-order van Hove singularity close to the experimental anisotropy and best describes the degree of flatness observed in the data. This further corroborates our hypothesis that this approach provides the best quantitative description of the spin dynamics in BLCTO.

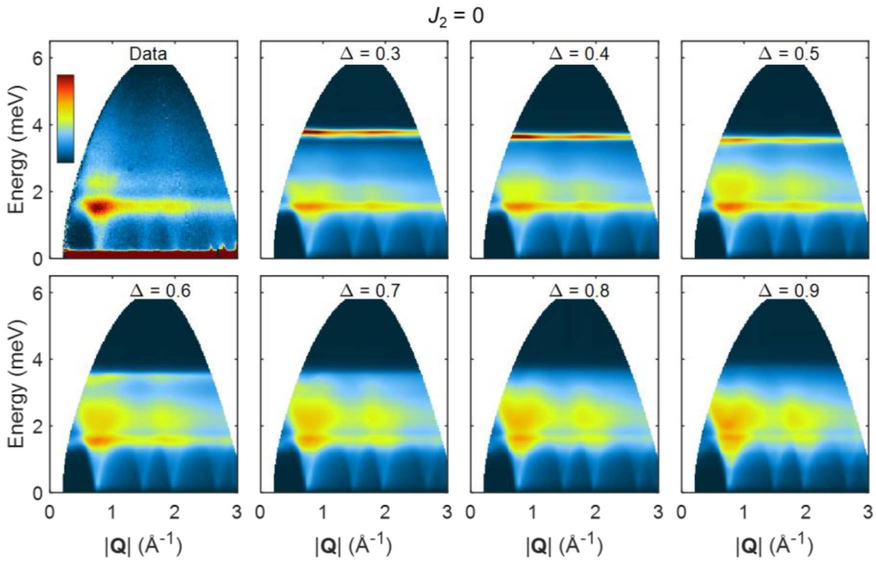

Fig. R1 Effects of Δ on a powder-averaged spinon spectrum from the Schwinger boson theory.

For a better understanding, we have revised the Supplementary Figure that shows the trend of flatness under different Δ in the Schwinger boson results, which is Supplementary Fig. 12 of the revised version (or see Fig. R1 above). It shows the Schwinger boson spectra for a wider range of Δ (0.3 ~ 0.9), like Supplementary Figs. 10–11 in the new SI. While the magnetic spectrum obtained from Schwinger boson theory contains flat profiles around the top of the two-spinon bound states (= one-magnon branches) for an entire range of anisotropies, the overall one-magnon profile is the flattest around the critical anisotropy, $\Delta = 0.6$, corresponding to the higher order van Hove singularity. On the other hand, the one-magnon branches become progressively "less" flat as Δ is tuned away from this critical value, especially around the K and Γ points (see Supplementary Figs. 12 and 14). This observation supports the hypothesis that the flat features originate from the higher-order van Hove singularity and further demonstrates their robustness as they remain observable for a finite window around the critical anisotropy. We note that the situation is completely analogous for LSWT and NLSWT; the flat profiles survive to larger values of the anisotropy but are flattest at $\Delta = 0.3$ (see Supplementary Figs 10–11), i.e., precisely where LSWT and NLSWT predict the higher order van Hove singularity.

Comment #2: Note that Supplementary Figs. 7, 8, 9, 10, and 16 were not referred in any place. ("Supplementary Fig. 16" in the second paragraph of page 7 in Supplementary Information (SI) should be " Supplementary Fig. 17".)

Reply: We thank the referee for pointing this out. Supplementary Figs. 7~10 and 16 are now referred to in the main text. We note that with additional Supplementary Figures their numbering has changed from the originally submitted manuscript.

Comment #3: Though the main text is well organized and connivingly described, there remains statements in the abstract, which are incoherent with the main text. In the abstract, it was mentioned that "a spinon confinement length that markedly exceeds the lattice spacing". However, there is no statement and information on the confinement length not only in the main text abut also in SI. Similarly, the word "a quantum melting point" in the abstract does not appear in the main text (only appears in SI). These terms in the abstract are a kind of key words of this manuscript. Therefore, they should be adequately explained in the main text.

Reply: We thank the referee for pointing out this discrepancy. We have revised our discussions on the spinons in the main text so that the keywords "spinon confinement length that markedly exceeds the lattice spacing" (or equivalently, significant weakening of spinon confinement) and "quantum melting point" are included and properly explained.

Comment #4: In Fig. 2e, a vertical broken line indicating BLCTO is located at delta/lambda_SCO=2. However, it mentioned that delta/lambda_SCO=1.854 in Supplementary Notes. They should be consistent each other.

Reply: While the blue dashed line indicating the location of BLCTO was correctly at δ/λ = 1.854, we admit that the previous version of Fig. 2e did not make this clear enough for the reader. In the new figure, we have explicitly noted the *x*-axis value of the vertical broken line (= 1.854) and increased the tick length for better clarity.

First Report of Referee 2

General Comment: This paper treats the magnetic excitations of Ba2La2CoTe2O12, abbreviated BLCTO. This compound is magnetically described as a spin-1/2 XXZ model on a triangular lattice. I read this manuscript with great interest. The authors investigated the excitation spectra of BLCTO powder by inelastic neutron scattering. They first evaluated the exchange anisotropy as D=0.56, applying LLD simulations to the excitation data above TN. Their result shows that the LLD simulation is a powerful tool for estimating the exchange anisotropy of the powder sample. Next, the authors analyzed the excitation spectrum below TN, using the Schwinger boson approach for spinons. The theoretical spectrum appears to capture the characteristics of the experimental spectrum. Their result shows that the spinon is an elementary excitation in the spin-1/2 triangular antiferromagnet. Since the spinon is the elementary excitation characteristic of quantum spin liquid, their result implies that the ground state has a significant spin liquid-like component. This manuscript is interesting and instructive. Thus, I recommend publication. However, I have some questions and comments as follows.

Reply: We appreciate the referee's positive evaluation and recommendation of our work.

Comment #1: In Supplementary Information, the authors described the anisotropy of the g-factors. However, there is little description of the g-factor's anisotropy in the text, and hence, readers may misunderstand that the authors assume the isotropic g-factor.

Reply: We have added a sentence to the main text (page 8) mentioning the usage and importance of anisotropic g-factors for our calculations.

Comment #2: As shown in Fig. 4, the best fit using the Schwinger boson calculation is given by *J*1=1.77 meV, D=0.56, and *J*2=0. Are these parameters consistent with the saturation fields measured in Ref. [29]? Any statement on the issue is needed because the saturation fields are another constraint for the exchange parameters.

Reply: We thank the referee for raising an excellent point. Indeed, the experimental saturation field values reported in Ref. 29 provide a good opportunity to crosscheck the exchange parameters. The values we have obtained for the Hamiltonian energy scales agree within the 10% level with the saturation fields previously determined for BLCTO. An abbreviated form of the analysis and discussion we provide here has been added to the Supplementary Notes for the manuscript.

The saturation fields perpendicular (H_s^{ab}) and parallel (H_s^c) to the *c*-axis were extracted to be 32 T and 41 T, respectively, in Ref. 25. However, there is some ambiguity in these values due to the smooth variation of the *M*–*H* curve around the saturation, likely due to powder averaging in conjunction with anisotropic *g*-factors and strong planar XXZ anisotropy ($\Delta < 1$). A fairer estimation of H_s^{ab} and H_s^c considering these uncertainties and based on the *dM*/d*H* curve would be approximately 30±2 T and 40±2 T.

H_s^c and H_s^{ab} for the J_1–Δ model are estimated as follows using the linear spin-wave theory (see Ref. 25):

$$H_s^{ab} = \frac{J_1 S\big(9 + 4(J_c\,/\,J_1)\big)}{g_{ab}\mu_{\mathrm{B}}} \tag{R1}$$

$$H_s^c = \frac{J_1 S\big(3 + 6\Delta + 2(1+\Delta)(J_c\,/\,J_1)\big)}{g_c\mu_{\mathrm{B}}}, \tag{R2}$$

where J_c is the interlayer coupling strength, g_{ab} (g_c) is the *g*-factor perpendicular to (along) the *c*-axis, and $\mu_{\mathrm{B}} = 0.05788\ \mathrm{meV/T}$. Using the anisotropic *g*-factors presented in the previous Supplementary Note and the optimal parameter set [J_1 = 1.77 meV, Δ = 0.56, J_2 = 0] with J_c = 0 (J_1 = 1.77 meV is from the Schwinger Boson calculation result in Fig. 4), we obtain H_s^{ab} = 27.9 T and H_s^c = 35.7 T, respectively. A more realistic assumption, $J_c \sim 0.05J_1$, yields H_s^{ab} = 28.5 T and H_s^c = 36.6 T. These values are 90~95 % of the experimental saturation fields.

Several factors may contribute to the 5~10% underestimation of H_s^{ab} and H_s^c. introducing $J_2 = 0.03J_1$ to the model, as suggested by our LLD analysis, increases optimal J_1 derived from the Schwinger Boson theory to 1.87 meV, 6 % larger than J_1 = 1.77 meV (see Supplementary Fig. 13 in the new SI). While this will likely reduce the underestimation of the saturation fields, further quantitative estimation is limited by unclear effects of J_2 on the *M*–*H* curve. Another factor could be the influence of $J_{z\pm}$ on the saturation field, which may range from $-0.15J_1$ to $0.15J_1$ according to our LLD analysis (see Supplementary Notes). Indeed, this effect was not included in either the Schwinger Boson calculation or the theoretical estimation of the *M*–*H* curve. Thus, given the simplicity of the J_1–Δ model suffering from systematic errors, achieving 90~95 % consistency in the saturation field analysis is reasonable.

In addition, the position of the 1/3-magnetization plateau in the *M*–*H* curve can provide additional diagnostic insights into our parameter set, although it is subject to greater systematic errors compared to the saturation field analysis. Please refer to our reply to Comment #6 from Referee 3 for additional details on this point.

Comment #3: Some readers will be interested in how the excitation spectrum varies with decreasing D. Some explanations for this point are desired.

Reply: We thank the referee for the helpful suggestion. We have supplemented the relevant explanations in the main text, which can be summarized as follows:

1) Reducing Δ from 1 gradually suppresses quantum fluctuations as implied in Fig. 1 (the

quantum spin liquid eventually disappears around $\Delta \sim 0.35$). Please note the vertical axis of Fig. 1 is shown as ($1-\Delta$). Thus, we expect that the spin dynamics close to the XY limit ($\Delta \sim 0$) is better described by the non-linear spin-wave theory, rather than the Schwinger Boson theory (pages 3 & 7 of the main text). However, this work confirms experimentally that quantum fluctuations are still significant down to $\Delta = 0.56$, and therefore, the noticeable suppression of quantum fluctuations is anticipated to be found at Δ much smaller than 0.5.

2) Decreasing Δ from 1 generally modifies magnetic excitations to be flat, independent of the theoretical model. Please see Supplementary Figs. 10–12 in the new SI. This flatness becomes most pronounced at the critical value Δ_0 which ranges from 0.2 to 0.6 depending on the calculation methods, and it corresponds to the onset of a higher-order van Hove singularity. We have improved the relevant explanations in the main text (pages 9–10).

First Report of Referee 3

General Comment: The well-written paper shows the large extent of analysis possible from powder ToF INS data when using multiple incident wavelengths and temperature regimes. Indeed, examples of real TLAF with various types of anisotropy are missing to validate the proposed phase diagram. The presented paper is very valuable in this regard. My comments below are aimed at making the paper more consistent and on making the data analysis process more transparent.

Reply: We are grateful for the referee's appreciation of the value of our work. We also thank the referee for the thoughtful comments from a careful review. We hope that the point-by-point responses below have satisfactorily addressed the referee's questions and concerns.

Comment #1: Often the authors are quick to pose the hypothesis of the significant role of quantum fluctuations without augmenting. No other hypotheses are considered for explaining the models/data disagreement (see eg. pg.7, but more places in the text).

Reply: We acknowledge that our manuscript initially may have appeared to preferentially consider quantum fluctuations without sufficiently discussing alternative reasons. We have reinforced our main text and SI in response to this comment.

To address this concern, it is pertinent to clarify the three primary interpretations that could account for the continuum scattering signal: 1) intrinsic quantum fluctuations, 2) substantial structural disorder, and 3) multiple scattering signals in neutron experiments. We have examined the possibility of disorder effects and concluded that this scenario can be robustly excluded for the following reasons. Firstly, our bulk characterization results (Fig. 2f–i) are consistent with a previous study known for its fine sample quality (Ref. 29), specifically in that it does not possess any noticeable site disorder or impurities. Secondly, we have an additional powder neutron diffraction result that substantiates the good quality of the samples we used. The profile does not exhibit any notable impurity peaks, and the Rietveld refinement result is consistent with the refined structure suggested in Ref. 29. This result has been included in the revised manuscript (Supplementary Fig. 2 and Table 2).

Multiple scattering is also unlikely to yield the continuum scattering observed in our measurements. Typically, the dominant form of multiple scattering resulting in a signal at finite energy transfers involves at least one elastic event (either incoherent or Bragg) plus scattering from an excitation. Since the observed continuum scattering follows the magnetic form factor, we can further conclude that any such inelastic processes involved must be a magnetic excitation (i.e., magnons). Such processes cannot yield continuum scattering at energies exceeding those of one-magnon processes and thus can be excluded as a possible explanation for the observed continuum. Alternatively, continuum-like multiple scattering in the high-energy region may arise from two inelastic processes, which should have much smaller cross-sections than a single inelastic process. Contrary to this expectation, the observed continuum exhibits spectral weight comparable to the one-magnon spectrum beneath it. Therefore, it is also unlikely that the observed continuum results from multiple-scattering involving two inelastic processes. Even if our experimental data were to include such a continuum-like multiple scattering signal, it would be substantially weaker than the total spectral weight of the observed continuum.

Finally, as already noted above and shown in Fig. 4m–o of the revised manuscript, the intensity of the continuum scattering signal follows nearly the same |**Q**| dependence to that of the one-magnon bands: it forms peaks at the magnetic zone centers (|**Q**| ~ 0.8 A^{-1} and 2.0 A^{-1}) and decreases at higher |**Q**| due to a magnetic form factor. This indicates that the continuum scattering signal is an intrinsic magnetic signal from BLCTO.

Given the exclusion of disorder and multiple scattering as contributing factors, we maintain that the strong continuum scattering signal observed has to do with significant quantum fluctuations. Regardless of whether these quantum fluctuations are best described through spinons or magnons, the implication of their strong presence is unmistakably supported by the large spectral weight of the continuum signal. Thus, the dominance of strong quantum fluctuations is well justified from our observation.

Comment #2: Often the authors argue the agreement with the model is excellent (eg. page 6) when it only describes a narrow region of the S(E). It is not clear how good it is at describing S(Q).

Reply: Following the suggestions given here and Comment # 8, we have added a new figure in the Supplementary Information that presents a comprehensive comparison between the four experimental and theoretical datasets in Fig. 3a-c, in terms of both S(|**Q**|) and S(*E*). Please find this information summarized in Supplementary Fig. 5. We also note that the nice agreement in terms of *S*(|**Q**|) was already indicated by Supplementary Fig. 6a (Supplementary Fig. 8a in the new version) and Supplementary Fig. 7 (Supplementary Fig. 9 in the new version).

In addition, we would like to acknowledge one error in Fig. 3d: in the previous version, the best-fitted simulation results, plotted as a solid orange line, was in fact the result from an incorrect parameter set [J_1 = 2.62 meV, **Δ = 0.36**] due to a mistake made in our computer code. We apologize for any confusion this may have caused. The new Fig. 3d shows the result from the correct optimal parameter set [J_1 = 2.62 meV, **Δ = 0.56**], indeed demonstrating much better agreement than the previous Fig. 3d.

Comment #3: What is the relation between the J1 obtained in the paramagnetic regime (2.6meV) and the one in the ordered phase (1.55-1.7meV). Why are they different, why is the paramagnetic larger? Would other exchange interactions increase in magnitude below TN? Would they contribute to LRO? They should discuss the relation between these two different results.

Reply: The inconsistency between the J_1 values obtained at $T > T_N$ by LLD simulations and $T < T_N$ by the spin-wave theory or Schwinger boson theory is due to the inability of our "classical" LLD methodology to account for the effective "quantum" spin length ($\langle \hat{\boldsymbol{S}}^2 \rangle = S(S+1)$) at high temperature. Consequently, the LLD result underestimates the excitation energy by a factor of at most $\left(\frac{S(S+1)}{S^2}\right)^{1/2}$ (i.e., about 1.732 for $S = 1/2$). This eventually leads to an over-estimation of J_1 by the same factor as J_1 is an overall scale factor of the excitation energy. Thus, one should be cautious about the interpretation of the J_1 value obtained from the paramagnetic phase (2.6 meV): it should not be interpreted as "the magnitude of exchange interactions are changing as a function of temperature". Please note that the above explanation was already included in the main text (pages 7–8).

However, it is important to note that this limitation does not affect our estimation of other parameters based on LLD, since they were all defined and fitted in a relative scale to J_1 (e.g., Δ and J_2/J_1). Thus, this does not affect the finding of strong planar XXZ anisotropy quantified by Δ. A recent study on $KYbSe_2$ (*Nat. Phys.* **20**, 74–81 (2024)) – another promising S = 1/2 triangular lattice antiferromagnet – used a similar approach to estimate the spin Hamiltonian, and stated that the estimation of other exchange parameters based on a relative scale to J_1 remains valid, despite the technical obstacle to estimating J_1 accurately.

Comment #4: Below TN, a continuum is seen above the magnons, it can partially be explained with Schwinger boson theory (the best option in the paper) but the contribution of two(N)-magnon continuum is not considered in the model. It is however discussed in the paper (see page 8). The authors should at least mention the energy range the two-magnon continuum should occupy perhaps by adding a sign/line in fig. supplementary 10e-g.

Reply: The continuum nature of these studies stands out as one of their most intriguing aspects. We have incorporated more explanations on our Schwinger boson theory and the two(N)-magnon continuum in the revised version of the manuscript. As outlined in the manuscript, numerical investigations, including Density Matrix Renormalization Group (DMRG) studies, suggest that triangular lattice antiferromagnets with nearest neighbor exchange are on the verge of a continuous phase transition toward a spin liquid phase. Remarkably, the quantum critical point, or quantum melting point, arises when the second neighbor exchange is merely 7% of the nearest neighbor exchange. This observation implies that magnons, the collective modes of the ordered magnet, should progressively deconfine into pairs of spinons as the system approaches the quantum phase transition into the spin liquid state.

Consequently, one anticipates that magnons are more accurately described as two-spinon bound states, and the conventional two-magnon continuum evolves into a multispinon continuum. This fundamental insight motivates the utilization of the Schwinger boson approach, which effectively captures this physics, over a semi-classical approach that can only interpret the continuum as a two-magnon continuum. It is noteworthy that this scenario parallels with the behavior observed in magnetically ordered weakly coupled chains, where the continuum spectrum is aptly described by a two-spinon continuum despite the underlying low-energy collective modes being magnons, i.e., two-spinon bound states.

The omission of the two-magnon contribution in the Schwinger boson (SB) calculation is due to its appearance at higher order in the 1/N expansion (N is the number of bosonic flavors). Our current approach incorporates corrections up to order 1/N. However, we appreciate the suggestion to mention the expected energy range of the two-magnon continuum, which is now included in page 7 of the main text and Supplementary Fig. 12. Since the two-magnon process is a pair creation of two two-spinon bound states under momentum and energy conservations, its energy should be extended to 2*E*, where *E* (= 1.6 meV in Fig. 4d & 4h) is the bandwidth of two-spinon bound states.

Comment #5: Considering the Schwinger boson result on the magnitude of the magnetic exchange interactions. How can the authors re-interpret heat capacity and susceptibility data? Would they be able to describe the field dependency of the heat capacity data?

Reply: The key features in the heat capacity and magnetic susceptibility data, T_N = 3.26 K and the maximum susceptibility position T_{max} = 5 K (see Fig. 2f–i), are in agreement with the exchange parameter J_1 = 1.77 meV derived from the Schwinger boson theory. While the most direct method of validation is to conduct theoretical calculations of the susceptibility and heat capacity, this would require a separate major effort outside the scope of the present work. Indeed, accurate calculations of these physical properties in a S = 1/2 triangular lattice, characterized by strong quantum fluctuations, represent a significant challenge and would necessitate extensive efforts, such as performing quantum Monte Carlo simulations.

As an alternative approach, we have validated the consistency between the exchange parameters and the bulk properties by comparing our system, BLCTO, with a similar S=1/2 TLAF where spin Hamiltonian and heat capacity/susceptibility curves are well-characterized. The best system of such is $Ba_3CoSb_2O_9$, exhibiting T_N = 3.8 K, T_{max} = 5.7 K, J_1 = 1.7 meV, and Δ = 0.92. The similarity between these two materials in terms of their magnetic energy scale is also evident in the comparison of their excitation spectra: see Fig. R2 below. Indeed, J_1 = 1.77 meV obtained by the Schwinger boson theory aligns closely with the case of $Ba_3CoSb_2O_9$, while J_1 = 2.6 meV derived from the LLD simulations does not.

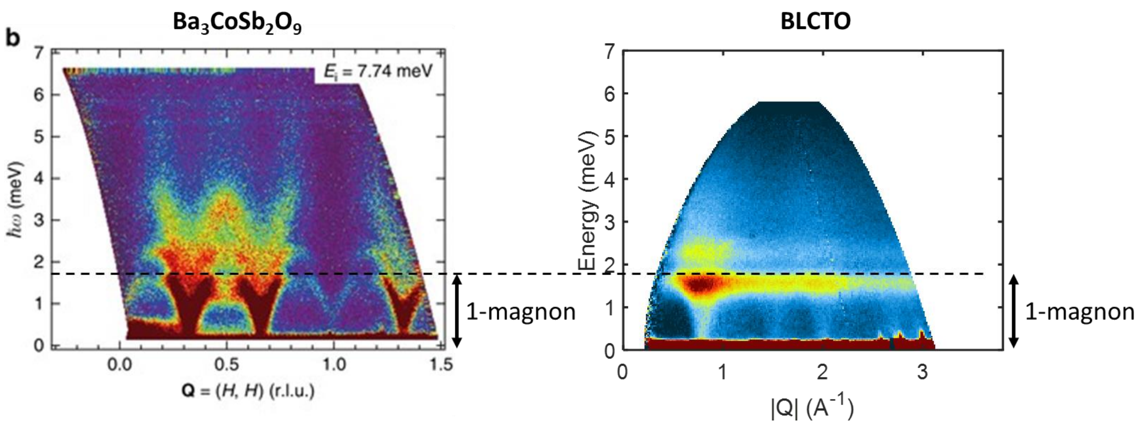

Fig. R2 Excitation spectrum of (left) single-crystal $Ba_3CoSb_2O_9$ and (right) BLCTO. The left panel was adapted from Ref. 14. These two materials share a very similar scale of excitation energies.

The field dependence of the heat capacity in our study can be qualitatively understood through the typical phase diagram of XXZ S = 1/2 TLAFs under a magnetic field. Figure R2 below shows the experimental temperature-field phase diagram of $Ba_3CoSb_2O_9$ (Phys. Rev B **92**, 014414 (2015)), which, again, has slight planar XXZ anisotropy and comparable strength of J_1. As shown in Fig. R3, the transition temperature increases noticeably under an in-plane field and decreases slightly with an out-of-plane field. This corresponds well with our observations in the heat capacity data for powder BLCTO under 9 T: one peak appears at a notably higher temperature compared to the zero-field condition, while the temperature of the other peak is marginally lower. However, the powder-averaged nature of the heat capacity curve limits a quantitative interpretation of the peak positions, as it represents a complex mixture of the two phase diagrams depicted in Fig. R3.

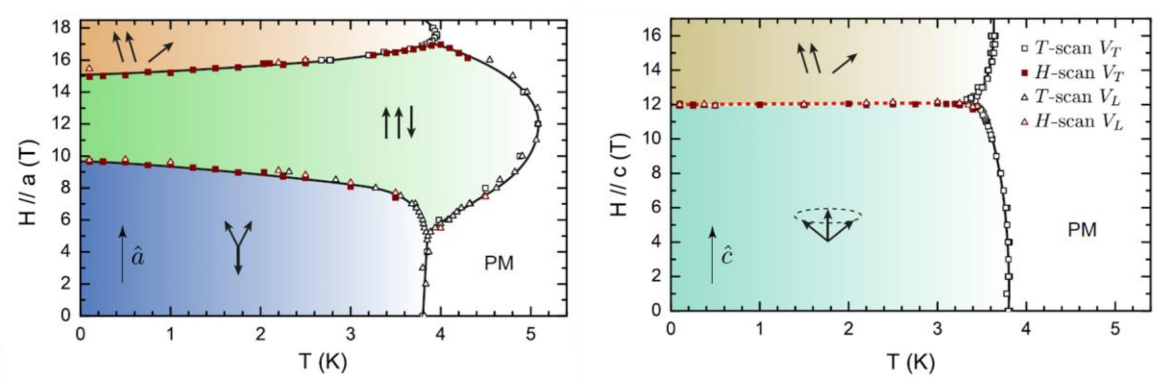

Fig. R3 Field-temperature phase diagrams of $Ba_3CoSb_2O_9$, an XXZ S=1/2 TLAF similar to BLCTO. Adapted from Phys. Rev B **92**, 014414 (2015).

Comment #6: Considering the models, can the authors interpret the two critical magnetic fields (~9 and 16T) found in Kojima's magnetization data?

Reply: This is a good point worth more explanation. As the referee may already know, 9 T and 16 T correspond to the two ends of the 1/3 magnetization plateau (a collinear up-up-down phase) region, a characteristic phase of S=1/2 triangular lattice antiferromagnets driven by quantum fluctuations. A *rough* crosscheck of J_1 can be made by comparing the experimental (9 T < H < 16 T) and *simplified* theoretical ($H_{1/3} \sim 3J_1S / g_{ab}\mu_B$, see Ref. 25) field positions of this plateau phase. A combination of J_1 = 1.77 meV and g_{ab} presented in Supplementary Notes gives $H_{1/3}$ = 9.3 T, aligning with the experimental plateau range.

However, more quantitative interpretations are not only subject to systematic errors but would also require a separate major effort that is beyond the scope of this work. From the theory perspective, planar XXZ anisotropy is known to affect both the position and the width of the plateau phase for $\mathbf{H} \perp \mathbf{c}$, which indeed requires cumbersome calculation techniques: see Ref. 25 and J. Phys.: Condens. Matter **3**, 69 (1991). Calculating the plateau with planar XXZ anisotropy and **H** // **c** is even more challenging: please find the phase diagram (= Fig. 1) calculated by DMRG in Phys. Rev. B **91**, 081104(R) (2015). Therefore, interpreting the powder-averaged M–H curve, which contains both field directions in it, is quite non-trivial.

Also, some systematic errors are expected from the simplicity of the J_1–Δ model, as described in our response to Comment # 2 from Referee 2. From the experiment's perspective, the smoothly varying *M*–*H* curve gives rise to ambiguity in determining the precise range of the plateau phase (it is not perfectly flat). Also, the *M*–*H* curve in Ref. 29 was taken at a finite temperature ($0.4T_N$), which could affect the plateau position and result in discrepancies with the theoretical estimation at $T = 0$ (see Ref. 26 of Supplementary Information).

Comment #7: Can the authors comment on what are they doing to integrate the ToF data at low wavevectors extending in energy all the way to the max. transfer energy? What are they doing for the points outside the ToF E-Q window?.

Reply: For all the *S*(E) plots shown in Figs. 3 and Fig. 4, the integration along |**Q**| (0.5 A^{-1} < |**Q**| < 2.2 A^{-1} for Fig. 3d and 0.5 A^{-1} < |**Q**| < 1.5 A^{-1} for Fig. 4i–j) was done by averaging dynamical structure factor values at the same *E* but different |**Q**|. Data points outside the ToF E-|**Q**| window are processed as "NaN" values (i.e., null data), which are automatically omitted during the averaging process and therefore do not participate in the *S*(E) plots. This was performed by using Mantid Workbench software. The simulation results corresponding to the *S*(E) plots in Figs. 3 and 4 were processed in the same way after converting the values outside the ToF coverage to "NaN".

Comment #8: The elastic line is cut off the data (see eg. fig.3), are they just not plotting the elastic line or has this been subtracted by using higher temperature data? This can drastically change fig 3d below 1meV, the most crucial comparison cut. Why not do the S(E) comparison using all three available data sets (Ei=6,10, 1.5meV -last from supplementary?)

Reply: The screened elastic region is not relevant to higher-temperature data subtraction, which we have not conducted with any dataset shown in the main text. Fig. 3 simply does not plot the region around 0 meV. More generally, the region not plotted in Fig. 3, except for the area outside the energy-momentum coverage of time-of-flight powder neutron spectroscopy, denotes the masked region for the χ^2 analysis (Fig. 3e) due to the presence of extrinsic signals not relevant to the sample. This involves spurious signals, a sizable quasi-elastic background, and noises in the vicinity of the detector coverage. We have added a relevant explanation in Methods.

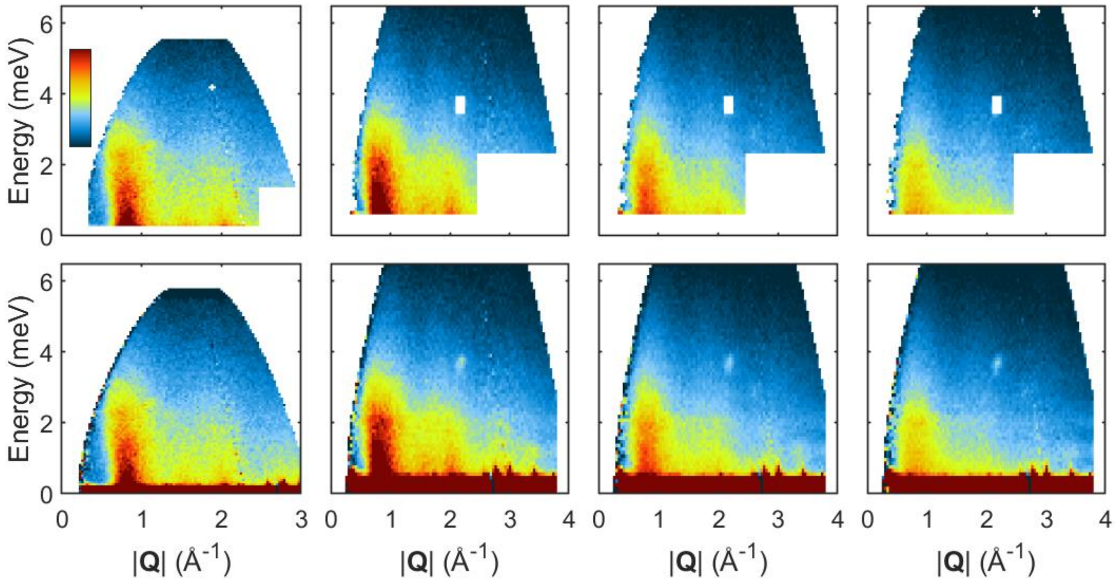

Fig. R4 INS data with bad regions dominated by extrinsic signals being excluded (same as Fig. 3a in the main text). (Bottom) The same INS data without masking the excluded regions.

Fig. R4 above shows the raw data without any masking (bottom row). As the referee can see, the energy range close to 0 meV (which the referee called "the elastic line") possesses a strong quasi-elastic background signal, thereby not providing any meaningful information and should be excluded from the goodness-of-fit analysis. However, the strong quasi-elastic background in the masked region does not affect the data plot in Fig. 3d. The lowest energy value plotted in Fig. 3d (= 0.38 meV) is still higher than the masked energy range in the leftmost panel of Fig. 3a ($0 < E < 0.28$ meV) so that the comparison cut of Fig. 3d is apart from the masked energy region due to the background signal.

Finally, we have extended our comparison of S(E) to other datasets from different incident energies and temperatures. Note that the measurement with E_i = 1.5 meV was only done at $T = 0.5T_N$, and Fig. 3a shows all datasets we collected in the paramagnetic phase. This full comparison has been included in Supplementary Information (Supplementary Fig. 5), together with the S(|**Q**|) analysis suggested in Comment #2.

Comment #9: The main paper should show the q-dependency of the intensity for the data below TN for all the main features (arrows in fig 4 e), and show how these compare to the three models. In general, q-dependencies are missing in the whole data analysis.

Reply: Following the referee's suggestion, the |**Q**|-dependence plots of the three main features are added in Fig. 4 (Fig. 4m–o). Here, LSWT results are not plotted since they would not convey any useful information (they have zero intensity above the one-magnon energies).

Comment #10: Can the authors include in the discussion an explanation (for the experimentalist) of the origin of the continuum in the Schwinger boson theory?

Reply: We thank the referee for the constructive suggestion. In the Schwinger Boson theory, magnons manifest as two-spinon bound states, while the continuum arises from excitations involving two or more spinons. Although two-magnon excitations anticipated to emerge from order $1/N^2$ not considered in our calculation (N is the number of bosonic flavors), their spectral weight is expected to be smaller than the multi-spinon continuum. More explanations can be found in our response to Comment #4 or on pages 2 and 7 of the main text.

Comment #11: Finally, can the authors comment on the advantages/disadvantages of studying an anisotropic system using only powder samples?

Reply: We appreciate the opportunity to discuss the advantages and disadvantages of using powder samples, which is one of the challenges that we have been trying to address more broadly. Our approach based on powder samples primarily hinges on the ability to rapidly examine intriguing spin systems with considerably less experimental effort than single-crystal measurements. Notably, the synthesis of many quantum materials, not only for magnetic materials with strong anisotropy, is still predominantly achievable only in powder form, and it takes significant effort to prepare a single-crystalline form large enough to conduct inelastic neutron scattering. Thus, the capability of utilizing polycrystals to study anisotropic spin models significantly broadens the scope of materials that can be explored experimentally. This eventually allows the rapid dissemination of information obtained through inelastic neutron scattering to the scientific community.

The aforementioned capability, however, is usually challenged by loss of information by powder averaging of momentum transfer, which is the biggest disadvantage of using a powder sample. In this work, this loss has been compensated by analyzing the energy-resolved diffuse scattering. While analyzing energy-integrated diffuse scattering provides significant insights as shown in Fig. S9, adding the energy axis to the analysis makes the differences between the spin dynamics of various models more qualitatively apparent and reduces the associated uncertainties.

Nevertheless, we acknowledge the necessity of carefully interpreting the powder measurement result and validating it, if possible, with single-crystal measurements. Less information compared to single-crystal measurements leads to greater parameter uncertainty, which becomes worse as the number of parameters increases. The analysis result on BLCTO with the generalized spin Hamiltonian (J_1–J_2–Δ–$J_{\pm\pm}$–$J_{z\pm}$ model) is a good example: it has yielded a large uncertainty of $J_{z\pm}$ roughly ranging from -0.15J_1 to 0.15J_1 (see Supplementary Notes). Thus, obtaining four-dimensional $S(\mathbf{Q},\omega)$ by single-crystal measurements is required to improve this uncertainty, as pointed out in the Supplementary Notes.

Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
The comments raised by the present reviewer have been properly reflected in the revised manuscript. In particular, new figures in the supplementary information (Supplementary Figs. 10, 11, and 12) systematically demonstrate the similarities and differences among the LSWT, NLSWT, and Schwinger boson. Discussions about a quantum melting point are also included properly in the main text. Therefore, I recommend the publication of the present manuscript in Nature Communications.

Reviewer #2:
Remarks to the Author:
The authors responded reasonably to all the referees' questions and comments and corrected the manuscript. Hence, I recommend publication.

Reviewer #3:
Remarks to the Author:
The new version of the manuscript is very comprehensive. Thank you for the detailed answer to all referees' comments. I recommend the editors to publish the manuscript.

Second Report of Referee #1

General Comment: The comments raised by the present reviewer have been properly reflected in the revised manuscript. In particular, new figures in the supplementary information (Supplementary Figs. 10, 11, and 12) systematically demonstrate the similarities and differences among the LSWT, NLSWT, and Schwinger boson. Discussions about a quantum melting point are also included properly in the main text. Therefore, I recommend the publication of the present manuscript in Nature Communications.

Reply: We thank for their thorough review and recommendation of our work for publication.

Second Report of Referee #2

General Comment: The authors responded reasonably to all the referees' questions and comments and corrected the manuscript. Hence, I recommend publication.

Reply: We appreciate the referee’s recommendation of our work for publication.

Second Report of Referee #3

General Comment: The new version of the manuscript is very comprehensive. Thank you for the detailed answer to all referees' comments. I recommend the editors to publish the manuscript.

Reply: We again thank the referee for their time in reviewing the manuscript and for their recommendation for publication.

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