# nature portfolio

## Peer Review File

REVIEWER COMMENTS


Reviewer #1 (Remarks to the Author):


Dear Editor and Authors,


Using parameters obtained from DFT calculations as well as MC and PFFRG theoretical calculations, this paper argues that K2Ni2(SO4)3 is very likely close to a 3D spin liquid-like pocket in the phase diagrams spanned by relevant exchange interactions. Its magnetic properties could reflect this proximity. This conclusion is supported by the fact that above the ordering temperature, the instantaneous correlations (estimated from an energy integration of INS data) are well reproduced by the calculations.


This is a very nice article, and the results deserve to be published in some form, so that they can be known by the condensed matter physics community. However, to my opinion, the writing is biased towards demonstrating a fashionable result, in the present case, the existence of a proximate 3D spin liquid state. Indeed, as far as I understand, experiments show that K2NI2(SO4)3 does order, and certainly has a spectrum consisting in spin waves. Furthermore, the conclusion of the paper is based on a comparison betwen calculations and integration of INS data taken at 2K, i.e. in the paramagnetic regime of K2Ni2(SO4)3. I feel this reasoning is misleading. Indeed, what counts above all is the energy structure, the INS technique being first and foremost a spectroscopic measurement. The Q structure, in reciprocal space, of course provides very interesting information, but it is above all sensitive to interference effects linked to atomic positions. It is therefore not in itself a validation of energy parameters, such as those involved in the Hamiltonian of a given system.


Given the context, I can't imagine that the authors do not have low temperature data. Why don't the authors compare this information with calculations? With such a complex theoretical arsenal at hand (MC, PFFRG), calculating spin waves should be simple? Phys. Rev. Lett. 127, 157204 also reports powder INS data. What about the proposed parameters? Do they match experiment?

A detailed comparison of calculations and spectra taken below Tc is definitely lacking. In these conditions, how to be confident that DFT calculations provide exchange parameters compatible with experiment? It could be that K2NI2(SO4)3 "simply" has a spectrum of classical magnons superimposed on a spectrum of longtudinal fluctuations, i.e. a physics still far from the intricated states expected in QSL. This being said, I have no doubt that the "QSL" region investigated from a theoretical point of view is correct. I thus strongly encourage the authors to show low temperature data and compare them with MC calculations of the spin dynamics.

In addition, even if the writing is excellent, as is the quality of the theoretical and experimental results, I still have comments and questions :

1) Please detail the nature of the ordered state arising below 1.1 K. I'm also quite curious about the magnetic entropy release. Could the authors comment on the origin of the poor spin freezing fraction? Line 160 : what do the authors mean by "q-vectors"? propagation vectors? A multi-k structure?

2) I note that Ce2Hf2O7 and Ce2Sn2O7 could be also 3D spin liquid candidates.

3) According to literature, application of B = 4T magnetic field leads to a fully dynamical state down to the lowest temperatures. What is the situation from a theoretical point of view ? Would the application of the field drive the system into the "liquid region" reported here?

4) What are J1 and J2?

5) In the limit J3, J5 → 0, the calculations predict a semi-classical AFM state. The limit J4 → 0 describes two independent trillium lattices, leading to a variant of the 120 degrees order. If surrounded by "classical" phases, how do the authors can conclude about the proximity to an "island of liquidity" ? A this point, the narrative is not clear to me.

6) Line 180 : "INS is featureless". This is not that surprising given the fact that the data have been taken above Tc, so we just expect quasi-elastic scattering as usually observed close to a transition. What about below Tc?

7) Line 187 -> 205. What is the purpose of this paragraph?

8) Line 274 : "K2Ni2(SO4)3 lies very close to a so-far unexplored region of genuine spin-liquid behaviour, centred around a particular point defined by J3 = 0, J4 = J5." Furthermore:

Line 294 : "For S = 1, PFFRG indicates a very narrow range around J3 = 0";

Line 328 : "cMC calculations show no ordering tendencies in a large J5/J4 region around the tetratrillium lattice when J3 = 0."

Line 333 : "On the other hand, even the smallest non zero value of J3 results in a finite critical temperature".

Given these sentences, I understand that the liquid region is very narrow. But DFT points to J5=0.5 J4 and J3= 0.1 J4, which is not "so" close. How can the reader then appreciate? How to conclude that the spin liquid region is relevant in understanding K2Ni2(SO4)3?

9) Line 297 : what is the purpose of considering S=1/2 (Ni having a spin S=1) ?

10) Line 307 : "many of the features observed in the spin structure factors can be traced back to the tetra-trillium lattice limit." Many? which ones?

11) Line 358 : e(T) is not defined

14) Line 335 : "This fine line of pure spin-liquid behavior can be understood from the Hamiltonian containing only J4 and J5, which can be written as a disjoint sum of squared

total spin in each tetrahedron (just as for the pyrochlore lattice". How is this remark related to the experimental entropy release?

Reviewer #2 (Remarks to the Author):

The authors employ a combination of inelastic neutron scattering, pseudo-fermion functional renormalization group, and classical Monte Carlo calculations to explore the complex properties of $K_2Ni_2(SeO_4)_3$, a compound composed of two 3D nets of corner-sharing triangles. The authors' efforts in integrating numerical calculations and experimental results are appreciated. However, the analysis and strategy of studying a frustrated magnet is not convincing. Therefore, I do not recommend Nature Communications publishing this manuscript.

Most importantly, I have reservations about the strategy employed to determine the exchange coupling constants. Given the highly dynamic ground state mentioned in the manuscript, it may be advisable to investigate the system by driving it into an ordered state with a magnetic field and subsequently fitting the spin wave excitations. This approach, as demonstrated in Reference [1], can provide more precise determinations of the exchange parameters.

Furthermore, the manuscript appears to introduce its own interpretation of "quantum spin liquids" (QSL) and "dynamics." In a journal like Nature Communications, it is vital to align with established scientific terminology and understanding [2]. I suggest addressing the following concerns:

1. Clarify whether the ground state is dynamic or not by examining the static moment at the base temperature. If there is no clear evidence indicating that the ordered moment is negligible, it is natural to assume that the ordered moment represents the full moment, suggesting that the ground state is not dynamic.

2. Reevaluate the phase diagram to ascertain if the compound indeed hosts a QSL state. It is possible that the field-polarized state is being misinterpreted as a QSL state. It is worth considering the transition to a field-polarized state, as proposed in Reference [3].

In summary, the manuscript is intriguing, especially in its observations that excitation in the ordered state does not exhibit typical spin wave behavior, indicating the presence of strong frustrations. To enhance the study's quality, I recommend the following actions:

1. Delve into the crystal field of Ni, which, while complex, is essential for a comprehensive understanding of the system. This aspect is currently missing from the manuscript.

2. Refine the diffraction data to determine precise ordered moments in both zero-field and high-field states.

3. Investigate the exchange interactions by fitting the spin-wave excitations under magnetic fields.

4. Employ numerical methods to reproduce the zero-field excitations based on the interactions determined.

For the current manuscript, I think it could be published in a more specialized journal after changing QSL-related arguments. I hope my comments and recommendations prove helpful in further refining the manuscript.

Reference:

[1] Ross, Kate A., et al. "Quantum excitations in quantum spin ice." Physical Review X 1.2 (2011): 021002.

[2] Broholm, C., et al. "Quantum spin liquids." Science 367.6475 (2020): eaay0668.

[3] Li, Han, et al. "Identification of magnetic interactions and high-field quantum spin liquid in α-RuCl3." Nature Communications 12.1 (2021): 4007.

Reviewer #3 (Remarks to the Author):

Dear Editor,

I believe that the manuscript "Dynamics of K2Ni2(SO4)3 governed by proximity to a 3D spin liquid model," by Gonzalez et al., is suitable for publication in Nature Communications after revisions. I believe the revisions I recommend require no further experiments or numerical simulations, but they will require important changes to the text.

In this paper, the authors consider the material K2Ni2(SO4)3, which has previously been found to show high magnetic entropy at low temperatures and spin-liquid like behavior under small applied fields. They authors find an approximate Heisenberg model for the material, with coupling strengths determined using density functional theory (DFT). They then study this model and some approximations to it (leaving out certain small spin-spin interactions), using both classical Monte Carlo simulations at finite temperature and quantum pseudo-fermion functional renormalization group (PFFRG) calculations at 0 temperature. They compare the results, for example for the spin structure factor, with experimental inelastic neutron scattering data. The key finding is that some spin models, which are close in coupling strengths to the one describing the real material, apparently exhibit spin liquid ground states, giving a possible explanation for the experimentally observed behavior of K2Ni2(SO4)3.

The agreement between numerical simulations and experimental data is remarkable, especially as shown in Fig. 1c and Fig. 3 a-f. While I am not an expert on either of the specific theoretical methods employed by the authors, I believe the numerical results presented give strong evidence for the quality of the calculations and the applicability of the methods to this problem. The authors also provide reasonable discussions of both methods in the Supplementary Information.

Overall, the authors address an important question that will be of interest to the condensed matter physics community, namely the origin of spin liquid-like behavior in 3D materials. They make a convincing case that the behavior is due to proximity in parameter-space to a model that seems to have a spin liquid phase, as identified by PFFRG calculations. They also provide a demonstration of the interesting correspondence between quantum and classical fluctuations.

All of that being said, the authors need to make some important revisions to improve the clarity of the paper, which I describe below. The points are roughly in order from most to least important.

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1. More explanation is needed regarding the choice of T for the cMC calculations and Lambda for the PFFRG calculations, and of why the cMC calculations are needed at all.

My impression is that the logic goes like this: (1) the goal is to simulate the T=0 (or T=2K, which I gather is in the ground state phase, though this should be clarified) behavior of the model; (2) in PFFRG, follow the renormalization group flow from large Lambda until hitting an instability that indicates ordering; (3) do quantum calculations at that Lambda to get correlation functions; (4) select a T for cMC that gives the most similar possible behavior to the PFFRG calculation. Sometimes such a T can be found (Fig. 3) and sometimes not (Fig. 4b, top half).

Some questions that come up:

- Does the cMC temperature have some physical meaning, apart from the size of (quantum) fluctuations? It is apparently an order of magnitude above physical temperature, e.g. for Fig. 3 the physical temperature is seemingly below a transition at 0.048 J4, while the cMC temperature is 0.35 J4.

- What is the purpose of the cMC calculation if the PFFRG is needed anyway in order to find the correct temperature for cMC? Is there a way to predict T given Lambda?

- I suspect the answer to the previous question is related to Fig. 4c, which shows data from sweeping the cMC temperature from 2 J4 to 0.001 J4. This figure shows that the behavior of the model is closely related to the behavior of spins on individual tetrahedra, but does the temperature actually have any physical meaning?

It appears to me that the main purpose of the cMC calculations is to demonstrate the quantum-to-classical correspondence, ie that the fluctuations in the quantum ground state either are or are not classical in nature. In other words, for a given set of couplings Ji,

the fluctuations in the ground state either can be reproduced by cMC, in which case the state is "less quantum," or cannot be reproduced, in which case the state is "more quantum." In other words, if cMC cannot reproduce the PFFRG results, as in Fig. 4b, is that an indication of a spin liquid?

The authors must explain why the cMC results are useful (as opposed to just using PFFRG) and whether the cMC temperature has any physical meaning.

As a less important point, I think for readers (including me) who are less familiar with PFFRG, some additional discussion of why the calculations should be done at precisely the Lambda at which the RG flow breaks down would be helpful. (This could be in the Supplementary Information, augmenting the current discussion there.)

2. Computation of S(q): the discussion around the integration in (1) needs to be improved. In particular, how are the limits omega1 and omega2 chosen? Also, these parameters currently appear in (1) but are not discussed at all until the end of the page, which I found confusing. They should be discussed, and the choice of values explained, immediately after the equation. This is especially important since the choice of integration limits is cited at the end of page 4 as a possible reason for discrepancies between the curves in Fig. 3 g-i.

3. I have some confusions about Fig. 2, so the authors could be clearer about the following:

- Clarify precisely what quantity is actually indicated by the color. Some measure of particle flux/intensity?

- Do white regions mean that the maximum of the scale was exceeded? Then why are there also white spots in the upper part of 2c?

- Also, my understanding of the phrase "streaks of intensity without any apparent q-dependence" is that it refers to regions in 2a-c where the intensity (ie color) is constant on some horizontal line. That only appears to be true at the bottom of the figure, but if white is off-scale, then how do we conclude that the intensity is constant?

The authors should clarify the answers to these questions, or, if I have fundamentally misunderstood something, they should write a clearer explanation of the data in Fig 2 to prevent this misunderstanding for future readers.

4. I am confused by the statement "Most importantly, PFFRG contains the correct large-N (spin flavour) and large-S (spin size) limits, making it ideal to study quantum systems." in the Supplementary Information. The authors should add some short additional clarification.

5. Minor point: inconsistent use of Q and q

6. Minor point: Fig 4c should have T/J4 as the x label

7. Minor point: why is cMC T=0.001 selected for 4b upper left? Just because it's the smallest value?

8. Minor point: many of the figures, although clear in color, are confusing in black and white. The authors should improve the appearance of the figures so that they are clear when the paper has been printed in grayscale. This is specifically a problem with: Fig 1a, Fig1b, Fig 3g-i, the stars in Fig 4a.

We thank the reviewers for their critical reading of the manuscript and comments. Below, we provide a detailed response to their concerns/questions and provide a list of the corresponding amendments made to the manuscript.

**REPLY TO REFEREES**

The text written by Referees is coloured blue, the text from the manuscript is coloured red while the changes and additions to the manuscript are coloured green.

**Reviewer 1**

(No 1.1) Using parameters obtained from DFT calculations as well as MC and PFFRG theoretical calculations, this paper argues that K2Ni2(SO4)3 is very likely close to a 3D spin liquid-like pocket in the phase diagrams spanned by relevant exchange interactions. Its magnetic properties could reflect this proximity. This conclusion is supported by the fact that above the ordering temperature, the instantaneous correlations (estimated from an energy integration of INS data) are well reproduced by the calculations.

We thank the Referee for a succinct description of the main message of our manuscript.

(No 1.2) This is a very nice article, and the results deserve to be published in some form, so that they can be known by the condensed matter physics community. However, to my opinion, the writing is biased towards demonstrating a fashionable result, in the present case, the existence of a proximate 3D spin liquid state.

We are grateful that the Referee is appreciative of our article and considers the results to be of value to the wider condensed matter physics community. However, we beg to disagree with the negative connotation ascribed to ours being a "fashionable result" and the assessment that writing is biased towards demonstrating the existence of a proximate 3D spin liquid state. Firstly, it is perfectly natural for scientific studies to focus on topics which are in vogue, and a journal such as Nature Communications is an excellent platform to disseminate results which are likely to attract much attention. Secondly, while arguing for a "proximate" 3D spin liquid state, we have been cautious in our language and used appropriate qualifiers. We would like to take this opportunity to clarify the notion that the phrase "proximate spin liquid" conveys and the context in which it has been used by us.

The Referee later argues that our claims about "a proximate 3D spin liquid state" are not correct, mostly basing his/her reasoning on the fact that $K_2Ni_2(SO_4)_3$ orders, and therefore the dynamic response that we report *above* its ordering temperature has nothing to do with spin liquid behaviour. We would like to clarify that whether a system should be considered to be in a spin liquid regime or not, is not determined by its ordering temperature. Rather, it is governed by the characteristic energy scale of the system, set by the values of exchange interactions which, in most cases, is well represented by its Curie-Weiss temperature $\theta_{CW}$. For $T < \theta_{CW}$, the correlations between spins start to develop and in the absence of geometric frustration, lead to the onset of long-range order at $T_N \sim \theta_{CW}$. However, in the presence of frustration, the ordering temperature gets suppressed, and it is precisely in the temperature window $T_N < T < \theta_{CW}$, that a *cooperative paramagnetic* phase –a highly correlated soup of spins featuring a nontrivial pattern of long-range spin correlations– is stabilized. This is a well-known feature ubiquitous to many frustrated Heisenberg magnetic models and materials, such as the celebrated kagome lattice antiferromagnet where this cooperative paramagnetic/spin liquid phase is associated with pinch points and bowties in the static structure factor. It is this very "cooperative paramagnetic" regime that is referred to as a spin-liquid. This terminology has been widely used, please see for example Nature Physics **3**, 487 (2007) "Order-by-disorder and spiral spin-liquid in frustrated diamond-lattice antiferromagnets", where, by now the well-established terminology of "spiral spin liquid" was introduced referring to the paramagnetic phase above the ordering temperature in the $J_1$-$J_2$ diamond lattice Heisenberg model. The existence of such a spin liquid phase was also reported in experiments, see Nature Physics **13**, 157 (2017).

Here, it might be appropriate to include a sentence from 'Quantum spin liquids: a review', written by Lucile Savary and Leon Balents (Ref. [2] in our manuscript):

> *Over time, the idea of a QSL has been conflated with the more conventional notion of a liquid in the classical sense: a spin liquid is a system of spins which are highly correlated with one another due to their mutual interactions, yet do not order even at very low or zero temperature.*

In the case of $K_2Ni_2(SO_4)_3$, $\theta_{CW} \approx -20\,K$, thus our experiment performed at $2\,K$ is well within the correlated/spin-liquid region. A similar example of a 'proximate' spin-liquid was recently published in Nature Physics (Scheie, A.O. et al., Proximate spin liquid and fractionalization in the triangular antiferromagnet KYbSe2. Nat. Phys. **20**, 74 (2024)) so our claims of a spin-liquid regime at $2\,K$ in $K_2Ni_2(SO_4)_3$ are very well justified. Hence, we would like to respectfully disagree with the Referee's assessment, that our writing is biased towards arguing for 'a fashionable result'. On the contrary, it is rather a truthful interpretation of the obtained results, interpreted and written within a framework and language which is by now well accepted in the community.

Similarly, if one asks whether a given material is a solid, a liquid, or a gas, it depends on which conditions (temperature, pressure, composition,...) it is observed. The fact that $H_2O$ is solid under some conditions does not mean it cannot be a liquid at some other. Most magnetic materials are not frustrated and therefore their 'liquid' region (with correlations but without long-range order) is either very narrow or practically non-existent, they go from a 'gas' phase (a paramagnet) to a 'solid' phase (a long-range ordered state). Geometrically frustrated systems are in focus because they allow an extended temperature region where 'liquid' properties can be investigated. Showing that a material does not order down to $T = 0$ is a bit philosophical since practically it is not possible to reach $T = 0$ and establishing liquid properties at (say) 50mK does not say anything about the eventual long-range order at even lower temperatures. Therefore, investigations at elevated temperatures are perfectly aligned with a widely accepted notion of spin-liquid behaviour.

To ameliorate the understanding for a wider audience of readers of Nature Communications, we have added the following text at the end of the paragraph starting at line 61 and ending at line 80:

> Although often it is found that QSL candidate materials order at some finite temperature $T_N$, their dynamic response is strongly influenced by the proximity to a quantum critical point where the order completely disappears. The quantum critical regime, emanating from the quantum critical point, leaves the characteristic fingerprint in the dynamic response even at finite temperatures, allowing to test the theoretical predictions of the particular QSL in real materials [19, 20].
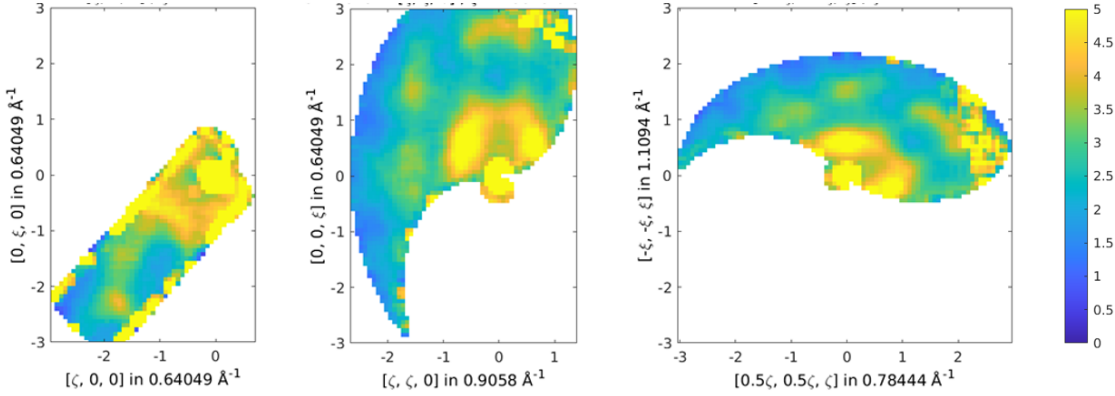
(No 1.3) Indeed, as far as I understand, experiments show that K2NI2(SO4)3 does order, and certainly has a spectrum consisting in spin waves.

First, let us reiterate that the case where the system orders at some temperature $T_N$ much smaller than the Curie temperature $\theta_C$, and develops spin-wave dispersions below $T_N$, does not in any way invalidate the existence of spin correlations in the region $T_N < T < \theta_C$. If the system is close (in parameter space) to a fully liquid behaviour at $T = 0$ (regions highlighted in Figure 4), then the characteristic spin correlations will persist in a wide region in the parameter space.

Typically, moving away from the region of liquid behaviour at $T = 0$ leads to the appearance of long-range order at $T_N$ and spin-waves can (in principle) be detected. In those cases, it is often imperative to go *above* $T_N$ to observe spin-spin correlations that characterize the spin-liquid in question. If one measures it below $T_N$, then a significant fraction of the scattering spectral weight might be found in spin waves and the analysis and the eventual comparison with theory that tries to establish the features of the spin-liquid will be severely hampered. In that context, the Referee's statement ... K2NI2(SO4)3 does order, and certainly has a spectrum consisting in spin waves is in no way in collision with our analysis of spin-liquid behaviour above $T_N$.

Going back to the actual behaviour of $K_2Ni_2(SO_4)_3$, we did perform some neutron scattering at very low temperatures (nominally 50mK), to try to see those spin waves, and, surprisingly, we saw

the same diffuse type of correlations that we present in Figure 3 of our manuscript:



This confirms that the 'ordered' phase of K2Ni2(SO4)3 is a very peculiar, unconventional state, where one should be careful with relying too much on assumptions that have been developed in the context of more 'classical' systems. It does, however, go along the same lines with results of only $\sim 1\%$ of the total entropy being released at $T_N$, and that $\mu$SR experiments indicate a fully dynamic system (no wiggles). This opens up an interesting possibility that the putative long-range magnetic order is (barely) static on the time-scale of neutrons ($10^{-15}$ s) while being dynamic on the time-scale of muons ($10^{-6}$ s). At the moment it is not clear how 'order' and dynamics exactly coexist in this material and the analysis of the above results will be a part of a future study. In the current manuscript, we are focusing solely on establishing the spin-liquid properties of $K_2Ni_2(SO_4)_3$ and how this is related to the proximity of a pure spin-liquid region whose properties we tried to disseminate in Figure 4.

(No 1.4) Furthermore, the conclusion of the paper is based on a comparison between calculations and integration of INS data taken at 2K, i.e. in the paramagnetic regime of K2Ni2(SO4)3. I feel this reasoning is misleading. Indeed, what counts above all is the energy structure, the INS technique being first and foremost a spectroscopic measurement. The Q structure, in reciprocal space, of course provides very interesting information, but it is above all sensitive to interference effects linked to atomic positions. It is therefore not in itself a validation of energy parameters, such as those involved in the Hamiltonian of a given system.

The emphasis of the Referee that 2 K is 'in the paramagnetic regime' and that 'this reasoning is misleading' has been addressed above [point (No 1.2)]. We reiterate that at 2 K, the system is not a 'boring' conventional thermal paramagnet, since the spins are strongly correlated in a nontrivial manner as the temperature is significantly below the Curie-Weiss energy scale of the system. It is common and justified to call these phases as spin-liquids, sometimes also correlated/cooperative paramagnets (following Villain, 1979), but it needs to be emphasized that they are not paramagnets in a classical sense, i.e., trivial random disordered states above Néel/Curie temperature. This has been demonstrated already in the first paper (PRL **127**, 157204 (2021), Ref. [19] in this manuscript) where a Brillouin function fails to describe $M$ vs $H$ data even at 20 K.

We agree that a thorough comparison of the complete energy structure is more appropriate than a comparison of the Q structure since the latter is an integrated quantity of the former. However, for 3D frustrated magnets, more so with quantum $S = 1$ spins and large unit cells, a computation of the dynamical structure factor remains out of reach of all current theoretical approaches, such that one has to settle for the Q maps. Nonetheless, the validation of the energy (Hamiltonian) parameters does not depend solely on these comparisons. Indeed, it is well-established that an estimation of the Hamiltonian couplings based on a fit of the spin structure factor S(Q,E) does not yield a unique set, and there generally exists a sizeable window in parameter space which provides equally good fits. We thus adopt a different route based on ab-initio approach which has widely proven itself to be reliable in uniquely determining Hamiltonian parameters, being concurrently or subsequently validated by experiment [see Nat. Commun. 11, 2348 (2020), Phys. Rev. B 101, 220408(R) (2020), npj Quantum Mater. 4, 63 (2019), Phys. Rev. B 98, 064427 (2018)]. Firstly,

these parameters are obtained via DFT energy mappings, a technique that only depends on the atomic positions and the Curie-Weiss temperature. Secondly, we show that cMC only finds the correct Q-ordering for this set of parameters and that even small deviations in couplings lead to states with larger magnetic unit cells, and thus very different signatures in Q-space. Subsequently, we find an excellent agreement of the Q structure between experiments and theoretical calculations, which is also found to be sensitive to changes in the energy (Hamiltonian) parameters (as shown in the Supplemental Information).

(No 1.5) Given the context, I can't imagine that the authors do not have low temperature data. Why don't the authors compare this information with calculations? With such a complex theoretical arsenal at hand (MC, PFFRG), calculating spin waves should be simple? Phys. Rev. Lett. 127, 157204 also reports powder INS data. What about the proposed parameters? Do they match experiment?

As elaborated above, the low-temperature data do not show any trace of spin waves, therefore, we cannot compare it to calculations. Furthermore, the Referee's suggestion that 'calculating spin waves should be simple', is true for simple magnetic structures. However, in the current context where there exist a multitude of ordering vectors and a tripling of the unit cell (each magnetic unit cell has $3 \times 3 \times 3 \times 8 = 216$ spins), the comparison is likely to be far more complicated. More importantly, by construction, spin wave theory is applicable for small deviations from a classical order, however, here the ordered moment is tiny such that the deviation from classical order is huge, rendering it outside the scope of a reliable application of spin wave theory.

The Referee also refers to the published powder INS data in Phys. Rev. Lett. **127**, 157204 (2021) and the proposed parameters therein. We would like to emphasize that the parameters proposed in Phys. Rev. Lett. **127**, 157204 (2021) are very close to the parameters proposed in this manuscript, they differ slightly only because the crystal structure used for the calculations in Phys. Rev. Lett. **127**, 157204 (2021) was obtained at room temperature, while for the current manuscript, we used the structure at $100\,\mathrm{K}$, thus being closer to the relevant temperature range. They were determined using the same approach (by the very same coauthors) and the reason why we believe the new set is closer to describing the system is because the old set leads to a quadrupling of the unit cell below the ordering temperature, while the new set leads to tripling, with the latter being in agreement with experimental observations.

(No 1.6) A detailed comparison of calculations and spectra taken below Tc is definitely lacking. In these conditions, how to be confident that DFT calculations provide exchange parameters compatible with experiment? It could be that K2NI2(SO4)3 "simply" has a spectrum of classical magnons superimposed on a spectrum of longtudinal fluctuations, i.e. a physics still far from the intricated states expected in QSL. This being said, I have no doubt that the "QSL" region investigated from a theoretical point of view is correct. I thus strongly encourage the authors to show low temperature data and compare them with MC calculations of the spin dynamics.

The questions raised in the first part of this paragraph have been addressed in points (No 1.4) and (No 1.5).

What we find perplexing is the statement "I have no doubt that the "QSL" region investigated from a theoretical point of view is correct". As Figure 3 in our manuscript demonstrates, there is a remarkable agreement between experiment and theory. With all the criticisms raised by the Referee related to (i) the system being a conventional paramagnet (which it is not as we have explained above), (ii) raising doubts about whether INS is an appropriate technique to address QQ-maps (which it is designed to capture), and (iii) expressing concerns on the accuracy of DFT-obtained parameters, the Referee never provided a reasoning as to how could such a perfect qualitative and quantitative 'coincidence' of different theoretical approaches with experiment appear. If the system was indeed truly a paramagnet, there would be no structure, let alone a well-defined chirality observed in spin-spin correlations, obtained in INS. Also, since the ordered moment is vanishingly small, the distinction between longitudinal and transverse fluctuations does not matter, and basing oneself on a picture of classical magnons which have well-defined dispersions dressed by longitudinal fluctuations can hardly account for the observed INS features. Similarly, QQ maps have been widely used in numerous articles investigating spin liquids (enlisted below).

- Sibille, R., Lhotel, E., Ciomaga Hatnean, M. *et al.* Coulomb spin liquid in anion-disordered pyrochlore $Tb_2Hf_2O_7$. Nat. Commun. 8, 892 (2017).

- Scheie, A.O., Ghioldi, E.A., Xing, J. *et al.* Proximate spin liquid and fractionalization in the triangular antiferromagnet $KYbSe_2$. Nat. Phys. 20, 74 (2024).

- S. Gao, M. A. McGuire, Y. Liu, D. L. Abernathy, C. de la Cruz, M. Frontzek, M. B. Stone, and A. D. Christianson, Spiral Spin Liquid on a Honeycomb Lattice, Phys. Rev. Lett. 128, 227201 (2022).

- T. Fennell, M. Kenzelmann, B. Roessli, M. K. Haas, and R. J. Cava, Power-Law Spin Correlations in the Pyrochlore Antiferromagnet $Tb_2Ti_2O_7$, Phys. Rev. Lett. 109, 017201 (2012).

- Sibille, R., Gauthier, N., Yan, H. *et al.* Experimental signatures of emergent quantum electrodynamics in $Pr_2Hf_2O_7$. Nat. Phys. 14, 711 (2018).

- Arnab Banerjee *et al.*, Neutron scattering in the proximate quantum spin liquid $\alpha$-$RuCl_3$. Science 356, 1055 (2017).

Finally, as we have already demonstrated in the Supplementary Material, even the slightest tweak to the DFT Hamiltonian parameters proves sufficient to visibly modify the QQ structure, and destroy the agreement. Thus, if any of these aspects were questionable such a degree of agreement would not be possible. Therefore, it is only when all three aspects are 'true', that we can reach the remarkable agreement between experiment and theory demonstrated in this paper.

(No 1.7) In addition, even if the writing is excellent, as is the quality of the theoretical and experimental results, I still have comments and questions :

1) Please detail the nature of the ordered state arising below 1.1 K. I'm also quite curious about the magnetic entropy release. Could the authors comment on the origin of the poor spin freezing fraction? Line 160 : what do the authors mean by "q-vectors"? propagation vectors? A multi-k structure?

Firstly, we would like to mention that our results relating to the magnetic structure are not obtained from the refinement of data but rather from classical Monte Carlo simulations based on DFT Hamiltonian parameters. This provides another validation of the values deduced from DFT as being quite close to the 'real' values: a small change in parameters (like seen by going from $100\,K$ structure to the room-temperature structure) results in a change of the periodicity of the magnetic structure (from $3n$ to $4n$). It is found to be a multi-**k** structure, governed by multiple **k** vectors. Correspondingly, we have elaborated on the following discussion on the ordering vectors in the main text

The calculated magnetic structure for $L = 3n$, shown in Figure 1d, comprises several **q** vectors and results in a rather complicated pattern, devoid of any particular spin textures. In Figure 1d, we show a view along the (111) plane for a $L = 6$ system, where black arrows on each node represent the direction of the net magnetic moment within each unit cell. The angles between neighbouring cells are found to be close to $120°$.

The calculated magnetic structure for $L = 3n$, shown in Figure 1d, comprises propagation vectors $q_1 = (1/3, 0, 0)$, $q_2 = (1/3, 1/3, 0)$ and $q_3 = (1/3, 1/3, 1/3)$. The resulting pattern is devoid of any particular spin textures like skyrmions and merons. In Figure 1d, we show a view along the (111) plane for a $L = 6$ system, where black arrows on each node represent the direction of the net magnetic moment within each unit cell. The angles between neighbouring cells are found to be close to $120°$. We do not expect a significant change in the calculated structure if the exchange parameters are slightly modified, as long as the tripling of the unit cell is preserved.

The origin of the poor spin fraction is not a trivial topic, and it deserves a dedicated study. However, if we take into account that all spin liquids are surrounded by classical phases, the adiabatic progression from a pure spin-liquid phase towards a classical phase (via changes in values of exchange interactions) will entail the appearance and growth towards a purely classical value of the order

5

parameter. The deviation of the order parameter from its classical value will reflect the closeness of a given material to the purely spin-liquid phase but will also inevitably be influenced by the type and properties of the dynamical background. At the moment it is not possible to take into account those effects (not just by us, but by anyone in the community), and therefore anything further than what is written above would be pure speculation, which would inadvertently shift the focus from the main story of the manuscript. This is of course a very important topic but it will require a different, properly conceived set of experiments to address it.

(No 1.8) 2) I note that Ce2Hf2O7 and Ce2Sn2O7 could be also 3D spin liquid candidates.

We thank the Referee for mentioning these promising 3D spin liquid candidates. We have now included an additional reference for the Ce-based pyrochlore spin-liquid candidate.

(No 1.9) 3) According to literature, application of B = 4T magnetic field leads to a fully dynamical state down to the lowest temperatures. What is the situation from a theoretical point of view ? Would the application of the field drive the system into the "liquid region" reported here?

A fully quantum mechanical theoretical treatment of the behaviour in a magnetic field is outside the scope of the current analysis and methodological state-of-the-art available from PFFRG (though recently developed, the application to the current system remains out of bounds). It is, however, strongly linked with the puzzling nature of the ordered component. We have started the investigation of in-field tendencies and so far it has been revealed by Monte Carlo calculations that indeed the experimentally observed decrease of the ordering temperature is replicated. At the moment we are not in a position to claim anything further.

From a general point of view, $4\,\mathrm{T}$ magnetic field is well below the characteristic energy scale of the system such that the dynamic part is not expected to be influenced appreciably, as has been also previously reported through specific heat measurements up to $14\,\mathrm{T}$. It remains an open question if the ordered fraction 'joins' the dynamic ensemble or if it remains polarized.

(No 1.10) 4) What are J1 and J2?

$J_1$ and $J_2$ couple two trillium lattices in a slightly different way than $J_4$. Since their values are significantly smaller compared to other $J$s, we have not included those bonds in the illustration for reasons of legibility. In the Supplementary Information, we have shown the full network of interactions in Supplementary Figure 1 (now), as also elaborated in the previous publication Phys. Rev. Lett. 127, 157204 (2021) (same colour scheme).

(No 1.11) 5) In the limit J3, J5 → 0, the calculations predict a semi-classical AFM state. The limit J4 → 0 describes two independent trillium lattices, leading to a variant of the 120 degrees order. If surrounded by "classical" phases, how do the authors can conclude about the proximity to an "island of liquidity" ? A this point, the narrative is not clear to me.

All the networks that support geometrical/parametric frustration are inevitably surrounded by 'semi-classical' phases. A square lattice, with a nearest-neighbour $J_1$ coupling, leads to a semi-classical AFM Néel state, however, the introduction of a diagonal coupling $J_2$ induces frustration, driving the system into a spin-liquid for $J_2/J_1 \sim 0.5$. Similarly, for the triangular lattice $S = 1/2$ Heisenberg antiferromagnet which has a $120°$ semi-classical magnetic ordered ground state with a considerably suppressed moment, it is sufficient to include $J_2 \sim 0.06J_1$ to destabilize the ordered state into a spin liquid. Thus, an island of liquidity can always be lurking around the corner proximate to semi-classical magnetic orders, and even small competing couplings can prove sufficient to melt ordered states into spin liquids even in 3D, e.g., as happens for the diamond lattice $J_1$-$J_2$ Heisenberg antiferromagnet, Phys. Rev. Lett. 120, 057201 (2018).

The essential manifestation of geometrical frustration is the reduction of the ordering temperature due to enhanced fluctuations. The degree of proximity of a given set of parameters to a region of spin-liquid behaviour is a delicate question. From a practical perspective, the important point is the extent of the fluctuation-dominant region. We show that $K_2Ni_2(SO_4)_3$ is dominated by dynamics and that these dynamics originate from the discovered 'island of liquidity'.

(No 1.12) 6) Line 180 : "INS is featureless". This is not that surprising given the fact that the data

have been taken above Tc, so we just expect quasi-elastic scattering as usually observed close to a transition. What about below Tc?

This remark is a continuation of the Referee's assumption that the system at $2\,\mathrm{K}$ is a conventional paramagnet. Line 180 states:

> The energy dependence of the intensity of the scattering signal for various $\mathbf{q}$-points (that present streaks) is featureless, with approximately linear decrease with energy (see panel (d) in Figure 2).

We would refrain from extrapolating this observation to a sweeping statement that 'INS is featureless', firstly by the very presence of vertical streaks appearing at certain q-values. We have emphasized that it is only the energy dependence which is relatively featureless, being monotonically decreasing towards higher energies. The presence of these streaks is a ubiquitous feature also observed in other QSL candidates such as the $S = 1$ pyrochlore material $NaCaNi_2F_7$ [Nat. Phys. **15**, 54 (2019)] and the $S = 1/2$ hyperhyperkagome systems $PbCuTe_2O_6$ [Nat. Comm. **11**, 2348 (2019)]. This in no way implies that the INS is featureless, as observed from the QQ maps. For a 'boring' paramagnet, the energy dependence would be both q- and E-independent.

Furthermore, the Referee continues with 'we just expect quasi-elastic scattering as usually observed close to a transition'. This has nothing to do with the observed streaks presented in Fig. 2. The streaks are centred around 0.75 and 1.8 inverse units, while the ordering occurs at multiples of 1/3. Additionally, the observed streaks are very broad and observed both below the ordering temperature and fade in intensity very slowly toward high temperatures.

(No1.13) 7) Line 187 $\rightarrow$ 205. What is the purpose of this paragraph?

Since we are aiming for a very general audience, the purpose of this paragraph was to explain that $S(Q)$ carries a fingerprint of the correlations living on a particular geometrically frustrated network. Seeing that our contribution is focused on revealing a new geometrically frustrated network, not studied before, we thought it is important to emphasize that the features revealed in Fig. 3 are unique, compared to the previously discovered pinch-point, half-moon, star and other features.

However, on the insistence of Referees 1 and 3, we have decided to remove this paragraph.

(No1.14) 8) Line 274 : "K2Ni2(SO4)3 lies very close to a so-far unexplored region of genuine spin-liquid behaviour, centred around a particular point defined by J3 = 0, J4 = J5." Furthermore: Line 294 : "For S = 1, PFFRG indicates a very narrow range around J3 = 0"; Line 328 : "cMC calculations show no ordering tendencies in a large J5/J4 region around the tetratrillium lattice when J3 = 0." Line 333 : "On the other hand, even the smallest non zero value of J3 results in a finite critical temperature".

Given these sentences, I understand that the liquid region is very narrow. But DFT points to J5=0.5 J4 and J3= 0.1 J4, which is not "so" close. How can the reader then appreciate? How to conclude that the spin liquid region is relevant in understanding K2Ni2(SO4)3?

Even if the zero-temperature spin liquid region obtained with PFFRG for $S = 1$ is narrow, the critical temperature around this region is expected to be very small, which is not that uncommon for 3D frustrated magnets. On top of that, even if a system is a bit distant from the island of liquidity, where weak ordered states appear, the effects of spin-liquid dynamics are very much present. As shown in Phys. Rev. B 98, 184403 (2018) and Nat. Phys. 20, 74 (2024), the existence of a quantum melting point at $T = 0$ for a given set of parameters affects the dynamical responses all around it, and also at finite temperatures, in what is referred to as the quantum critical regime. From this point of view, the dynamical features of $K_2Ni_2(SO_4)_3$ provide evidence of proximity to an island of liquidity at $T = 0$. Furthermore, the revealed phase diagram is only 'theoretical' and it should serve as a guiding principle when discussing real materials. It is possible that the 'real island of liquidity' is wider.

(No1.15) 9) Line 297 : what is the purpose of considering $S = 1/2$ (Ni having a spin $S = 1$) ?

As the spin length increases, the behaviour of the system progressively becomes more 'classical'

$-S = 1/2$ is, therefore, an ultimate quantum limit. We confirmed our expectation that by going from $S = 1$ to $S = 1/2$, the island of liquidity gets wider in parameter space. This has implications for potential $S = 1/2$ materials, for instance, the Cu-based $K_2Cu_2(SO_4)_3$, which has, so far, not been reported.

In the opposite limit $(S \gg 1)$, it is expected that the island would shrink since quantum fluctuations get progressively weaker. Surprisingly, it was revealed in Ref. [21], that even a $S = 5/2$ system on the same magnetic lattice does not show order to the lowest temperature investigated. Hence, it is not trivial to assess the impact of quantum fluctuations on the spin dynamics based on general considerations alone and one has to take into account the specificity of the model and lattice under consideration.

(No1.16) 10) Line 307 : "many of the features observed in the spin structure factors can be traced back to the tetra-trillium lattice limit." Many? which ones?

This phrase refers to the fact that the overall Q structure observed in PFFRG for the relevant model of $K_2Ni_2(SO_4)_3$ and that in the tetra-trillium limit does not present any abrupt or discontinuous changes, showing that they can be considered proximate in parameter space. To clarify this, we have changed the sentence:

The set of parameters calculated with DFT for $K_2Ni_2(SO_4)_3$, indicated by a green star in Figure 4a, lies close to these paramagnetic regions and many of the features observed in the spin structure factors can be traced back to the tetra-trillium lattice limit.

The set of parameters calculated with DFT for $K_2Ni_2(SO_4)_3$, indicated by a green star in Figure 4a, lies close to these purely spin-liquid regions. This becomes evident when the PFFRG spin structure factor (at finite $\Lambda$) is compared to the results obtained in the tetra-trillium lattice limit (at the smallest $\Lambda$), as many of the features can be put in correspondence (see Figure 4b and Supplementary Information). Generally, the peaks become more smeared in the tetra-trillium limit, but there is no indication of a strong difference between the two phases. This provides evidence that the properties of $K_2Ni_2(SO_4)_3$ are governed by its proximity to the quantum spin liquid phase in the tetra-trillium lattice [19,20].

(No1.17) 11) Line 358 : e(T) is not defined

We thank the Referee for pointing this out. We have changed:

...the calculations of 2e(T) for a single tetrahedron...

...the calculations of (twice) the energy as a function of the temperature, 2e(T), for a single tetrahedron...

(No1.18) 14) Line 335 : "This fine line of pure spin-liquid behavior can be understood from the Hamiltonian containing only J4 and J5, which can be written as a disjoint sum of squared total spin in each tetrahedron (just as for the pyrochlore lattice". How is this remark related to the experimental entropy release?

While it is true that the classical spin liquid in the tetra-trillium lattice probably has a non-zero entropy at $T = 0$, we do not see a direct connection between this and the entropy release in $K_2Ni_2(SO_4)_3$. For the tetra-trillium lattice limit, the discussion revolves around a pure spin-liquid regime while for $K_2Ni_2(SO_4)_3$, the entropy release is related to the partial order on top of the dynamic background.

**Reviewer 2**

(No2.1) The authors employ a combination of inelastic neutron scattering, pseudo-fermion functional renormalization group, and classical Monte Carlo calculations to explore the complex properties of K2Ni2(SeO4)3, a compound composed of two 3D nets of corner-sharing triangles. The authors' efforts in integrating numerical calculations and experimental results are appreciated. However, the analysis and strategy of studying a frustrated magnet is not convincing. Therefore, I do not recommend Nature Communications publishing this manuscript.

Most importantly, I have reservations about the strategy employed to determine the exchange coupling constants. Given the highly dynamic ground state mentioned in the manuscript, it may be advisable to investigate the system by driving it into an ordered state with a magnetic field and subsequently fitting the spin wave excitations. This approach, as demonstrated in Reference [1], can provide more precise determinations of the exchange parameters.

We do appreciate the approach to determine exchange coupling constants employed in Physical Review X 1, 021002 (2011). It is certainly the most important tool to determine exchange interactions for $4f$ magnets where density functional theory has problems. However, we beg to disagree with the viewpoint that spin wave theory fits the magnetic dispersion measured in the field-induced polarized state is now the only possible approach to the problem for all spin systems. In our particular case, the experimental approach to the Hamiltonian favoured by the Referee suffers from several shortcomings:

- Linear spin wave theory is only approximate for spin 1 systems

- Numerical fits become more and more ambiguous as the number of parameters increase, therefore reducing the reliability and increasing the error bars

- Most notably, the question of the energy scale is not taken into consideration. In Physical Review X 1, 021002 (2011), the authors deal with $Yb_2Ti_2O_7$, a $4f$ system whose Curie-Weiss temperature is $< 1\,K$ and the field-polarized state is reached with a very accessible $5\,T$. In the case of $K_2Ni_2(SO_4)_3$, the energy scale is $\sim 20x$ larger, requiring fields above $25\,T$. This is confirmed by our recently acquired pulsed-magnetic-field experiments:
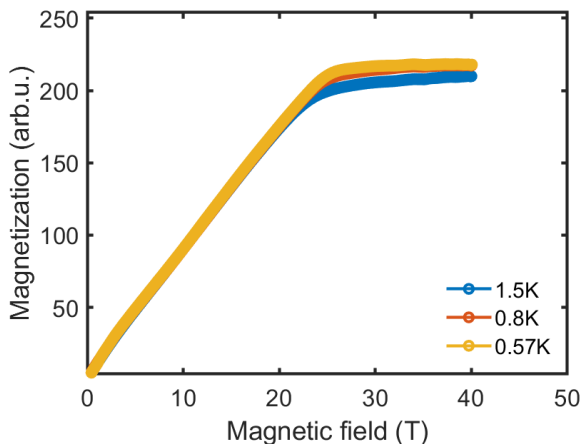


Currently, there does not exist any neutron facility that would allow reaching $30\,T$ and measuring dispersions in a fully polarized state. Therefore, the suggested approach is completely unfeasible.

We would like to point out that the excellent comparison we can achieve between INS spectra and susceptibilities calculated from the DFT energy mapping Hamiltonian suggests that the method is very reliable here, for Ni $3d$ electrons. As we have pointed out in (No 1.6) to Referee 1, the agreement necessitates all the steps to be correct, including the values of exchange parameters. Additionally, only a narrow set of exchange parameters results in tripling of the unit cell (the room-temperature set suggested in Phys. Rev. Lett. 127, 157204 (2021) leads to quadrupling), so we dare to claim that no other set will be significantly better in describing the system under investigation.

More generally, narrowing down the set of parameters that describe any material serves to position

this particular system within a well-established phase diagram. Our contribution is not ONLY to determine the set of parameters for $K_2Ni_2(SO_4)_3$ but also to reveal for the first time the phase diagram and identify the source of its highly dynamic behaviour for this novel frustrated magnetic network. Within that phase diagram, the behaviour of $K_2Ni_2(SO_4)_3$ will not change qualitatively if some exchange parameters are off by a few percent. This is well within an error bar that can be expected from any neutron-scattering-based method.

Since the Referee does not specify what they think does not align with the established terminology, we can only speculate on the thoughts he/she might have. Hence, we take this opportunity to clarify and reiterate a few points that we think might help clarify the terminology used. As previously elaborated in (No 1.2), any magnetic system that shows correlations (i.e., it is not a conventional paramagnet), while at the same time is not ordered due to geometrical frustration, is considered a spin-liquid. If one was to adopt the Referee's reasoning, then we are afraid that it would exclude *all* materials which the community has so far identified to be promising spin liquid candidates. Adopting this assessment would imply that even at the lowest accessible temperatures of $\sim 50\,\text{mK}$, any 'dynamic' response would be invalidated because the material might order at lower temperatures. Rather, what *is* relevant is the energy scale of spin interactions, which for $K_2Ni_2(SO_4)_3$ is $20\,\text{K}$ so at $2\,\text{K}$, without long-range order, the system is deep into a correlated spin-liquid regime. Indeed, this is precisely the notion of "cooperative paramagnetic" behavior arising in the intermediate temperature regime $T \ll J$ which, for example, in the kagome Heisenberg antiferromagnet shows pinch-points and bowties (indicative of non-trivial correlations) in the Coulomb spin liquid regime, before it disappears at even lower temperatures with the onset of a coplanar regime (please see Fig. 3 in Phys. Rev. B 78, 094423 (2008)). The fact that upon cooling, a very faint order emerges does not invalidate the correlation effects at $2\,\text{K}$. Moreover, the dynamic part remains the same even below $1\,\text{K}$, there is no redistribution of spectral weight towards magnon dispersions. The magnetic response of $K_2Ni_2(SO_4)_3$ is thus truly dynamic in every sense of the word below and above this (unconventional) ordered state.

We would like to mention that the Referee's assessment is based on several assumptions which have been taken for granted and extrapolated to the case of $K_2Ni_2(SO_4)_3$ (similar to those made by Referee 1 in No 1.3) without taking into account the evidence presented by us in Phys. Rev. Lett. 127, 157204 (2021) and the current manuscript pointing to an unusual behaviour in this material. The question of the static moment has been addressed in the previous publication but because of a very small signal it was not possible to refine the structure, and therefore it was not possible to extract the size of the static moment. Moreover, the results of $\mu$SR experiments revealed no oscillations, indicating a fully dynamic state. Finally, with only 1% of the entropy freezing at $T_N$, it is not reasonable at all to assume that the ordered moment represents the full moment. We have shown under point No 1.3, that the results at the lowest accessible temperature show that the response is equally dynamic as observed at $2\,\text{K}$. We would like to reiterate that given the observed features at $50\,\text{mK}$, the dynamic features at 2K *are* representative of spin-liquid behaviour characteristic for this particular lattice and are relevant for comparison with theoretical predictions.

We would like to clarify that the field-induced QSL state reported in our previous work [Phys. Rev. Lett. 127, 157204 (2021)] is not to be confused with a field-polarized state. It is well known from

studies on other materials, e.g., Nat. Commun. 12, 5559 (2021) that the application of a magnetic field can induce QSL behaviour at small to intermediate fields. In our case, a 4 T magnetic field well-places the system in this regime and not in a field-polarized state. If the application of a 4 T magnetic field has induced a field-polarized state, this would be reflected in the behaviour of the specific heat. However, in our previous publication Phys. Rev. Lett. 127, 157204 (2021), we have shown that at 14 T the specific heat behaviour is practically identical to the 0 T results. We would like to mention that it would require a field of at least 30 T to reach full polarization (confirmed with pulsed-field data (No 2.1)).

(No 2.5) In summary, the manuscript is intriguing, especially in its observations that excitation in the ordered state does not exhibit typical spin wave behavior, indicating the presence of strong frustrations.

Indeed, while the data we show has been taken at 2 K which is just above $T_N$, it is nevertheless true that even below $T_N$, an astonishingly similar excitation spectrum is observed without any clear evidence of magnons. While this feature does deserve further investigation, it is not part of our manuscript, and we have only shown this data in response to No 1.3.

(No 2.6) 1. Delve into the crystal field of Ni, which, while complex, is essential for a comprehensive understanding of the system. This aspect is currently missing from the manuscript.

We would respectfully beg to disagree with the Referee on this point. May we please take this opportunity to refer to the book *Transition Metal Compounds (Cambridge University Press)* (Daniel I. Khomskii) where the crystal field effects are discussed in glorious detail in Chapter 3. In particular, on page 77 it is explained that for $Ni^{2+}$ due to completely occupied $t_{2g}$ levels the complex effects that the Referee is indicating are much weaker. The crystal field of a $3d$ ion in an octahedral environment is thus not complex, as suggested by the Referee. A $d^8$ configuration of $Ni^{2+}$ leaves no uncertainties as to which spin-state is formed and a $S = 1$ state is quite robust in that it is found in every possible environment of a $Ni^{2+}$ ion, regular or irregular octahedron, pyramidal, tetragonal, or any other.

On the other hand, the complexities of the crystal field are regularly investigated in $4f$ systems. Often crystal field level population is temperature-dependent, leading to $T$-dependent magnetic properties. While these comments may be relevant for $4f$ systems, they cannot be extrapolated to be used for a $3d$ system.

(No 2.7) 2. Refine the diffraction data to determine precise ordered moments in both zero-field and high-field states.

We appreciate the Referee's suggestion and we would like to mention that this has already been done in our previous work Phys. Rev. Lett. 127, 157204 (2021). A very small ordered moment was found, but due to multiple ordering vectors and the high symmetry of the unit cell, a refinement was not possible (i.e., one could not achieve a unique solution).

(No 2.8) 3. Investigate the exchange interactions by fitting the spin-wave excitations under magnetic fields.

We would like to reiterate that this idea is not feasible for any experimental facility in the world, as it is not possible to perform neutron scattering experiments with fields of 30 T needed to fully polarize $K_2Ni_2(SO_4)_3$.

(No 2.9) 4. Employ numerical methods to reproduce the zero-field excitations based on the interactions determined.

The suggested utilization of numerical methods to reproduce the zero-field calculations has already been addressed in the reply to No 1.5. Currently, there exist no theoretical/numerical quantum many-body methods to calculate dynamical spin structure factors in 3D, and more so for such complicated lattice geometries with $S = 1$ magnetic moments.

(No2.10) For the current manuscript, I think it could be published in a more specialized journal after changing QSL-related arguments. I hope my comments and recommendations prove helpful

Based on our replies to the aforementioned questions of the Referee, in particular, those clarifying the QSL-related arguments, we believe that we have now alleviated his/her concerns which led the Referee to this recommendation. In particular, we point out that the field polarized approach towards estimating the exchange parameters cannot be applied to $K_2Ni_2(SO_4)_3$ due to the absence of any neutron scattering facility capable of performing it in fields of $30\,T$. On the other hand, the field-polarized approach is by no means the only available route towards estimating the exchange parameters, and evidence from many more perspectives can be shown to instil confidence in the reliability of exchange parameters determined from, say, ab-initio approaches. For one, compared to the field-polarized approach, ab-initio modelling provides an insight into the energy scales involved, and the uncertainty in the fits is comparatively much lesser than those from the field-polarized approach which still leaves us with many choices, thus yielding a non-unique set of exchange parameters. The energy mapping approach does not suffer from this uncertainty and yields a unique solution for a given Curie-Weiss temperature. The confidence in our magnetic Hamiltonian can be read from the excellent comparison between theory and experiment, which is rarely observed for such complicated Hamiltonians on intricate 3D lattices. We would like to point out that similar modelling yielded a quantitative agreement between theory and experiment for the $S = 1/2$ $Cu^{2+}$ hyperhyperkagome material $PbCuTe_2O_6$ Nat Commun 11, 2348 (2020). Such an agreement heavily depends on the accuracy of exchange interactions deduced by DFT, for both QQ maps and for the correct prediction of the magnetic unit cell. Therefore, even if a slight change of interactions is eventually found, its position in the phase diagram of Figure 4 will be (maybe) slightly different but ultimately the main result is that $K_2Ni_2(SO_4)_3$ is highly frustrated and close (in parameter space) to a novel type of magnetic frustrated lattice, a tetra-trillium lattice which governs its physics –this is an irrefutable observation and one of our highlights as it reveals the novel frustration mechanism at play in stabilizing a QSL, and one which is different from that (naively) expected out of trillium lattice physics, thus pinning the origin of the spin liquid. This emergence, we believe, has not been acknowledged and appreciated by the Referee. Overall, these results, have much wider ramifications than solely determining the exchange parameters of a given material, to which our work should not be reduced to. We would like to point out that the introduction of a new effective lattice, namely, the tetra-trillium geometry, which has been unreported so far combined with strong evidence of it determining the physics of the material is not a scenario that is widely encountered, if at all. Unfortunately, the Referee has restricted himself/herself to be too narrow in scope and thus overlooked this new overarching paradigm, which provides the strongest evidence in support of the correctness of our exchange parameters. Thus, we are confident that when the whole scope of our manuscript is taken into account, it *does* constitute a contribution relevant and worthy of publication in Nature Communications.

**Reviewer 3**

(No 3.1) I believe that the manuscript "Dynamics of K2Ni2(SO4)3 governed by proximity to a 3D spin liquid model," by Gonzalez et al., is suitable for publication in Nature Communications after revisions. I believe the revisions I recommend require no further experiments or numerical simulations, but they will require important changes to the text.

In this paper, the authors consider the material K2Ni2(SO4)3, which has previously been found to show high magnetic entropy at low temperatures and spin-liquid like behavior under small applied fields. They authors find an approximate Heisenberg model for the material, with coupling strengths determined using density functional theory (DFT). They then study this model and some approximations to it (leaving out certain small spin-spin interactions), using both classical Monte Carlo simulations at finite temperature and quantum pseudo-fermion functional renormalization group (PFFRG) calculations at 0 temperature. They compare the results, for example for the spin structure factor, with experimental inelastic neutron scattering data. The key finding is that some spin models, which are close in coupling strengths to the one describing the real material, apparently exhibit spin liquid ground states, giving a possible explanation for the experimentally observed behavior of K2Ni2(SO4)3.

We appreciate the positive consideration and accurate description of our work made by the Referee, and, in particular, his/her appreciation of the broad scope of the work.

(No 3.2) The agreement between numerical simulations and experimental data is remarkable, especially as shown in Fig. 1c and Fig. 3 a-f. While I am not an expert on either of the specific theoretical methods employed by the authors, I believe the numerical results presented give strong evidence for the quality of the calculations and the applicability of the methods to this problem. The authors also provide reasonable discussions of both methods in the Supplementary Information.

We appreciate the positive consideration and accurate description of our methodology made by the Referee. We especially acknowledge that he/she finds confidence in our magnetic Hamiltonian based on the excellent agreement found between theory and experiment.

(No 3.3) Overall, the authors address an important question that will be of interest to the condensed matter physics community, namely the origin of spin liquid-like behavior in 3D materials. They make a convincing case that the behavior is due to proximity in parameter-space to a model that seems to have a spin liquid phase, as identified by PFFRG calculations. They also provide a demonstration of the interesting correspondence between quantum and classical fluctuations.

All of that being said, the authors need to make some important revisions to improve the clarity of the paper, which I describe below. The points are roughly in order from most to least important.

1. More explanation is needed regarding the choice of T for the cMC calculations and Lambda for the PFFRG calculations, and of why the cMC calculations are needed at all.

In PFFRG, the Lambda parameter is akin to temperature and its flow may be viewed as an annealing process. The value of Lambda is chosen above but close to the flow breakdown, i.e., still in the paramagnetic phase just before entering the incipient magnetic order. The resulting spin structure factor does not change significantly (the qualitative profile is the same) as long as these conditions are met. The flow breakdowns, indicating the onset of magnetic order, are detected as kinks or peaks in the evolution of the susceptibility maxima as a function of Lambda. For Lambda below the breakdown, the results are numerically unstable and can no longer be trusted, or in other words, the RG flow cannot be carried over "into" the magnetically ordered phase. This was previously mentioned in the manuscript:

> Within PFFRG, the renormalization-group flow breaks at $\Lambda = 0.582 \ J_4$, indicating the presence of a magnetically ordered ground state. Static spin correlations are measured at this point and, ...

We have clarified the choice of Lambda by changing the text to

The choice of the value of $T$ in cMC is solely determined empirically to obtain the best comparison with results from PFFRG. While we find an excellent qualitative agreement with PFFRG over a wide temperature range above $T_N$, we choose a $T$ value (scanning in steps of 0.05 $J_4$) that gives the best quantitative agreement. The insights and complementary support that cMC results provide can be summed up as (i) they shed light on the existence of the puzzling phenomena of quantum-to-classical correspondence of structure factors in 3D, known so far mainly for 2D models Phys. Rev. B 101, 035132, (ii) Our cMC results further validate the exchange parameters obtained from DFT energy mappings, by providing a comparison to the measured ordering wave vector which is seen to be in excellent agreement. We would like to emphasize that the new set of exchange couplings are the only ones that allow cMC to obtain the same ordering wave vector (the tripling of the magnetic unit cell) as observed in experiments, and even a small tweak leads to spin configurations with a different periodicity –thus providing a high degree of confidence in the reliability of the exchange parameters. The other two sets of parameters give different results (see Supplemental Information).

To make the selection of temperature clearer, we modified the following part:

Thus, in an attempt to simulate the experimental data within a classical approach, we perform cMC calculations above the finite-temperature phase transition. Particularly, for T = 0.35 J4 , we find an excellent agreement with the PFFRG calculations (see left and right panels separated by black lines in panels (d)-(f) of Figure 3).

Thus, in an attempt to simulate the experimental data within a classical approach, we perform cMC calculations above the finite-temperature phase transition. We find an excellent agreement with the PFFRG calculations for a large range of temperatures and, particularly for T = 0.35 J4, we find the best quantitative agreement (see left and right panels separated by black lines in panels (d)-(f) of Figure 3).

(No 3.4) My impression is that the logic goes like this: (1) the goal is to simulate the T=0 (or T=2K, which I gather is in the ground state phase, though this should be clarified) behavior of the model; (2) in PFFRG, follow the renormalization group flow from large Lambda until hitting an instability that indicates ordering; (3) do quantum calculations at that Lambda to get correlation functions; (4) select a T for cMC that gives the most similar possible behavior to the PFFRG calculation. Sometimes such a T can be found (Fig. 3) and sometimes not (Fig. 4b, top half).

The logic is right but we are not trying to simulate the ground state but the state at finite temperatures which is measured by INS. We can only add that a finite Lambda acts similarly as a finite $T$ (however an exact functional relation between both quantities likely does not exist). So, when cMC agrees with PFFRG, it is always a comparison between results at finite temperatures. A threshold temperature is always expected for the quantum-to-classical correspondence: below a given temperature, fluctuations are too quantum and cannot be reproduced by classical thermal fluctuations. This is what happens in the top half of Fig. 4b: the PFFRG results are taken at Lambda $\sim 0$, which is the authentic $T = 0$ result, and are therefore impossible to simulate and reproduce with classical thermal fluctuations.

(No 3.5) Some questions that come up: - Does the cMC temperature have some physical meaning, apart from the size of (quantum) fluctuations? It is apparently an order of magnitude above physical temperature, e.g. for Fig. 3 the physical temperature is seemingly below a transition at 0.048 J4, while the cMC temperature is 0.35 J4.

The classical temperature has no particular physical meaning in the sense that a certain physical phenomenon is to be expected in the material at this temperature. Yet, a general consistency of our energy and temperature scales can be observed: The INS experiments are performed at 2 K, which in terms of $J_4$ is $T = 0.36\ J_4$, very similar to the temperature of 0.35 $J_4$ used for cMC.

We do not think there is such a "correct temperature" for cMC. Without the PFFRG calculations, we would have found a large range of cMC temperatures that give a spin structure factor that is in perfect agreement with the experiments. On the other hand, there is no way to predict $T$ given Lambda, but we can mention that there is another related method, PMFRG, that uses Majorana auxiliary particles instead of fermions for the mapping of spin operators (Phys. Rev. B 103, 104431 (2021)). In that approach, Lambda can be implemented such that it is identical to the temperature $T$. It would be certainly interesting to compare the quantum and classical temperatures for which the quantum-to-classical correspondence exists. However, this is beyond the scope of this work. Finally, the idea of selecting a temperature for cMC that agrees with the PFFRG calculations has the sole purpose of showing the quantum-to-classical correspondence, an interesting phenomenon in itself.

The temperature in Fig. 4c is the physical temperature of a classical spin system on the tetra-trillium lattice (or a single tetrahedron) and, hence, has a physical meaning. However, the temperature range investigated in Fig. 4c does not have direct relevance for $K_2Ni_2(SO_4)_3$. Rather, the purpose of Fig. 4c is to demonstrate that classical spin models on the tetra-trillium lattice, despite their complexity, inherit some of their properties from simple isolated tetrahedra.

The cMC calculations are used to demonstrate the quantum-to-classical correspondence, but also to determine the correct magnetic unit cell (something which cannot be done with PFFRG), as well as for the nature of the spin liquid in the tetra-trillium limit.

There is still some confusion present in the Referee's terminology. The *ground state* is the state of the system at $T = 0$. At finite temperatures, states are just thermodynamic equilibrium states. The quantum-to-classical correspondence is never fulfilled for ground states because quantum fluctuations can never be reproduced by entropic thermal fluctuations. In other words, the fluctuations in the quantum ground state are always quantum in nature (because $T = 0$). This is why the PFFRG result in the top of Fig. 4b cannot be reproduced by cMC. At finite temperatures, however, entropy plays a role in the state of the system. If thermal fluctuations are strong enough, the same state may be simulated by cMC calculations at a different temperature. This is the quantum-to-classical correspondence. This is what happens in the bottom part of Fig. 4b, where PFFRG results at finite Lambda (equivalent to some unknown finite temperature) can be reproduced by cMC calculations at finite temperature. This also happens for the model of the compound, where calculations are exclusively performed at finite temperature and Lambda to reproduce the experimental measurements performed at finite temperature.

To answer the question directly: If cMC cannot reproduce the PFFRG results, as in Fig. 4b (top part), it is an indication that quantum calculations are performed at too low temperatures, such that classical thermal fluctuations cannot reproduce the inherent quantum fluctuations. However, this is no indication of a spin liquid. On the other hand, the fact that the PFFRG flow continues down to Lambda $= 0.01$ without breaking is an indication of a quantum spin liquid. Furthermore, the fact that the heat capacity $c_v$ in cMC continues to $T = 0$ without any sharp peak is an indication of a classical spin liquid.

and whether the cMC temperature has any physical meaning.

We hope that the previous answers have illustrated correctly why the cMC results are useful for building the whole picture of the physics related to the model compound.

To re-elaborate, the purpose of the cMC results is not only to compare with the INS experiments, something which could have been done with PFFRG alone. The cMC calculations also validate the set of parameters from DFT by finding the correct magnetic unit cell (which does not work for other sets of parameters). This identification of the correct magnetic unit cell is technically too challenging for the PFFRG. The cMC results also provide a classical phase diagram, in which the spin liquid in the tetra-trillium lattice is at the center. This allows one to partially explain the complex physical phenomena of our quantum tetra-trillium system with a simpler classical system. Finally, we repeat that while the cMC temperature is a physical temperature, it has no direct relevance for comparison with experiments.

(No3.10) As a less important point, I think for readers (including me) who are less familiar with PF-FRG, some additional discussion of why the calculations should be done at precisely the Lambda at which the RG flow breaks down would be helpful. (This could be in the Supplementary Information, augmenting the current discussion there.)

We have clarified in the Supplemental Information that the selection of Lambda corresponds to the lowest Lambda value that can be calculated (after the flow breaks, results cannot be trusted any longer). Ideally, one would like to obtain results at Lambda=0, but PFFRG cannot easily access symmetry-broken phases. Results above the flow breakdown typically do not change abruptly, such that it is not necessary to calculate the correlations exactly at the flow breakdown; it can be done anywhere close to it.

(No3.11) 2. Computation of S(q): the discussion around the integration in (1) needs to be improved. In particular, how are the limits omega1 and omega2 chosen? Also, these parameters currently appear in (1) but are not discussed at all until the end of the page, which I found confusing. They should be discussed, and the choice of values explained, immediately after the equation. This is especially important since the choice of integration limits is cited at the end of page 4 as a possible reason for discrepancies between the curves in Fig. 3 g-i.

Since the intensity of streaks shown in Fig2a-c is monotonously decreasing towards larger energies, without any visible dispersion (i.e. there is no visible q-dependence), in theory, one could integrate across the whole energy range and perform the comparison. In practice, on the low-energy side, we are limited by the presence of an incoherent scattering, which extends up to $0.5 \, \mathrm{meV}$. On the high-energy side, the signal from streaks merges with noise, therefore we have tried to find an optimum upper limit which would maximize our signal-to-noise ratio.

The text is now changed:

In the top row of Figure 3 we show $S_{\mathrm{exp}}(\mathbf{q})$ along three different planes in reciprocal space, obtained by integrating the intensity in the energy range $\omega_1 \sim E_{\mathrm{min}} = 0.5 \, \mathrm{meV}$ and $\omega_2 \sim E_{\mathrm{max}} = 1 \, \mathrm{meV}$. The patterns depicted carry fingerprints from the underlying magnetic lattice composed of two interconnected trillium lattices, with a hexagonal galaxy-like motif reflecting the chirality of the crystal structure (see panel (c)), arising from the non-centrosymmetric space group.

In the top row of Figure 3 we show $S_{\mathrm{exp}}(\mathbf{q})$ along three different planes in reciprocal space, obtained by integrating the intensity in the energy range $\omega_1 \sim E_{\mathrm{min}} = 0.5 \, \mathrm{meV}$ and $\omega_2 \sim E_{\mathrm{max}} = 1 \, \mathrm{meV}$. The lower limit is given by the extent of the incoherent scattering while the upper limit is found as an optimal value to maximize the signal-to-noise ratio. The patterns depicted carry fingerprints from the underlying magnetic lattice composed of two interconnected trillium lattices, with a hexagonal galaxy-like motif reflecting the chirality of the crystal structure (see panel (c)), arising from the non-centrosymmetric space group.

Due to the Referee's suggestion to explain these limits immediately after Eq. (1), we had to com-

pletely remove the paragraph which sits in-between and where some general remarks on how S(Q) behaves in various long-range ordered and spin-liquid phases are given.

<span style="color:blue">(No3.12) 3. I have some confusions about Fig. 2, so the authors could be clearer about the following: - Clarify precisely what quantity is actually indicated by the color. Some measure of particle flux/intensity?</span>

We have used one single label $S(q, \omega)$ for both the colour bar and the 'y' axis of the (d) panel. We understand that this is confusing. Therefore, we separated the two axes and indicated the labels separately. We also changed it to $S_{\text{exp}}(\mathbf{q}, \omega)$ in agreement with the main text, and introduced the name at the beginning of the paragraph where the dynamical structure factor is introduced. We hope that now the text and figures are clearer.

<span style="color:blue">- (No3.13) Do white regions mean that the maximum of the scale was exceeded? Then why are there also white spots in the upper part of 2c?</span>

Those few points are part of the experimental result but should be regarded as artefacts (for instance a cosmic ray hitting a neutron detector would typically saturate its electronics).

<span style="color:blue">- (No3.14) Also, my understanding of the phrase "streaks of intensity without any apparent q-dependence" is that it refers to regions in 2a-c where the intensity (ie color) is constant on some horizontal line. That only appears to be true at the bottom of the figure, but if white is off-scale, then how do we conclude that the intensity is constant?</span>

<span style="color:blue">The authors should clarify the answers to these questions, or, if I have fundamentally misunderstood something, they should write a clearer explanation of the data in Fig 2 to prevent this misunderstanding for future readers.</span>

We thank the Referee for pointing out this potential confusion. Indeed, the intended meaning is not to refer to any horizontal line with a constant intensity. The streaks that we refer to are those bright-coloured (yellow/orange/red) vertical regions seen at several q-vector values. Each streak is rather broad, with the only dependence being that on energy, seen monotonously decreasing as shown in panel (d). Our claim that there is no q-dependence refers to the fact that for each streak the intensity remains centered at the same q-vector value.

We changed the text as follows:

> <span style="color:red">Energy-momentum plots along three principal directions show streaks of intensity without any apparent **q**-dependence, indicating a non-dispersive type of excitations.</span>

> <span style="color:green">Energy-momentum plots along three principal directions show vertical streaks of intensity, four in each panel, indicating a non-dispersive type of excitations.</span>

Hopefully, these changes will avoid any confusion for future readers.

<span style="color:blue">(No3.15) 4. I am confused by the statement "Most importantly, PFFRG contains the correct large-N (spin flavour) and large-S (spin size) limits, making it ideal to study quantum systems." in the Supplementary Information. The authors should add some short additional clarification.</span>

We have added an extended explanation of the meaning of this phrase in the Supplemental Information. We changed:

> <span style="color:red">Most importantly, PFFRG contains the correct large-N (spin flavour) and large-S (spin size) limits, making it ideal to study quantum systems. Higher values of $S > 1/2$ are obtained coupling copies of the $S = 1/2$ fermionic spins at each site [S6] (in the present case for $S = 1$ only two are needed).</span>

> <span style="color:green">Most importantly, PFFRG contains the correct large-$N$ and large-$S$ limits [S6,S7]. This means that the PFFRG equations can be generalized for a Heisenberg model with $\text{SU}(N)$ symmetry, where $N$ is the number of spin flavours, and they become exact on a mean-field level for $N \to \infty$. In this limit, spin systems usually do not develop magnetic</span>

long-range order, such that the PFFRG's ability to describe quantum paramagnetic phases is intrinsically guaranteed by its exactness at $N \to \infty$. On the other hand, the PFFRG equations can also be extended for arbitrary spin magnitudes $S$, and in the limit $S \to \infty$ they can be solved analytically to obtain exact classical mean-field results. Since in the classical large $S$ limit, spin models usually develop magnetic long-range order, the exactness in this limit ensures an accurate treatment of static magnetism. Hence, with PFFRG's ability to describe both types of spin phases, non-magnetic ones and magnetically long-range ordered ones, it is an ideal approach for studying quantum spin systems. In practice, higher values of $S > 1/2$ are obtained coupling copies of the $S = 1/2$ fermionic spins at each site [S6]

(No3.16) 5. Minor point: inconsistent use of Q and q

We thank the Referee for pointing this out. We have made the notation uniform by switching to q always.

(No3.17) 6. Minor point: Fig 4c should have T/J4 as the x label

We thank the Referee for pointing this out. In this case, $J_4 = 1$ so that we left it out, but it is more general to keep it. We have changed it as suggested.

(No3.18) 7. Minor point: why is cMC T=0.001 selected for 4b upper left? Just because it's the smallest value?

Yes, this is the smallest value of temperature reached, and it is sufficiently small so that no qualitative changes are expected for even smaller temperatures.

(No3.19) 8. Minor point: many of the figures, although clear in color, are confusing in black and white. The authors should improve the appearance of the figures so that they are clear when the paper has been printed in grayscale. This is specifically a problem with: Fig 1a, Fig1b, Fig 3g-i, the stars in Fig 4a.

We thank the Referee for pointing this out. We have updated the colours and line types of Fig. 3g-i and changed one of the stars in Fig. 4a for a circle. The figures are now comprehensible in grayscale.

**SUMMARY OF CHANGES**

<u>Added text</u>

Page 1:

Although often it is found that QSL candidate materials order at some finite temperature $T_N$, their dynamic response is strongly influenced by the proximity to a quantum critical point where the order completely disappears. The quantum critical regime, emanating from the quantum critical point, leaves the characteristic fingerprint in the dynamic response even at finite temperatures, allowing to test the theoretical predictions of the particular QSL in real materials [19, 20].

<u>Replaced text</u>

Page 3:

The calculated magnetic structure for $L = 3n$, shown in Figure 1d, comprises several **q** vectors and results in a rather complicated pattern, devoid of any particular spin textures. In Figure 1d, we show a view along the (111) plane for a $L = 6$ system, where black arrows on each node represent the direction of the net magnetic moment within each unit cell. The angles between neighbouring cells are found to be close to 120°.

The calculated magnetic structure for $L = 3n$, shown in Figure 1d, comprises propagation vectors $q_1 = (1/3, 0, 0)$, $q_2 = (1/3, 1/3, 0)$ and $q_3 = (1/3, 1/3, 1/3)$. The resulting pattern is devoid of any particular spin textures like skyrmions and merons. In Figure 1d, we show a view along the (111) plane for a $L = 6$ system, where black arrows on each node represent the direction of the net magnetic moment within each unit cell. The angles between neighbouring cells are found to be close to 120°. We do not expect a significant change in the calculated structure if the exchange parameters are slightly modified, as long as the tripling of the unit cell is preserved.

Page 3:

Energy-momentum plots along three principal directions show streaks of intensity without any apparent **q**-dependence, indicating a non-dispersive type of excitations.

Energy-momentum plots along three principal directions show vertical streaks of intensity, four in each panel, indicating a non-dispersive type of excitations.

Page 3:

In the top row of Figure 3 we show $S_{\text{exp}}(\mathbf{q})$ along three different planes in reciprocal space, obtained by integrating the intensity in the energy range $\omega_1 \sim E_{\text{min}} = 0.5\,\text{meV}$ and $\omega_2 \sim E_{\text{max}} = 1\,\text{meV}$. The patterns depicted carry fingerprints from the underlying magnetic lattice composed of two interconnected trillium lattices, with a hexagonal galaxy-like motif reflecting the chirality of the crystal structure (see panel (c)), arising from the non-centrosymmetric space group.

In the top row of Figure 3 we show $S_{\text{exp}}(\mathbf{q})$ along three different planes in reciprocal space, obtained by integrating the intensity in the energy range $\omega_1 \sim E_{\text{min}} = 0.5\,\text{meV}$ and $\omega_2 \sim E_{\text{max}} = 1\,\text{meV}$. The lower limit is given by the extent of the incoherent scattering while the upper limit is found as an optimal value to maximize the signal-to-noise ratio. The patterns depicted carry fingerprints from the underlying magnetic lattice composed of two interconnected trillium lattices, with a hexagonal galaxy-like motif reflecting the chirality of the crystal structure (see panel (c)), arising from the non-centrosymmetric space group.

Page 3:

Within PFFRG, the renormalization-group flow breaks at $\Lambda = 0.582\ J_4$, indicating the presence of a magnetically ordered ground state. Static spin correlations are measured at this point and, ...

Within PFFRG, the renormalization-group flow breaks at $\Lambda = 0.582\ J_4$, indicating the presence of a magnetically ordered ground state. As is standard for the PFFRG calculations (see SI), the static spin correlations are measured slightly above the flow breakdown and, ...

Page 4:

Thus, in an attempt to simulate the experimental data within a classical approach, we perform cMC calculations above the finite-temperature phase transition. Particularly, for T = 0.35 J4 , we find an excellent agreement with the PFFRG calculations (see left and right panels separated by black lines in panels (d)-(f) of Figure 3).

Thus, in an attempt to simulate the experimental data within a classical approach, we perform cMC calculations above the finite-temperature phase transition. We find an excellent agreement with the PFFRG calculations for a large range of temperatures and, particularly for T = 0.35 J4, we find the best quantitative agreement (see left and right panels separated by black lines in panels (d)-(f) of Figure 3).

Page 5:

... and extends up to the sole trillium lattice limit $J_3 = J_4 = 0$.

... and extends up to larger $J_5/J_4$ below the sole trillium lattice limit $J_3 = J_4 = 0$.

Page 5:

The set of parameters calculated with DFT for $K_2Ni_2(SO_4)_3$, indicated by a green star in Figure 4a, lies close to these paramagnetic regions and many of the features observed in the spin structure factors can be traced back to the tetra-trillium lattice limit.

The set of parameters calculated with DFT for $K_2Ni_2(SO_4)_3$, indicated by a green star in Figure 4a, lies close to these purely spin-liquid regions. This becomes evident when the PFFRG spin structure factor (at finite $\Lambda$) is compared to the results obtained in the tetra-trillium lattice limit (at the smallest $\Lambda$), as many of the features can be put in correspondence (see Figure 4b and Supplementary Information). Generally, the peaks become more smeared in the tetra-trillium limit, but there is no indication of a strong difference between the two phases. This provides evidence that the properties of $K_2Ni_2(SO_4)_3$ are governed by its proximity to the quantum spin liquid phase in the tetra-trillium lattice [19,20].

Page 5 (Figure 4a caption):

PFFRG paramagnetic region for $S = 1/2$ (red+blue) and $S = 1$ (red).

PFFRG correlated paramagnetic region, indicating no long-range order at $T = 0$, for $S = 1/2$ (red+blue) and $S = 1$ (red).

We have also removed 'PM' from the label of the figure, to avoid confusion.

Page 6:

...the calculations of 2e(T) for a single tetrahedron...

...the calculations of (twice) the energy as a function of the temperature, 2e(T), for a single tetrahedron...

Supplement, page 3:

Most importantly, PFFRG contains the correct large-N (spin flavour) and large-S (spin size) limits, making it ideal to study quantum systems. Higher values of $S > 1/2$ are obtained coupling copies of the $S = 1/2$ fermionic spins at each site [S6] (in the present case for $S = 1$ only two are needed).

Most importantly, PFFRG contains the correct large-$N$ and large-$S$ limits [S6,S7]. This means that the PFFRG equations can be generalized for a Heisenberg model with $SU(N)$ symmetry, where $N$ is the number of spin flavours, and they become exact on a mean-field level for $N \to \infty$. In this limit, spin systems usually do not develop magnetic long-range order, such that the PFFRG's ability to describe quantum paramagnetic phases is intrinsically guaranteed by its exactness at $N \to \infty$. On the other hand, the PFFRG equations can also be extended for arbitrary spin magnitudes $S$, and in the limit $S \to \infty$ they can be solved analytically to obtain exact classical mean-field results. Since in the classical large $S$ limit, spin models usually develop magnetic long-range order, the exactness in this limit ensures an accurate treatment of static magnetism. Hence, with PFFRG's ability to describe both types of spin phases, non-magnetic ones and magnetically long-range ordered ones,

it is an ideal approach for studying quantum spin systems. In practice, higher values of $S > 1/2$ are obtained coupling copies of the $S = 1/2$ fermionic spins at each site [S6]

Removed text

Page 3:

The magnetic structure factor $S(\mathbf{q})$ in semiclassically ordered states displays sharp and well-defined peaks, alongside magnonic excitations in the dynamic structure factor. At $E = 0$, the latter originate from the Goldstone modes that recover the broken symmetry of the magnetic phase. On the other hand, quantum spin liquids display more spread and diffuse features in the static part, where clear peaks are absent and the dynamic part is dominated by continua corresponding to fractional excitations [24,25]. In many cases, however, features from both limits such as magnon branches and fractionalization coexist [26-29]. It is worth emphasizing that fluctuations in spin liquids are far from being just incoherent and trivial disorder, and often generate specific patterns that can be associated with the characteristics of spin liquids in question, like pinch points observed in $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, and $Nd_2Zr_2O_7$ spin ices [4,30,31], or bow ties, half moons, and other theoretically predicted structures [32-34].

# Report on "Dynamics of K2Ni2(SO4)3 governed by proximity to a 3D spin liquid model"

March 18, 2024

Dear Editor,

First of all, I would like to thank the authors for this new version, as well as for the discussion and for the documented and clear response. This article has many merits, not least of which is the very detailed study of a compound based on an unconventional crystal structure. Again, I have no doubt that these results should be published in some form. The theoretical calculations seem to me to be of very good quality, and offer a very elegant example of what is now possible with numerical methods. I think our discussion has been very useful for me, and, I hope, may also help the authors to further improve their words. I fully agree with many of the points that are developed in the authors' response, however, I see that we don't understand each other. In what follows, I've selected a few points to continue the discussion and try to better explain my position.

What do I have at hand, as a referee? From an *experimental* point of view, the results are essentially shown in Figures 1 and 2. These are the elastic line at 100 mK in 1c and the inelastic scattering in Figure 2 taken at 2K, above $T_N$, but in the correlated region (I have no doubts about that). Is this data enough to call this material a QSL? I sincerely think not. It seems to me that any reader could call this "weak". The only other indication we have is the paragraph page 1 line 99: "Recently, it has been shown that a new 3D magnetic network exhibits a highly dynamical ground state. K2Ni2(SO4)3, a member of the langbeinite family, develops spin correlations below 20K between S = 1 moments, with a peculiar ordered state... "

As a result, authors will understand why I'm asking about the ordered phase, simply to obtain additional information. In this respect, the authors' response is very interesting, since it demonstrates, but with the help of data **not shown in the manuscript**, that the ordered phase is itself unusual. I quote their answer:

- "Going back to the actual behaviour of K2Ni2(SO4)3, we did perform some neutron scattering at very low temperatures (nominally 50mK), to try to see those spin waves, and, surprisingly, we saw the same diffuse type of correlations that we present in Figure 3 of our manuscript: This confirms that the ordered phase of K2Ni2(SO4)3 is a very peculiar, unconventional state, where one should be careful with relying too much on assumptions that have been developed in the context of more classical systems. It does, however, go along the same lines with results of only 1% of the total entropy being released at TN, and that SR experiments indicate a fully dynamic system (no wiggles). This opens up an interesting possibility that the putative long-range magnetic order is (barely) static

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on the time-scale of neutrons (1015 s) while being dynamic on the time-scale of muons (106 s). At the moment it is not clear how order and dynamics exactly coexist in this material and the analysis of the above results will be a part of a future study. In the current manuscript, we are focusing solely on establishing the spin-liquid properties of K2Ni2(SO4)3 and how this is related to the proximity of a pure spin-liquid region whose properties we tried to disseminate in Figure 4."

That changes the perspective and I thank very much the authors for this. But why don't such solid arguments appear in the body of the text?

- "The streaks are centred around 0.75 and 1.8 inverse units, while the ordering occurs at multiples of 1/3. Additionally, the observed streaks are very broad and observed both below the ordering temperature and fade in intensity very slowly toward high temperatures."

Again, this is a very interesting argument. But how could I have known? I'm sorry not to read it in the manuscript. Even the anomalous Q-position of the streaks is not mentioned in the main text. This is new, very interesting and convincing information that I hope will find its way into a revised version of the article.

- "This in no way implies that the INS is featureless, as observed from the QQ maps. For a boring paramagnet, the energy dependence would be both q- and E-independent.

I take a different view, based on my own experience of INS measurements: the signal just above $T_N$ is usually centered around the values of Q where magnetic order will appear, and often takes the form of a quasi-elastic spectrum in energy.

- The authors continue their reasoning with : "As elaborated above, the low-temperature data do not show any trace of spin waves, therefore, we cannot compare it to calculations. Furthermore, the Referees suggestion that calculating spin waves should be simple, is true for simple magnetic structures. However, in the current context where there exist a multitude of ordering vectors and a tripling of the unit cell (each magnetic unit cell has 3 3 3 8 = 216 spins), the comparison is likely to be far more complicated. More importantly, by construction, spin wave theory is applicable for small deviations from a classical order, however, here the ordered moment is tiny such that the deviation from classical order is huge, rendering it outside the scope of a reliable application of spin wave theory."

These are perfectly valid arguments. But they don't appear in the revised version, which confines itself to arguments that alone, in my opinion, are not sufficient.

- "What we find perplexing is the statement I have no doubt that the QSL region investigated from a theoretical point of view is correct.

There's no need to be puzzled, it was probably a clumsy way of paying a compliment.

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- "As Figure 3 in our manuscript demonstrates, there is a remarkable agreement between experiment and theory. With all the criticisms raised by the Referee related to (i) the system being a conventional paramagnet (which it is not as we have explained above),

  I agree that there is a very nice agreement. But I feel that the authors' response sheds much more light on the physics of this compound than the manuscript, by developing solid arguments, without which, however, I spontaneously express reservations. Again, I would encourage them to use these arguments in the main text.

  "(ii) raising doubts about whether INS is an appropriate technique to address QQ-maps (which it is designed to capture),

  I have with a different opinion : INS is also designed to address energy spectra.

  "and (iii) expressing concerns on the accuracy of DFT-obtained parameters, the Referee never provided a reasoning as to how could such a perfect qualitative and quantitative coincidence of different theoretical approaches with experiment appear.

  I may have misspoken. I readily acknowledge the considerable efforts made by the authors and the quality of the theoretical calculations compared with the experiments.

- What I'm essentially criticizing, and consider dangerous, is the idea that J1-J5 can be "measured" from S(q) maps, leaving aside the energy spectra (please define J1 and J2 not only in the Sup info). Indeed, the manuscript explains, **without any particular precautions**, that exchange parameters can be measured or validated by examining instantaneous correlation maps in reciprocal space, which, in my opinion, is misleading. The authors are well aware of this danger, and indeed prove it in their response: "We agree that a thorough comparison of the complete energy structure is more appropriate than a comparison of the Q structure since the latter is an integrated quantity of the former. However ... ". So why not explain this better in the text? The reservations and difficulties they express there take nothing away from the quality of the paper, and I find it more honest.

  In addition, I note that : *Sibille, R., Lhotel, E., Ciomaga Hatnean, M. et al. Coulomb spin liquid in anion-disordered pyrochlore Tb2Hf2O7. Nat. Commun. 8, 892 (2017)* and *T. Fennell, M. Kenzelmann, B. Roessli, M. K. Haas, and R. J. Cava, Power-Law Spin Correlations in the Pyrochlore Antiferromagnet Tb2Ti2O7, Phys. Rev. Lett. 109, 017201 (2012).* do not attempt to determine exchange integrals on the basis of S(q).

In conclusion :

a) **On the sole basis of the arguments presented in the main text and in Figure 2**, I don't see any experimental evidence likely to show anything abnormal, unexpected, spectacular or exciting in the dynamics. I maintain that a large number of ordered magnetic systems exhibit a spectrum of this type just above their ordering temperature, but this does not mean that they are QSL candidates. However, I agree

that the new arguments developed in the response letter (see above) are quite convincing and should, in my opinion, be integrated into the text. Please include the discussion about the absence of spin wave in the ordered phase, the anomalous Q position of the streaks, ...

b) As is well known, S(q) is a lattice Fourier transform, weighted by equal time correlations on different bonds. Any information about the energy parameters (in K or meV) has vanished. This is, in essence, very different from a direct comparison between calculated /measured spin wave spectrum. To me, this would be a proper validation of the proposed parameters, as the structure in energy of the dispersion is a direct measure of those parameters. The authors have explained in their response that this is unfortunately not possible. It would be nice to read this discussion in a revised version.

With these modifications, I would definitely recommend publication of the manuscript.

Reviewer #2 (Remarks to the Author):

The manuscript explores Quantum Spin Liquids (QSL) within the paramagnetic phases of ordered systems, a topic often met with skepticism within the community. Both Reviewer1 and I share reservations from this perspective. It does not hold back the authors from publishing this manuscript, but certainly represents a loud voice in the community, which should be appreciated. I do not recommend Nature Communications publish this manuscript in its current form. The authors failed to clarify the distinctions between QSL, classical spin liquids, and what experimentalists have traditionally referred to as quantum spin liquids. The conflation of these concepts could potentially mislead the community.

(No. 2.2) "As previously elaborated in (No 1.2), any magnetic system that shows correlations (i.e., it is not a conventional paramagnet), while at the same time is not ordered due to geometrical frustration, is considered a spin-liquid." The authors' characterization of spin liquids aligns somewhat with the pre-2010 understanding of quantum spin liquids, as in examples like $ZnCr_2O_4$ and $NiGa_2S_4$, where systems exhibit correlations without ordering due to geometrical frustration and/or competing interactions. However, it is not suitable to include $K_2Ni_2(SO_4)_3$ in the discussion, as it does magnetically order.

Furthermore, the manuscript extensively discusses Quantum Spin Liquids (QSLs), a state characterized by long-range entanglement and cannot be transformed into a product state through short-range unitary transformations. While numerical methods like quantum Monte Carlo (MC) and Density Matrix Renormalization Group (DMRG) have identified regions, such as $J_1 - J_2$ models on triangular lattices, where QSLs may occur, the authors' reliance on FRG poses limitations. FRG lacks the capability to provide detailed insights into wave functions, making it unrealistic to identify the disordered phase as a QSL. These regions could also represent classical spin liquids, which are characterized by product states, as in examples like the Heisenberg model on a pyrochlore lattice and spin ice.

The authors demonstrate a comparison between excitations in the paramagnetic phase and simulations from FRG and classical Monte Carlo (CMC) methods. However, it raises questions about how CMC simulates static or dynamic spin correlations within a QSL. A good alignment between the data and CMC simulations may suggest that the system is closer to a classical spin liquid, not a QSL.

While the authors' experimental efforts are commendable, and their data and simulations are on the threshold for publication in journals like Nature Communications, the manuscript suffers from poor clarity and potential misinterpretation. Therefore, significant revisions are necessary to address these concerns before considering publication in any journal.

Reviewer #3 (Remarks to the Author):

Dear Editor,

After revisions, I continue to believe that the manuscript "Dynamics of K2Ni2(SO4)3 governed by proximity to a 3D spin liquid model" by Gonzalez et al. is suitable for eventual publication in Nature Communications. I also appreciate that the authors made a number of changes I suggested, improving the clarity of figures and of the discussion.

However, there are two key issues that need to be addressed:

- The authors need to clarify how they identify a phase as being a quantum spin liquid or not

- More generally, to answer the questions that I and the other referees asked, almost 20 pages of explanation were required, yet the changes to the text were minimal in comparison. If so much explanation was needed to answer questions raised by referees who have read the whole paper carefully, shouldn't a significant portion of the new explanatory material also be available to the broader audience of future readers of the paper? The author must make the paper stand on its own by incorporating some of the arguments they have made in their response letter.

I detail both issues below, and I also suggest some more minor revisions

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The key piece that the authors have not addressed in the revision is the question of how they identify states as belonging to a quantum spin liquid phase. I did not previously raise any direct objection about the authors' use of the term "spin liquid" since I think that there is a strong argument to be made that the presented data do support the classification as a spin liquid. However, my confidence that the authors have thought about this carefully actually decreased rather than increased after reading their response letter.


The main point of the other two referees that I feel the authors have missed is that a quantum spin liquid (QSL) is *not* merely a disordered low-temperature quantum state, but that from a modern perspective spin liquids have positive signatures such as fractionalized quasiparticles, topological ground state degeneracy, and/or gapless Dirac cones. The authors try to dismiss this suggestion using a quote from Savary and Balents, but that quote means precisely the opposite of what the authors claim; Savary and Balents go on to say "Here, we will take the point of view that the essential ingredient of a QSL is not the lack of order but the presence of an anomalously high degree of entanglement, or massive quantum superposition." Thus it is a valid question whether the phase to which the model on the tetra-trillium lattice belongs is indeed a quantum spin liquid, and the authors need to address this question rather than dismissing it.


Fortunately, I think that the authors' data does possibly support this conclusion, although my confidence has decreased since my previous review. After seeing the authors' response, I am convinced that in the paper the authors need to add a very explicit argument of how/why their data indicate a spin liquid.


To me, a possible argument that the Heisenberg model on the pure tetra-trillium lattice has a spin liquid ground state could have the following key pieces: (1) The authors rigorously establish (I haven't checked the precise spin flip procedure they describe, but the argument is sound) that the ground state of the classical Heisenberg model on this lattice is a spin ice with macroscopic ground state degeneracy. [Note: the authors do not use the term spin ice, which I think would be the appropriate term for the classical ground state they describe.] (2) They show that the quantum ground state is also disordered, since there is no singularity in the RG flow in PFFRG down to Lamba -> 0. (3) They show that the spin

structure factors at Lambda -> 0 for PFFRG and at T -> 0 for cMC are different, suggesting that the quantum ground state is different from the classical one, so the ground state is distinct from the classical spin ice. In other words, (1) – (3) suggest that the quantum ground state is disordered, comes from adding quantum fluctuations to a classical spin ice, and is distinct from the classical ground state; taken together, these point towards the ground state being a QSL.

However, (3) is a possible sticking point here, where the argument might fall apart. In my previous report, I asked the following question:

"It appears to me that the main purpose of the cMC calculations is to demonstrate the quantum-to-classical correspondence, ie that the fluctuations in the quantum ground state either are or are not classical in nature. In other words, for a given set of couplings Ji, the fluctuations in the ground state either can be reproduced by cMC, in which case the state is "less quantum," or cannot be reproduced, in which case the state is "more quantum." In other words, if cMC cannot reproduce the PFFRG results, as in Fig. 4b, is that an indication of a spin liquid?"

The authors replied that because the quantum ground state will always feature some fluctuations beyond the T=0 classical state, the mismatch between structure factors from PFFRG and cMC is actually *not* an indication of a quantum spin liquid. In that case, if the authors want to maintain their strong claim that the tetra-trillium ground state is a QSL, how do they know it is distinct from a classical spin ice? As an analogy, consider the ground state of the Heisenberg model on the square lattice. The ground state of the classical model is the Neel antiferromagnet; the quantum ground state is the same but with a reduced magnetic moment. I believe this quantum state belongs to the same phase as the classical one (ie they are adiabatically connected by tuning the XXZ limit from the Heisenberg limit to the Ising limit). In that sense, how do the authors know that the ground state of the quantum model on the tetra-trillium lattice is indeed a QSL and not simply a spin ice with reduced moments from quantum fluctuations?

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For the second point, the authors have explained a number of key ideas in their response but not in the paper:

- Why is it reasonable to look at T=2K data? The argument is basically that if there were a quantum spin liquid at T=0 (even if nearby in parameter space and not precisely for the model describing the true material), a disordered state at low-but-finite T would be closely related. This is not stated clearly enough in the paper.

- The idea that the key contribution from cMC is to establish the magnetic unit cell, which is apparently not feasible to do in PFFRG, is currently not clear from the manuscript alone

- Add something from the answer to (No 3.7) to the paper about the purpose of Fig 4c [since the figure doesn't have direct relevance for K2Ni2(SO4)3]

- I agree with the authors' response in (No 1.11), however this is an example of the kind of explanation that needs to be also added to the paper. If a referee has this question, so will readers.


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Additional minor suggestions:

- In the paragraph beginning on line 212, the authors need to add a very basic explanatory statement at the start of the paragraph just stating that PFFRG is an RG method controlled by a flow parameter Lambda that represents temperature, before they start giving values of Lambda. This sentence already basically exists later in the paragraph, lines 222-225. So really the paragraph just needs to be rearranged for clarity.

- Lines 351-355: it should be made clear that this statement is about the classical model, not the quantum one

- (No 3.15): I appreciate the additional information added here, the picture is much clearer to me. However, I still don't understand the relevance of the types of states PFFRG is capable of representing in the large-N and large-S limits, since my understanding is that the authors do not use either of these limits in their calculations. Why, for example, does the fact that large-S tends to favor magnetic order tell us anything about the validity of PFFRG for S=1/2? Some more explanation is needed for this point (and in the Supplement itself, not just in the response letter!)

Dear Editor,

We thank the reviewers for their critical reading of the manuscript and comments. Below, we provide a detailed response to their concerns/questions, accompanied with changes to the manuscript. We emphasize that these are not the only changes we have made, and therefore we provide also a document where differences are explicitly highlighted.

**REPLY TO REFEREES**

The text written by Referees is coloured blue and the changes and additions to the manuscript are coloured green, while all line numbering is done concerning the stricken-through version.

**Reviewer 1**

First of all, I would like to thank the authors for this new version, as well as for the discussion and for the documented and clear response. This article has many merits, not least of which is the very detailed study of a compound based on an unconventional crystal structure. Again, I have no doubt that these results should be published in some form. The theoretical calculations seem to me to be of very good quality, and offer a very elegant example of what is now possible with numerical methods. I think our discussion has been very useful for me, and, I hope, may also help the authors to further improve their words. I fully agree with many of the points that are developed in the authors' response, however, I see that we don't understand each other. In what follows, I've selected a few points to continue the discussion and try to better explain my position.

We thank the Referee for noting the merits of our work and appreciation of our response to the many points discussed therein. We also thank the Referee for fully agreeing with many of the points in our response and are sorry that we did not understand each other better from the start. All the discussions we had so far have improved our understanding of the issues that a general reader might encounter while reading our article. Therefore, we have now poured the results of our discussions into the manuscript, resulting in a new improved version, hoping that the Referee finds it suitable for publication in Nature Communications.

What do I have at hand, as a referee? From an experimental point of view, the results are essentially shown in Figures 1 and 2. These are the elastic line at 100 mK in 1c and the inelastic scattering in Figure 2 taken at 2K, above $T_N$, but in the correlated region (I have no doubts about that). Is this data enough to call this material a QSL? I sincerely think not. It seems to me that any reader could call this "weak". The only other indication we have is the paragraph page 1 line 99: "*Recently, it has been shown that a new 3D magnetic network exhibits a highly dynamical ground state. K2Ni2(SO4)3, a member of the langbeinite family, develops spin correlations below 20K between S = 1 moments, with a peculiar ordered state...* "

As a result, authors will understand why I'm asking about the ordered phase, simply to obtain additional information. In this respect, the authors' response is very interesting, since it demonstrates, but with the help of data not shown in the manuscript, that the ordered phase is itself unusual. I quote their answer:

"*Going back to the actual behaviour of K2Ni2(SO4)3, we did perform some neutron scattering at very low temperatures (nominally 50mK), to try to see those spin waves, and, surprisingly, we saw the same diffuse type of correlations that we present in Figure 3 of our manuscript: This confirms that the ordered phase of K2Ni2(SO4)3 is a very peculiar, unconventional state, where one should be careful with relying too much on assumptions that have been developed in the context of more classical systems. It does, however, go along the same lines with results of only 1% of the total entropy being released at TN, and that SR experiments indicate a fully dynamic system (no wiggles). This opens up an interesting possibility that the putative long-range magnetic order is (barely) static on the time-scale of neutrons (1015 s) while being dynamic on the time-scale of muons (106 s). At the moment it is not clear how order and dynamics exactly coexist in this material and the analysis of the above results will be a part of a future study. In the current manuscript, we are focusing solely on establishing the spin-liquid properties of K2Ni2(SO4)3 and how this is related to the proximity of a pure spin-liquid region whose properties we tried to disseminate in Figure 4.*"

(No1.1) We thank the Referee for acknowledging the nuances of associating the material behavior below 2K with magnetic order since it is devoid of conventionally expected signatures such as presence of spin waves, wiggles in muons and subtleties of similar ilk. In order not to give an impression that we are claiming $K_2Ni_2(SO_4)_3$ to be a realization of QSL behavior in the sense of remaining liquid-like to the lowest temperatures, we have addressed the issue at the following two instances in the manuscript. In the second version of the manuscript there are two instances where 'QSL' has been related to $K_2Ni_2(SO_4)_3$. One is in the abstract:

> *Although magnetically ordered, $K_2Ni_2(SO_4)_3$ has been found to exhibit a highly dynamical and correlated state which can be driven into a pure quantum spin liquid under magnetic fields of only $B \simeq 4$ T.*

This is a reference to the previously published results. The other one is in the first sentence of the last paragraph:

> *The evidence for QSL behaviour in $K_2Ni_2(SO_4)_3$ opens a window of possibilities in search of exotic quantum phases born out of complex 3D lattice geometries beyond the iconic pyrochlore and hyperkagome lattices.*

Admittedly, the second example could be understood in a wrong way, that the QSL state is present at all (low) temperatures in $K_2Ni_2(SO_4)_3$. However, that was not our intention. What we meant is the dynamic aspects seen in $K_2Ni_2(SO_4)_3$ *above* the ordering temperature are linked (have their origins) to the 'island of liquidity' that we unveil around the tetra-trillium lattice limit. Therefore, we have changed the sentence to clarify and accurately convey this message (line 611):

> The evidence of strong dynamics seen in $K_2Ni_2(SO_4)_3$ opens a window of possibilities in search of exotic quantum phases born out of complex 3D lattice geometries beyond the iconic pyrochlore and hyperkagome lattices.

Indeed, the dynamic response we observe above the ordering temperature is undoubtedly related to the spin-liquid model we have demonstrated to arise around the tetra-trillium lattice limit. A very similar discussion can be found in a recent work on a triangular lattice material (Nat. Phys. **20**, 74 (2024)). It is important to note that their approach breaks down at the ordering temperature, clearly demonstrating that the order is detrimental for spin-liquid characteristics seen in materials. Therefore, it makes all the sense to study and discuss the spin-liquid properties at 2K, *above* the ordering tendencies seen in $K_2Ni_2(SO_4)_3$.

The fact that $K_2Ni_2(SO_4)_3$ exhibits a very dynamic ordered state (as far as the inelastic neutron scattering is concerned, but also no wiggles in $\mu$SR) requires additional experiments to clarify. The spectrum is NOT identical to that at 2K and how much of the difference comes from the effects of this peculiar order is a non-trivial problem. In this respect, $K_2Ni_2(SO_4)_3$ seems to be in an uncharted territory and more than one study is needed to try to answer many questions arising from the peculiar interplay between static and dynamic aspects below the ordering temperature. However, we stress again that unknowns about the ordered state do not invalidate the discussion about the correlations above it and how those correlations are related to the tetra-trillium lattice. We do not call $K_2Ni_2(SO_4)_3$ a QSL but the correlations at 2K are related to the tetra-trillium spin-liquid behavior, in the same way that Nat. Phys. **20**, 74 (2024) discusses the correlations seen in $KYbSe_2$ in connection to a proximate triangular lattice spin-liquid.

We have tried to include a mention of these peculiarities in the manuscript. Our opinion is that a lengthy discussion would not add much to the current manuscript, and could rather prove distracting. Additionally, we decided to include a sketch of the quantum criticality in Figure 1f, which should make clear how the correlations above the ordering temperature are connected with a pure spin-liquid behaviour.

The changes introduced can be found in lines (218-242):

We should point out that the agreement between cMC and the weak magnetic Bragg scattering serves only to narrow down the appropriate set of $J$s. The ordered state in $K_2Ni_2(SO_4)_3$ remains highly dynamic down to the lowest temperatures [?, ?] and exactly how the weak static component emerges from a highly dynamic background remains to be investigated in future studies. A similarly strong dynamic ground state, with faint features of spin-glass order, has been observed in $Tb_2Hf_2O_7$ [?], with spin-liquid-like dynamics observed to the lowest temperatures. In what follows, we focus on temperatures above the ordering ($T = 2\,K$) but significantly below the characteristic energy of the system ($\theta_{CW} = 18\,K$). In this case, the dynamic features above the ordering trace their origin to a region proximate in parameter space that exhibits purely spin-liquid characteristics and shows no magnetic order down to $T = 0$, as illustrated in Figure ??f. Although the dynamics observed below the ordering temperature in $K_2Ni_2(SO_4)_3$ could potentially be also dominated by the spin-liquid-like features, as found for $Tb_2Hf_2O_7$, it is also important to realize that entering the ordered state can significantly redistribute the scattering spectral weight, and often completely remove the spin-liquid features, as in the recently studied case of $KYbSe_2$ [?].

and lines (272-286):

We note that a comparison on the level of $S(\mathbf{q}, \omega)$ would be far from trivial, even within the ordered state due to multiple ordering vectors and 216 spins per magnetic unit cell. Additionally, spin-wave theory is by construction applicable for small deviations from a classical order. In the case of $K_2Ni_2(SO_4)_3$ the deviations from the classical order cannot be taken as small perturbations, rendering it practically outside of the scope of a reliable application of a spin-wave theory. Finally, the ordered state of $K_2Ni_2(SO_4)_3$ proves to be rather peculiar, marked by only 1% of entropy release and weak magnetic Bragg scattering [?]. Other characteristic features of classical order, like oscillations in $\mu$SR and magnon dispersions, are completely absent [?, ?].

*"The streaks are centred around 0.75 and 1.8 inverse units, while the ordering occurs at multiples of 1/3. Additionally, the observed streaks are very broad and observed both below the ordering temperature and fade in intensity very slowly toward high temperatures."*

Again, this is a very interesting argument. But how could I have known? I'm sorry not to read it in the manuscript. Even the anomalous Q-position of the streaks is not mentioned in the main text. This is new, very interesting and convincing information that I hope will find its way into a revised version of the article.

(No1.2) We appreciate the Referee for ascribing these features as novel, very interesting and more so convincing evidence for QSL behaviour. We have now added modifications to the revised manuscript (lines 254-260):

The streaks are centred around 0.75 and 1.8 inverse units, and are very broad, excluding the possibility of a quasi-elastic scattering from ordering that would appear in a much denser grid at multiples of 1/3 of the unit cell [?, ?].

*"This in no way implies that the INS is featureless, as observed from the QQ maps. For a boring paramagnet, the energy dependence would be both q- and E-independent."*

I take a different view, based on my own experience of INS measurements: the signal just above TN is usually centered around the values of Q where magnetic order will appear, and often takes the form of a quasi-elastic spectrum in energy.

(No1.3) We agree that in some systems, just above $T_N$ a quasi-elastic signal appears as a precursor of ordering. We emphasize that this is typically seen very close to $T_N$. Our 2K experiment is performed at $\sim$ 2x larger temperature than $T_N$. It is very hard to argue that such precursor features would be seen at such a significantly higher temperature than $T_N$. We again note that the dynamics above the ordering temperature in $K_2Ni_2(SO_4)_3$ are a reflection of a spin-liquid state at $T = 0$ found around the tetra-trillium lattice and not as a precursor to ordering. Ordering in $K_2Ni_2(SO_4)_3$ is

too weak and its fingerprints are overwhelmed by the spin-liquid-type correlations.

Modified text can be found in lines (249-250):

> In Figure **??**, we present experimental results for the dynamical structure factor $S_{\exp}(\mathbf{q}, \omega)$ with the incident energy $E_i = 2.8\,\mathrm{meV}$ obtained at $2\,\mathrm{K}$, significantly above the appearance of order to avoid possible quasi-elastic scattering but well within the dynamical state.

and (256-262):

> The streaks are centred around 0.75 and 1.8 inverse units, and are very broad, excluding the possibility of a quasi-elastic scattering from ordering that would appear in a much denser grid at multiples of 1/3 of the unit cell [**?**, **?**].

*The authors continue their reasoning with :* "*As elaborated above, the low-temperature data do not show any trace of spin waves, therefore, we cannot compare it to calculations. Furthermore, the Referees suggestion that calculating spin waves should be simple, is true for simple magnetic structures. However, in the current context where there exist a multitude of ordering vectors and a tripling of the unit cell (each magnetic unit cell has 3 3 3 8 = 216 spins), the comparison is likely to be far more complicated. More importantly, by construction, spin wave theory is applicable for small deviations from a classical order, however, here the ordered moment is tiny such that the deviation from classical order is huge, rendering it outside the scope of a reliable application of spin wave theory.*"

*These are perfectly valid arguments. But they don't appear in the revised version, which confines itself to arguments that alone, in my opinion, are not sufficient.*

(No1.4) We thank the Referee for suggesting we add these arguments to the manuscript. We have now done so in the lines (152-163):

> A more standard approach of determining the values of exchange interactions by measuring magnon dispersions within the ordered state or magnetic-field-polarized state is not applicable in the case of $K_2Ni_2(SO_4)_3$. The ordered state seems to remain predominantly dynamic, with the persistence of the diffuse scattering and absence of magnon dispersions [**?**, **?**]. On the other hand, the required polarizing magnetic field can be estimated to reach 25-30 T, therefore it is out of the reach of the current state-of-the-art facilities. Finally, the magnetic unit cell contains 216 spins, which represents a significant challenge for any spin-wave modelling approach.

and (272-286):

> We note that a comparison on the level of $S(\mathbf{q}, \omega)$ would be far from trivial, even within the ordered state due to multiple ordering vectors and 216 spins per magnetic unit cell. Additionally, spin-wave theory is by construction applicable for small deviations from a classical order. In the case of $K_2Ni_2(SO_4)_3$ the deviations from the classical order cannot be taken as small perturbations, rendering it practically outside of the scope of a reliable application of a spin-wave theory. Finally, the ordered state of $K_2Ni_2(SO_4)_3$ proves to be rather peculiar, marked by only 1% of entropy release and weak magnetic Bragg scattering [**?**]. Other characteristic features of classical order, like oscillations in $\mu$SR and magnon dispersions, are completely absent [**?**, **?**].

*"What we find perplexing is the statement I have no doubt that the QSL region investigated from a theoretical point of view is correct.*

*There's no need to be puzzled, it was probably a clumsy way of paying a compliment.*

(No1.5) We thank the Referee for this compliment on the accuracy of our theoretical analysis. May we take this chance to reiterate the message of our manuscript from a theoretical point of view.

The same theoretical approach that describes the spin-liquid behaviour around the tetra-trillium lattice Heisenberg antiferromagnet manages to describe the behaviour of a real material proximate in parameter space, to a rarely-seen level of agreement. Our claims about $K_2Ni_2(SO_4)_3$ do not go any further, we state that the dynamics of this material above the ordered state are governed by the proximity of the spin-liquid model, true to the title. We do not attempt to extend that claim to the lowest temperature, $K_2Ni_2(SO_4)_3$ is not a QSL (in the sense of remaining disordered to the lowest measurable temperatures), but its dynamics above the order (which is extremely feeble and unconventional in many aspects) is related to the proximate spin-liquid region. The main contribution is to address the source of these dynamics identifying the frustration mechanism at play, and we find it in a hitherto unreported geometry, namely a tetra-trillium lattice. In a similar spirit, almost 30 years ago, the pyrochlore lattice was suggested as a centre-stage in being a source of spin dynamics (at that time leading to the spin-ice physics), and as such it is still studied today, with all the variations due to the size of spins, various anisotropies, couplings, disorder, etc. We believe that with our contribution we are placing the tetra-trillium lattice in the focus of future studies in a similar vein as it shows classical and quantum spin liquid behaviour in the $S \to \infty$ and $S = 1/2, 1$ limits, respectively.

The ordering has its own quirks which will need to be addressed in the future but whatever happens there, it is not affecting any of our conclusions at 2K. An identical scenario occurs in $KYbSe_2$ [Nat. Phys. **20**, 74 (2024)] mentioned previously (and many other spin-liquid materials) which orders, and they still discuss the spin-liquid behaviour *above* its ordered state.

*"As Figure 3 in our manuscript demonstrates, there is a remarkable agreement between experiment and theory. With all the criticisms raised by the Referee related to (i) the system being a conventional paramagnet (which it is not as we have explained above),*

I agree that there is a very nice agreement. But I feel that the authors' response sheds much more light on the physics of this compound than the manuscript, by developing solid arguments, without which, however, I spontaneously express reservations. Again, I would encourage them to use these arguments in the main text.

(No1.6) We are happy that we have managed to substantially illuminate the physics of this compound in our response letter and that the Referee finds the arguments therein to be solid. We have now tried to implement all of these arguments in the manuscript, making it 'stand on its own', as one of the Referees pointed out.

*"(ii) raising doubts about whether INS is an appropriate technique to address QQ-maps (which it is designed to capture),*

I have with a different opinion : INS is also designed to address energy spectra.

(No1.7) We agree that INS is predominantly used to capture energy spectra. On the other hand, if there is no energy dependence of the observed excitations, as seen in Fig.2., then integrating out the energy dependence should not be considered as something out of the ordinary. In fact, it is the only case where such an integration is physically meaningful. There is no question that integrating out a full set of magnon dispersions to obtain an S(q) would make little sense. We repeat again that there are multiple articles on spin-liquid candidate materials, with energy-independent excitations, where S(q) is used to compare the predictions of a theoretical model to experimental results.

*"and (iii) expressing concerns on the accuracy of DFT-obtained parameters, the Referee never provided a reasoning as to how could such a perfect qualitative and quantitative coincidence of different theoretical approaches with experiment appear.*

I may have misspoken. I readily acknowledge the considerable efforts made by the authors and the quality of the theoretical calculations compared with the experiments.

(No1.8) We thank the Referee for clarifying his/her stance and appreciate his/her positive evaluation of the quality of the agreement between theory and experiment.

What I'm essentially criticizing, and consider dangerous, is the idea that J1-J5 can be "measured" from S(q) maps, leaving aside the energy spectra (please define J1 and J2 not only in the Sup info).

(No1.8) Respectfully, but we must express our surprise with this statement that somehow we are "measuring" $J_1$-$J_5$ from S($\mathbf{q}$) maps. The Referee placed the quotes there so it is not completely clear in which way this should be read. We would like to clarify that we do not measure $J_1$-$J_5$ from S($\mathbf{q}$) maps in any way. The values $J_1$-$J_5$ are determined solely by DFT calculations, which only take into account the types of atoms and their positions within the structural unit cell, and the Curie-Weiss temperature. We then compare the experimental results to simulations which have been performed using DFT-calculated $J_1$-$J_5$. There is an explicit statement in the manuscript for the classical approach ("We performed classical Monte Carlo (cMC) calculations using the new set of exchange interactions for several system sizes ...") and for PFFRG, although not explicitly stated, it is very much clear that we use the same approach. Additionally, in our first reply, we have argued in several places why a usual approach of extracting $J$s from dispersions, either in the ordered state or field-polarized state, does not work, and that we use a different approach, based on *calculating* $J$s and then obtaining the numerical results based on those $J$s. This approach cannot be interpreted as "measuring" $J$s from $S(q)$ maps.

The observed agreement presented in Fig.3 can only be obtained by using a relatively narrow set of $J_1$-$J_5$ values, which are calculated by DFT, not extracted from $qq$-maps in any way. If we use some other sets, as extensively presented in the supplement, the agreement deteriorates (again, we have argued the same in our first reply). So there is a certain level of confidence that those values of $J_1$-$J_5$ are rather well selected. Again, we do not 'fit' a model to experimental data to obtain $J_1$-$J_5$, we use the values obtained by DFT. This cannot in any way be interpreted as "measuring" them.

On the other hand, it is true that the agreement is 'only' established with the instantaneous correlators and not the full energy resolved spectra. But the experiment at 2K clearly shows the excitations do not have any energy dependence so what actually is there to compare in S(q,$\omega$), even if there would be a theoretical framework that could provide the full spectrum? Again, the Referee here is focused on securing that the set of exchange interactions is 'correct'. There is nothing wrong with that approach but one needs to look at a bigger picture. The main message of our manuscript is not to pinpoint the full set of $J$s to a never-before-seen level of accuracy. The phase diagram presented in Figure 4a clearly shows that $K_2Ni_2(SO_4)_3$ lies very close but outside of the full spin-liquid region. In the $J_3 - J_4 - J_5$ parameter space the 'closeness' is an arbitrary parameter, so if the green circle representing $K_2Ni_2(SO_4)_3$ is moved a bit along any axis, it will not change any of the conclusions of our manuscript. The set of $J$s that we use in our calculations shows an excellent agreement in both static (Figure 1d) and dynamic regions (Figure 3a-f), and all that is based on DFT calculations from structural data. This is a very, very rare example where practically from first principles one could calculate material's properties to this level. As we asked also in the first reply, why the insistence on a 'conventional' approach (which we argued multiple times is not applicable in the case of $K_2Ni_2(SO_4)_3$) when the agreement is already achieved?

Taking into account the Referee's criticism, in the revised manuscript we are more careful to mention that such an agreement with the instantaneous correlations lends some degree of confidence but is not a complete validation of the model Hamiltonian. We would also like to note that the discussion about the ordering characteristics presented in Fig.1 and the discussion about different sets of $J_1$-$J_5$ leading to different magnetic unit cell sizes is also there to provide arguments in favour of the currently used values of exchange interactions and thus to effectively rule out parameter sets which yield obvious inconsistencies.

We have modified the appropriate part of the text, see lines (377-393):

> A more thorough comparison would require a full theoretical description of $S(\mathbf{q}, \omega)$.
> However, given the highly frustrated 3D network, the large unit cell, and the spin value

In addition, I note that : Sibille, R., Lhotel, E., Ciomaga Hatnean, M. et al. Coulomb spin liquid in anion-disordered pyrochlore $Tb_2Hf_2O_7$. Nat. Commun. 8, 892 (2017) and T. Fennell, M. Kenzelmann, B. Roessli, M. K. Haas, and R. J. Cava, Power-Law Spin Correlations in the Pyrochlore Antiferromagnet Tb2Ti2O7, Phys. Rev. Lett. 109, 017201 (2012). do not attempt to determine exchange integrals on the basis of S(q).

(No1.9) Taking into account our response to the previous comment, we hope we made it clear that we are also NOT attempting to determine exchange integrals on the basis of $S(\mathbf{q})$. Additionally, there are numerous papers where a direct comparison of experimental and theoretical $S(\mathbf{q})$ has been presented, e.g., for $PbCuTe_2O_6$ in Nat. Commun. 11, 2348 (2020), for $Tb_2Ti_2O_7$ in Nat. Commun. 7, 11572 (2016).

Interestingly, both articles mentioned by the Referee use the comparison of $S(\mathbf{q})$ between the experiment and a model. For example by Sibille *et al.* on $Tb_2Hf_2O_7$:
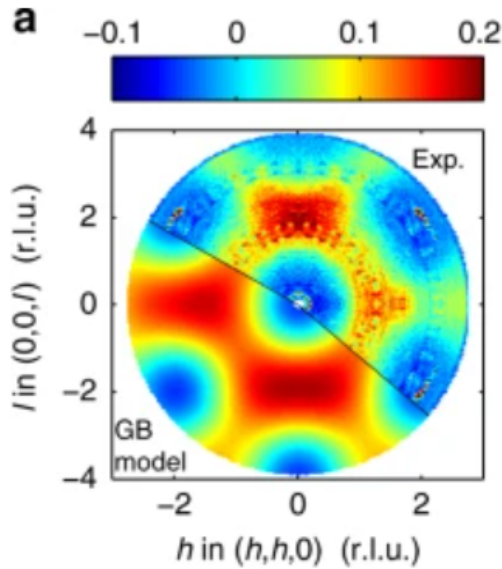


Moreover, the comparison is performed at 1.7K, where they state:

> *At the low temperature of this experiment (T=1.7K), the diffuse magnetic scattering typical of a **spin liquid** is observed, without any magnetic Bragg peaks that would indicate long-range ordered magnetic moments.*

Note that they discuss spin-liquid properties at 1.7K, while at the same time $Tb_2Hf_2O_7$ exhibits a spin-glass ordering around 0.7K:
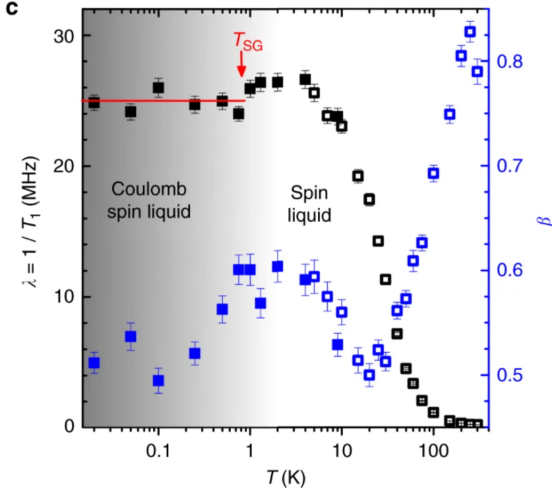
Additionally, note that even within the spin-glass phase the dynamics is actually that of a spin-liquid type, the moments are not completely frozen. This is very similar to what we know so far about the behavior of $K_2Ni_2(SO_4)_3$ below 1K.

We have iterated this line of reasoning also in our first reply, that to have such a level of agreement between experimental results and theory, both steps need to be correct (DFT calculations and numerical simulations). We are comparing those results with the experiment and based on the observed agreement (backed up by the results presented in Fig. 1 about the ordering tendencies) conclude that the values of exchange parameters cannot be too far off from what we are using. Additionally, the main message of our manuscript is not ONLY to determine the exchange parameters for $K_2Ni_2(SO_4)_3$, but to address why it is so frustrated. If the values of $J$s in $K_2Ni_2(SO_4)_3$ are found to be slightly different (we know they cannot be too different since the magnetic unit cell will not be $3\times$ larger than the crystallographic one) that would move the position of $K_2Ni_2(SO_4)_3$ in the phase diagram presented in Fig. 4 slightly but it will not change the main conclusion of our manuscript, that the geometrical frustration at the core of the dynamic behaviour in $K_2Ni_2(SO_4)_3$ has its origins in the proximate tetra-trillium lattice.

In conclusion :

a) On the sole basis of the arguments presented in the main text and in Figure 2, I don't see any experimental evidence likely to show anything abnormal, unexpected, spectacular or exciting in the dynamics. I maintain that a large number of ordered magnetic systems exhibit a spectrum of this type just above their ordering temperature, but this does not mean that they are QSL candidates. However, I agree that the new arguments developed in the response letter (see above) are quite convincing and should, in my opinion, be integrated into the text. Please include the discussion about the absence of spin wave in the ordered phase, the anomalous Q position of the streaks, ...

(No1.10) We thank the Referee for appreciating and highlighting the new arguments we made in our first reply as convincing evidence for unconventional dynamic behaviour governed by the proximate spin liquid in the tetra-trillium lattice. We have now modified the manuscript with arguments that have been presented in our reply.

b) As is well known, S(q) is a lattice Fourier transform, weighted by equal time correlations on different bonds. Any information about the energy parameters (in K or meV) has vanished. This is, in essence, very different from a direct comparison between calculated /measured spin wave spectrum. To me, this would be a proper validation of the proposed parameters, as the structure in energy of the dispersion is a direct measure of those parameters. The authors have explained in their response that this is unfortunately not possible. It would be nice to read this discussion in a revised version.

(No1.11) We would like to address specifically the statement that " Any information about the energy parameters (in K or meV) has vanished." Indeed, if we take a simple AFM dimer system

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and calculate its S(q), we will get the same S(q) no matter which $J$ is used. However, if we start adding multiple AFM dimers, with their own $J_1$, $J_2$, $J_3$,..., now the intensity pattern will reflect the relative ratio of various $J$s. So in principle, the above statement is not really (completely) correct. And Supplementary Figure 5 shows exactly that, the patterns are changing as we slightly change the set of parameters used for calculations.

We hope that we have convinced the Referee that the discussion about the exactness of the values of exchange parameters is not really central to our manuscript. Of course, there is no question that it would be better to have an experimentally confirmed (based on the full energy spectra comparison) approach validating those values. On the other hand, as we elaborated now multiple times, a large number of spins in a magnetic unit cell, coupled with $S = 1$ makes a prospect of a trustworthy spin-wave theory very speculative. Additionally, with 5 different exchange parameters, the error bars on the values determined by a 'usual' approach, comparing the full $S(\mathbf{q},\omega)$ determined by INS with some theoretical model (non-existant!), would be rather large and therefore not lead to a much better positioning of $K_2Ni_2(SO_4)_3$ within the phase diagram and how 'close' this material really is to the tetra-trillium limit.

With these modifications, I would definitely recommend publication of the manuscript.

We have now addressed each and every point of the Referee and incorporated all his/her suggestions in the revised manuscript. We are confident that the manuscript has now been greatly improved by our discussion with the Referee, and that she/he will recommend its publication in Nature Communications.

**Reviewer 2 (Remarks to the Author)**

The manuscript explores Quantum Spin Liquids (QSL) within the paramagnetic phases of ordered systems, a topic often met with skepticism within the community. Both Reviewer1 and I share reservations from this perspective. It does not hold back the authors from publishing this manuscript, but certainly represents a loud voice in the community, which should be appreciated. I do not recommend Nature Communications publish this manuscript in its current form. The authors failed to clarify the distinctions between QSL, classical spin liquids, and what experimentalists have traditionally referred to as quantum spin liquids. The conflation of these concepts could potentially mislead the community.

(No. 2.2) "*As previously elaborated in (No 1.2), any magnetic system that shows correlations (i.e., it is not a conventional paramagnet), while at the same time is not ordered due to geometrical frustration, is considered a spin-liquid.*" The authors' characterization of spin liquids aligns somewhat with the pre-2010 understanding of quantum spin liquids, as in examples like ZnCr2O4 and NiGa2S4, where systems exhibit correlations without ordering due to geometrical frustration and/or competing interactions. However, it is not suitable to include K2Ni2(SO4)3 in the discussion, as it does magnetically order.

(No2.1) Here we must again emphasize that we do not claim that $K_2Ni_2(SO_4)_3$ is a spin-liquid in the sense that it remains not ordered down to the lowest temperatures. As elaborated above in the answer to Referee 1, the spin-liquid-like dynamics seen *above* the ordering is related to the frustration-induced spin-liquid behaviour centred around the tetra-trillium lattice which does extend to the lowest (theoretical) temperatures. Our discussion about the results presented in Fig. 4 is about spin-liquid properties of the tetra-trillium lattice, not of $K_2Ni_2(SO_4)_3$. Again, our approach is nothing new, since many highly dynamic magnetic systems do order, but the proximity to a certain point in the parameter space makes their dynamics *above* the order related to a spin-liquid behaviour of that 'ideal' point. Both examples that we used extensively in our reply to Referee 1, $KYbSe_2$ and $Tb_2Hf_2O_7$, do eventually order but in both references (Nat. Phys. **20**, 74 (2024), Nat. Commun. 8, 892 (2017)) their dynamics above the order is described as spin-liquid or spin-liquid-like. Therefore, the Referee's implied objection that the language that we use to describe $K_2Ni_2(SO_4)_3$ is somehow out of the ordinary and that our contribution could 'mislead the community' is not correct. Our approach is not 'pre-2010' but very much aligned with current trends in the community.

We reiterate here what we also wrote in our first reply. The objection that a spin-liquid language is not suitable for systems that do show order is ignoring the fact that any material can only be cooled to a finite temperature. Therefore, there will never be a definite proof that a material shows no order because one cannot test it at low enough temperatures. Taking the Referee's objection into account, not a single material could be discussed in the context of spin-liquid. On the other hand, the community does discuss, extensively, spin-liquid behavior in various materials, whether they order or not, and we have presented more than one example ($KYbSe_2$, $Tb_2Hf_2O_7$). If they order, then the discussion is focused to the temperatures above the order, if the order happens at a significantly lower energy scale compared to those present in the system. All of these attributes are present also in $K_2Ni_2(SO_4)_3$ so we do not agree that we are in any way misleading the community.

Furthermore, the manuscript extensively discusses Quantum Spin Liquids (QSLs), a state characterized by long-range entanglement and cannot be transformed into a product state through short-range unitary transformations. While numerical methods like quantum Monte Carlo (MC) and Density Matrix Renormalization Group (DMRG) have identified regions, such as J1 – J2 models on triangular lattices, where QSLs may occur, the authors' reliance on FRG poses limitations. FRG lacks the capability to provide detailed insights into wave functions, making it unrealistic to identify the disordered phase as a QSL. These regions could also represent classical spin liquids, which are characterized by product states, as in examples like the Heisenberg model on a pyrochlore lattice and spin ice.

(No2.2) Here, the Referee wonders whether the system discussed in our manuscript could also be a classical spin liquid. We would first like to clarify that a classical spin liquid is not a product state but a classical superposition of macroscopically many degenerate classical ground states. Hence, in

contrast to a product state which decomposes into decoupled blocks, a classical spin liquid usually has long-range correlations (e.g. in classical spin ice they are of long-range dipolar nature).

A classical spin liquid in our $S = 1$ Heisenberg tetra-trillium model would be very unusual and quite exceptional. Classical spin liquids occur in systems with large spin length $S$ and/or classical Ising interactions, where frustration generates an extensive ground state degeneracy (i.e. where the number of degenerate ground states scales *exponentially* in the volume). Examples are classical spin ice materials with Ising interactions such as $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ where the spins are as large as $S = 15/2$ and $S = 8$, respectively. There, the strong classical nature prevents the systems from quantum tunnelling between the classically degenerate ground states and, hence, they remain *classical* spin liquids. In our model, we have demonstrated that we also have an extensive ground state degeneracy in the classical Ising limit of the tetra-trillium model. However, our system is rather far away from that limit since we have $S = 1$ and the interactions are of Heisenberg type. The latter means that the Hamiltonian consists of terms $S_i^z S_j^z + (S_i^+ S_j^- + S_i^- S_j^+)/2$ where the second term in brackets induces spin flips, hence tunneling between degenerate classical ground states. This tunneling is what drives a system away from a classical spin liquid and may induce a macroscopic *quantum* superposition of (formerly degenerate) classical states such as quantum spin liquid behavior. This is also the scenario in quantum spin ice materials where the spin is reduced compared to the aforementioned large value in spin ice materials (for example the promising quantum spin ice material $Ce_2Zr_2O_7$ has effective $J^z = \pm 3/2$ $z$-component degrees of freedom of the spins). The survival of a classical spin liquid in our system would mean that the reduced spin length in combination with the spin flip terms in the Hamiltonian still fail to induce quantum tunneling, which appears very unusual and counter-intuitive. If that was nevertheless the case, it would actually be very interesting on its own (implying 'avoided quantum tunneling', or 'emergent classical liquidity', maybe also worth a high-impact publication). However, the arguments above and the observation that our quantum PFFRG calculation at $T = 0$ agrees nicely with neutron experiments speak against a classical spin liquid. Furthermore, the system releases its full entropy without any residual entropy contribution which also speaks against a classical spin liquid and makes the quantum spin liquid scenario more plausible.

Of course, our experiments for $K_2Ni_2(SO_4)_3$ are performed at finite temperatures of $T = 2$ K and, in principle, that could re-introduce classical behavior. However, 2 K is almost an order of magnitude smaller than the Curie-Weiss temperature of 18 K which corresponds to the energy scale of the interactions and marks the onset of the quantum correlated regime at smaller temperatures. Hence, we do not expect that the finite temperatures in experiment change the reasoning above.

The Referee is right that PFFRG does not have access to actual wave functions. On the other hand, this approach is very powerful in calculating spin correlations and it's Fourier transform, which can be directly compared with the spin structure factor measured in neutron scattering experiments. Furthermore, there is practically no other quantum method which can deal with such complicated frustrated three-dimensional spin-1 models. In other words, our theoretical analysis is already based on state-of-the art numerical methods and we hardly see any possibility to go beyond that.

Based on the above arguments, we have substantially modified the manuscript surrounding Figure 4 and the follow-up discussion.

The authors demonstrate a comparison between excitations in the paramagnetic phase and simulations from FRG and classical Monte Carlo (CMC) methods. However, it raises questions about how CMC simulates static or dynamic spin correlations within a QSL. A good alignment between the data and CMC simulations may suggest that the system is closer to a classical spin liquid, not a QSL.

(No2.3) It is important to note that the Quantum to Classical correspondence only holds at the level of static susceptibilities and not for the full dynamical spectrum of the spin model. This correspondence has been established for three different two-dimensional spin-1/2 frustrated Heisenberg models which host a robust quantum spin liquid ground state, namely, the kagome lattice Heisenberg antiferromagnet, the square lattice $J_1$-$J_2$ model and the anisotropic triangular lattice antiferromagnet [see Phys. Rev. B 101, 035132 (2020)]. Thus, a good agreement between the data and CMC simulations of the static susceptibility does not, in any way, imply that the system is

closer to a classical spin liquid.

We have added a corresponding modification to lines (341-362):

This quantum to classical correspondence at the level of static correlators has been previously reported in several frustrated 2D and 3D spin-1/2 Heisenberg models at finite temperatures [?, ?, ?], some of which host a QSL ground state. However, it is important to note that this correspondence does not hold for the full dynamical spectrum of the models, which are different in quantum and classical systems. Furthermore, a thorough theoretical explanation of why the quantum-to-classical correspondence emerges only on the level of static correlations at finite temperature is still lacking, making it hard to extract conclusions from its appearance in this case.

While the authors' experimental efforts are commendable, and their data and simulations are on the threshold for publication in journals like Nature Communications, the manuscript suffers from poor clarity and potential misinterpretation. Therefore, significant revisions are necessary to address these concerns before considering publication in any journal.

We thank the Referee for recognizing our commendable efforts on the experimental side, and for acknowledging that our simulations are on the threshold for publication in Nature Communications. We have now, hopefully, addressed in detail the response to all the concerns raised by Referee 2. This led to extensive revisions in the manuscript and we hope the Referee will find the new version clear of potential misinterpretations and suitable for publication in Nature Communications.

**Reviewer 3 (Remarks to the Author)**

We thank the Referee for (still) considering our article suitable for (eventual) publication in Nature Communications, as well as for acknowledging the changes introduced to improve the clarity of the manuscript.

The Referee is right that our lengthy response in the previous round did not correspond with the necessary modifications to the manuscript. We amend this issue in the current version, hoping that now the manuscript stands on its own and becomes suitable for publication in Nature Communications.

We are sorry to hear that the Referee's confidence in our argument has decreased after reading our response letter. In what follows, we hope to restore his/her trust in our results.

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appropriate term for the classical ground state they describe.] (2) They show that the quantum ground state is also disordered, since there is no singularity in the RG flow in PFFRG down to Lamba -¿ 0. (3) They show that the spin structure factors at Lambda -¿ 0 for PFFRG and at T -¿ 0 for cMC are different, suggesting that the quantum ground state is different from the classical one, so the ground state is distinct from the classical spin ice. In other words, (1) – (3) suggest that the quantum ground state is disordered, comes from adding quantum fluctuations to a classical spin ice, and is distinct from the classical ground state; taken together, these point towards the ground state being a QSL.

(No3.1) The Referee's train of thought is correct, and we appreciate his/her clear layout of the justification. This argument goes in line with our reply to Referee 2's concern, which we have addressed both in response No2.2 and the manuscript. This led to significant changes in and around Figure 4, which we invite the Referee to read from line 461 to the end.

There are a couple of things that we want to mention to clarify our results. First, our arguments about the existence of a ground state and the possibility of making many more out of it by flipping spin clusters justify the existence of a spin liquid state in the Ising model on the tetra-trillium lattice (which could be called spin ice). These solutions, of course, are also ground states for the classical Heisenberg model. This, supported by the fact that we do not observe any phase transition down to $T = 0$, confirms the existence of a classical spin liquid in the classical Heisenberg model. In the quantum case, as stated by the Referee, PFFRG calculations show that the ground state is also disordered. In fact, what PFFRG evidences is that there is no symmetry breaking. Finally, as mentioned by the Referee, the spin structure factors of the classical and quantum cases are fundamentally different, showing that the ground states are not the same. This makes sense, as explained in the response to Referee 2, since the classical states are not eigenstates of the quantum Hamiltonian. Therefore, tunnelling between states is to be expected, leading to a quantum superposition of classical states: a quantum spin liquid. Not only we have added these arguments to the manuscript, but we have also modified Figure 4 by including the direct comparison between $T = 0$ classical and quantum spin structure factors in panels (c)-(e).

However, (3) is a possible sticking point here, where the argument might fall apart. In my previous report, I asked the following question:

"*It appears to me that the main purpose of the cMC calculations is to demonstrate the quantum-to-classical correspondence, ie that the fluctuations in the quantum ground state either are or are not classical in nature. In other words, for a given set of couplings Ji, the fluctuations in the ground state either can be reproduced by cMC, in which case the state is "less quantum," or cannot be reproduced, in which case the state is "more quantum." In other words, if cMC cannot reproduce the PFFRG results, as in Fig. 4b, is that an indication of a spin liquid?*"

The authors replied that because the quantum ground state will always feature some fluctuations beyond the T=0 classical state, the mismatch between structure factors from PFFRG and cMC is actually *not* an indication of a quantum spin liquid. In that case, if the authors want to maintain their strong claim that the tetra-trillium ground state is a QSL, how do they know it is distinct from a classical spin ice? As an analogy, consider the ground state of the Heisenberg model on the square lattice. The ground state of the classical model is the Neel antiferromagnet; the quantum ground state is the same but with a reduced magnetic moment. I believe this quantum state belongs to the same phase as the classical one (ie they are adiabatically connected by tuning the XXZ limit from the Heisenberg limit to the Ising limit). In that sense, how do the authors know that the ground state of the quantum model on the tetra-trillium lattice is indeed a QSL and not simply a spin ice with reduced moments from quantum fluctuations?

(No3.2) We apologize for the confusion that our previous response may have caused. What we wanted to state is the following: the fact that quantum-to-classical correspondence cannot be achieved for a generic system at $T = 0$ is not proof (alone) of a spin liquid ground state. A good example of this is the square lattice antiferromagnet mentioned by the Referee: at $T = 0$ the peaks will be in the same place for quantum and classical spin structure factors, but the heights will be different due to the reduced magnetic moment. Even though they are different, there is no quantum spin liquid. Our case is completely different since the ground states are disordered.

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Then, the Referee wonders whether our system could also be classical spin liquid. For this, we refer the Referee to our answers to No2.2 and No3.1.

Additionally, we would like to comment on the Referee's comparison with a classical Néel antiferromagnet. This is an interesting thought, however, we believe this comparison is misleading in our case. A classical Néel antiferromagnet is (up to states related by symmetry transformations) a single, non-degenerate state. The usual effect of quantum fluctuations acting on such a state is the formation of a quantum superposition of the perfect Néel state and some admixtures from energetically 'nearby' states. The term 'nearby' here refers to states where the perfect antiferromagnetic arrangement is interrupted by small ferromagnetic domains which are classically higher in energy. These admixtures reduce the effective magnetic moment. In contrast, in our case, before adding quantum fluctuations we have a *macroscopically degenerate* classical ground state manifold in the tetra-trillium model, in which case quantum fluctuations act differently. More specifically, the primary effect of quantum fluctuations is to create quantum superpositions (via tunnelling) between these extensively degenerate classical states. This macroscopic quantum superposition (which typically has no longer large degeneracies) may give rise to long-range entangled quantum spin liquid behaviour. On the other hand, the aforementioned admixtures from higher energy states are a secondary quantum effect: If one introduces quantum fluctuations perturbatively, e.g., via small transverse XY interactions on top of Ising interactions, these contributions are suppressed in a perturbative expansion. We hope this convinces the Referee that in our system quantum fluctuations act very differently than in classically ordered states, where small effects of quantum fluctuations just lead to some admixtures of mildly perturbed states but do not create macroscopic and long-range entangled superpositions.

For the second point, the authors have explained a number of key ideas in their response but not in the paper:

Why is it reasonable to look at T=2K data? The argument is basically that if there were a quantum spin liquid at T=0 (even if nearby in parameter space and not precisely for the model describing the true material), a disordered state at low-but-finite T would be closely related. This is not stated clearly enough in the paper.

(No3.3) The Referee is correct that this was not properly included in the manuscript. We have now introduced a new panel (f) in Figure 1, illustrating the key concept of proximity to a quantum spin liquid, with an accompanying text in lines (218-242):

> We should point out that the agreement between cMC and the weak magnetic Bragg scattering serves only to narrow down the appropriate set of $J$s. The ordered state in $K_2Ni_2(SO_4)_3$ remains highly dynamic down to the lowest temperatures [?, ?] and exactly how the weak static component emerges from a highly dynamic background remains to be investigated in future studies. A similarly strong dynamic ground state, with faint features of spin-glass order, has been observed in $Tb_2Hf_2O_7$ [?], with spin-liquid-like dynamics observed to the lowest temperatures. In what follows, we focus on temperatures above the ordering ($T = 2\,K$) but significantly below the characteristic energy of the system ($\theta_{CW} = 18\,K$). In this case, the dynamic features above the ordering trace their origin to a region proximate in parameter space that exhibits purely spin-liquid characteristics and shows no magnetic order down to $T = 0$, as illustrated in Figure ??f. Although the dynamics observed below the ordering temperature in $K_2Ni_2(SO_4)_3$ could potentially be also dominated by the spin-liquid-like features, as found for $Tb_2Hf_2O_7$, it is also important to realize that entering the ordered state can significantly redistribute the scattering spectral weight, and often completely remove the spin-liquid features, as in the recently studied case of $KYbSe_2$ [?].

Also, the changes in Figure 4 make it clearer that the spin structure factor of the real material can be traced to the quantum ground state of the tetra-trillium lattice.

The idea that the key contribution from cMC is to establish the magnetic unit cell, which is apparently not feasible to do in PFFRG, is currently not clear from the manuscript alone

(No3.4) We have now stated explicitly the importance of cMC in obtaining the comparison shown in Figure 1(c) and narrowing down the possible couplings $J$s by finding the right magnetic unit cell. The changes have been introduce in lines (164-208):

Before addressing the dynamic properties of the system, we focus briefly on the peculiarities of the magnetic order and how it changes depending on the different sets of exchange parameters $J_1$-$J_5$ proposed for $K_2Ni_2(SO_4)_3$. For this purpose, we perform cMC calculations since this method is best suited to detect classical magnetic orders in the absence of quantum fluctuations, allowing us to validate/invalidate different Hamiltonians by comparing them to neutron scattering measurements. All our classical calculations indicate a transition to a magnetically ordered phase at finite temperatures. However, the lowest ground-state energy is only reached for $L = 3n$ lattices (with critical temperature $T_c^{cMC}/J_4 = 0.048(2)$), while all other sizes ($L \neq 3n$) give higher energies at $T = 0$, implying that the magnetic order is frustrated by periodic boundary conditions. The magnetic orders obtained for $L = 12, 9, 6$, and 3 are all identical, indicating a tripling of the magnetic unit cell and a perfect agreement with the Bragg points observed experimentally (Figure ??d). Interestingly, if the DFT Hamiltonian corresponding to the room temperature structure is used, $L = 4n$ magnetic order has the lowest energy (only 0.09% below $L = 3n$), indicating a very sensitive landscape of complex magnetic configurations. Another recently suggested set of values with $J_3 = 0$ [?] leads to a quintupling of the magnetic unit cell, exhibiting the lowest ground-state energies for $L = 5n$ lattices. These calculations evidence not only that it is not trivial to capture the experimentally observed Bragg structure, but also give a certain level of confidence for the set of exchange interactions determined based on the structure at 100 K. The cMC magnetic structure for $L = 3n$, shown in Figure ??e, comprises propagation vectors $q_1 = (1/3, 0, 0)$, $q_2 = (1/3, 1/3, 0)$ and $q_3 = (1/3, 1/3, 1/3)$.

and lines (218-220):

We should point out that the agreement between cMC and the weak magnetic Bragg scattering serves only to narrow down the appropriate set of $J$s.

Add something from the answer to (No 3.7) to the paper about the purpose of Fig 4c (since the figure doesn't have direct relevance for K2Ni2(SO4)3)

(No3.5) We have added two modifications to clarify this: on one hand, we reduced the space occupied in Figure 4 to non-material related things by removing the classical energy calculations which are redundant once the specific heat is shown. This left more space to include the spin structure factor on all planes in the tetra-trillium lattice limit, which is more relevant to compare with the experiments and the results on the DFT Hamiltonian. On the other hand, we mentioned explicitly in the text what is the purpose of showing the specific heat, which illustrates the absence of a phase transition at finite temperatures and shows the reduced value of $c_v(T = 0)$. This resulted in a large section surrounding Figure 4 being changed.

I agree with the authors' response in (No 1.11), however this is an example of the kind of explanation that needs to be also added to the paper. If a referee has this question, so will readers.

(No3.6) The Referee is right and we have added response No 1.11 from the previous round in the main text in order to avoid further confusion, see lines (394-408):

Due to the complexity of the underlying 3D magnetic network, with the classical limits for $J_3, J_5 \rightarrow 0$ and $J_4 \rightarrow 0$, it is far from evident why it is so geometrically frustrated in the case of $K_2Ni_2(SO_4)_3$. Simpler networks with similar semi-classically ordered ground states, like the square or triangular lattices, are very susceptible to the introduction of additional frustrating couplings which eventually drive them to spin-liquid states ($J_2/J_1 \sim 0.5$ and $J_2/J_1 \sim 0.06$ for the square [?, ?, ?, ?, ?, ?] and triangular lattice [?, ?, ?, ?, ?, ?], respectively, even though other non-magnetic states have been proposed for the square case [?]). In the context of a large variety of different compounds with

Additional minor suggestions:

In the paragraph beginning on line 212, the authors need to add a very basic explanatory statement at the start of the paragraph just stating that PFFRG is an RG method controlled by a flow parameter Lambda that represents temperature, before they start giving values of Lambda. This sentence already basically exists later in the paragraph, lines 222-225. So really the paragraph just needs to be rearranged for clarity.

(No3.7) The Referee is right. We have now added a proper explanation of the parameter $\Lambda$ and the PFFRG method before plunging into the actual numbers. Lines (315-321) now read:

> Within PFFRG, the renormalization-group flow is controlled by the cutoff frequency $\Lambda$, which is lowered from the interactions-free limit $\Lambda = \infty$ where the solution is known (see SI for further details). In this case, the flow breaks at $\Lambda = 0.582 \, J_4$, indicating the presence of a magnetically ordered ground state.

Lines 351-355: it should be made clear that this statement is about the classical model, not the quantum one

(No3.8) We have now made it clearer that these arguments (and the whole paragraph) correspond to the classical model alone. This is part of all the modifications surrounding Figure 4. The relevant part is now on lines 461-527:

> In analogy to quantum spin ice, where a QSL arises out of a classical spin liquid (CSL) when adding quantum fluctuations, we address here the question of whether the classical tetra-trillium lattice also hosts a CSL which can potentially turn into a QSL when adding quantum fluctuations. The first indication is that cMC calculations show no ordering tendencies down to the lowest temperatures. However, a more conclusive proof of the CSL ground state in the classical limit can be constructed as follows. First, the Hamiltonian can be written as a disjoint sum of squared total spin in each tetrahedron (just as for the pyrochlore lattice). With this re-writing, the classical ground-state energy can be found exactly given that a solution with zero net magnetic moment in each tetrahedron exists. This solution cannot exist for $J_5/J_4 < 1/3$, providing a theoretical limit (which we confirmed with cMC calculations). Second, for the tetra-trillium lattice, we confirmed that a collinear solution exists (which is also a solution of the Ising model) and clusters of 18 spins can be systematically found and flipped while conserving the property of vanishing total spin in each tetrahedron (see SI). This allows the system to move through an extensive ground-state manifold and confirms the liquidity of this phase. Compared to the pyrochlore lattice, more spins are needed to construct a minimal flippable cluster because of the ramifications that occur at sites shared by three tetrahedra in the tetra-trillium lattice. By adding other couplings such as $J_1$, $J_2$ or $J_3$, perfect squares cannot be completed, and therefore the freedom to fluctuate is restricted. The cMC calculations show that even the smallest non-zero value of $J_3$ results in a finite critical temperature (however small). On the other hand, for $J_5 > J_4$, no ordering is observed at least up to $J_5/J_4 = 3.5$. The CSL in the tetra-trillium lattice exhibits a different spin structure factor than the quantum case, as shown in Figure ??a-c where we compare the PFFRG spin structure factors at $\Lambda = 0.01 \, J_4$ with the classical counterpart calculated from cMC at $T = 0.001 \, J_4$.

(No 3.15): I appreciate the additional information added here, the picture is much clearer to me. However, I still don't understand the relevance of the types of states PFFRG is capable of representing in the large-N and large-S limits, since my understanding is that the authors do not use either of these limits in their calculations. Why, for example, does the fact that large-S tends to favor magnetic order tell us anything about the validity of PFFRG for S=1/2? Some more explanation is needed for this point (and in the Supplement itself, not just in the response letter!)

We have now done so in the Supplemental Material, and thank the Referee for suggesting adding these clarifications.

We hope that all requests made by the Referee have now been properly addressed in the manuscript and that all doubts regarding our results and their relevance have been alleviated. With this, we hope that the Referee considers that our article is now ready for publication in Nature Communications.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

Dear Editor

I have carefully read the authors' reply, and I think they have responded to my remarks in an interesting manner and have then modified their manuscript accordingly. I regret, however, that the authors did not choose to rethink the structure of their manuscript, upon reading the referee reports. I think the reasoning would undoubtedly have benefited from such a reorganization. It is clear indeed that the referees have pointed out more or less the same things. Instead, we can see the trace of the discussions during the reading. Despite this, I do think that the overall quality of this work merits publication in Nat Comm.

yours sincerely

Reviewer #2 (Remarks to the Author):

The authors have addressed my concerns by demonstrating that they have explored all possible avenues. This is fine. Experimentally, driving the system to a well-ordered state for spin waves is unrealistic, as evidenced by the magnetization data presented in the first revision. Numerically, methods such as QMC or DMRG cannot be applied to this lattice at present.

Given that QSL likely does not exhibit definitive "smoking-gun" features and that methods like cMC and FRG cannot conclusively determine whether a state is a QSL, I, along with other referees, suggest further softening the QSL-related arguments. From my perspective, the merit of this work lies in the determination of the Hamiltonian using cMC and FRG, despite the experimental challenges.

It is encouraging to see that the authors have become more precise with terminology. (No. 2.2) they clarify that a CSL is not a product state but a classical superposition of many degenerate ground states. Indeed, while a CSL may be described as a product state, there are subtleties in the correlations. However, there exist a bunch of claims written in a haphazard manner including "... can be driven into a pure QSL under magnetic fields of only B~4 T." They are not relevant to the best part of this work and, in fact, undermine its strength.

Reviewer #3 (Remarks to the Author):

Dear Editor,

After the second round of revisions, I am happy to recommend publication of the manuscript "Dynamics of K2Ni2(SO 1 4)3 governed by proximity to a 3D spin liquid model" by Gonzalez et al. in Nature Communications.

The authors have addressed my key concerns to my satisfaction. Most importantly, they now address explicitly and at length the question of why one should believe the disordered phase on the tetra-trillium lattice is specifically a quantum spin liquid. The discussion of the nature of the classical spin liquid is clearer than before (and it was already relatively clear). Then the discussion of the S=1 quantum version is substantially improved, including a specific comment addressing my comparison in the previous report with the case of classical magnetic order being reduced but not fundamentally altered by quantum fluctuations. While the authors' current arguments do not definitively prove that the disordered state on the tetra-trillium lattice is a QSL, I find the arguments convincing, and I also believe that the arguments are now clear enough that readers will be able to make an

informed judgement for themselves on the basis of the data/evidence shown by the authors.

Beyond this, the authors have added explanations of various points from their previous reply letter to the manuscript, making it more complete and better able to stand on its own.

The authors have also addressed all of my more minor points to my satisfaction. I especially appreciate the further explanation of the importance of the large-S and large-N limits being captured by PFFRG.

I will point out one small change that I would like to recommend: in lines 522-527, the authors comment on the difference between structure factors for the classical and quantum calculations. This is an important comment and should be included in the paper, but it seems to be in the wrong place here - this is right in the middle of the discussion of the CSL, and it would be better for the comparison with the quantum case to come after the authors start discussing the quantum case (somewhere around 590). Maybe at 522 the authors can just say that the CSL structure factors are shown on the left half of 4c-e. (Note also that the sentence currently refers to 4a-c, which is incorrect.)

In this submission we address the final remarks from reviewers.

Reviewer 1 did not have a specific comment, but it was argued that the text could be better organized. We tried to compensate by improving the flow of the text with some small changes and phrasing.

Reviewer 2 asked to 'soften' further the statements about QSL. The statement mentioned explicitly ('can be driven into a pure QSL under magnetic fields of only $B \sim 4$ T') has been removed completely from the abstract due to the overall shortening of the abstract (150 words limit). At another place we have replaced the word 'observed' to 'potential', in line with Reviewer's suggestion.

> *An exciting task for future theoretical studies will be to identify possible structures of emergent gauge theories on the tetratrillium lattice that could underlie the potential QSL behaviour.*

Reviewer 3 suggested a reshuffling of a part of the text in the discussion, which we duly implemented.

Additionally, a new paper was submitted to arxiv which directly addresses the quantum-to-classical correspondence, and mentions our work on K2Ni2 compound. We have included the reference to the paper (ref37 in the new submission) with a short sentence (starting at line 167).

On behalf of all the authors, Ivica Zivkovic.

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