Peer Review File

Manuscript Title: Quantum spin nematic phase in a square-lattice iridate

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:
Referee #1 (Remarks to the Author):

The authors study the material Sr2IrO4 using a diverse set of experimental techniques to reveal a spin quadrupolar order intertwined with a Neel antiferromagnetic order. Moreover, it is shown that the critical temperature for the quadrupolar ordering is distinct and higher than the Neel temperature. This is done using X-ray diffraction, Raman spectroscopy, and magneto-optical Kerr measurements. This finding is of particular interest since the magnetic ion has a local spin S=1/2 moment, which a priori cannot have a quadrupole moment, as opposed to S=1 ions. Instead, the quadrupolar order seems to be a genuine many-body phenomenon and likely is due to strong electron correlations. I would thus qualify the authors' findings as quite groundbreaking. To the best of my knowledge, this would make Sr2IrO4 to be among the first compounds to show spin nematic order without having higher spin local moments.

The experimental data is presented well and it is quite easy to follow the arguments of the authors. I particularly, like the brief descriptions of the experiments in the main text, and the more elaborate coverage in the supplementary material. From what I can tell, the experimental data, in particular, the Raman spectroscopy data for showing the intensity peaks in the Eg, A1g, and B2g representations are very convincing. The plots are very tidy and bring across the key results in a convincing way.

I do have several comments, though, which require to be addressed.

1) One major critique I have is repeated, in my opinion unwarranted, mentioning of the resonating valence bond (RVB) picture by Anderson. The RVB state has unfortunately often been presented as a picture whenever "something weird beyond classical" is seen. Too often in literature, no precise statements are why any RVB picture would be needed to explain an observation, and I am afraid the authors do the same thing here. I think the author's finding of a spin nematic state is more than interesting enough, even without having a connection to the obscure RVB picture. I suggest the authors remove mention of the RVB picture completely since it does not contribute anything to their findings and it appears to have been added to "add some theoretical spice". However, if the authors are convinced otherwise, they should elaborate in more detail on how spin singlets would be resonating and how the observed experimental data can be explained by this. This should be thorough though. I would not accept adding one or two sentences. Instead, a full theory with predictions should be developed, which I assume would be a significant amount of work. The related statements about the RVB picture should not be published in a high-impact journal such as Nature. Hence, either they are completely removed or a substantial theory should be developed. As is, the discussion of RVB states is a bit unscientific.

2) Accordingly, I suggest removing the RVB picture from Fig 1.

3) Instead of relying on the RVB picture, there exist several computational works, which discuss the possibility of having spin nematic order in S=1/2 systems, in particular close to ferromagnetic phases. I suggest the authors give a more thorough account of the prior models which have been understood to show similar phenomenology by adding several references.

Since the finding is genuine, novel, and in my opinion rather groundbreaking, I do think that the manuscript should eventually be published in Nature. However, my concerns need to be properly addressed before I can finally commit to giving an according recommendation.

Referee #2 (Remarks to the Author):

A. Summary of the key results
In this manuscript the authors present an experimental study of the square-lattice iridate Sr2IrO4. Sr2IrO4 is the prototypical Spin-Orbital Mott Insulator, and one of the best studied model systems for spin-orbit-driven magnetism. The authors use a combination of resonant x-ray diffraction/circular dichroism, Raman spectroscopy, and resonant inelastic x-ray scattering to investigate this compound. These measurements are supported by detailed analysis and a simple two-site S=1/2 model calculation. The authors propose three key results:
(1) Sr2IrO4 displays spin nematic (i.e. magnetic quadrupolar) order.
(2) This magnetic quadrupolar order develops at higher temperatures (Tc ~263K) than canted antiferromagnetic order (Tn ~ 230K) as shown by Raman spectroscopy. It then persists into the antiferromagnetic state, as shown by resonant x-ray diffraction.
(3) There is a breakdown of coherent magnon excitations at the (Pi,0) position in reciprocal space, which can be attributed to RVB-like quantum entanglement in the canted antiferromagnetic state.

B. Originality and significance
Sr2IrO4 is a well-studied model system. It has previously been studied using all 3 of these techniques (RXD, Raman, and RIXS), but the authors present new and original measurements using these probes. In the case of RXD, there is a new focus on circular dichroism, which can be sensitive (albeit very subtly) to the presence of quadrupolar order. In the case of Raman, the authors employ a different geometry, illuminating the short/narrow side of a thin plate-like crystal. In the case of RIXS, the authors focus on polarization-dependence measurements at three high symmetry positions in reciprocal space.

Because of the level of interest in Sr2IrO4, proof of spin nematic/magnetic quadrupolar order would be a significant result. It is not entirely unprecedented, since several previous studies have reported "hidden order" above Tn in Sr2IrO4 (see Refs 51, 52, and 53). However, the authors claim to have observed a different symmetry breaking state (i.e. a different type of hidden order), which can be definitively attributed to spin nematic ordering.

C. Data and methodology

The authors present a significant amount of new experimental data. The data quality appears to be good, and the figures are well-presented. It is worth noting that the RXD measurements are quite challenging (i.e. to obtain good azimuthal dependence of magnetic peaks), and the CD effects are relatively weak. However, Fig. 2 is quite convincing for showing the CD effect. The authors also provide extensive supplementary information with this manuscript. In particular, the SI contains a more detailed description of the representational analysis (for RXD) and polarization dependence (for RIXS). The methodology also seems sound: RXD and Raman are both sensitive probes for detecting weak or subtle symmetry-breaking, and RIXS is well-suited to the study of magnetic excitations in Sr2IrO4.

D. Appropriate use of statistics and treatment of uncertainties

The authors include appropriate experimental uncertainties and error bars when discussing the RXD (Fig. 2), Raman (Fig. 3), and RIXS data (Fig. 4). The data analysis appears to be thorough and technically sound.

E. Conclusions

I do have some concerns regarding the interpretation of the data, and the robustness/reliability of the conclusions. My biggest concern is in Section IV, when the authors discuss the polarization-dependence of the RIXS spectra. In particular, the authors claim that there is no magnon mode at the (Pi,0) position in reciprocal space (Fig. 4(d)). The absence of the magnon mode is the basis of the claim that the quadrupolar order results from RVB-like quantum entanglement. My issue with this statement is that there is a clear magnon peak at ~200 meV. The full dispersion of this mode has previously been mapped out in Ref. 48, and the energy of the magnon is known to be ~200 meV at (Pi,0). While the polarization-dependence of this feature may not match the authors' expectations, the peak at 200 meV cannot simply be ignored. This would seem to undermine the only piece of evidence for key result #3 (breakdown of coherent magnon excitations at short-wavelength scales).

I am also concerned that the authors have not attempted to reconcile their "hidden order" state above Tn, with previous reports based on Second Harmonic Generation (Refs. 51 and 55), polarized neutron diffraction (Ref. 52), and torque magnetometry/elastoresistance (Ref. 53). In particular, the elastoresistance measurements in Ref. 53 seem to closely match the 263K temperature-scale identified in this study. The differences in reported symmetry-breaking also seem to merit further discussion (especially since the reported breaking of inversion-symmetry could affect the arguments used to interpret the RXD data). I was not convinced by the claim that these discrepancies could be explained by the existence of two different, unrelated hidden order states.

Lastly, one argument presented in favor of spin nematic order in this compound is that the SN state has been theoretically shown to emerge on a frustrated 2D square lattice with ferromagnetic (FM) nearest-neighbor (nn) couplings, and antiferromagnetic (AF) next-nearest-neighbor (nnn) coupling. It should be noted that this scenario does not apply to Sr2IrO4, which has strong AF nn coupling and weaker FM nnn coupling. Do the authors know of any theoretical support for SN order under the conditions relevant to this compound?

F. Suggested improvements

I would recommend that the authors attempt to reconcile the current results with previous reports of hidden order in Sr2IrO4. How can they conclusively demonstrate that this hidden order corresponds to spin nematic order and not anapole or loop-current order? Should signatures of spin nematic order have been observable in these previous studies? What can account for the discrepancies in reported time-reversal, rotational, and inversion symmetry breaking? To make a strong statement on the existence of spin nematic order, I think this issue needs to be addressed.

I would also recommend that the authors remove or heavily edit Section IV. The polarization-dependent RIXS data is fairly limited (only 3 Q-points) and I do not believe that it supports the claim that the coherent magnon mode breaks down at short-wavelength scales (Pi,0). This section is not essential to the case for spin nematic order, which is largely based on Sections I and III.

I believe it is also worth pointing out to the reader that the magnetic field used to control the ordered moment direction (B = 0.3 T) will also change the magnetically ordered structure below Tn (as originally reported in Ref. 8). This point is noted in the SI, but it is an important enough detail to justify mention in the main text.

G. References
The authors have provided an extensive list of references, and they appear to have cited all of the most relevant previous work. As noted above, my main concern is that the results from previous studies (i.e. experimental studies of hidden order, theoretical studies of the frustrated square lattice) are not fully integrated or reconciled with the current study.

H. Clarity and context
This manuscript is well-organized and reasonably well-written, with a clear abstract/summary. The main text is quite technical (particularly in Section I), which may pose a challenge for non-specialists. The discussion section is somewhat brief, and (as noted above) there are several important points which would be helpful to discuss in more detail.

Referee #3 (Remarks to the Author):

In their manuscript, Kim et al report the discovery of a quantum spin nematic (SN) phase in the Sr2IrO4 material.

Using resonant x-ray diffraction (RXRD), the authors detect a prominent circular dichroism (CD) in the (0 0 21) magnetic reflection and interpret this as evidence for a quadrupolar order interfering with the magnetic order.
Using temperature-dependent Raman spectroscopy, they show that the low-energy magnetic fluctuations appear well above the Neel temperature. Using a special experimental configuration, they also report a novel excitation emerging at Tc = 263 K that they assign to a collective mode associated with the breaking of rotational symmetry. They also show, using magneto-optical Kerr effect, that time-reversal symmetry is preserved for TN<T<Tc.

Using resonant inelastic x-ray scattering (RIXS) at the Ir L3-edge, they report a deviation from the linear spin wave theory of the pseudospin excitation at [pi,0], where the signal in the longitudinal and transverse channels becomes isotropic. They interpret this as evidence of quantum entanglement of near-neighbor pseudospins.
The experimental approach comprises different techniques and the work is of high overall quality. The manuscript is well-written and the content is original. On the other hand, I still have some criticism that I elaborate on here below.

The RXRD data consist of only one temperature point. The SN order is detected through the coupling with the Neel order and it is therefore impossible to observe a full suppression of the CD by sweeping the temperature. Nonetheless, measuring a few temperature points and comparing them with the Raman data would have strengthened the interpretation.
The Raman data clearly show the presence of a previously unknown thermodynamic transition at ≈263 K but it does not fully clear whether this transition is actually the SN order.
Finally, using RIXS, the authors report an anomaly in the pseudospin wave mode at Q = [pi,0] that is striking, despite the data being somewhat noisy. This finding will spark further investigations for, and not only, the similarity with the [pi,0] anomaly observed in cuprates. Similar to the case of RXRD, also here it would have been beneficial to collect a few temperatures. Indeed, the spin-wave dispersion in Sr2IrO4 persists above TN [see Lupascu et al PRL 112, 147201 (2014)] and this would have probably allowed the authors to reach up to the critical temperature.

The SN phase has been observed in other materials and the theory seems to be established, however, this is the first report in a 5d material. Moreover, the authors show how resonant x-ray techniques are sensitive to SN order and in general to local spin entanglement which is of great interest to the condensed matter community.
The iridates are widely studied due to their exotic physics and the many similarities to the high-Tc cuprates. In this context, the present report of a SN phase adds an important piece to this puzzle that will motivate further research and therefore deserves publication.
Overall, the data support the interpretation but it requires a rather involved analysis, making the manuscript more suitable for expert readers rather than a general audience. Indeed, one definitely needs to read the supplementary information and the authors themselves refer to the supplement many times throughout the text. This does not diminish the scientific validity of the manuscript but in my opinion, makes it more suitable for a more specialist journal such as Nature Physics or others.

In summary, my suggestion is to transfer this manuscript to a more specialized journal.

In the case there will be a second review round, I would request the authors to address the following specific points:

(1) In the RXRD data, the authors rule out any source of dichroism other than the interference between a dipole and a quadrupole in the E1-E1 channel. I wonder whether no other more trivial mechanism for a CD signal could be at play. Have you tried measuring the CD signal in any other RXRD peak or even in the XAS spectrum? For instance, could the material be birefringent in this energy range [see Joly Phys. Rev. B 86, 220101]?

(2) In the derivation of eq. 2, explained in the supplement, it is not clear why the structure factor is such that f1 = +/- f2. This notation is also ambiguous. Perhaps the authors can expand on this and give a better description.

(3) In Fig. 3(d), the spectral weight of the A1g mode shows an anomaly at 100 K. Below that temperature, the spectral weight drops by about 25% and becomes the same as for the B2g mode. Can the authors comment on this effect?

(4) In line 246, the authors say that the A1g mode becomes gapped in reference to the data of Fig. 3(f). I do not understand how this information is obtained from the data.

(5) In Raman studies of Sr2IrO4, the Fano lineshape of certain phonon modes was interpreted as an indication of strong pseudospin-lattice interactions, and the asymmetry was reported to be temperature dependent and renormalized at TN [e.g. Gretarsson Phys. Rev. Lett. 116, 136401 (2016); Samanta Appl. Phys. Lett. 114, 152402 (2019)]. Can you comment on the peak asymmetry and whether this has a temperature dependence in your dataset?

(6) In the supplement section S4, equation S12 is not sufficiently described.

(7) The RIXS measurement is not directly polarization-resolved and so, I would expect the statistics to be comparable with other published Ir L3-edge RIXS data. Conversely, the data looks rather noisy. Perhaps the reason is self-absorption due to the quasi-normal incidence geometry. Can you please comment on that?

(8) What is the peak appearing at 300 meV in the L-channel at [pi/2, pi/2] in Fig.4(c)? Is that a glitch or is it a real signal? Its spectral weight is higher than the one of the spin wave.

(9) In Fig. 4(d), what is the origin of the finite spectral weight in the L-channel around E=0, including the energy gain side? I guess this could be an artifact due to the summation/subtraction of the raw data or a difficult extraction of the elastic line. In that case, I would enlarge the error bars. In general, I think it would be beneficial to show the raw RIXS spectra somewhere in the text or the supplement.

Author Rebuttals to Initial Comments:

Reply to Reviewers

Referee #1 (Remarks to the Author):

Comment #1

"The authors study the material Sr_2IrO_4 using a diverse set of experimental techniques to reveal a spin quadrupolar order intertwined with a Neel antiferromagnetic order. Moreover, it is shown that the critical temperature for the quadrupolar ordering is distinct and higher than the Neel temperature. This is done using X-ray diffraction, Raman spectroscopy, and magneto-optical Kerr measurements. This finding is of particular interest since the magnetic ion has a local spin S=1/2 moment, which a priori cannot have a quadrupole moment, as opposed to S=1 ions. Instead, the quadrupolar order seems to be a genuine many-body phenomenon and likely is due to strong electron correlations. I would thus qualify the authors' findings as quite groundbreaking. To the best of my knowledge, this would make Sr_2IrO_4 to be among the first compounds to show spin nematic order without having higher spin local moments.

The experimental data is presented well and it is quite easy to follow the arguments of the authors. I particularly, like the brief descriptions of the experiments in the main text, and the more elaborate coverage in the supplementary material. From what I can tell, the experimental data, in particular, the Raman spectroscopy data for showing the intensity peaks in the Eg, A_{1g}, and B_{2g} representations are very convincing. The plots are very tidy and bring across the key results in a convincing way.

I do have several comments, though, which require to be addressed."

Our reply:
We greatly appreciate Referee #1 for his/her strong and positive appraisal of our work. We share the same view with Referee #1 that the quadrupolar order must be a genuine many-body phenomenon as a S=½ moment cannot have a quadrupole moment on its own, and this makes Sr_2IrO_4 to be among the first compounds to show spin nematic order without having spin local moments.

Comment #2

"1) One major critique I have is repeated, in my opinion unwarranted, mentioning of the resonating valence bond (RVB) picture by Anderson. The RVB state has unfortunately often been presented as a picture whenever "something weird beyond classical" is seen. Too often in literature, no precise statements are why any RVB picture would be needed to explain an observation, and I am afraid the authors do the same thing here. I think the author's finding of a spin nematic state is more than interesting enough, even without having a connection to the obscure RVB picture. I suggest the authors remove mention of the RVB picture completely since it does not contribute anything to their findings and it appears to have been added to "add some theoretical spice". However, if the authors are convinced otherwise, they should elaborate in more detail on how spin singlets would be resonating and how the observed experimental data can be explained by this. This should be thorough though. I would not accept adding one or two sentences. Instead, a full theory with predictions should be developed, which I assume would be a significant amount of work. The related statements about the RVB picture should not be published in a high-impact journal such as Nature. Hence, either they are completely removed or a substantial theory should be developed. As is, the discussion of RVB states is a bit unscientific."

Our reply:
We agree with the Referee's comment that our finding of a spin nematic state is more than interesting enough, even without having a connection to the obscure RVB state. In the revised manuscript, we thus have removed all of our previous claims/suggestions on the possible connections to RVB. Although we have shown through a simple two-site model that quadrupoles can be generated by "canting" the singlets in angle phi, admittedly this does not prove any connection to RVB.

Comment #3

"2) Accordingly, I suggest removing the RVB picture from Fig 1."

Our reply:
The main message of the Fig. 1 is that quantum entanglement between neighboring spins can manifest as quadrupoles on the bonds, which is a new feature only allowed for a canted AF. As we have shown through a two-site model, quadrupoles of t_{2g} symmetry are not allowed for a collinear AF. Thus, we believe that RVB in Fig. 1 provides a good contrast between the conventional form of spin entanglement (RVB) with the new form (quadrupoles) discovered in our work. As we have replied above, we have carefully revised the manuscript to NOT hint at a possible connection to RVB.

Comment #4
"3) Instead of relying on the RVB picture, there exist several computational works, which discuss the possibility of having spin nematic order in S=1/2 systems, in particular close to ferromagnetic phases. I suggest the authors give a more thorough account of the prior models which have been understood to show similar phenomenology by adding several references."

Our reply:
Following the Referee #1's suggestion, we have included the references that discuss spin nematic order arising from the condensation of two-magnon bound states in the vicinity of the ferromagnetic phase (i) for the frustrated spin chain model [L. Kecke *et al.*, PRB **76,** 060407(R) (2007); T. Hikihara *et al.*, PRB **78,** 144404 (2008)] and (ii) the frustrated S=½ square lattice model [H. T. Ueda *et al.*, PRB **80,** 014417 (2009); S. Jiang *et al.*, PRL **130,** 116701 (2023)]. Additionally, we have supplemented the discussion section by citing more references that demonstrate the importance of four-spin exchange J_c in the spin nematic order (Ref. 2 and Ref. 6), which can be relevant to the case of Sr_2IrO_4.

Comment #5
"Since the finding is genuine, novel, and in my opinion rather groundbreaking, I do think that the manuscript should eventually be published in Nature. However, my concerns need to be properly addressed before I can finally commit to giving an according recommendation."

Our reply:
We thank Referee #1 for evaluating our work as "genuine", "novel" and "groundbreaking". We believe that we have addressed all of the concerns raised by Referee #1.

Referee #2 (Remarks to the Author):

Comment #1
"A. Summary of the key results
In this manuscript the authors present an experimental study of the square-lattice iridate Sr2IrO4. Sr2IrO4 is the prototypical Spin-Orbital Mott Insulator, and one of the best studied model systems for spin-orbit-driven magnetism. The authors use a combination of resonant x-ray diffraction/circular dichroism, Raman spectroscopy, and resonant inelastic x-ray scattering to investigate this compound. These measurements are supported by detailed analysis and a simple two-site S=1/2 model calculation. The authors propose three key results:
(1) Sr2IrO4 displays spin nematic (i.e. magnetic quadrupolar) order.
(2) This magnetic quadrupolar order develops at higher temperatures (Tc ~263K) than canted antiferromagnetic order (Tn ~ 230K) as shown by Raman spectroscopy. It then persists into the antiferromagnetic state, as shown by resonant x-ray diffraction.
(3) There is a breakdown of coherent magnon excitations at the (Pi,0) position in reciprocal space, which can be attributed to RVB-like quantum entanglement in the canted antiferromagnetic state."

Our reply:
We highly appreciate Referee #2's careful and accurate assessment of our work.

Comment #2

"B. Originality and significance

Sr2IrO4 is a well-studied model system. It has previously been studied using all 3 of these techniques (RXD, Raman, and RIXS), but the authors present new and original measurements using these probes. In the case of RXD, there is a new focus on circular dichroism, which can be sensitive (albeit very subtly) to the presence of quadrupolar order. In the case of Raman, the authors employ a different geometry, illuminating the short/narrow side of a thin plate-like crystal. In the case of RIXS, the authors focus on polarization-dependence measurements at three high symmetry positions in reciprocal space.

Because of the level of interest in Sr2IrO4, proof of spin nematic/magnetic quadrupolar order would be a significant result. It is not entirely unprecedented, since several previous studies have reported "hidden order" above Tn in Sr2IrO4 (see Refs 51, 52, and 53). However, the authors claim to have observed a different symmetry breaking state (i.e. a different type of hidden order), which can be definitively attributed to spin nematic ordering."

Our reply:

We thank Referee #2 for acknowledging the novelty of our work relative to previous studies in the literature.

As pointed out by Referee #2, there have been several previous studies reporting a hidden order in Sr_2IrO_4, mostly interpreted in terms of loop current orders, which are distinct from our spin nematic/magnetic quadrupolar order. As Referee #2 has asked us in his/her Comment #8 to attempt to reconcile our result with previous reports of hidden order in Sr_2IrO_4, we address this issue in detail in our reply to Comment #8 below.

Comment #3

"C. Data and methodology

The authors present a significant amount of new experimental data. The data quality appears to be good, and the figures are well-presented. It is worth noting that the RXD measurements are quite challenging (i.e. to obtain good azimuthal dependence of magnetic peaks), and the CD effects are relatively weak. However, Fig. 2 is quite convincing for showing the CD effect. The authors also provide extensive supplementary information with this manuscript. In particular, the SI contains a more detailed description of the representational analysis (for RXD) and polarization dependence (for RIXS). The methodology also seems sound: RXD and Raman are both sensitive probes for detecting weak or subtle symmetry-breaking, and RIXS is well-suited to the study of magnetic excitations in Sr2IrO4."

Our reply:

We thank Referee #2 for his/her very positive evaluation on our data quality, presentation, and methodology. In particular, we are glad that Referee #2 finds "Fig. 2 is quite convincing for showing the CD effect". Indeed, the CD effects in our case arise from high-order RXD processes and thus are quite challenging to measure.

Comment #4

"D. Appropriate use of statistics and treatment of uncertainties

The authors include appropriate experimental uncertainties and error bars when discussing the RXD (Fig. 2), Raman (Fig. 3), and RIXS data (Fig. 4). The data analysis appears to be thorough and technically sound."

Our reply:

We thank Referee #2 for his/her comment that the data analysis appears to be thorough and technically sound.

Comment #5

"E. Conclusions

I do have some concerns regarding the interpretation of the data, and the robustness/reliability of the conclusions. My biggest concern is in Section IV, when the authors discuss the polarization-dependence of the RIXS spectra. In particular, the authors claim that there is no magnon mode at the (Pi,0) position in reciprocal space (Fig. 4(d)). The absence of the magnon mode is the basis of the claim that the quadrupolar order results from RVB-like quantum entanglement. My issue with this statement is that there is a clear magnon peak at

~200 meV. The full dispersion of this mode has previously been mapped out in Ref. 48, and the energy of the magnon is known to be ~200 meV at (Pi,0). While the polarization-dependence of this feature may not match the authors' expectations, the peak at 200 meV cannot simply be ignored. This would seem to undermine the only piece of evidence for key result #3 (breakdown of coherent magnon excitations at short-wavelength scales)."

Our reply:
Referee #2 raises an important question about our claim on the absence of the magnon mode at (π, 0). This is also related to his/her Comment #8 that "polarization-dependent RIXS data is fairly limited (only 3 Q-points)." We understand that Referee #2 does not find our claim convincing partly because of the limited data set. To address this issue, as shown in Fig. R1, we have performed additional polarization-resolved RIXS to measure 6 Q-points along the magnetic zone-boundary, which show in greater detail how the spectra evolve on going from (π/2, π/2) to (π, 0). Our new data, much improved by the high quality of the sample without Pt impurities [see Phys. Rev. Materials **6**, 103401 (2022)], shows a resolution-limited single magnon mode at (π/2, π/2) that appears dominantly in the transverse channel as expected, and consistent with Figure 4. This magnon mode gradually loses spectral weight as (π, 0) is approached, where there is no detectable difference between transverse and longitudinal spectra.

As Referee #2 has pointed out the peak in the (π,0) spectrum in previous RIXS studies on Sr_2IrO_4 [J. Kim *et al.*, Phys. Rev. Lett. **108,** 177003 (2012)] was assigned to a single magnon. However, the energy resolution then in 2012 was only ~150 meV, and the measured peak widths of (π/2, π/2) and (π, 0) spectra appeared to be comparable. Now, with the energy resolution improved by a factor of 5, together with significant improvement in the sample quality, it is clear that the peak widths are very much different: the peak widths of (π/2, π/2) and (π, 0) are, respectively, <28 meV (resolution limited) and ~100 meV. It is still possible that the (π, 0) peak has a much broader width due to its higher energy and a much wider phase space for the single magnon to decay into. To investigate this possibility, we have analyzed the inelastic neutron scattering spectra of La_2CuO_4 measured by N. S. Headings *et al.* [Phys. Rev. Lett. **105,** 247001 (2010)], which is shown in Fig. R3. We note that the similarity between La_2CuO_4 and Sr_2IrO_4 in terms of their lattice and spin structures has been previously pointed by many papers [see, e.g., Annu. Rev. Condens. Matter Phys. **10,** 315 (2019)]. In La_2CuO_4, the peak widths do not differ much between (π/2, π/2) and (π, 0) and both are much narrower than in our case. (Although this paper does not provide any information about their energy and momentum resolution, we suspect the peak widths are limited more by the momentum resolution than the energy resolution.)

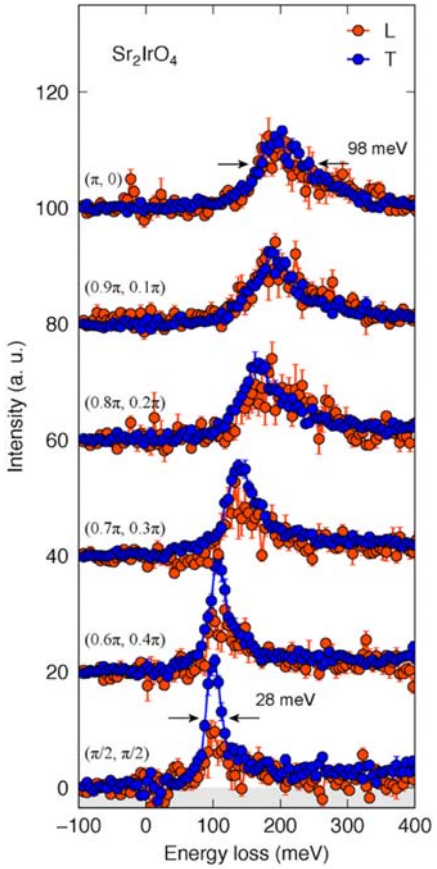


Figure R1. RIXS spectra along the zone boundary from (π/2, π/2) to (π, 0). The scheme shown in Fig. 4 was used to resolve spin components transverse (T) and longitudinal (L) to the ordered AF moments.

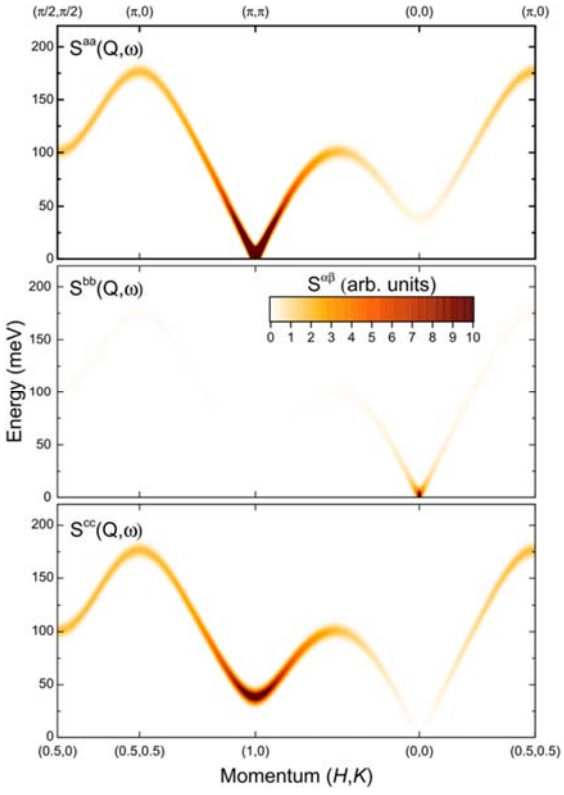


Figure R2. Dynamic structure factors for Sr_2IrO_4 taken from [Phys. Rev. B **99,** 085125 (2019)]

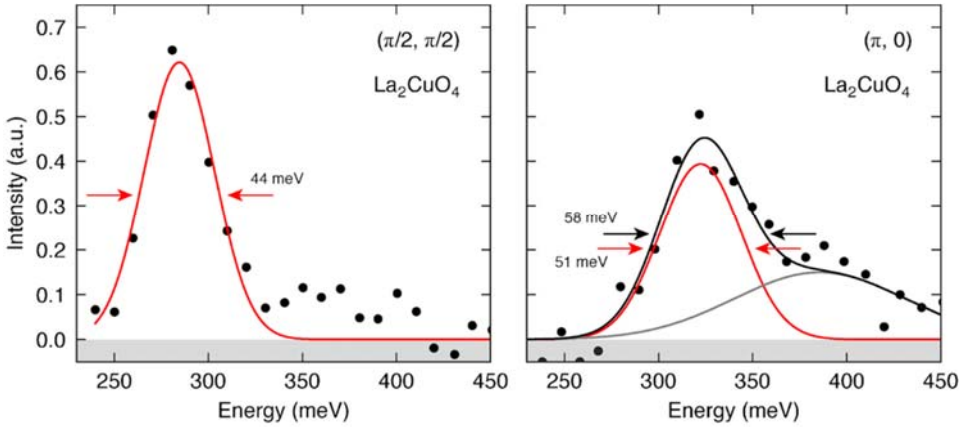


Figure R3. Our analysis of the inelastic neutron spectra of La_2CuO_4 measured at (π/2, π/2) and (π, 0) reported in [Phys. Rev. Lett. **105,** 247001 (2010)]. The (π/2, π/2) spectrum is fitted with a single Gaussian peak, while an additional Gaussian peak is used for the (π, 0) spectrum to account for a high-energy tail.

Further, the polarization dependence cannot be reconciled with the single magnon scenario. It is not only our expectation but also a well-established fact that single magnons are excited when spin components transverse to the ordered moments are measured. Thus, magnon modes cannot be insensitive to the direction of the ordered moment. Our virtually identical transverse and longitudinal spectra imply that the dynamic spin structure is insensitive to the direction of the ordered moment, and hence cannot be attributed to single magnon excitations. This can be explicitly seen, for example, from the dynamic structure factor calculated for Sr_2IrO_4 under the linear spin-wave approximation shown in Fig. R2 from J. Porras *et al.*, Phys. Rev. B **99,** 085125 (2019). In this figure, the magnon mode intensity dominantly appears in S_{aa} and S_{cc} which are the in-plane and out-of-plane transverse channels, respectively, when the magnetic moments are aligned along the *b*-axis. Small intensity in the longitudinal channel (S_{bb}) comes from the canting of moments by ~11 degrees from the *b*-axis. Indeed, as we have shown in the main figure 4, at (π, π) most of the spectral weight is in the transverse channel, and single magnons are well resolved at (π/2, π/2). According to this calculation, the single magnon at (π, 0) should also have dominant intensity in the transverse channel, which is clearly not the case.

In the context of copper-based square-lattice S=½ AFs, '(π, 0) anomaly' is observed across many different compounds with varying degrees of the single magnon weight. There, the transfer of spectral weight from transverse to isotropic "background" is widely taken as evidence for the magnon decay [see, for example, D. Piazza *et al.*, Nat. Phys. **11,** 62-68 (2015); N. Ma *et al.*, Phys. Rev. B **98,** 174421 (2018)]. Thus, we believe that the concept that a fully isotropic response cannot be due to a single magnon is well accepted in the community.

Comment #6

"I am also concerned that the authors have not attempted to reconcile their "hidden order" state above Tn, with previous reports based on Second Harmonic Generation (Refs. 51 and 55), polarized neutron diffraction (Ref. 52), and torque magnetometry/elastoresistance (Ref. 53). In particular, the elastoresistance measurements in Ref. 53 seem to closely match the 263K temperature-scale identified in this study. The differences in reported symmetry-breaking also seem to merit further discussion (especially since the reported breaking of inversion-symmetry could affect the arguments used to interpret the RXD data). I was not convinced by the claim that these discrepancies could be explained by the existence of two different, unrelated hidden order states."

Our reply:

We fully agree with Referee #2 that it is very important to discuss our results in the context of previous reports of symmetry breakings in iridates. Below, we have summarized the previous reports based on second harmonic generation (SHG), polarized neutron diffraction (PND), torque magnetometry (TM), and elastoresistance (ER) measurements, and compared them with our results.

	Observation	Interpretation	Inconsistency with our data
SHG	• Breaking of inversion and rotational symmetries at T_{HO}~243 K in undoped Sr_2IrO_4, and at T_{HO}~210 K for 4% Rh-doped sample.	• Symmetry consistent with magneto-electric loop current • Reinterpreted as due to surface magnetization induced electric-dipole SHG process based on additional experiment in magnetic field (for undoped sample)	• T_{HO} and our T_C different by ~20 K (for undoped sample). • Inconsistent with PND and TM (ferroic stacking along *c* axis).
PND	• Spin-flip scattering at Q=(1, 1, 2) setting in at T_{HO}~240±30 K for both	• Symmetry consistent with magneto-electric loop current	• Q vector different from our x-ray data; CD is observed at (0, 0, odd).

	parent and 7% Rh-doped samples.	(but of different type from the one suggested by SHG)	• T_{HO}~240 K is much lower than our T_C~263 K. • Our magneto-optical Kerr effect data rules out any time-reversal breaking order.
TM	• Broken C_4 symmetry at T_{HO}~270 K for the parent compound.	• Nematic phase transition (but distinct from nematic orders reported in other systems, which have even parity)	•Consistent with our data. •T_{HO} ~ T_C.
ER	• Absence of divergent behavior in the nematic susceptibility, which suggests odd parity.	• Together with the magnetic torque data, the hidden order is suggested to be a loop current order.	• The data is available only for Rh-doped samples.

Table R1. Summary of symmetry breakings reported in iridates.

Second harmonic generation (SHG): we note that the authors of the SHG measurements [L. Zhao *et al.*, Nat. Phys. **12,** 32-36 (2016)], who were the first to claim the orbital loop current in Sr_2IrO_4, have reinterpreted their data in terms of the surface magnetization induced electric dipole SHG process, because their additional measurements under magnetic field [K. L. Seyler *et al.*, Phys. Rev. B **102,** 201113 (2020)] clearly showed inconsistency with the original proposal (linear coupling to magnetic field in the absence of electric field). These authors have commented that "while our work does not rule out the existence of such a hidden order, it firmly establishes that the anomalous SHG signal in parent Sr_2IrO_4 is instead dominated by a process induced by surface magnetization." Therefore, there is no evidence for a bulk, Inversion symmetry breaking order from SHG as far as undoped Sr_2IrO_4 is concerned.

Polarized neutron diffraction (PND): a time-reversal symmetry breaking order, interpreted as a loop current order, was reported by J. Jeong *et al.*, Nat. Commun. **8,** 15119 (2017). Our data cannot be reconciled with this observation because of two following reasons: (i) their observation is made at Q=(1,1,2) which is distinct from Q = (0,0,21) where we observed CD, implying their different stacking patterns; (ii) our high-sensitivity Kerr measurement shown in Extended Data Fig. 2 does not show any indication of time-reversal symmetry breaking at T_c within our experimental resolution of 3.4x10^-5 bohr magneton/ion.

Torque magnetometry I: the data reported by H. Murayama *et al.*, Phys. Rev. X **11,** 011021 (2021) shows a breaking of four-fold rotational symmetry. The authors of this paper refer to this as "nematic". This is the only observational fact for the undoped sample, and the data itself is consistent with our SN and the onset temperature (270 K) is also close to our T_c (263 K). Their interpretation in terms of a loop current order is based on the absence of a divergent behavior in elastoresistance, which however was only measured on Rh-doped samples. Further, we think that while a divergent behavior in elastoresistance may be evidence for even-parity nematic, its absence does not necessarily imply odd parity.

In summary, the only evidence that supports a loop current order in undoped Sr_2IrO_4 is from PND, but we do not find any indication of time-reversal symmetry breaking above T_N in our sample within our experimental resolution of 3.4x10^-5 bohr magneton/ion. Further, there is no indication of inversion symmetry breaking in the SHG data in the temperature range 250 K < T < 270 K reported in [L. Zhao *et al.*, Nat. Phys. **12,** 32-36 (2016); K. L. Seyler *et al.*, Phys. Rev. B **102,** 201113 (2020)], which conclusively rule out the possibility of a loop current as the origin of our observation.

Comment #7

"Lastly, one argument presented in favor of spin nematic order in this compound is that the SN state has been theoretically shown to emerge on a frustrated 2D square lattice with ferromagnetic (FM) nearest-neighbor (nn) couplings, and antiferromagnetic (AF) next-nearest-neighbor (nnn) coupling. It should be noted that this scenario does not apply to Sr_2IrO_4, which has strong AF nn coupling and weaker FM nnn coupling. Do the authors know of any theoretical support for SN order under the conditions relevant to this compound?"

Our reply:

To the best of our knowledge, the possibility of realizing SN in systems with strong anisotropic exchange interactions has not been investigated theoretically in any detail. In Sr_2IrO_4, the large canting angle of the spins allows the yz/zx quadrupoles to coexist with the AF order, which suggests that anisotropic interactions somehow play a role in stabilizing the SN phase. However, it is not clear at this point if SN can in theory be stabilized in systems with AF nn coupling if anisotropic interactions are sufficiently strong, and we hope that our observation will stimulate future theoretical investigations along this direction.

Comment #8

"F. Suggested improvements

I would recommend that the authors attempt to reconcile the current results with previous reports of hidden order in Sr_2IrO_4. How can they conclusively demonstrate that this hidden order corresponds to spin nematic order and not anapole or loop-current order? Should signatures of spin nematic order have been observable in these previous studies? What can account for the discrepancies in reported time-reversal, rotational, and inversion symmetry breaking? To make a strong statement on the existence of spin nematic order, I think this issue needs to be addressed."

Our reply:

In our reply to Referee #2's Comment #6 above, we have argued that our SN order cannot be reconciled with anapole or loop-current order, which is odd under both space inversion and time reversal. As our SN order is parity even and time-reversal even, we do not expect it to show any signal in SHG or PND in the spin-flip channel, and indeed so far there is no evidence of either inversion or time-reversal symmetry breaking from any experiment close to our T_C.

Comment #9

"I would also recommend that the authors remove or heavily edit Section IV. The polarization-dependent RIXS data is fairly limited (only 3 Q-points) and I do not believe that it supports the claim that the coherent magnon mode breaks down at short-wavelength scales (Pi,0). This section is not essential to the case for spin nematic order, which is largely based on Sections I and III."

Our reply:

As we have discussed in detail above, we hope that our additional data can convince Referee #2 that the coherence magnon mode breaks down at (π, 0). While we agree this section is not essential for establishing the spin nematic order, and thus may be removed, we have kept it because of the Referee #3's comment that "this finding will spark future investigations for, and not only, the similarity with the [pi,0] anomaly observed in cuprates".

Comment #10

"I believe it is also worth pointing out to the reader that the magnetic field used to control the ordered moment direction (B = 0.3 T) will also change the magnetically ordered structure below Tn (as originally reported in Ref. 8). This point is noted in the SI, but it is an important enough detail to justify mention in the main text."

Our reply:

Following Referee #2's suggestion, we now mention in the main text that applying a magnetic field of 0.3 T changes the magnetically ordered structure. We have also added the below figure R4 to SI to show that this has no appreciable effect on the RIXS spectra.

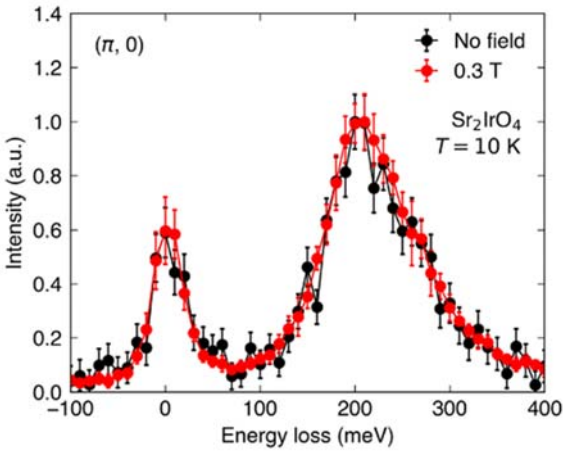


Figure R4. (π, 0) spectra of Sr_2IrO_4 measured with and without an external magnetic field of 0.3 T. Polarization components are not resolved.

Comment #11

"G. References

The authors have provided an extensive list of references, and they appear to have cited all of the most relevant previous work. As noted above, my main concern is that the results from previous studies (i.e. experimental studies of hidden order, theoretical studies of the frustrated square lattice) are not fully integrated or reconciled with the current study."

Our reply:

In the Supplementary Information (Note 7) of our revised manuscript, we discuss the results from previous studies in more detail and provide the Table R1.

Comment #12

"H. Clarity and context

This manuscript is well-organized and reasonably well-written, with a clear abstract/summary. The main text is quite technical (particularly in Section I), which may pose a challenge for non-specialists. The discussion section is somewhat brief, and (as noted above) there are several important points which would be helpful to discuss in more detail."

Our reply:

We thank Referee #2 for overall very positive evaluation of our presentation. Admittedly, Section I of the main text is somewhat technical, but we would like to emphasize that our work not only establishes a SN phase but also reveals its microscopic structure for the first time. The symmetry analysis inevitably involves some technical details not readily accessible to non-specialists. As acknowledged by Referee #1, we have tried to organize the paper in such a way that a concise description of the experiment is provided in the main text and a more elaborate coverage in the Supplementary information, so that non-specialists can follow our paper without going through technical details. We have also reinforced the discussion section to address the issues raised by Referee #2.

Referee #3 (Remarks to the Author):

Comment #1

"In their manuscript, Kim et al report the discovery of a quantum spin nematic (SN) phase in the Sr_2IrO_4 material.

Using resonant x-ray diffraction (RXRD), the authors detect a prominent circular dichroism (CD) in the (0 0 21) magnetic reflection and interpret this as evidence for a quadrupolar order interfering with the magnetic order. Using temperature-dependent Raman spectroscopy, they show that the low-energy magnetic fluctuations appear well above the Neel temperature. Using a special experimental configuration, they also report a novel excitation emerging at Tc = 263 K that they assign to a collective mode associated with the breaking of rotational symmetry. They also show, using magneto-optical Kerr effect, that time-reversal symmetry is preserved for TN<T<Tc.
Using resonant inelastic x-ray scattering (RIXS) at the Ir L3-edge, they report a deviation from the linear spin wave theory of the pseudospin excitation at [pi,0], where the signal in the longitudinal and transverse channels becomes isotropic. They interpret this as evidence of quantum entanglement of near-neighbor pseudospins.
The experimental approach comprises different techniques and the work is of high overall quality. The manuscript is well-written and the content is original. On the other hand, I still have some criticism that I elaborate on here below."

Our reply:
We thank Referee #3 for his/her careful review and high appraisal of our work, and also for many constructive comments to strengthen our paper. Below, we provide point-by-point response to Referee #3's comments.

Comment #2
"The RXRD data consist of only one temperature point. The SN order is detected through the coupling with the Neel order and it is therefore impossible to observe a full suppression of the CD by sweeping the temperature. Nonetheless, measuring a few temperature points and comparing them with the Raman data would have strengthened the interpretation."

Our reply:
Following Referee #3's suggestion, we have performed an additional CD-RXD experiment to collect a few temperature points as shown below. In particular, in reply to the Comment #9 below where the anomalous spectral weight decrease of the A_{1g} mode below 100 K is pointed out, we have investigated if the spectral weight decrease can also be seen through the CD. Within our error bars, we could not see a similar spectral weight decrease in CD. We note, however, that the CD effects are weak, and the error bars are rather large. This is due to the fact that the CD effects appear only at (0 0 odd) reflections, whose intensities are weaker by two orders of magnitude compared to main (1 0 L) magnetic reflections; we note that even the temperature dependence of (0 0 odd) reflections have not been previously reported because of their very small intensities close to T_N. Further, the CD signals are weaker by another two orders of magnitude due to the high-order process of RXD involved as explained in the section 4 of Supplementary Information. Under these circumstances, we were able to collect in a week of beamtime (including setting up a diamond phase plate for CD measurements) data at the azimuth angle of 45 degrees where the CD signal is maximal at five different temperatures, as shown below.

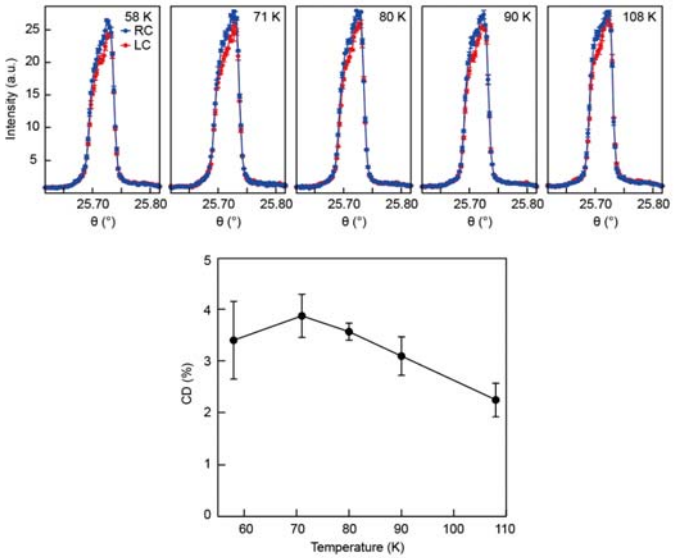


Figure R5. Rocking curves measured at (0 0 21) reflection with left and right circular incident x-ray polarizations and the resulting CD.

Comment #3

"The Raman data clearly show the presence of a previously unknown thermodynamic transition at ≈263 K but it does not fully clear whether this transition is actually the SN order."

Our reply:

For Mott insulators, it is well established that Raman operators have leading order terms bilinear in spin operators [B. S. Shastry et al., Phys. Rev. Lett. **65,** 1068 (1990)]. In the Z(YY)X scattering geometry, $S_i^z S_j^x + S_i^x S_j^z$ is the only symmetry allowed combinations of spin operators that can couple to the relevant photon polarizations. Thus, Raman can directly detect fluctuations of quadrupoles and their condensation. Given that there are no other degrees of freedom active in the ~meV energy scale, it is hard to imagine an electronic order of different origin. Note that both "orbital" ($J_{1/2}$ -> $J_{3/2}$) and charge excitations are of the order of ~0.5 eV. As we have shown through the magneto-optical Kerr effect, the time-reversal symmetry is preserved across the transition, and because the order parameter of E_g symmetry breaks rotational symmetries, by definition it must be a nematic order of spin origin.

Comment #4

"Finally, using RIXS, the authors report an anomaly in the pseudospin wave mode at Q = [pi,0] that is striking, despite the data being somewhat noisy. This finding will spark further investigations for, and not only, the similarity with the [pi,0] anomaly observed in cuprates. Similar to the case of RXRD, also here it would have been beneficial to collect a few temperatures. Indeed, the spin-wave dispersion in Sr_2IrO_4 persists above TN [see Lupascu et al PRL 112, 147201 (2014)] and this would have probably allowed the authors to reach up to the critical temperature."

Our reply:

Following Referee #3's suggestion, we performed an additional RIXS experiment to collect a few temperature points. However, we did not perform polarization analysis, since the Q=(π, 0) spectrum is already isotropic at the base temperature of 10 K. We were not able to see a significant difference between the spectra measured above and below T_C.

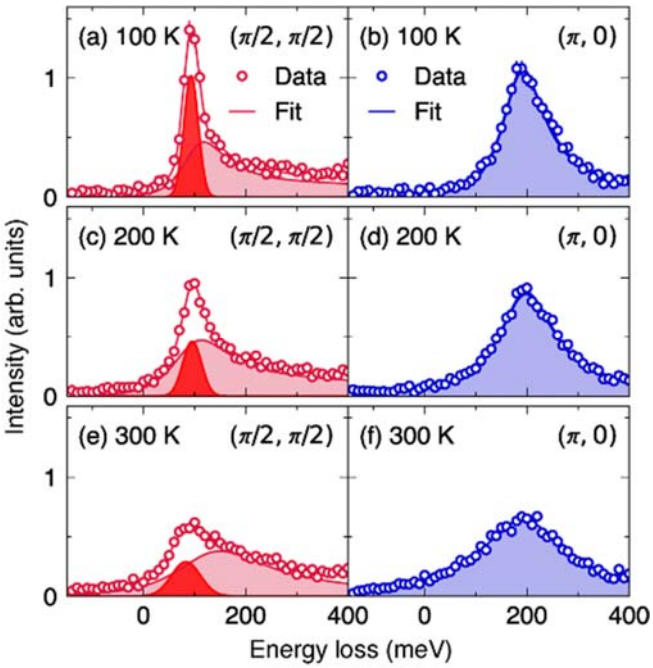


Figure R6. Temperature-dependent RIXS spectra of Sr_2IrO_4. The magnon modes in the (π/2, π/2) spectra are fitted with a Gaussian function, and the high-energy continuum in both (π/2, π/2) and (π, 0) spectra are fitted by antisymmetric Lorentzian function. Polarization components are not resolved for all the spectra.

Comment #5

"The SN phase has been observed in other materials and the theory seems to be established, however, this is the first report in a 5d material. Moreover, the authors show how resonant x-ray techniques are sensitive to SN order and in general to local spin entanglement which is of great interest to the condensed matter community."

Our reply:

In our view, the existence of a SN phase has not been established, both in experiment and theory. The best-studied scenario is SN phase emerging in a very high magnetic field close to saturation by condensation of two magnons, in strongly frustrated 1D and 2D systems with ferro NN and AF NNN couplings. The SN phase appears in a very narrow field range (<1T) at very high magnetic fields (~50 T), where difficulties of experimental access obstruct precise characterization needed to identify the SN phase. The report of SN is mostly based on the absence of a dipolar order transverse to the applied field, lacking direct evidence for phase transition or identification of the symmetry of the order parameter. For example, the SN being indirectly inferred from nuclear magnetic resonance [A. Orlova *et al.*, Phys. Rev. Lett. **118,** 247201 (2017)] or thermodynamic measurements [Y. Kohama *et al.*, PNAS **116,** 10686 (2019)]. Even in theory, the phase boundary of the SN phase is still under debate; a very recent paper [S. Jiang et al., PRL **130,** 116701 (2023)] shows that the phase space occupied by SN is much smaller than previously suggested. Thus, we believe there is not yet a firmly established case for a SN.

Comment #6

The iridates are widely studied due to their exotic physics and the many similarities to the high-Tc cuprates. In this context, the present report of a SN phase adds an important piece to this puzzle that will motivate further research and therefore deserves publication.

Overall, the data support the interpretation but it requires a rather involved analysis, making the manuscript more suitable for expert readers rather than a general audience. Indeed, one definitely needs to read the supplementary information and the authors themselves refer to the supplement many times throughout the text. This does not diminish the scientific validity of the manuscript but in my opinion, makes it more suitable for a more specialist journal such as Nature Physics or others.

In summary, my suggestion is to transfer this manuscript to a more specialized journal.

In the case there will be a second review round, I would request the authors to address the following specific points:"

Our reply:
Admittedly, Section I of the main text is somewhat technical, but we would like to emphasize that our work not only establishes a SN phase but also reveals its microscopic structure for the first time. The symmetry analysis inevitably involves some technical details not readily accessible to non-specialists. As acknowledged by Referee #1, we have tried to organize the paper in such a way that a concise description of the experiment is provided in the main text and a more elaborate coverage in the Supplementary information, so that non-specialists can follow our paper without going through technical details.

Comment #7
"(1) In the RXRD data, the authors rule out any source of dichroism other than the interference between a dipole and a quadrupole in the E1-E1 channel. I wonder whether no other more trivial mechanism for a CD signal could be at play. Have you tried measuring the CD signal in any other RXRD peak or even in the XAS spectrum? For instance, could the material be birefringent in this energy range [see Joly Phys. Rev. B 86, 220101]?"

Our reply:
We thank Referee #3 for bringing up this important point. We agree that CD can arise from an extrinsic origin such as birefringence. To rule out this possibility, we have measured the resonance profiles of the CD signals at (1 0 18) magnetic reflections, which have similar scattering geometries/angles as (0 0 19) reflection. Unlike the (0 0 21) reflection exhibiting a clear CD at the resonance, we found no indication of CD from (1 0 18) reflection. This clearly rules out the possibility of birefringence. We have included this discussion in our Supplementary Information.

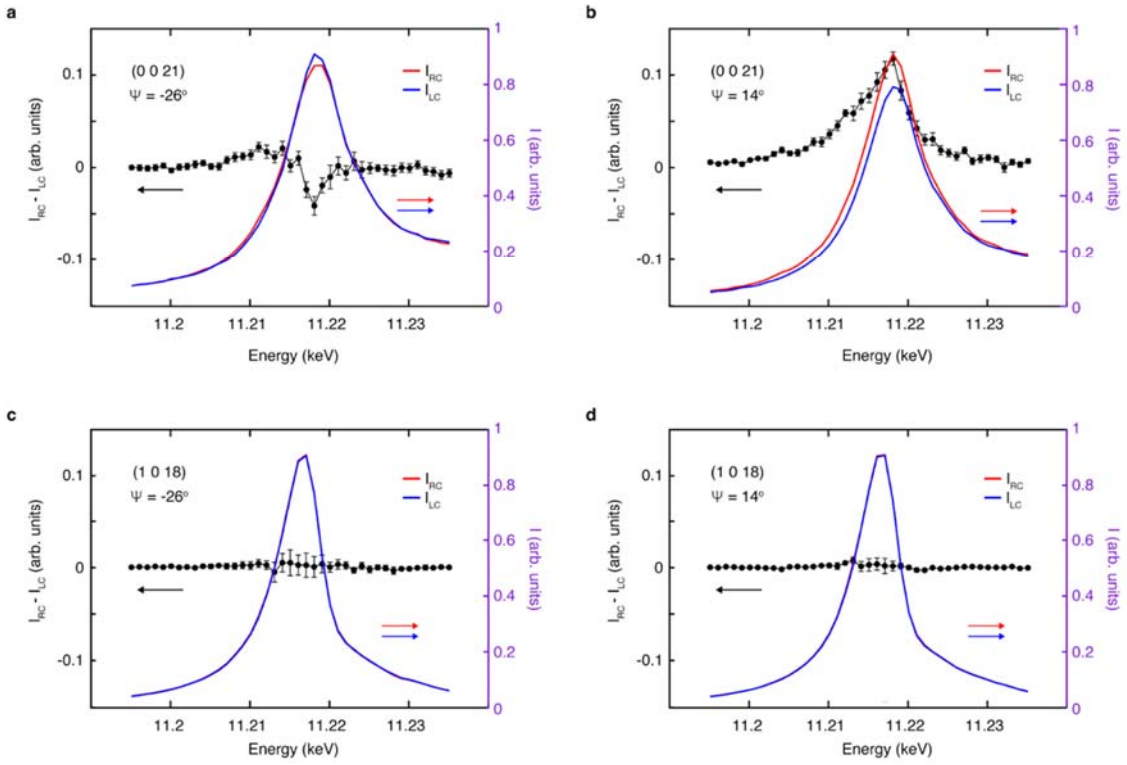


Figure R7. CD-RXD on different magnetic reflections. a,b, (0 0 odd) reflections exhibit a clear CD through interference between dipolar and quadrupolar moments **c,d,** whereas the CD signals are not manifested for (1 0 4n+2) reflections of similar scattering geometry.

Comment #8

"(2) In the derivation of eq. 2, explained in the supplement, it is not clear why the structure factor is such that f1 = +/- f2. This notation is also ambiguous. Perhaps the authors can expand on this and give a better description."

Our reply:

We thank Referee #3 for pointing this out. We have revised Supplementary Note 1 to make our notation more clear. The fact that F1 = +/- F2 in our case is explicitly confirmed in Table S4 for all possible structures. To rationalize this result, we first note that the x-ray phase factor $\exp(iQ \cdot r_j)$ at the eight bond centers (j=1,..,8) listed in Fig. S2 is {1,-1,i,-i,-i,i,-1,1}; that is, the phase is uniform within a layer and the phase difference between adjacent layers is +-i. This phase is multiplied by the atomic form factor and summed over j to yield the structure factor. Since all possible structures have atomic form factors such that they are same within the layer (ferroquadrupolar order), and can only have sign change between layers, the sum either cancels out to zero or have equal real part and the imaginary part up to their signs. A similar logic holds for the magnetic structure.

This is a bit difficult to explain in words, so we have added another column in Table S4 to show the x-ray phase factor, so that one can easily do the math described above.

Comment #9

"(3) In Fig. 3(d), the spectral weight of the A1g mode shows an anomaly at 100 K. Below that temperature, the spectral weight drops by about 25% and becomes the same as for the B2g mode. Can the authors comment on this effect?"

Our reply:

We thank Referee #3 for raising this interesting question. The downturn of the A_{1g} intensity reminds us of the magnetic anomaly at a similar temperature reported in [S. Chikara *et al.*, Phys. Rev. B **80,** 140407(R) (2009)]. In Sr_2IrO_4, they observed a magnetization downturn and anomalous behaviors of the dielectric constant at $T_M \approx 100$ K. As this feature is absent in other samples (Ref. 54), we have first tried to reproduce our result in other samples. As shown below, the A_{1g} downturn was reproduced, suggesting that it is an intrinsic effect. We speculate that the downturn could be due to competition between the SN and the AF orders, but more experiments are required to clarify its origin. We hope to address this issue in a future study.

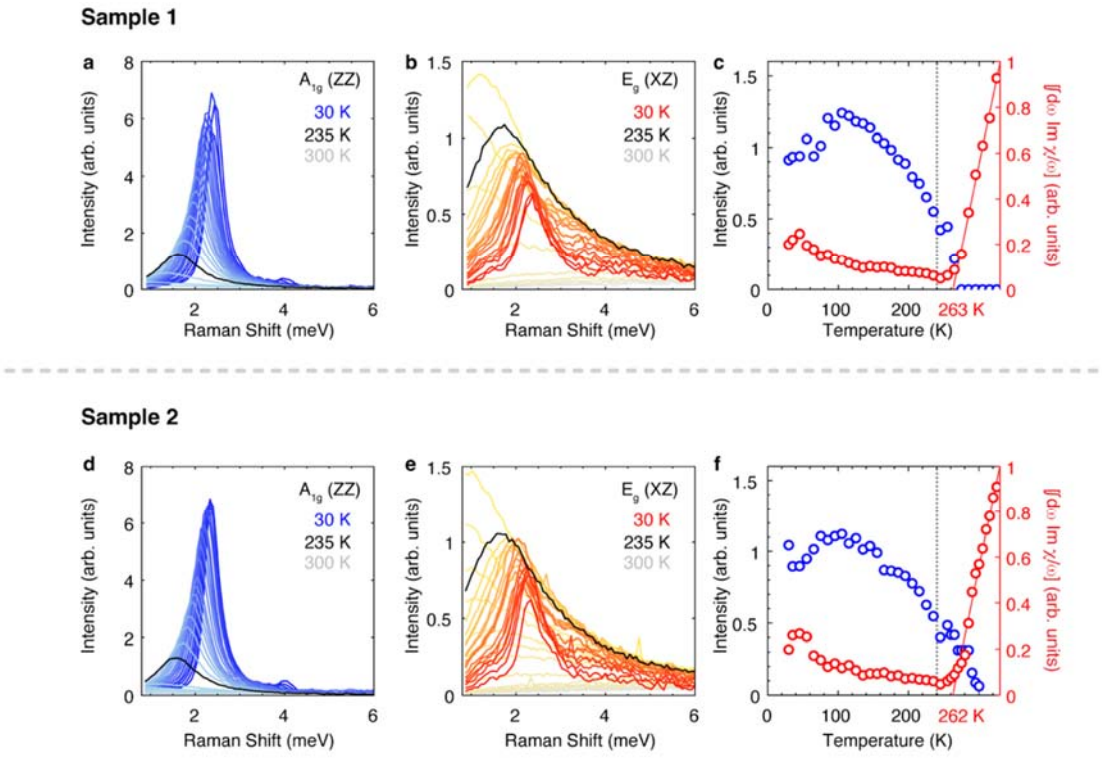


Figure R8. Sample dependence of the Raman measurement results. Compared to the main text data (Sample 1), the Curie-Weiss behavior of the E_g mode and the emergence of the A_{1g} mode are reproduced in Sample 2 synthesized by the same method. In Sample 2, the downturn of the A_{1g} mode intensity occurs at a lower temperature of ~ 70 K.

Comment #10

"(4) In line 246, the authors say that the A1g mode becomes gapped in reference to the data of Fig. 3(f). I do not understand how this information is obtained from the data."

Our reply:

In line 246, we simply stated the experimental observation that the A_{1g} mode acquires a finite excitation energy near T_N, as clearly shown by the blue circles in Fig. 3f. In our Raman setup, we can detect signals above our low-energy cutoff of ~10 cm^{-1} and the very fact that the peak maxima can be seen means that the mode has non-zero energy.

Comment #11

"(5) In Raman studies of Sr_2IrO_4, the Fano lineshape of certain phonon modes was interpreted as an indication of strong pseudospin-lattice interactions, and the asymmetry was reported to be temperature dependent and renormalized at TN [e.g. Gretarsson Phys. Rev. Lett. 116, 136401 (2016); Samanta Appl. Phys. Lett. 114, 152402 (2019)]. Can you comment on the peak asymmetry and whether this has a temperature dependence in your dataset?"

Our reply:

In our scattering geometry (see Fig. R9 below), there is no Raman-active phonon mode below 100 cm^{-1}, where we observe the emergent mode associated with the SN order, to analyze the asymmetry with.

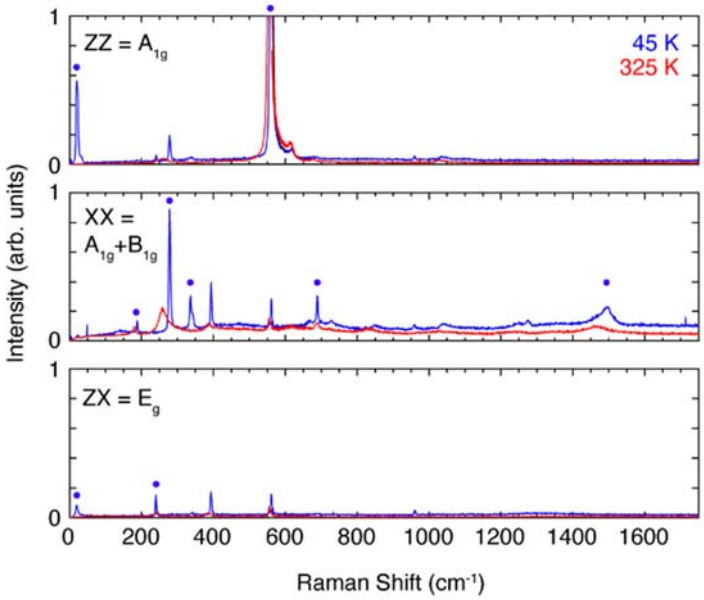


Figure R9. Raman spectrum obtained on the side surface of Sr_2IrO_4

Comment #12

"(6) In the supplement section S4, equation S12 is not sufficiently described."

Our reply:

Following Referee #3's comment, we have revised section S4 to describe the RIXS process in more detail by labeling each step of the RIXS process in Fig. S4 and explaining all the parameters in Eq. S12.

Comment #13

"(7) The RIXS measurement is not directly polarization-resolved and so, I would expect the statistics to be comparable with other published Ir L3-edge RIXS data. Conversely, the data looks rather noisy. Perhaps the reason is self-absorption due to the quasi-normal incidence geometry. Can you please comment on that?"

Our reply:

We thank Referee #3 for pointing this out. The rather poor statistics of our data is due to a very tight mask (~10 mm) used to achieve a high momentum resolution (~0.08 $Å^{-1}$ or ~0.02π in the in-plane direction), which in retrospect was unnecessary. Thus, we have replaced Fig. 4 with higher statistics data measured with a 75 mm mask (see also Fig. R1).

Comment #14

"(8) What is the peak appearing at 300 meV in the L-channel at [pi/2, pi/2] in Fig.4(c)? Is that a glitch or is it a real signal? Its spectral weight is higher than the one of the spin wave.

(9) In Fig. 4(d), what is the origin of the finite spectral weight in the L-channel around E=0, including the energy gain side? I guess this could be an artifact due to the summation/subtraction of the raw data or a difficult extraction of the elastic line. In that case, I would enlarge the error bars. In general, I think it would be beneficial to show the raw RIXS spectra somewhere in the text or the supplement."

Our reply:

The peak appearing at 300 meV in Fig. 4(c) and the finite spectral weight in the L-channel around E = 0 in Fig. 4(d) in the original manuscript must be glitches or artifacts, since they are not observed in the updated Fig. 4 in the main text and Fig. R1 which is based on our new data set with higher statistics.

Summary of changes made

#1. Paragraph 1 in the revised version
In response to Referee #1's comment #2, we have revised as follows

Original version
"Further, we find using resonant inelastic x-ray scattering a complete breakdown of coherent magnon excitations at short-wavelength scales, suggesting a resonating-valence-bond-like quantum entanglement in the AF state."

Revised version
"Further, we find using resonant inelastic x-ray scattering a complete breakdown of coherent magnon excitations at short-wavelength scales, suggesting a many-body quantum entanglement in the AF state."

#2. Paragraph 9 in the revised version
In response to Referee #1's comment #2, we have deleted following discussion on RVB:

Original version
"This raises an intriguing possibility that a SN may arise from a smooth deformation of a RVB state that has non-zero singlet correlation for every NN pair spins. For instance, recent studies find that rotating one of the spins by 180° in every dimer in a singlet RVB leads to a triplet RVB, which inherits some of the generic properties of the RVBs, such as quasiparticles with fractional quantum numbers[29, 30]."

Revised version
Deleted

#3. Paragraph 14 in the revised version
In response to Referee #1's comment #2, we have revised as follows

Original version
"Rather, attributing the isotropic continuum to deconfined spinons in turn suggests that the quadrupolar order may be a byproduct of a RVB-like state."

Revised version
Deleted

#4. Paragraph 14 in the revised version
In response to Referee #1's comment #4, we have revised as follows

Original version
"Theoretically, SNs are predicted to arise in certain spin models with competing interactions[1,43-46]. For example, in a square-lattice, four-spin exchange (J_c) competing with ferromagnetic (FM) NN Heisenberg exchange (J) can lead to a SN[4,5]."

Revised version
"Theoretically, SNs are predicted to arise in certain spin models with competing interactions[1,41-48]. For example, in a square-lattice, four-spin exchange (J_c) competing with ferromagnetic (FM) NN Heisenberg exchange (J) can lead to a SN[2,5,6]."

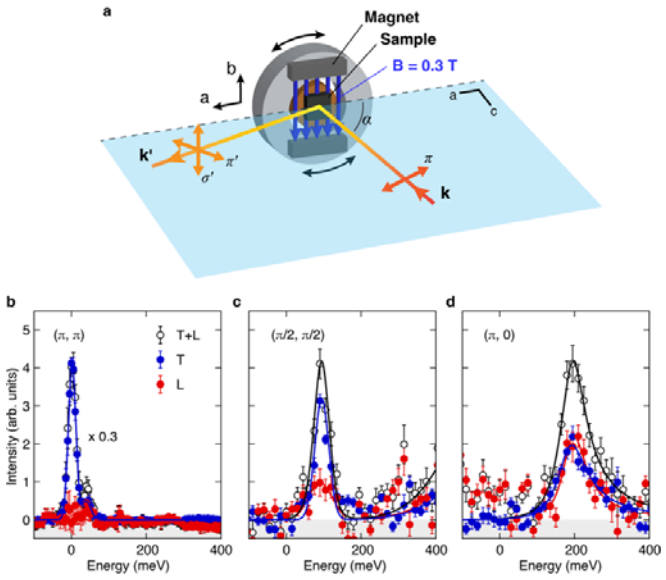
#5. Fig. 3 caption in the revised version
We have revised as follows

Original version
"**d,f,** Temperature dependence of the intensities (**d**) and energies (**f**) of A_{1g} and B_{2g} modes extracted by fitting the spectra to Fano profiles $I(\omega) = \frac{I_0}{\Gamma(1-q^2)}(1 - \frac{(q+\epsilon)^2}{1+\epsilon^2})$, where $\epsilon = (\omega - \omega_0)/\Gamma$, Γ is the linewidth, and q is the asymmetry parameter."

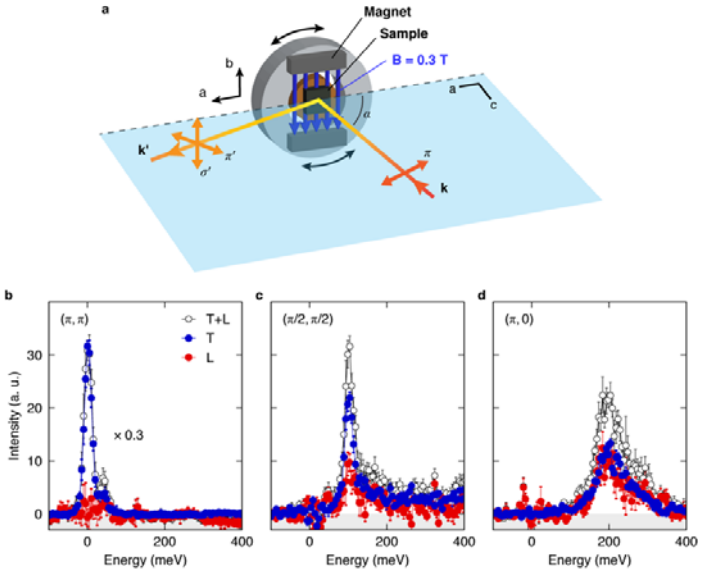
Revised version
"**d,f,** Temperature dependence of the intensities (**d**) and energies (**f**) of A_{1g} and B_{2g} modes extracted by fitting the spectra to Fano profiles $I(\omega) = \frac{I_0}{\Gamma(1-q^2)}(1 - \frac{(q+\epsilon)^2}{1+\epsilon^2})$, where $\epsilon = (\omega - \omega_0)/\Gamma$, Γ is the linewidth, and q is the asymmetry parameter. The intensity curves (**d**) are normalized by the low-temperature values."

#6. Fig. 4 in the revised version
In response to Referee #3's comments #13 and #14, Fig. 4 is replaced by higher statistics data.

Original version



Revised version



#7. Fig. 4 caption in the revised version
In response to Referee #2's comments #5 and #9, we have revised as follows

Original version
"The transverse mode (T) and the longitudinal mode (L) are displayed along with the sum of the two (T+L). Solid lines are guides to the eye. Each components are extracted from the raw spectra measured in different magnetic field directions with a fixed scattering geometry (see Extended Data Table 1 and Supplementary Note S6)."

Revised version
"The transverse mode (T) and the longitudinal mode (L) are displayed along with the sum of the two (T+L). Solid lines are guides to the eye. Each components are extracted from the raw spectra measured in different magnetic field directions with a fixed scattering geometry (see Extended Data Table 1 and Supplementary Note S6). The spectra measured at intermediate wavevectors between (π/2, π/2) and (π, 0) are displayed in Extended Data Fig. 3."

#8. Paragraph 13 in the revised version
In response to Referee #2's comment #10, we have revised as follows

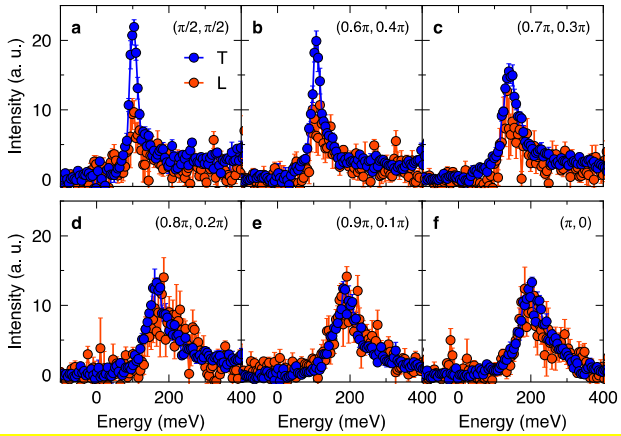
Original version
"By contrast, at (π, 0) the spectrum is completely isotropic and shows no sharp feature that can be identified as a magnon mode (Fig. 4d)."

Revised version
"By contrast, at (π, 0) the spectrum is completely isotropic and shows no sharp feature that can be identified as a magnon mode (Fig. 4d). We note that the zone-boundary RIXS spectra are hardly affected by the small magnetic field (see Supplementary Note S6). "

#9. Extended Data Fig. 3 in the revised version
In response to Referee #2's comments #5 and #9, we have added Extended Data Fig. 3 to the main text.

Original version
None

Revised version



Extended Data Fig. 3| Polarization-resolved RIXS spectra. a-f, RIXS spectra along the zone boundary from (π/2, π/2) to (π, 0) with the spin components transverse (T) and longitudinal (L) to the ordered AF moments resolved. The spectra were acquired with the same experimental setup in Fig. 4.

#10. Paragraph 3 of the Methods section in the revised version

In response to Referee #2's comments #5 and #9, and Referee#3's comments #13 and #14, we carried out additional RIXS experiments and accordingly modified the method section.

Original version

"RIXS spectra were measured at the 27-ID-B beamline of the Advanced Photon. Incident x-ray was tuned to the Ir L_3 edge (11.215 keV). Using a diamond (1 1 1) high-heat-load monochromator in combination with a Si (8 4 4) channel-cut monochromator reduced the energy bandpass down to 14.8 meV. The beam was then focused by a set of Kirkpatrick-Baez mirrors, producing a spot size of 40 x 10 (HxV) um^2 FWHM at the sample position. Scattered photons were analyzed by a Si (8 4 4) diced spherical analyzer with a radius of 2 m and with a mask of 2 cm diameter."

Revised version

"RIXS spectra were measured at the 27-ID-B beamline of the Advanced Photon Source and at the ID20 beamline of the European Synchrotron Radiation Facility. Incident x-ray was tuned to the Ir L_3 edge (11.215 keV). Using a diamond (1 1 1) high-heat-load monochromator in combination with a Si (8 4 4) channel-cut monochromator reduced the energy bandpass down to 14.8 meV. The beam was then focused by a set of Kirkpatrick-Baez mirrors, producing a spot size of 40 x 10 (HxV) um^2 FWHM at the sample position. Scattered photons were analyzed by a Si (8 4 4) diced spherical analyzer with a radius of 2 m and with a mask of 2 inch diameter for measurement at (π, π) and 3 inch for other momenta."

#11. Paragraph 15 in the revised version

In response to Referee #2's comment #6, #8, and #11, we have revised as follows

Original version

"In Sr_2IrO_4, evidence for a symmetry breaking order above T_N has been found in studies using second harmonic generation, inelastic neutron scattering, and magnetic torque measurements. These studies suggest loop currents as the possible order. While our result is inconsistent with such a time-reversal symmetry breaking order (see Extended Data Fig. 2), it is possible that these experiments are probing yet another order of different nature."

Revised version

"In Sr_2IrO_4, evidence for a symmetry breaking order above T_N has been found in studies using second harmonic generation, polarized neutron scattering, and magnetic torque measurements. These studies suggest loop currents as the possible order. While our result is inconsistent with such a time-reversal

symmetry breaking order (see Extended Data Fig. 2), it is possible that these experiments are probing yet another order of different nature (see Supplementary Note S7)."

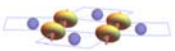
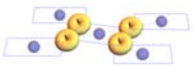
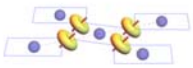
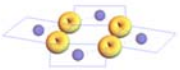
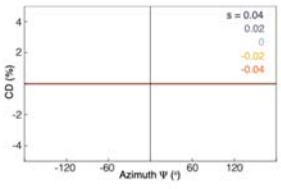
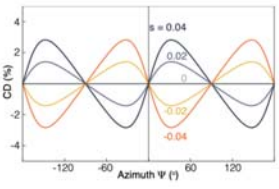
#12. Note S1 of the revised Supplementary Information
In response to Referee #3's comment #8, we have revised as follows

Original version
(Line 5) "where f is the tensorial atomic form factor"
(Line 10) "We omit the indices below for clarity."
(Line 12) "$I \propto |(F_1 + iF_2)(e_1 + ie_2)|^2$
(Line 28) "Finally, we note that $f_1^S(f_1^A)$ and $f_2^S(f_2^A)$ are interchangeable in Eq. S8 as in our case the structure factor is such that $f_1 = \pm f_2$."

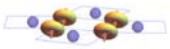
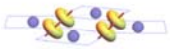
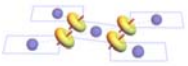
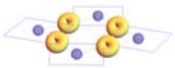
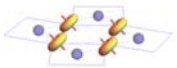
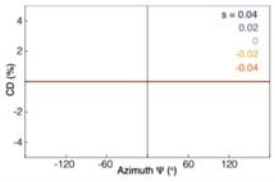
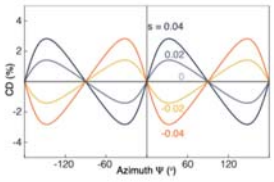
Revised version
(Line 5) "where f is the atomic form factor given by a 3 x 3 matrix"
(Line 10) "We omit the indices α and β below for clarity."
(Line 12) "$I \propto \left|F_{\alpha\beta}e_{\alpha\beta}\right|^2 = |(F_1 + iF_2)(e_1 + ie_2)|^2$
(Line 28) "Further simplification is possible when $F_1^S = \pm F_2^S$ and $F_1^A = \pm F_2^A$, in which case F_1^S (F_1^A) and F_2^S (F_2^A) can be interchanged. This is the case for $\mathbf{Q} = (0\ 0\ odd)$ reflections as explicitly confirmed in Table S4 for all possible quadrupole structures with non-zero structure factors."

#13. Fig. S4 of the revised Supplementary Information
In response to Referee #3's comment #8, we have revised as follows (a phase factor column $e^{i\mathbf{Q}\cdot\mathbf{r}}$ is added)

Original version

BC	Γ_{1u} (Q_{yz})		Γ_{1v} (Q_{zx})	
1	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, q_1)$	
2	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
3	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
4	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, q_1)$	
5	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, q_1)$	
6	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
7	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
8	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, q_1)$	
	$F^Q = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & c^*q_1 \\ 0 & c^*q_1 & 0 \end{pmatrix}$		$\begin{pmatrix} 0 & 0 & cq_1 \\ 0 & 0 & 0 \\ cq_1 & 0 & 0 \end{pmatrix}$	

Revised version

BC	$e^{i\mathbf{Q}\cdot\mathbf{r}}$	Γ_{1u} (Q_{yz})		Γ_{1v} (Q_{zx})	
1	1	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, q_1)$	
2	-1	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
3	i	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
4	-i	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, q_1)$	
5	-i	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, q_1)$	
6	i	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
7	-1	$(0, 0, 0, -q_1, 0)$		$(0, 0, 0, 0, -q_1)$	
8	1	$(0, 0, 0, q_1, 0)$		$(0, 0, 0, 0, q_1)$	
		$F^Q = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & c^*q_1 \\ 0 & c^*q_1 & 0 \end{pmatrix}$		$\begin{pmatrix} 0 & 0 & cq_1 \\ 0 & 0 & 0 \\ cq_1 & 0 & 0 \end{pmatrix}$	

#14. Note S2 of the revised Supplementary Information
In response to Referee #3's comment #7, we have revised as follows

Original version
"Here, we discuss other known origins of CD in RXD and show that they are irrelevant to our case. There are three physically distinct origins for CD: (i) a chiral lattice structure or a helical magnetic structure, where CD arises from a single type of scattering entity; (ii) interference between two distinct types of scatterers within E1-E1 process; (iii) higher-order processes such as electric dipole-magnetic dipole (E1-M1)."

Revised version
"Here, we discuss other known origins of CD in RXD and show that they are irrelevant to our case. In Fig. S1, we first compare the CD response of ferromagnetic (0 0 odd) reflections to that of antiferromagnetic (1 0 4n+2) reflections. The reflections (0 0 21) and (1 0 18) are chosen as to have similar scattering geometries, and the CD differences $I_{RC} - I_{LC}$ are normalized by the magnetic diffraction peak intensity. Unlike (0 0 21) reflection exhibiting a clear CD at the resonance, we find no indication of CD on (1 0 18) reflection. This rules out extrinsic sources of CD, such as birefringence.

As for intrinsic mechanisms, there are three physically distinct origins for CD: (i) a chiral lattice structure or a helical magnetic structure, where CD arises from a single type of scattering entity; (ii) interference between two distinct types of scatterers within E1-E1 process; (iii) higher-order processes such as electric dipole-magnetic dipole (E1-M1)."

#15. Fig. S1 of the revised Supplementary Information
In response to Referee #3's comment #7, we have added a new Fig. S1 to Supplementary Note S2

Original version
None

Revised version

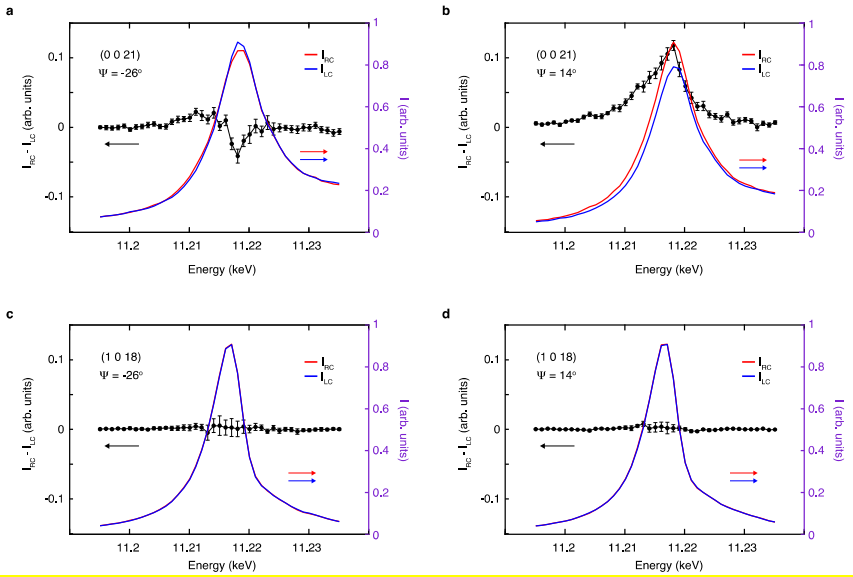
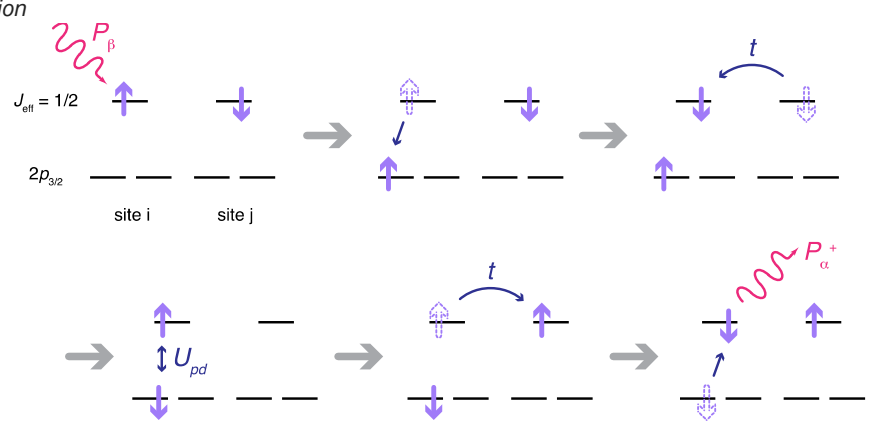


Fig. S1. **CD-RXD on magnetic reflections. a,b,** (o o odd) magnetic reflections exhibit a clear CD through interference between dipolar and quadrupolar moments, **c,d,** whereas the CD signals are not manifested for (1 o 4n+2) reflections of similar scattering geometry.

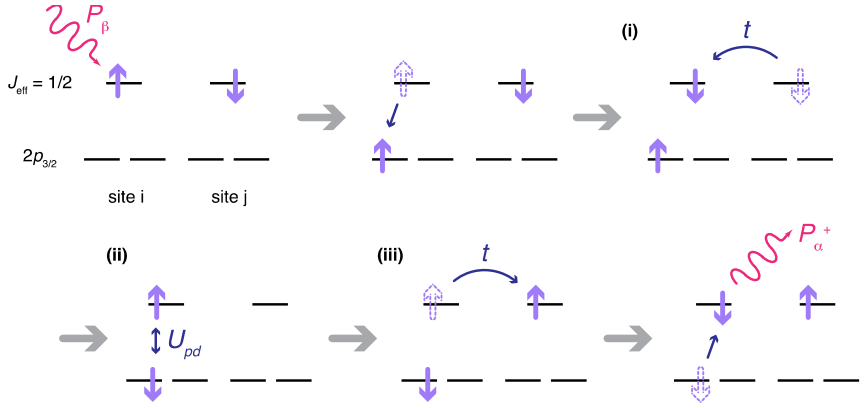
#16. Fig. S5 of the revised Supplementary Information

In response to Referee #3's comment #12, we have revised the figure as follows

Original version



Revised version



#17. Note S4 of the revised Supplementary Information

In response to Referee #3's comment #12, we have revised as follows

Original version

"The lowest-order RIXS process sensitive to bond-centered quadrupoles is depicted in Fig. S3, which involves hopping to a neighboring site, exchange scattering between 2*p* core hole and 5*d* valence electron, and hopping back to the original site, thus having an amplitude smaller than the on-site processes by a factor ~ $t^2 \frac{U_{pd}}{\Gamma^3}$:

$$\hat{R}_{\alpha\beta} \propto \sum_{\{m,m'\}} \left\langle i \middle| \hat{P}_\alpha^+ \frac{\hat{t}}{\Gamma} \middle| m \right\rangle \left\langle m \middle| \frac{\hat{U}_{pd}}{\Gamma} \middle| m' \right\rangle \left\langle m' \middle| \frac{\hat{t}}{\Gamma} \hat{P}_\beta \middle| i \right\rangle,$$

where $\hat{t}$ denotes electron hopping, $\hat{U}_{pd}$ denotes exchange Coulomb interaction between core and valence holes, and Γ is a core-hole lifetime. Alternatively, quadrupoles may be detected through inter-site photon-induced transitions (see Fig. S4), as discussed in Refs. 15, 16."

Revised version

"The lowest-order RIXS process sensitive to bond-centered quadrupoles is depicted in Fig. S4, which involves (i) hopping to a neighboring site, (ii) exchange scattering between 2*p* core hole and 5*d* valence electron, and (iii) hopping back to the original site, thus having an amplitude smaller than the on-site processes by a factor ~ $t^2 \frac{U_{pd}}{\Gamma^3}$. Specifically, the RIXS operator $\hat{R}_{\alpha\beta}$ reads:

$$\hat{R}_{\alpha\beta} \propto \sum_{\{m,m'\}} \left\langle i \middle| \hat{P}_\alpha^+ \frac{\hat{t}}{\Gamma} \middle| m \right\rangle \left\langle m \middle| \frac{\hat{U}_{pd}}{\Gamma} \middle| m' \right\rangle \left\langle m' \middle| \frac{\hat{t}}{\Gamma} \hat{P}_\beta \middle| i \right\rangle,$$

where $\hat{P}_\beta(\hat{P}_\alpha^+)$ corresponds to the dipole transition operator for x-rays with polarizations β (α); $|i\rangle$ is the initial state, and $|m\rangle, |m'\rangle$ are intermediate states; $\hat{t}$ denotes electron hopping, $\hat{U}_{pd}$ denotes exchange Coulomb interaction between core and valence holes, and Γ is a core-hole lifetime. The energy denominators of this perturbation expansion are replaced by Γ using the fast-collision approximation [16, 17]. Alternatively, quadrupoles may be detected through inter-site photon-induced transitions (see Fig. S4), as discussed in Refs. 15, 16."

#18. Note S6 of the revised Supplementary Information

We have revised as follows

Original version (paragraphs 5 and 6)

"In our experiment, we choose to measure RIXS in a normal-incidence geometry varying the field direction. We fixed the scattering geometry because changing the scattering geometry significantly varies the x-ray footprint on the sample which makes a direct comparison of intensities between the spectra difficult. Also, since two spectra obtained in a grazing-incidence geometry practically provide the same polarization components (Extended Data Table 1), we decided to conduct RIXS in a normal geometry which can provide two spectra of T+T′ and T′+L. Since the in-plane (T) and out-of-plane (T′) transverse modes at short wavelengths are indistinguishable for both magnons and fractionalized excitations, we can approximate T+T′ and T′+L to 2T and T+L, respectively. Then, the pure transverse (T) and longitudinal mode can be obtained by subtraction/addition of the raw spectra.

We choose **q** vectors around (3 0 28.5) in a unit cell with lattice parameters of a = b = 5.5 A and c = 25.8 A; (3 0 25.8), (3.5 0 25.8) and (3.5 0.5 25.8) are selected for (π, π), (π/2, π/2) and (π, 0), respectively. It should be noted that the two-dimensional wavevectors are expressed in a tetragonal unit cell to match the notations to those of cuprate compounds as in previous RIXS studies on Sr_2IrO_4. A large H value is required to make the incident angle close to normal (90°). For these **q** vectors, the incident angle was around 75°, which gives ~ 6.7% of unwanted leakage ($\sin^2(15°)$ ~ 0.067) to our raw RIXS spectra. Therefore, we calculate the portions of the polarization components (S_{aa}, S_{bb}, S_{cc}) from polarization channels, π x π′ and π x σ′, for each raw spectrum,"

Revised version

"In our experiment, we choose to measure RIXS in a normal-incidence geometry varying the field direction. We fixed the scattering geometry because changing the scattering geometry significantly varies the x-ray footprint on the sample which makes a direct comparison of intensities between the spectra difficult. Also, since two spectra obtained in a grazing-incidence geometry practically provide the same polarization components (Extended Data Table 1), we decided to conduct RIXS in a normal geometry which can provide two spectra of

T+T′ and T′+L. Since the in-plane (T) and out-of-plane (T′) transverse modes at short wavelengths are virtually identical, we can approximate T+T′ and T′+L to 2T and T+L, respectively. Then, the pure transverse (T) and longitudinal mode can be isolated by subtraction/addition of the raw spectra.

We choose **q** vectors around (3 0 28.5): (3 0 25.8), (3.5 0 25.8) and (3.5 0.5 25.8) are selected for (π, π), (π/2, π/2) and (π, 0), respectively. A large H value is required to make the incident angle close to normal (90°). For these **q** vectors, the incident angle was around 75°, which gives ~ 6.7% leakage ($\sin^2(15°)$ ~ 0.067) in our raw RIXS spectra. Thus, the contributions to each components (S_{aa}, S_{bb}, S_{cc}) from the two polarization channels, π x π′ and π x σ′, are calculated as"

#19. Fig. S8 of the revised Supplementary Information
In response to Referee #3's comment #14, we have added a new figure to Supplementary Note S6 and revised the note accordingly

Original version
None

Revised version

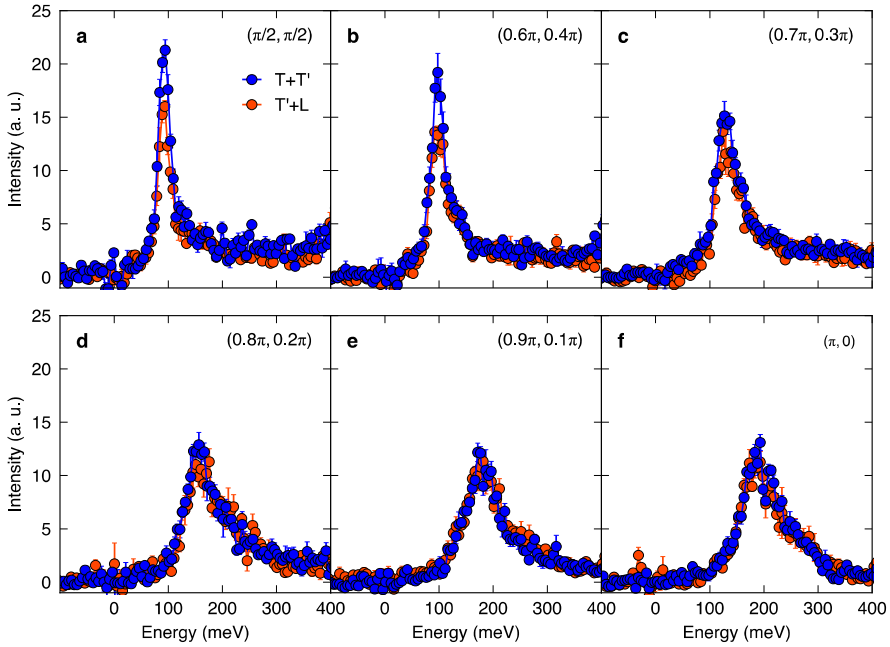


Fig. S8. **Raw RIXS spectra of Sr_2IrO_4. a-f,** The raw RIXS spectra measured along the magnetic zone boundary from (π/2, π/2) to (π, 0). Elastic peaks and constant backgrounds have been removed by fittings. All spectra are normalized to the intensity of electronic excitations (10*Dq*). The error bars are the standard errors of multiple measurements.

Figure S8 shows the raw RIXS spectra after removing the elastic peaks by fitting. With the vertical (horizontal) magnetic field, the spectrum approximately measures the T+T′ (T+L) components. From the (π/2, π/2) spectra, it is seen that the peak in the T+T′ channel is much larger than that in the T+L channel due to the single magnon in the T channel. Upon approaching (π, 0), the difference between the two channels gradually decreases, and eventually become identical at (π, 0). The error bars in Fig. 4 in the main text were determined by the standard propagation of uncertainty.

#20. Fig. S9 of the revised Supplementary Information
In response to Referee #2's comment #10, we have added a new figure to Supplementary Note S6 and revised the note accordingly

Original version
None

Revised version

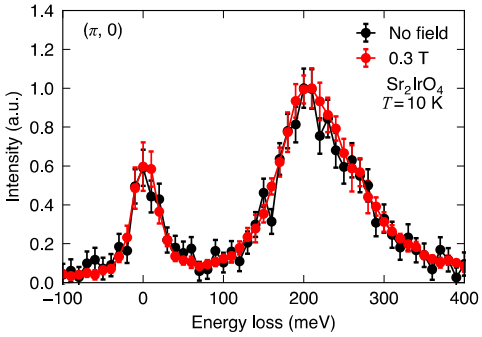


Fig. S9. **Field dependence of RIXS spectrum.** The (π, 0) spectrum of Sr_2IrO_4 is measured with (black) and without (red) an external magnetic field of 0.3 T. The two raw spectra show no difference within the error bars.

We note that a small magnetic field we used here (~ 0.3 T) does not change magnetic excitations in RIXS measurements as demonstrated in Fig. S9, since the magnitude of the in-plane spin exchange (*J*) in Sr_2IrO_4 is about 60 meV while a field of 0.3 T corresponds to 57 ueV for a 1 uB spin.

#21. Note S7 of the revised Supplementary Information
In response to Referee #2's comments #6, #8, and #11, we have added a new supplementary note

Original version
None

Revised version
7. Spin nematic order and the hidden order
The SN order discovered in our work is symmetry distinct from that of a loop current (LC) order, originally suggested to account for the pseudogap in cuprates and much discussed recently in Sr2IrO4 as a possible origin of the hidden order. Below, we have summarized the previous reports based on second harmonic generation (SHG), polarized neutron diffraction (PND), torque magnetometry (TM), and elastoresistance (ER) measurements, and compared them with our results.

	Observation	**Interpretation**	**Inconsistency with our data**
SHG	• Breaking of inversion and rotational symmetries at T_{HO}~243 K in undoped Sr_2IrO_4, and at T_{HO}~210 K for 4% Rh-doped sample.	• Symmetry consistent with magneto-electric loop current • Reinterpreted as due to surface magnetization induced electric-dipole SHG process based on additional experiments in magnetic field (for undoped sample)	• T_{HO} and T_C different by ~ 20 K (for undoped sample). • Inconsistent with PND and TM (ferroic stacking along *c*-axis).
PND	• Spin-flip scattering at **Q** = (1,1,2) setting in at T_{HO} ~ 240 ± 30 K for both parent and 7% Rh-doped samples.	• Symmetry consistent with magneto-electric loop current (but of different type from the one suggested by SHG)	• **Q** vector different from our x-ray data; CD is observed at (0 0 odd) • T_{HO} ~ 240 K is much lower than our T_C ~ 263 K. • Our magneto-optical Kerr effect data rules out any time-reversal breaking order.

TM	• Broken C_4 symmetry at T_{HO} ~ 270 K for the parent compound.	• Nematic phase transitions (but distinct from nematic orders reported in other systems, which have even parity)	• Consistent with our data • T_{HO} ~ T_C
ER	• Absence of divergent behavior in the nematic susceptibility, which suggests odd parity.	• Together with the magnetic torque data, the hidden order is suggested to be a loop current order.	• The data is available only for Rh-doped samples.

TABLE S5. **Comparison to previous reports of hidden order in Sr_2IrO_4**

#22. Author list and acknowledgements

In response to Referees' comments, we carried out additional RIXS measurements at ESRF. The author list and the acknowledgements are modified accordingly.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

The authors have properly addressed my previous concerns. It is also worth pointing out to which extent the revised manuscript has now been improved to fit all the refere's comments and suggestions. I can, thus, recommend publication in Nature.

Referee #2 (Remarks to the Author):

I believe that Kim et al have provided a detailed and thorough response to the potential issues and concerns raised in the three original referee reports. I was very impressed by the comprehensiveness of the response, and in particular, I would like to acknowledge the considerable amount of work that the authors must have put into obtaining additional experimental data to support their arguments.

I believe that this revised manuscript is considerably stronger than the original. The revisions to the main text, combined with the expanded supplementary material, have addressed all of the major concerns raised in my report. I am happy to change my initial assessment and recommend this manuscript for publication.

Referee #3 (Remarks to the Author):

I have assessed the revised manuscript and the authors' reply to my comments.

In my previous report, I had concerns regarding:
(1) The amplitude of the dataset that had room for improvement
(2) The low quality of the RIXS data
(3) The insufficient description of technicalities, mainly in the supplement
(4) The possible presence of extrinsic effects in the RXRD signal

To my satisfaction, the authors answered extensively to all my comments and addressed all the concerns listed above.
In particular, this led to additional experimental efforts to extend the dataset, improve the data quality and rule out the presence of artifacts.

In my opinion, the manuscript is now improved and the data supports better the conclusions.
I can now recommend the manuscript for publication in Nature.