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Supplementary information

Angular momentum generation in nuclear fission

In the format provided by the authors and unedited

Peer Review File

Manuscript Title: Angular momentum generation in nuclear fission

Editorial Notes:

Redactions – reviewer opt-out

Parts of this Peer Review File have been redacted as indicated as we could not obtain permission to publish the reports of a reviewer.

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Reviewer #1 provided confidential comments to the editor. [REDACTED].

Referee #2 (Remarks to the Author):

The manuscript reports on an experimental investigation on the origin of angular momentum (spin) in fission fragments.
By measuring and identifying the gamma rays emitted by the fission fragments, the authors are able to reconstruct the average spin of the fragments as a function of their mass and charge. They investigate three reactions: neutron induced fission in 232Th and 238U, as well as 252Cf spontaneous fission. These are all actinides close to the valley of stability which are known to fission asymmetrically with a heavy and a light fragment.
The proposal is to use these average spin to identify the mechanism for angular momentum production in the fragments. This is certainly an important question in nuclear physics. In addition, the authors claim that this will impact applications in nuclear technologies is genuine.
Several theoretical scenarios have been proposed in the past as possible mechanism producing angular momentum in the fragments. Some models predict that the mechanism occurs before scission (eg, collective vibration of the parent nucleus on its way to scission), while others proposes post-scission mechanisms, such as Coulomb repulsion between the fragments.
Here, the proposal is to identify when the angular momentum is produced by investigating the correlations between the spins of the two fragments. If correlations are observed, then a pre-scission mechanism is more likely. This is done by setting conditions on the minimum spin of the populated states in one fragment while measuring the average spin of the other. The authors argue that their observations show that there is no correlation between the spins of the fragments and thus it must be originated after scission. Then they propose a simple mechanism which would explain this observation while reproducing the observed dependence of the spin versus the fragment mass.
If correct, this would be an important step towards our understanding of the fission mechanism. However I have several concerns with the present interpretations which would need to be addressed before I can form a recommendation on this work.

1) Figure 2 (wrongly labelled fig 1 in the manuscript) shows that the average spin of one fragment does not depend (in first order) on the minimum spin of the other fragment. The authors conclude that the spins of both fragments are uncorrelated. The minimum spin is determined by setting a gate in the gamma ray spectrum, thus ensuring that the fragment has indeed been produced with a larger (or equal) spin than the spin of the yrast state used in the gate. However, this minimum spin needs to be large enough for the correlation to be probed. Consider, as an example, that the fragment is produced with an average spin of 8, with a distribution ranging from 6 to 10. In this case, all the decays are expected to go through yrast states of spins 6 and lower. Thus, setting a

minimum spin between 0 and 6 is not expected to impact the distribution of spins of the other fragment. However, if the minimum spin is set in this example to be 8 or 10, then the correlations, if they exist, could be potentially observed with the average spin of a fragment deviating from a constant at the largest values of the minimum spin of its partner.
Let us look at each of the 6 fragments studied in Fig 2:
- 146Ba: its partner (90Kr) has $<I>\sim5.5$ but the largest minimum spin considered is 6 which may be too closed to $<I>$ to conclude.
- 140Xe: its partner (96Sr) has $<I>$ almost 6. As expected, the $<I>$ of 140Xe is flat until I_{min} reaches 6. If one draws a constant line across the first 3 points, we then see that the points at $I_{min}=8$ and 10 are lower than this line. This may not be significative due to the experimental uncertainties, but I would argue that this seems to indicate a possible correlation.
- 136Te and 100Zr are each other's partner with a $<I>\sim6$. Again, with respect to a constant line going through the first 3 points, we see a small deviation for the points at $I_{min}=8$ and 10. In this case the average spins seem to go up.
- 96Sr and 90Kr both have partners (respectively 140Xe and 146Ba) with $<I>\sim8$. So one would need to look at $I_{min}=10$ or greater to look for a correlation. As there is only one point (for 96Sr) obeying this condition, it is hard to conclude.
To summarise, (unless I missed something), I don't think there are enough points with large enough I_{min} to draw a definitive conclusion with respect to the correlations. I understand that statistics is a limiting factor for large I_{min}. However, without it, I don't see how one can conclude on the correlations.

2) My second point is in fact related to the first one. In the inset of Fig 2, it is shown that the slope of $<I>$ versus I_{min} of the partner is compatible with zero. As explained above, this is expected and, unless I am mistaken, it is not a signature for no correlation. A more sensitive observable to correlations would be the slope I_{min} greater than $<I>$ (both for the partner). I also don't see how the pre-scission mechanism (a) would lead to such a large slope.

3) The proposed mechanism is that fragment spins are generated post-scission by independent torques. As mentioned by the authors, the total angular momentum is conserved, and thus $I_1+I_2+I_0=0$ ($I_{1,2}$ are spins of the fragments and I_0 is the orbital angular momentum between the fragments). In terms of the corresponding quantum numbers, this requires $|I_1-I_2| < I_0 < I_1+I_2$. Thus I_0 could reach up to ~14hbar. If the spin of the fragment is "decided" post-scission, so must be I_0. I don't see how such a relatively large orbital angular momentum could be produced between 2 fragments which are not in contact anymore.

4) The simple model is based on the shape of the fragments at scission. The number of nucleons in the neck is in fact a measure of the deviation of the shape to a spherical equilibrated shape, and thus it quantifies the "restoring force". There are several theoretical tools which are able to predict the shape of a system at scission. This would certainly provide a better ground to the model than the simple assumption of a magic core plus neck nucleons. Moreover, there is a strong argument against the presence of 78Ni and 132Sn magic cores surrounded by neck nucleons: 78Ni has N/Z=1.79, 132Sn has N/Z=1.64, leaving 14 protons ands 14 neutrons in the neck with N/Z=1. Spherical magic shell effects are simply not strong enough to sustain such an asymmetric distribution of N/Z.

5) The message carried by Fig 4 is not clear to me. If I understand correctly, the predicted average spin is based on the model which itself has been fitted on the data. Thus the figure is expected to simply show a quality of the fit. However, this was already shown in Fig 1. Also, my understanding is that to get the pre-neutron emission (right panel), one shifts each point by a number (the same for both axis) corresponding to the amount of angular momentum carried away by the neutrons. Both panels have similar agreement between predicted and measured quantities so perhaps only one is needed.

To conclude the manuscript presents an interesting study to an important problem. I have no doubt that the measurements, in particular the data shown in Fig 1, will be of strong interest to the community. However, I have raised some concerns regarding the interpretation of the results. These concerns need to be addressed before I can make a recommendation.

Referee #3 (Remarks to the Author):

This manuscript discusses recent experimental results obtained at the ALTO facility in Orsay using the LICORNE neutron source, on gamma rays observed in the fission fragments produced in the spontaneous fission of 252Cf and in the neutron-induced fission reactions of 238U and 232Th for incident neutron energies centered at 1.9 MeV. Using a data analysis first developed at the University of Manchester in the late 1980s, dubbed "MSM" for "Manchester Spin Method", and using g-g coincidences, the authors infer an average angular momentum value as a function of the fission fragment mass. In the case of 252Cf (sf), the authors rely on the additional tagging of one of the two fission fragments, while for the (n,f) reactions on 238U and 232Th, they use a pulsed neutron beam to eliminate undesirable background due in particular to long-lasting beta-delayed gamma emissions. Three main results are presented as significant:

1. The reported coincidence data support the saw-tooth behavior of the average fragment spin <J> vs the mass of the fragment (after neutron emission), already reported in a limited set of experimental results since the 1970s.
2. Their results and conclusions are mostly independent of the fissioning system studied.
3. The observed lack of correlation between the inferred average angular momenta for the two complementary fragments is used to propose a simple model, and a fit to the experimental data, that explains the production of spin in the fragments post-scission only.

The data analysis of the very complex g-g correlation matrices is performed through the use of the (publicly available) Radware software package. This analysis is limited to about 30 even-even post-neutron emission fission fragments, for which the nuclear structure is much better known than for their odd-even and odd-odd neighbors.

While this work is certainly of interest to the nuclear fission community, it is difficult to make the case that it is of such "outstanding scientific importance" that it merits a publication in this journal. The claim that the present results would also impact the fields of nuclear reactor physics, nuclear structure for very neutron-rich isotopes, and super-heavy elements, is also difficult to support. That aside, I believe that the presented methodology and results lack the necessary rigor and assessment (and elimination) of potential systematic biases, including a serious attempt at quantifying uncertainties, that could impair the main conclusions of this work.

I will now go in some details to explain those specific technical points. In no particular order:

• While the authors briefly mention previous experimental results, there is no discussion on what could have caused those results to either agree or disagree with the present results. It would have been of great interest to the reader to understand why such a controversy still exists. The results on the saw-tooth behavior presented here are not original, but confirm earlier selected results.
• Figure 2 (mis-labelled Fig.1!): how are the (a) band and (b) line in the inset plot obtained? A vague sentence is given in the caption, but with no information on the assumptions and code used to obtain those results. The interpretation is not put into question as a slope consistent with zero should indeed correspond to a post-scission mechanism with uncorrelated spins, but more details would be useful to understand how this quantitative result was obtained.
• The simple model of the "snapping of a taut elastic band" introduced by the authors is interesting but obviously very schematic. It is very similar to the "random neck rupture" model introduced by Brosa et al to explain the saw-tooth behavior of the prompt neutron multiplicity as a function of the fragment mass. There, the physical intuition was based on the Rayleigh instabilities experienced by a fluid given a certain surface-to-volume ratio constraint. The physical intuition

presented here is less clear as there is no distinction made between the translation kinetic and rotational energies expected from the snapping of this stretched elastic. The authors offer the idea that fluctuations in the direction of motion of the nascent fragments are what cause the development of two independent torques. This is probable, similar to the idea presented in their reference [9], but the lack of discussion of translational vs rotational energies makes it difficult to analyze this idea further. While the kinetic energy of the fragments was not recorded in the present experiments, some results of TKE vs fragment mass are well known, and could have been used to discuss this interesting question.

• The idea briefly mentioned that "quantum entanglement" between the moving fragments could be witnessed is far-fetched, to say the least, given the exceptional conditions (and isolation from any disturbing environment) that the production of entangled elementary particles require.

• Under Consequences, the authors mention the issue of gamma-ray heating in a nuclear reactor as a potential issue that their current results addresses. They also mention "delayed g rays from isomeric states which contribute to the decay heat after reactor shutdown." This is true but not coming from the fission fragments prior to beta-decay, so it isn't relevant to the present discussion. To carry out the post-neutron emission calculation of the mass weighted averages shown in Fig.4 requires the knowledge of neutron multiplicity per fragment mass, which is well-known in the case of 252Cf (sf) only. What was used for the two other fissioning systems at the 1.9 MeV incident neutron energy for which no measurement exist (to my knowledge). What is of course of interest to the reactor physics community is essentially the heat/energy deposited by g rays produced in those fission reactors, which is a step beyond what is discussed here.

• One of the main limitations of the present approach is that only even-even fragments are considered, which in many cases, is a small subset of all fragments produced. The extension of (or at least an attempt to extend) the conclusion presented here to all fragments is lacking.

• "The major strength of the MSM is that it uses all the available g-ray intensity and coincidence data (…) and condenses this information into a single number, the average spin after neutron emission." I would not call this a "strength" since it means many details of the decay sequence are lost in the process, and several assumptions have to be made to get to this result. And no information on the distribution of spins can be obtained.

• The conclusion that the Yrast sequence of transitions is the only information needed to obtain an average initial spin is troubling, but could be correct for the simplified case of even-even fragments. This result is presented as the conclusion of a numerical sensitivity study but its interpretation is not pursued further.

• The validity of the MSM approach is "demonstrated" through the comparison of fusion-evaporation vs neutron transfer reactions. The spin difference in those cases is massive, and it is unclear how this example demonstrates that the MSM method is sensitive enough to distinguish average spin values between different fragmentations, separated by only a fraction of hbar.

• The crux of the analysis presented in this paper, and therefore its main conclusions, is obtained through the use of the Radware software package. A very large global fit is performed with "thousands of parameters." No discussion of uncertainties is presented at any point in the publication. Such a global fit should come with specific metrics, or figure of merit, and quantifiable uncertainties associated with those, included strong correlations among the many parameters. The authors only mention the "powerful visual comparisons" and "visual checks" that can be performed. While this is definitely useful, it is certainly impossible to go through all g-g correlation data obtained in this work. The systematic study and reporting of those uncertainties is simply lacking.

• Under the "Determination of the side-feeding of the first 2+ state," the authors mention a possible experimental bias undetected in their recent analysis of fission yield anomalies in the 238U(n,f) reaction. More than just the side feeding issue addressed here, other reasons such as biasing due to a high-multiplicity gating on near-spherical nuclei could also cause some misinterpretations. This was covered in a recent paper by Fotiades et al (Phys. Rev. C 99, 024606 (2019)). Similar assumptions are being made in the present paper, although the experimental setup is different and the conclusions probably not impacted as much by those assumptions. However, assuming that the ground-state feeding is zero for all fragments is certainly wrong. The quantitative impact of such assumptions is difficult to obtain without performing a comprehensive set of calculations that uses all known information about the nuclear structure and decay schemes of the fragments.

Author Rebuttals to Initial Comments:

Response to the referees

We would like to sincerely thank both referees for their thorough work in reviewing and evaluating our manuscript and for their detailed comments which are very pertinent and well justified. Their work has provided valuable tests of the strength and validity of the methods employed in the data analysis and the robustness of the conclusions and interpretations we draw from our experimental data. These comments have helped us to make a significantly improved and substantially revised manuscript.

During the revision process we have received additional theoretical support (see acknowledgements) and have developed a parallel microscopic description of the mechanism of intrinsic spin generation in fission which fits well our experimental data and gives us additional confidence in the interpretation. Before we address all the specific points and concerns raised we present an overview of the changes that have been made.

Overview of the revisions

(The colour scheme employed here corresponds to that of the highlighted changes in the manuscript)

Main article

- Introduction: addition of a new paragraph giving a brief history of previous experimental studies
- Discussion: Extensive revisions and presentation of a parallel microscopic theoretical description
- Discussion (end): A step-by-step summary describing the intrinsic spin generation mechanism incorporating both macroscopic and microscopic perspectives
- Figures: Application of corrections for the 0+ feeding to the data and fits in figure 1
- Figures: Revisions of figure 1 and figure 4

Methods:

- Application of the MSM to nu-ball data: Removal of text describing other reactions and the corresponding Data Table 1 to save space
- Fitting procedure and statistical uncertainties: Significant expansion of this section to include this crucial information on the analysis, uncertainties, and their propagation
- Addition of new section entitled “Sensitivity of the MSM”
- Ground state feeding: Revision of this section to explain new corrections to the data
- Non-inclusion of weak transitions: Additional quantitative information on the impact of inclusion/exclusion of non-yrast transitions

Extended Data:

- Addition of new figure 7 showing results from Monte-Carlo simulation of the experimental signature of correlated spins at scission
- Addition of new figure 8 showing examples of measured spin distributions and the fitted extrapolation of these distributions to extract values for the 0+ feedings

Supporting Information:

- Derivation of the functional form for the dependence of fragment spin on mass based on statistical theory and the Fermi gas model
- Description of the Monte-Carlo code developed to simulate the experimental signatures of correlated spins at scission

Referee #2 (Remarks to the Author):

The manuscript reports on an experimental investigation on the origin of angular momentum (spin) in fission fragments.
By measuring and identifying the gamma rays emitted by the fission fragments, the authors are able to reconstruct the average spin of the fragments as a function of their mass and charge. They investigate three reactions: neutron induced fission in 232Th and 238U, as well as 252Cf spontaneous fission. These are all actinides close to the valley of stability which are known to fission asymmetrically with a heavy and a light fragment.
The proposal is to use these average spin to identify the mechanism for angular momentum production in the fragments. This is certainly an important question in nuclear physics. In addition, the authors claim that this will impact applications in nuclear technologies is genuine.
Several theoretical scenarios have been proposed in the past as possible mechanism producing angular momentum in the fragments. Some models predict that the mechanism occurs before scission (eg, collective vibration of the parent nucleus on its way to scission), while others proposes post-scission mechanisms, such as Coulomb repulsion between the fragments.
Here, the proposal is to identify when the angular momentum is produced by investigating the correlations between the spins of the two fragments. If correlations are observed, then a pre-scission mechanism is more likely. This is done by setting conditions on the minimum spin of the populated states in one fragment while measuring the average spin of the other. The authors argue that their observations show that there is no correlation between the spins of the fragments and thus it must be originated after scission. Then they propose a simple mechanism which would explain this observation while reproducing the observed dependence of the spin versus the fragment mass.
If correct, this would be an important step towards our understanding of the fission mechanism. However I have several concerns with the present interpretations which would need to be addressed before I can form a recommendation on this work.

We are glad the referee agrees with the potential significance of the work and interest for the general reader and we are confident we can address his/her concerns expressed about the interpretations and the theoretical description of the mechanism.

1) Figure 2 (wrongly labelled fig 1 in the manuscript) shows that the average spin of one fragment does not depend (in first order) on the minimum spin of the other fragment. The authors conclude that the spins of both fragments are uncorrelated. The minimum spin is determined by setting a gate in the gamma ray spectrum, thus ensuring that the fragment has indeed been produced with a larger (or equal) spin than the spin of the yrast state used in the gate. However, this minimum spin needs to be large enough for the correlation to be probed. Consider, as an example, that the fragment is produced with an average spin of 8, with a distribution ranging from 6 to 10. In this case, all the decays are expected to go through yrast states of spins 6 and lower. Thus, setting a minimum spin between 0 and 6 is not expected to impact the distribution of spins of the other fragment. However, if the minimum spin is set in this example to be 8 or 10, then the correlations, if they exist, could be potentially observed with the average spin of a fragment deviating from a constant at the largest values of the minimum spin of its partner.
Let us look at each of the 6 fragments studied in Fig 2:
- 146Ba: its partner (90Kr) has ~5.5 but the largest minimum spin considered is 6 which may be too closed to to conclude.
- 140Xe: its partner (96Sr) has almost 6. As expected, the of 140Xe is flat until I_min reaches 6. If one draws a constant line across the first 3 points, we then see that the points at I_min=8 and 10 are lower than this line. This may not be significative due to the experimental uncertainties, but I would argue that this seems to indicate a possible correlation.
- 136Te and 100Zr are each other's partner with a ~6. Again, with respect to a constant line going through the first 3 points, we see a small deviation for the points at I_min=8 and 10. In this case the average spins seem to go up.
- 96Sr and 90Kr both have partners (respectively 140Xe and 146Ba) with ~8. So one would need to look at I_min=10 or greater to look for a correlation. As there is only one point (for 96Sr) obeying this condition, it is hard to conclude.
To summarise, (unless I missed something), I don't think there are enough points with large enough I_min to draw a definitive conclusion with respect to the correlations. I understand that statistics is a limiting factor for large I_min. However, without it, I don't see how one can conclude on the correlations.

The referees reasoning and conclusions would be correct, however, the example chosen of a fragment produced with an average spin of 8+/-2 ħ is not realistic. The spin distributions extend down to zero spin for almost all fragments. We have added figure 8 in the extended data to illustrate this. Significant 2+ and 4+ direct side-feedings are observed for almost all nuclei. The chosen example is also not consistent with what is already known about prompt fission gamma-ray multiplicity distributions which generally extend down towards zero. Spins of 6-10 units in both fragments would imply a minimum multiplicity of at least 8 gamma-rays assuming at least one statistical feeding transition and three stretched E2 transitions along the yrast line in each nucleus.

2) My second point is in fact related to the first one. In the inset of Fig 2, it is shown that the slope of versus I_min of the partner is compatible with zero. As explained above, this is expected and, unless I am mistaken, it is not a signature for no correlation. A more sensitive observable to correlations would be the slope I_min greater than (both for the partner). I also don’t see how the pre-scission mechanism (a) would lead to such a large slope.

The referee is right to question thoroughly the conclusions we draw from the lack of significant slopes observed in figure 2, and we ourselves were initially surprised by this result. Therefore, to understand fully what we would expect to see experimentally for correlated and spins at scission we performed Monte-Carlo simulations which are described in the supporting information section, along with a new figure 7 in the extended data. Hopefully, here it becomes clear that gating in one fragment is truncating its spin distribution at yrast and places greater and greater constraints on the distribution at scission which would strongly affect the yrast distribution in the partner if the spins were correlated.

3) The proposed mechanism is that fragment spins are generated post-scission by independent torques. As mentioned by the authors, the total angular momentum is conserved, and thus I_1+I_2+I_0=0 (I_1,2 are spins of the fragments and I_0 is the orbital angular momentum between the fragments). In terms of the corresponding quantum numbers, this requires | I_1-I_2 | < I_0 < I_1+I_2. Thus I_0 could reach up to ~14hbar. If the spin of the fragment is “decided” post-scission, so must be I_0. I don’t see how such a relatively large orbital angular momentum could be produced between 2 fragments which are not in contact anymore.

We agree. The potential for large orbital angular momentum generation is a natural consequence of a post-scission, uncorrelated mechanism. The range of quantum numbers for I_0 is indeed correct and furthermore we can deduce that the distribution of I_0 will have a tail extending to large values. The orbital angular momentum (i.e. the relative angular momentum of the fragments with respect to each other) does not require fragment contact to be defined. In fact, we argue that fragments in contact can only produce correlated spins (with $I_0 = 0$) as they push off each other. Orbital angular momentum will also be accompanied by a corresponding orbital rotational energy, E_0, although this will be very small due to the large moment of inertia of the pair of separated fragments. Since orbital angular momentum is an inferred rather than a measured quantity, we do not see the possibility of large values occurring as inherently problematic.

We also point out that in figure1, the observation of large average spin asymmetries of over 6 units which occur for certain fragment pairs provides additional evidence of both non-zero orbital angular momentum and independent spin generation. Finally, the observation that average spin does not depend on the mass of the partner fragment (which can differ by up to 25% between the 3 systems) also suggests a similar conclusion. The data presented in figure 2 confirms this.

4) The simple model is based on the shape of the fragments at scission. The number of nucleons in the neck is in fact a measure of the deviation of the shape to a spherical equilibrated shape, and thus it quantifies the “restoring force”. There are several theoretical tools which are able to predict the shape of a system at scission. This would certainly provide a better ground to the model than the simple assumption of a magic core plus neck nucleons.

We recognise that our double-cluster plus neck description (and elastic band analogy) is schematic and that is why we call it a parametrisation to fit the data rather than a model, although it has the advantage of being easily accessible to the general reader and contains the essential ingredients of the problem. We have now revised the discussion section (with additional support from theorists) to develop an additional parallel microscopic description and its equivalent parametrisation which we use to better fit the data. The functional form derived from statistical theory turns out the be very similar to that obtained from our schematic macroscopic description. Its derivation can be found in the supporting information. That the two different approaches produce similar results give us added confidence in the interpretation. Nonetheless, our work is still mainly experimental work, and it’s clear that none of the many theories previously developed to explain the origin of spin in fission appear to be confirmed by our results, which we argue is the principal impact. A step-by-step summary of the mechanism (highlighted in red) can now be found at the end of the discussion.

Moreover, there is a strong argument against the presence of 78Ni and 132Sn magic cores surrounded by neck nucleons: 78Ni has N/Z=1.79, 132Sn has N/Z=1.64, leaving 14 protons and 14 neutrons in the neck with N/Z=1. Spherical magic shell effects are simply not strong enough to sustain such an asymmetric distribution of N/Z.

Spherical shell effects do have the power to distort the N/Z ratio, but as the referee rightly points out, in the case of 78Ni, they are simply not strong enough to pull the fragment distribution to such an extreme N/Z. This is why we are very careful in the article to refer to “Sn-like” and “Ni-like” fragments. For the choice of the exact masses used in the fitting parametrisation, we used 78 and 130. To achieve the appropriate N/Z balance in the neck we could easily chose ^{82}Ge as the attractor for the lighter masses. Choosing slightly different values would not impact the conclusions of the paper. If we allow the two mass parameters to float freely we obtain fitted values of A_L =75.4 and A_H=129.5.

5) The message carried by Fig 4 is not clear to me. If I understand correctly, the predicted average spin is based on the model which itself has been fitted on the data. Thus the figure is expected to simply show a quality of the fit. However, this was already shown in Fig 1. Also, my understanding is that to get the pre-neutron emission (right panel), one shifts each point by a number (the same for both axis) corresponding to the amount of angular momentum carried away by the neutrons. Both panels have similar agreement between predicted and measured quantities so perhaps only one is needed.

We would like to thank the referee for pointing out these deficiencies in the final figure. To illustrate better the extension to the general case in the actinides and the connection with reactor physics, we have replaced the right panel in figure 4 with a plot of average spin vs average fragment mass which shows predictions for other fissioning systems.

To conclude the manuscript presents an interesting study to an important problem. I have no doubt that the measurements, in particular the data shown in Fig 1, will be of strong interest to the community. However, I have raised some concerns regarding the interpretation of the results. These concerns need to be addressed before I can make a recommendation.

Referee #3 (Remarks to the Author):

This manuscript discusses recent experimental results obtained at the ALTO facility in Orsay using the LICORNE neutron source, on gamma rays observed in the fission fragments produced in the spontaneous fission of 252Cf and in the neutron-induced fission reactions of 238U and 232Th for incident neutron energies centered at 1.9 MeV. Using a data analysis first developed at the University of Manchester in the late 1980s, dubbed “MSM” for “Manchester Spin Method”, and using g-g coincidences, the authors infer an average angular momentum value as a function of the fission fragment mass. In the case of 252Cf (sf), the authors rely on the additional tagging of one of the two fission fragments, while for the (n,f) reactions on 238U and 232Th, they use a pulsed neutron beam to eliminate undesirable background due in particular to long-lasting beta-delayed gamma emissions. Three main results are presented as significant:

1. The reported coincidence data support the saw-tooth behavior of the average fragment spin <J> vs the mass of the fragment (after neutron emission), already reported in a limited set of experimental results since the 1970s.
2. Their results and conclusions are mostly independent of the fissioning system studied.
3. The observed lack of correlation between the inferred average angular momenta for the two complementary fragments is used to propose a simple model, and a fit to the experimental data, that explains the production of spin in the fragments post-scission only.

The data analysis of the very complex g-g correlation matrices is performed through the use of the (publicly available) Radware software package. This analysis is limited to about 30 even-even post-neutron emission fission fragments, for which the nuclear structure is much better known than for their odd-even and odd-odd neighbors.

While this work is certainly of interest to the nuclear fission community, it is difficult to make the case that it is of such "outstanding scientific importance" that it merits a publication in this journal. The claim that the present results would also impact the fields of nuclear reactor physics, nuclear structure for very neutron-rich isotopes, and super-heavy elements, is also difficult to support. That aside, I believe that the presented methodology and results lack the necessary rigor and assessment (and elimination) of potential systematic biases, including a serious attempt at quantifying uncertainties, that could impair the main conclusions of this work.

I will now go in some details to explain those specific technical points. In no particular order:

• While the authors briefly mention previous experimental results, there is no discussion on what could have caused those results to either agree or disagree with the present results. It would have been of great interest to the reader to understand why such a controversy still exists. The results on the saw-tooth behavior presented here are not original, but confirm earlier selected results.

The referee is correct. To address this point we have added new paragraph to the main article giving a brief history of previous experimental attempts to understand the problem of intrinsic spin generation in nuclear fission. We describe the different techniques employed along with their limitations and highlight the contradictory or inconclusive nature of previous results.

• Figure 2 (mis-labelled Fig.1!): how are the (a) band and (b) line in the inset plot obtained? A vague sentence is given in the caption, but with no information on the assumptions and code used to obtain those results. The interpretation is not put into question as a slope consistent with zero should indeed correspond to a post-scission mechanism with uncorrelated spins, but more details would be useful to understand how this quantitative result was obtained.

Referee 2 has asked similar questions, and indeed we are sorry we could not include details in the methods of the original submission due to lack of space. We have now added a section in the supporting information giving details of our Monte-Carlo simulations entitled "Monte-Carlo code for correlated spins" and added a figure 7 in the extended data showing simulation results.

• The simple model of the "snapping of a taut elastic band" introduced by the authors is interesting but obviously very schematic. It is very similar to the "random neck rupture" model introduced by Brosa et al to explain the saw-tooth behavior of the prompt neutron multiplicity as a function of the fragment mass. There, the physical intuition was based on the Rayleigh instabilities experienced by a fluid given a certain surface-to-volume ratio constraint. The physical intuition presented here is less clear as there is no distinction made between the translation kinetic and rotational energies expected from the snapping of this stretched elastic.

We have added a sentence at the beginning of the discussion section to make the principle elements of the elastic band analogy clear. The analogy is both accessible to the general reader and contains essential ingredients of the problem. Indeed we reference the random neck rupture of Brosa, which describes the neck/band snapping position. We have now also developed a parallel microscopic model (see discussion) which goes beyond the limitations of this analogy, where the intuition is from statistical theory.

The authors offer the idea that fluctuations in the direction of motion of the nascent fragments are what cause the development of two independent torques. This is probable, similar to the idea presented in their reference [9], but the lack of discussion of translational vs rotational energies makes it difficult to analyze this idea further. While the kinetic energy of the fragments was not recorded in the present experiments, some results of TKE vs fragment mass are well known, and could have been used to discuss this interesting question.

The question of a direct correlation between TKE and E_{rot} is intriguing, especially in the light of the recent results by Chebboubi et al. (Physics Letters B 775 (2017) 190–195), where an anti-correlation between TKE and <I> is shown for ^{132}Sn.

We remark that the relation between TKE and E_{rot} is experimentally challenging to investigate directly as E_{rot} is small compared to TKE (around 2% of TKE), and to our knowledge, a suitable experimental setup does not currently exist, although we are able to extract values of average E_{rot} at the yrast line from our current data. The shape of <KE>(A) is known to be negatively correlated with fragment deformation at scission and is approximately constant for the light fragment, has a local maximum for A~132, and decreases for higher A. Since we observe a spin-sawtooth both for the light and heavy fragment, it seems no direct relationship between <I>(A) and <KE>(A) exists. E_{rot} increases with <I> and decreases with fragment deformation, and an increase in E_{rot} will not necessarily lead to a decrease in TKE, but rather in the excitation energy E_x. In the fission models FREYA and FIFRELIN, E_{rot} cannot influence TKE as E_{rot} is determined after TKE (taken from experiments) is removed. The same is true for Ref [9], where TKE is taken as the Coulomb energy of the two fragments and related to the compactness of the configuration at scission. However, Ref[9] predicts fragments at scission with the same spin. If the referee had something else in mind that we've missed, then we are happy to apply further changes to the manuscript.

• The idea briefly mentioned that "quantum entanglement" between the moving fragments could be witnessed is far-fetched, to say the least, given the exceptional conditions (and isolation from any disturbing environment) that the production of entangled elementary particles require.

We too think this idea is far-fetched. This is why we wrote "does not appear to apply" in our original submission. We brought this up because the eminent theoretician J. Dobaczewski recently proposed the idea (https://arxiv.org/abs/1910.03924), which was subsequently repeated in this recent review article on nuclear fission (M. Bender et al. J. Phys. G (2020), https://iopscience.iop.org/article/10.1088/1361-6471/abab4f). Our intent was to dampen speculation about something that we consider highly unlikely. However, we have now removed the sentences referring to this idea.

• Under Consequences, the authors mention the issue of gamma-ray heating in a nuclear reactor as a potential issue that their current results addresses. They also mention "delayed g rays from isomeric states which contribute to the decay heat after reactor shutdown." This is true but not coming from the fission fragments prior to beta-decay, so it isn't relevant to the present discussion.

Here we disagree with the referee. There is a definite contribution to the total decay heat coming from prompt population of isomeric states (~20 nuclei) in the time range relevant for modelling Fukushima-type reactor accidents (seconds, minutes, hours, days). Many of these isomers have strong prompt population (e.g. ^{133m}Te 11/2$^-$ 55.4 m, ^{136m}I 6$^-$ 47 s, ^{132m}Sb 8- 2.8 m, etc.) These isomeric decays are not the main source of decay heat, which mostly comes from beta-decays close to stability, but the population of these isomers nonetheless needs to be understood and decays included in the decay heat. We therefore think they are relevant in the discussion.

To carry out the post-neutron emission calculation of the mass weighted averages shown in Fig.4 requires the knowledge of neutron multiplicity per fragment mass, which is well-known in the case of 252Cf (sf) only. What was used for the two other fissioning systems at the 1.9 MeV incident neutron energy for which no measurement exist (to my knowledge).

The referee is correct. Prompt fission neutron (PFN) multiplicities are very challenging to measure for neutron-induced fission of fertile nuclei. Corrections are made using data from (Oberstedt et al.) where ^{238}U(n,f) PFN is inferred from measurements in the neighbouring fissile isotope ^{235}U(n,f). For ^{232}Th(n,f) the only available information in the literature comes from calculations (A. Tudora et al). The variation of PFN(A) between different systems is rather small, so the introduction of a slight model-dependence to make this small correction for ^{232}Th(n,f) is justified. We have now revised figure 4 in response to the suggestion of referee 2.

What is of course of interest to the reactor physics community is essentially the heat/energy deposited by g rays produced in those fission reactors, which is a step beyond what is discussed here.

This is true, but to get there requires a gamma-transport calculation with high-precision information on the source-terms from gamma-emitting processes, i.e. gamma-ray spectra and average multiplicities (measurements which are top of the high priority request list for the OECD nuclear energy agency). These are currently impossible to model accurately due to lack of information on fragment spin distributions, which we now provide. We would argue that all physicists, including reactor physicists, would be genuinely interested in a description of the mechanism behind these prompt gamma emissions (analogous to Kepler's laws) rather than just the nuclear data measurements (analogous to Tycho Brahe's cataloging of the night sky).

• One of the main limitations of the present approach is that only even-even fragments are considered, which in many cases, is a small subset of all fragments produced. The extension of (or at least an attempt to extend) the conclusion presented here to all fragments is lacking.

The method can be extended to extract average spins since the level schemes of odd nuclei are included in the global fit. If the same procedure can be applied and the ground state spin subtracted from the result to obtain <I>, then the results appear to follow the curves presented in figure 1 (e.g. ^{133}Sb 4.18(14), ^{135}I 5.75(14), ^{139}Xe 7.01(6), ^{143}Ba 8.1(3)) for the ^{238}U(n,f) reaction. However, we prefer to present only the even-even data which we are sure we can solidly defend.

For many odd nuclei one major problem is that the ground state spin is unknown/tentative (e.g. ^{135}Sb (7/2+), ^{147}La (3/2+), ^{99}Y (5/2+), etc.). Furthermore, if the ground state spin is high (e.g. 7/2) the spin distribution measured at/near yrast may not be representative of the spin distribution at entry because states with spin below the ground state are not properly sampled.

An interesting question which could be addressed by the inclusion of odd-nuclei would be the presence, or absence, of an odd-even staggering effect in average spin coming from an extra microscopic contribution from unpaired protons in two odd-Z fragments (Naik). At present, we are not confident that the MSM has sufficient sensitivity for odd nuclei to measure variations in spin to the sub-hbar levels which might be required to detect such an effect (see our subsequent response on the sensitivity of the technique).

• "The major strength of the MSM is that it uses all the available g-ray intensity and coincidence data (…) and condenses this information into a single number, the average spin after neutron emission." I would not call this a "strength" since it means many details of the decay sequence are lost in the process, and several assumptions have to be made to get to this result. And no information on the distribution of spins can be obtained.

The referee is correct. We have removed the non-neutral word "strength" and shortened this sentence.

• The conclusion that the Yrast sequence of transitions is the only information needed to obtain an average initial spin is troubling, but could be correct for the simplified case of even-even fragments. This result is presented as the conclusion of a numerical sensitivity study but its interpretation is not pursued further.

We include quantitative information on this study in the "methods: non-inclusion of weaker transitions" section along with an interpretation/explanation of why this turns out to be the case.

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The referee raises an excellent point and we have now added a new sub-section in methods entitled "Sensitivity of the MSM" where we quantify the sensitivity via two different methods. We also remove the text describing application of the MSM to other reactions to save on space.

• The crux of the analysis presented in this paper, and therefore its main conclusions, is obtained through the use of the Radware software package. A very large global fit is performed with "thousands of parameters." No discussion of uncertainties is presented at any point in the publication. Such a global fit should come with specific metrics, or figure of merit, and quantifiable uncertainties associated with those, included strong correlations among the many parameters. The authors only mention the "powerful visual comparisons" and "visual checks" that can be performed. While this is definitely useful, it is certainly impossible to go through all g-g correlation data obtained in this work. The systematic study and reporting of those uncertainties is simply lacking.

We now realize that we did a poor job explaining the fitting procedures used and the details of the statistical error analysis and apologise for the omission of this important information. We would like to assure the referee that uncertainty analysis is something we have taken very seriously. Some key information on Radware can be found in this article (D.C. Radford, Nucl. Instrum. Meth. A361 297 (1995)). However, to fully address the referees concerns we have completely revised and extended the methods section "Application of the MSM to the fission data", now entitled "Fitting procedures and statistical uncertainties". We have now made explicit every single step of the fitting/analysis procedure, how uncertainties are treated at each stage and report their median values. We show how they propagate through into the final result. Figure 8 in the extended data shows examples of the spin distributions behind each data point and allows visualization of the associated uncertainties on side-feedings which leads to the final results.

• Under the "Determination of the side-feeding of the first 2+ state," the authors mention a possible experimental bias undetected in their recent analysis of fission yield anomalies in the 238U(n,f) reaction. More than just the side feeding issue addressed here, other reasons such as biasing due to a high-multiplicity gating on near-spherical nuclei could also cause some misinterpretations. This was covered in a recent paper by Fotiades et al (Phys. Rev. C 99, 024606 (2019)). Similar assumptions are being made in the present paper, although the experimental setup is different and the conclusions probably not impacted as much by those assumptions.

The thorough work by Fotiades et al., demonstrates clearly the impact and importance of 2+ side-feedings on fission yield measurements using techniques of gamma-ray spectroscopy. Indeed the origin of our previously-published ^{238}U(n,f) yield anomaly is clearly related to the large increase in 2+ side-feeding for ^{132}Sn and its nearest neighbours. These 2+ side-feedings can now be measured directly in our current work due to the much higher resolving power of the ν-Ball spectrometer over our previous setup.

We can demonstrate that the potential biases due to high-multiplicity gating near spherical nuclei are negligible, since the ^{252}Cf fission tag enables us to precisely quantify these effects. Increasing the (offline) minimum trigger condition from $M_\gamma \geq 2$ to $M_\gamma \geq 6$ results in a global increase in average fragment spin of only 1.7%. The gamma-ray multiplicity distributions measured with ν-Ball when gating on different fragments turn out to be remarkably similar (including ^{132}Sn and neighbours). The saw-tooth pattern assures that on average, low gamma multiplicities from one nucleus are compensated by high multiplicities from its partner.

However, assuming that the ground-state feeding is zero for all fragments is certainly wrong. The quantitative impact of such assumptions is difficult to obtain without performing a comprehensive set of calculations that uses all known information about the nuclear structure and decay schemes of the fragments.

We agree with the referee. Our original assumption of zero direct ground-state feeding is incorrect. As we suggested in our initial submission, an extrapolation of the spin distribution to zero spin is possible. We initially hesitated to do this because we did not wish to be criticized for applying a correction which is certain to amplify the saw-tooth pattern.

To address the referees comment we have completely redone the data analysis and now fitted the spin distribution associated with each data point and used it to extract a value for the 0+ feeding. These values are then used to correct the average spin data presented in figure 1, assuming appropriately large uncertainties on the extrapolations. Figure 8 in the extended data illustrates many examples of the yrast spin distributions and the fits used to make the extrapolation. We have modified the section "Ground state feeding" in the methods section to describe the procedure.

Reviewer Reports on the First Revision:

Referee #2 (Remarks to the Author):

The authors have provided a comprehensive response to my criticisms.
In my first report, I had some concerns which required clarification.

- 1) and 2) referred to the interpretation of the data and the validity of the conclusion that the spin distributions in both fragments were uncorrelated. My point would have been valid in the hypothesis of a narrow spin distribution. However, as demonstrated in the new material (Figs 7 and 8), this is not the case. I am now convinced that the interpretation offered by the authors, i.e., that the fragment spin distributions are uncorrelated, is supported by the data.

- In 3), I raised a concern about the possibility for the system to produce relatively large values of orbital angular momentum between the fragments to ensure angular momentum conservation. This, I think, is a puzzling consequence of the spin independence scenario. Nevertheless, this seems to be the most likely scenario. The origin of the mechanism producing this orbital angular momentum remains an open question and it is likely to attract future studies.

- In 4), I raised concerns about the proposed simple model to explain the experimental observation of independent spins. The authors have provided an additional support to their interpretation using a statistical model. Although this gives better ground to the proposed mechanism,I am still not sure this is the right interpretation. However, as mentioned by the authors, other approaches are incompatible with their data. I expect this paper will generate theoretical works to confirm this mechanism, or propose alternative ones.

- My last point 5) was more minor and concerned the relevance of part of fig 4. It has been addressed and the corresponding figure has been replaced.

I mentioned in my first report that, if correct, this work would be an important step towards our understanding of fission mechanism and that these results are of general interest. Indeed, fission plays an important role both in nature (e.g., in nucleosynthesis) and for applications.
The experimental results are surprising, to say the least. Yet, I agree that the scenario in which the spins of the fragments are uncorrelated is probably the only viable explanation. The proposed mechanism to explain these independent distributions is one possible interpretation and I expect this work will attract new experimental and theoretical studies.
I therefore recommend its publication in Nature.

Referee #3 (Remarks to the Author):

This manuscript discusses recent experimental results obtained at the ALTO facility in Orsay using the LICORNE neutron source, on gamma rays observed in the fission fragments produced in the spontaneous fission of 252Cf and in the neutron-induced fission reactions of 238U and 232Th for incident neutron energies centered at 1.9 MeV. Using a data analysis first developed at the University of Manchester in the late 1980s, dubbed “MSM” for “Manchester Spin Method”, and using g-g coincidences, the authors infer an average angular momentum value as a function of the fission fragment mass. In the case of 252Cf (sf), the authors rely on the additional tagging of one of the two fission fragments, while for the (n,f) reactions on 238U and 232Th, they use a pulsed neutron beam to eliminate undesirable background due in particular to long-lasting beta-delayed gamma emissions. Three main results are presented as significant:

1. The reported coincidence data support the saw-tooth behavior of the average fragment spin <J> vs the mass of the fragment (after neutron emission), already reported in a limited set of experimental results since the 1970s.
2. Their results and conclusions are mostly independent of the fissioning system studied.
3. The observed lack of correlation between the inferred average angular momenta for the two complementary fragments is used to propose a simple model, and a fit to the experimental data, that explains the production of spin in the fragments post-scission only.

The data analysis of the very complex g-g correlation matrices is performed through the use of the (publicly available) Radware software package. This analysis is limited to about 30 even-even post-neutron emission fission fragments, for which the nuclear structure is much better known than for their odd-even and odd-odd neighbors.

While this work is certainly of interest to the nuclear fission community, it is difficult to make the case that it is of such “outstanding scientific importance” that it merits a publication in this journal. The claim that the present results would also impact the fields of nuclear reactor physics, nuclear structure for very neutron-rich isotopes, and super-heavy elements, is also difficult to support. That aside, I believe that the presented methodology and results lack the necessary rigor and assessment (and elimination) of potential systematic biases, including a serious attempt at quantifying uncertainties, that could impair the main conclusions of this work.

I will now go in some details to explain those specific technical points. In no particular order:

• While the authors briefly mention previous experimental results, there is no discussion on what could have caused those results to either agree or disagree with the present results. It would have been of great interest to the reader to understand why such a controversy still exists. The results on the saw-tooth behavior presented here are not original, but confirm earlier selected results.

> The referee is correct. To address this point we have added new paragraph to the main article giving a brief history of previous experimental attempts to understand the problem of intrinsic spin generation in nuclear fission. We describe the different techniques employed along with their limitations and highlight the contradictory or inconclusive nature of previous results.

>> The added paragraph is somewhat helpful but again fails to provide possible explanations as why previous studies would have been flawed. Some important points to emphasize would have been the difference between event-by-event vs fragment-by-fragment studies, as discussed for instance in Ref. [Marin et al, NIM A968 (2020) 163907], or the possible role of different time coincidence windows on the saw-tooth shape, especially in the A~130 region where long-lived (> ns) isomeric states are important. A quantitative comparison with previous results, and not just about the qualitative saw-tooth shape observed for the gamma-ray multiplicity vs fragment mass, is also lacking.

• Figure 2 (mis-labelled Fig.1!): how are the (a) band and (b) line in the inset plot obtained? A vague sentence is given in the caption, but with no information on the assumptions and code used to obtain those results. The interpretation is not put into question as a slope consistent with zero should indeed correspond to a post-scission mechanism with uncorrelated spins, but more details would be useful to understand how this quantitative result was obtained.

> Referee 2 has asked similar questions, and indeed we are sorry we could not include details in the methods of the original submission due to lack of space. We have now added a section in the supporting information giving details of our Monte-Carlo simulations entitled "Monte-Carlo code for correlated spins" and added a figure 7 in the extended data showing simulation results.

>> The sentence "To our knowledge no Monte Carlo code exists which simultaneously follows the decay from scission of both fragments on an event-by-event basis…" is very confusing, as it would mean ignoring a relatively large body of literature on fission in the past ~15 years that has seen the development of various such codes, e.g., GEF, FIFRELIN, CGMF, FREYA. This is even more confusing since the authors refer to FREYA and FIFRELIN further down in their response. What is actually missing in those codes? If it is about the emission of neutrons and gamma rays before the full acceleration of the fragments, then a much broader discussion of what they authors have implemented is warranted, as technical details (lacking at present) would matter tremendously.

>> That the authors developed their own Monte Carlo code is positive though, but it is difficult to judge the quality of the results given the lack of details on what this code does. For instance, it does not treat the statistical gamma decay, which is performed using the relatively recent RAINIER code. What does that mean for the competition between prompt neutrons and gammas? Are the known neutron data, including the dependence of the average neutron multiplicity (and P(nu)!) on the fragment mass, which would have a direct impact on the dependence of the gamma-ray multiplicity vs fragment mass, well reproduced? Limited but important comparisons between the above-mentioned codes have been performed. The same cannot be said of the present code. Results shown in Fig.7 are reassuring and expected though.

• The simple model of the "snapping of a taut elastic band" introduced by the authors is interesting but obviously very schematic. It is very similar to the "random neck rupture" model introduced by Brosa et al to explain the saw-tooth behavior of the prompt neutron multiplicity as a function of the fragment mass. There, the physical intuition was based on the Rayleigh instabilities experienced by a fluid given a certain surface-to-volume ratio constraint. The physical intuition presented here is less clear as there is no distinction made between the translation kinetic and rotational energies expected from the snapping of this stretched elastic.

> We have added a sentence at the beginning of the discussion section to make the principle elements of the elastic band analogy clear. The analogy is both accessible to the general reader and contains essential ingredients of the problem. Indeed we reference the random neck rupture of Brosa, which describes the neck/band snapping position. We have now also developed a parallel microscopic model (see discussion) which goes beyond the limitations of this analogy, where the intuition is from statistical theory.

>> I indeed appreciate the efforts of the authors to find some classical analogy that can be understood more broadly. Let me however focus on the "microscopic" model. I would first claim that there is very little "microscopic" in this model. Those are phenomenological formulae derived from statistical physics, and they are not obtained from nucleon-nucleon microscopic calculations. There are additional simplifications in this model (e.g., E_x proportional to A_N, where A_N is crudely defined from deviations from A=78 or 130) that make it even more phenomenological in nature. I do agree with the authors that the data are consistent with the expectations from "statistical theory for a post-scission, uncorrelated, spin generation mechanism." I would however

refrain from using the term microscopic.

The authors offer the idea that fluctuations in the direction of motion of the nascent fragments are what cause the development of two independent torques. This is probable, similar to the idea presented in their reference [9], but the lack of discussion of translational vs rotational energies makes it difficult to analyze this idea further. While the kinetic energy of the fragments was not recorded in the present experiments, some results of TKE vs fragment mass are well known, and could have been used to discuss this interesting question.

> The question of a direct correlation between TKE and Erot is intriguing, especially in the light of the recent results by Chebboubi et al. (Physics Letters B 775 (2017) 190–195), where an anti-correlation between TKE and <I> is shown for 132Sn.
We remark that the relation between TKE and Erot is experimentally challenging to investigate directly as Erot is small compared to TKE (around 2% of TKE), and to our knowledge, a suitable experimental setup does not currently exist, although we are able to extract values of average Erot at the yrast line from our current data. The shape of <KE>(A) is known to be negatively correlated with fragment deformation at scission and is approximately constant for the light fragment, has a local maximum for A~132, and decreases for higher A. Since we observe a spin-sawtooth both for the light and heavy fragment, it seems no direct relationship between <I>(A) and <KE>(A) exists. Erot increases with <I> and decreases with fragment deformation, and an increase in Erot will not necessarily lead to a decrease in TKE, but rather in the excitation energy Ex. In the fission models FREYA and FIFRELIN, Erot cannot influence TKE as Erot is determined after TKE (taken from experiments) is removed. The same is true for Ref [9], where TKE is taken as the Coulomb energy of the two fragments and related to the compactness of the configuration at scission. However, Ref[9] predicts fragments at scission with the same spin. If the referee had something else in mind that we've missed, then we are happy to apply further changes to the manuscript.

>> My main point is that the proposed model is simplistic and convenient to fit a rather limited set of data. According to this model, the two independent torques in the fragments originate in the snapping of the band, but that's only true if there are deviations from strictly aligned fragments, which is highly probable indeed. If we follow the analogy of the band, the torque is actually created when the band is twisted, and the larger the twist the larger the torques. The authors invoke the uncertainty principle (on fission fragments!) in a qualitative manner to generate some torque. That's all very generic and intuitive but rather non predictive. What would be the predictions from such model for more compact or more elongated fragments, which would strongly correlate with TKE and Erot?

• The idea briefly mentioned that "quantum entanglement" between the moving fragments could be witnessed is far-fetched, to say the least, given the exceptional conditions (and isolation from any disturbing environment) that the production of entangled elementary particles require.

> We too think this idea is far-fetched. This is why we wrote "does not appear to apply" in our original submission. We brought this up because the eminent theoretician J. Dobaczewski recently proposed the idea (https://arxiv.org/abs/1910.03924), which was subsequently repeated in this recent review article on nuclear fission (M. Bender et al. J. Phys. G (2020), https://iopscience.iop.org/article/10.1088/1361-6471/abab4f). Our intent was to dampen speculation about something that we consider highly unlikely. However, we have now removed the sentences referring to this idea.

>> Thank you. I don't think this idea should be taken too seriously, despite its natural appeal.

• Under Consequences, the authors mention the issue of gamma-ray heating in a nuclear reactor as a potential issue that their current results addresses. They also mention "delayed g rays from isomeric states which contribute to the decay heat after reactor shutdown." This is true but not coming from the fission fragments prior to beta-decay, so it isn't relevant to the present discussion.

> Here we disagree with the referee. There is a definite contribution to the total decay heat coming from prompt population of isomeric states (~20 nuclei) in the time range relevant for modelling Fukushima-type reactor accidents (seconds, minutes, hours, days). Many of these isomers have strong prompt population (e.g. 133mTe 11/2- 55.4 m, 136mI 6- 47 s, 132mSb 8- 2.8 m, etc.) These isomeric decays are not the main source of decay heat, which mostly comes from beta-decays close to stability, but the population of these isomers nonetheless needs to be understood and decays included in the decay heat. We therefore think they are relevant in the discussion.

>> Those isomers are already taken into account in decay heat calculations, as long as they are present in the decay data libraries. The exact decay paths have a very small impact on the final results, and as I mentioned earlier and as the authors confirm, most of the decay heat will come from beta decays. In more realistic simulations of reactor designs, such "details" should be taken into account indeed. Please see below for my more general comment on fundamental science vs applied technologies.

To carry out the post-neutron emission calculation of the mass weighted averages shown in Fig.4 requires the knowledge of neutron multiplicity per fragment mass, which is well-known in the case of 252Cf (sf) only. What was used for the two other fissioning systems at the 1.9 MeV incident neutron energy for which no measurement exist (to my knowledge).

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>> Thank you for the clarification, and indeed, I believe this is correct and that such corrections would be small indeed.

What is of course of interest to the reactor physics community is essentially the heat/energy deposited by g rays produced in those fission reactors, which is a step beyond what is discussed here.

> This is true, but to get there requires a gamma-transport calculation with high-precision information on the source-terms from gamma-emitting processes, i.e. gamma-ray spectra and average multiplicities (measurements which are top of the high priority request list for the OECD nuclear energy agency). These are currently impossible to model accurately due to lack of information on fragment spin distributions, which we now provide. We would argue that all physicists, including reactor physicists, would be genuinely interested in a description of the mechanism behind these prompt gamma emissions (analogous to Kepler's laws) rather than just the nuclear data measurements (analogous to Tycho Brahe's cataloging of the night sky).

>> Definitely a cheap shot addressed to reactor physicists… While I was not arguing about the obvious (in my mind) importance of fundamental research vs the more pragmatic and applied engineering R&D (also important!), justifying the former based on its supposed (long-term) benefits to the latter is a delicate proposition, which I often endorse, whenever appropriate. Unfortunately, it is often misused or abused, at the expense of developing long-lasting and trusting relationships between the two. If the authors want to make the present claim, I encourage

them to first discuss with nuclear reactor physicists and engineers to make sure they are behind it. A more subtle and appropriate statement would be to emphasize the role of advanced computer simulations of new reactor designs that may eventually benefit from more realistic nuclear data. This is a longer-term prospect, but definitely of importance to the nuclear energy community, but which will require significant developments beyond what is currently available. As for the OECD/NEA HPRL mentioned here, it is unfortunate that it is not updated more regularly and/or more thoroughly. It is a daunting task, and I am not blaming anyone for this state of affairs. I do recognize the need for fundamental science to piggy-back on well-funded shorter-term ventures, but let's not fool anyone in the process.

• One of the main limitations of the present approach is that only even-even fragments are considered, which in many cases, is a small subset of all fragments produced. The extension of (or at least an attempt to extend) the conclusion presented here to all fragments is lacking.

> The method can be extended to extract average spins since the level schemes of odd nuclei are included in the global fit. If the same procedure can be applied and the ground state spin subtracted from the result to obtain <I>, then the results appear to follow the curves presented in figure 1 (e.g. 133Sb 4.18(14), 135I 5.75(14), 139Xe 7.01(6), 143Ba 8.1(3)) for the 238U(n,f) reaction. However, we prefer to present only the even-even data which we are sure we can solidly defend.
For many odd nuclei one major problem is that the ground state spin is unknown/tentative (e.g. 135Sb (7/2+), 147La (3/2+), 99Y (5/2+), etc.). Furthermore, if the ground state spin is high (e.g. 7/2) the spin distribution measured at/near yrast may not be representative of the spin distribution at entry because states with spin below the ground state are not properly sampled. An interesting question which could be addressed by the inclusion of odd-nuclei would be the presence, or absence, of an odd-even staggering effect in average spin coming from an extra microscopic contribution from unpaired protons in two odd-Z fragments (Naik). At present, we are not confident that the MSM has sufficient sensitivity for odd nuclei to measure variations in spin to the sub-hbar levels which might be required to detect such an effect (see our subsequent response on the sensitivity of the technique).

>> I fully agree that the analysis is on more solid footing for the even-even nuclei than even-odd ones. The fact that the latter are still being considered in the analysis is hopefully good, and I'd hope the authors have checked that their results are not biased significantly either way.

• "The major strength of the MSM is that it uses all the available g-ray intensity and coincidence data (…) and condenses this information into a single number, the average spin after neutron emission." I would not call this a "strength" since it means many details of the decay sequence are lost in the process, and several assumptions have to be made to get to this result. And no information on the distribution of spins can be obtained.

> The referee is correct. We have removed the non-neutral word "strength" and shortened this sentence.

>> Good. It was a very small remark, but thank you for addressing it.

• The conclusion that the Yrast sequence of transitions is the only information needed to obtain an average initial spin is troubling, but could be correct for the simplified case of even-even fragments. This result is presented as the conclusion of a numerical sensitivity study but its interpretation is not pursued further.

> We include quantitative information on this study in the "methods: non-inclusion of weaker transitions" section along with an interpretation/explanation of why this turns out to be the case.

>> This explanation is satisfactory. Thank you.

• The validity of the MSM approach is “demonstrated” through the comparison of fusion-evaporation vs neutron transfer reactions. The spin difference in those cases is massive, and it is unclear how this example demonstrates that the MSM method is sensitive enough to distinguish average spin values between different fragmentations, separated by only a fraction of hbar.

> The referee raises an excellent point and we have now added a new sub-section in methods entitled “Sensitivity of the MSM” where we quantify the sensitivity via two different methods. We also remove the text describing application of the MSM to other reactions to save on space.

>> It is nice to see more details regarding this point. In discussing the unknown ~10% spread of the data points beyond the statistical trend, the authors mention two “local” reasons both related to our limited knowledge of the nuclear structure for some fragments. Those local effects are in fact important when studying trends since they relate to only specific parts of the phase space, as opposed to being a systematic bias or unrecognized but fully correlated error. This effect may not be strong enough to impact the reported saw-tooth shape, but could certainly disrupt it.

• The crux of the analysis presented in this paper, and therefore its main conclusions, is obtained through the use of the Radware software package. A very large global fit is performed with “thousands of parameters.” No discussion of uncertainties is presented at any point in the publication. Such a global fit should come with specific metrics, or figure of merit, and quantifiable uncertainties associated with those, included strong correlations among the many parameters. The authors only mention the “powerful visual comparisons” and “visual checks” that can be performed. While this is definitely useful, it is certainly impossible to go through all g-g correlation data obtained in this work. The systematic study and reporting of those uncertainties is simply lacking.

> We now realize that we did a poor job explaining the fitting procedures used and the details of the statistical error analysis and apologise for the omission of this important information. We would like to assure the referee that uncertainty analysis is something we have taken very seriously. Some key information on Radware can be found in this article (D.C. Radford, Nucl. Instrum. Meth. A361 297 (1995)). However, to fully address the referees concerns we have completely revised and extended the methods section “Application of the MSM to the fission data”, now entitled “Fitting procedures and statistical uncertainties”. We have now made explicit every single step of the fitting/analysis procedure, how uncertainties are treated at each stage and report their median values. We show how they propagate through into the final result. Figure 8 in the extended data shows examples of the spin distributions behind each data point and allows visualization of the associated uncertainties on side-feedings which leads to the final results.

>> This added section, including Fig.8, is definitely helpful. It still lacks many quantitative details that would be useful to grasp the full procedure and in particular the treatment of correlations, which seems to be lacking. This is however a complicated topic that would require much more space than can reasonably be used by such a manuscript. A general concern is that most global fitting procedures fail to consider local deviations. A global Chi^2/d.o.f. becomes of little use when studying trends, as seen in various nuclear mass predictions using semi-classical and microscopic approaches.

• Under the “Determination of the side-feeding of the first 2+ state,” the authors mention a possible experimental bias undetected in their recent analysis of fission yield anomalies in the 238U(n,f) reaction. More than just the side feeding issue addressed here, other reasons such as biasing due to a high-multiplicity gating on near-spherical nuclei could also cause some misinterpretations. This was covered in a recent paper by Fotiades et al (Phys. Rev. C 99, 024606 (2019)). Similar assumptions are being made in the present paper, although the experimental setup is different and the conclusions probably not impacted as much by those assumptions.

> The thorough work by Fotiades et al., demonstrates clearly the impact and importance of 2+ side-feedings on fission yield measurements using techniques of gamma-ray spectroscopy. Indeed the origin of our previously-published 238U(n,f) yield anomaly is clearly related to the large increase in 2+ side-feeding for 132Sn and its nearest neighbours. These 2+ side-feedings can now be measured directly in our current work due to the much higher resolving power of the ν-Ball spectrometer over our previous setup.
We can demonstrate that the potential biases due to high-multiplicity gating near spherical nuclei are negligible, since the 252Cf fission tag enables us to precisely quantify these effects. Increasing the (offline) minimum trigger condition from $M_{\gamma}\geq 2$ to $M_{\gamma}\geq 6$ results in a global increase in average fragment spin of only 1.7%. The gamma-ray multiplicity distributions measured with ν-Ball when gating on different fragments turn out to be remarkably similar (including 132Sn and neighbours). The saw-tooth pattern assures that on average, low gamma multiplicities from one nucleus are compensated by high multiplicities from its partner.

>> Interesting, and somewhat unexpected. I am not sure I understand the number of 1.7% correctly though. If this is a "global increase", then it drowns the local effect that could be occurring near spherical fragments, and again a global, systematic perturbation would not impact the saw-tooth shape, but local ones would. I believe the authors have done their due diligence here, but this explanation still remains rather opaque to me.

However, assuming that the ground-state feeding is zero for all fragments is certainly wrong. The quantitative impact of such assumptions is difficult to obtain without performing a comprehensive set of calculations that uses all known information about the nuclear structure and decay schemes of the fragments.

> We agree with the referee. Our original assumption of zero direct ground-state feeding is incorrect. As we suggested in our initial submission, an extrapolation of the spin distribution to zero spin is possible. We initially hesitated to do this because we did not wish to be criticized for applying a correction which is certain to amplify the saw-tooth pattern.
To address the referees comment we have completely redone the data analysis and now fitted the spin distribution associated with each data point and used it to extract a value for the 0+ feeding. These values are then used to correct the average spin data presented in figure 1, assuming appropriately large uncertainties on the extrapolations. Figure 8 in the extended data illustrates many examples of the yrast spin distributions and the fits used to make the extrapolation. We have modified the section "Ground state feeding" in the methods section to describe the procedure.

>> That's helpful. I would expect the error bars in Fig.1 to be significantly higher near A~130, based on the results shown in Fig.8, but it might just be the optical illusion of having a relative error applied smaller quantities. However, it also reflects the fact that the authors use the Bethe formula to fit their data, even though the data are really lacking near A~130.

Author Rebuttals to First Revision:

We would again like to thank both referees for their further very helpful comments. We have fully responded to the points raised and provide the following summary (highlighted in green in the document) of the principal changes made to the manuscript:

- Insertion of a caveat before our summary of the spin mechanism by clarifying it is just our proposition and other interpretations based on our current results may be possible.
Insertion of "we suggest" in several places (Discussion section)
- Clarification of the limitations of the statistical model (Discussion section)

- Revision of the title to “Angular momentum generation in nuclear fission”
- Extension to the methods section “fitting procedure” with further methodological details on fitting and fit convergence
- Creation of a new Methods subsection entitled “uncertainty propagation and variance-covariance” containing information on correlated sources of uncertainty and application of appropriate methods to best determine the uncertainties on average spin values
- Inclusion of additional sentences about conflicting results from previous experiments, and a suggestion about how this conflict may now be resolved in the light of our new results (beginning of the Results section, end of Results section)
- Clarification of the technical details of our Monte-Carlo code and provision of further information about its precise purpose (Supporting Information section)

Referees' comments:

Referee #2 (Remarks to the Author):

The authors have provided a comprehensive response to my criticisms.
In my first report, I had some concerns which required clarification.

- 1) and 2) referred to the interpretation of the data and the validity of the conclusion that the spin distributions in both fragments were uncorrelated. My point would have been valid in the hypothesis of a narrow spin distribution. However, as demonstrated in the new material (Figs 7 and 8), this is not the case. I am now convinced that the interpretation offered by the authors, i.e., that the fragment spin distributions are uncorrelated, is supported by the data.

- In 3), I raised a concern about the possibility for the system to produce relatively large values of orbital angular momentum between the fragments to ensure angular momentum conservation. This, I think, is a puzzling consequence of the spin independence scenario. Nevertheless, this seems to be the most likely scenario. The origin of the mechanism producing this orbital angular momentum remains an open question and it is likely to attract future studies.

- In 4), I raised concerns about the proposed simple model to explain the experimental observation of independent spins. The authors have provided an additional support to their interpretation using a statistical model. Although this gives better ground to the proposed mechanism,I am still not sure this is the right interpretation. However, as mentioned by the authors, other approaches are incompatible with their data. I expect this paper will generate theoretical works to confirm this mechanism, or propose alternative ones.

- My last point 5) was more minor and concerned the relevance of part of fig 4. It has been addressed and the corresponding figure has been replaced.

I mentioned in my first report that, if correct, this work would be an important step towards our understanding of fission mechanism and that these results are of general interest. Indeed, fission plays an important role both in nature (e.g., in nucleosynthesis) and for applications.

The experimental results are surprising, to say the least. Yet, I agree that the scenario in which the spins of the fragments are uncorrelated is probably the only viable explanation. The proposed mechanism to explain these independent distributions is one possible interpretation and I expect this work will attract new experimental and theoretical studies. I therefore recommend its publication in Nature.

We would like to thank the referee for their positive recommendation. We have modified the manuscript accordingly to acknowledge that our proposed mechanism is only a suggestion and that other interpretations are possible.

Referee #3 (Remarks to the Author):

This manuscript discusses recent experimental results obtained at the ALTO facility in Orsay using the LICORNE neutron source, on gamma rays observed in the fission fragments produced in the spontaneous fission of 252Cf and in the neutron-induced fission reactions of 238U and 232Th for incident neutron energies centered at 1.9 MeV. Using a data analysis first developed at the University of Manchester in the late 1980s, dubbed “MSM” for “Manchester Spin Method”, and using g-g coincidences, the authors infer an average angular momentum value as a function of the fission fragment mass. In the case of 252Cf (sf), the authors rely on the additional tagging of one of the two fission fragments, while for the (n,f) reactions on 238U and 232Th, they use a pulsed neutron beam to eliminate undesirable background due in particular to long-lasting beta-delayed gamma emissions. Three main results are presented as significant:

1. The reported coincidence data support the saw-tooth behavior of the average fragment spin <J> vs the mass of the fragment (after neutron emission), already reported in a limited set of experimental results since the 1970s.
2. Their results and conclusions are mostly independent of the fissioning system studied.
3. The observed lack of correlation between the inferred average angular momenta for the two complementary fragments is used to propose a simple model, and a fit to the experimental data, that explains the production of spin in the fragments post-scission only.

The data analysis of the very complex g-g correlation matrices is performed through the use of the (publicly available) Radware software package. This analysis is limited to about 30 even-even post-neutron emission fission fragments, for which the nuclear structure is much better known than for their odd-even and odd-odd neighbors.

While this work is certainly of interest to the nuclear fission community, it is difficult to make the case that it is of such “outstanding scientific importance” that it merits a publication in this journal. The claim that the present results would also impact the fields of nuclear reactor physics, nuclear structure for very neutron-rich isotopes, and super-heavy elements, is also difficult to support. That aside, I believe that the presented methodology and results lack the necessary rigor and assessment (and elimination) of potential systematic biases, including a serious attempt at quantifying uncertainties, that could impair the main conclusions of this work.

I will now go in some details to explain those specific technical points. In no particular order:

• While the authors briefly mention previous experimental results, there is no discussion on what could have caused those results to either agree or disagree with the present results. It would have been of great interest to the reader to understand why such a controversy still exists. The results on the saw-tooth behavior presented here are not original, but confirm earlier selected results.

> The referee is correct. To address this point we have added new paragraph to the main article giving a brief history of previous experimental attempts to understand the problem of intrinsic spin generation in nuclear fission. We describe the different techniques employed along with their limitations and highlight the contradictory or inconclusive nature of previous results.

>> The added paragraph is somewhat helpful but again fails to provide possible explanations as why previous studies would have been flawed. Some important points to emphasize would have been the difference between event-by-event vs fragment-by-fragment studies, as discussed for instance in Ref. [Marin et al, NIM A968 (2020) 163907], or the possible role of different time coincidence windows on the saw-tooth shape, especially in the A~130 region where long-lived (> ns) isomeric states are important. A quantitative comparison with previous results, and not just about the qualitative saw-tooth shape observed for the gamma-ray multiplicity vs fragment mass, is also lacking.

We apologize if the added paragraph was too concise; we were concerned about the total word count. To answer why previous results might agree or disagree, our observation of clear saw-tooth shapes in average spin for multiple systems indicates that the early measurements of gamma yields by Armbuster et al. and Pleasonton et al. were not in fact flawed, as suggested later by Glassel et al.

Glassel pointed out that previous experimental difficulties such as the time window employed, which rejected early and late gammas, might create an artificial saw-tooth shape due to the loss in yield from isomeric decays around mass A~130 in particular. Our experiment does not have these difficulties.

Glassel used a different technique, relying on a comparison of Doppler-shifted gamma spectra, to separate the fragment contributions. However, if Glassel measured event-by-event correlations then no saw-tooth observation would be expected if fragment spins are independent of each other (a key finding of our work shown in figure 2). Saw-tooths would then only appear for fragment-by-fragment measurements (as we show in figure 1). Glassel et al. may not have understood the implications of what they had observed, and the absence of saw-tooth would confirm, rather than contradict our results. To explain this potential resolution of the historical controversy surrounding previous results we have now added new sentences at the beginning and the end of the Results section.

Direct quantitative comparison of our results with those from gamma-yield and isomeric-yield-ratio measurements are difficult because our results are the first direct measurement of average spin values for these systems, whereas previous measurements are only of indirect, spin-sensitive quantities. Such comparisons require transformation of the indirect quantities, either using assumptions about average spin per gamma transition or the invocation of a particular model, respectively. Additionally, such a comparison would involve different systems to ours (e.g. 235U(n_{th},f) by Armbuster et al.). We would be very happy to include a new figure in the manuscript making such a comparison if the Editor deems it necessary.

• Figure 2 (mis-labelled Fig.1!): how are the (a) band and (b) line in the inset plot obtained? A vague sentence is given in the caption, but with no information on the assumptions and code used to obtain those results. The interpretation is not put into question as a slope consistent with zero should indeed correspond to a post-scission mechanism with uncorrelated spins, but more details would be useful to understand how this quantitative result was obtained.

> Referee 2 has asked similar questions, and indeed we are sorry we could not include details in the methods of the original submission due to lack of space. We have now added a section in the supporting information giving details of our Monte-Carlo simulations entitled "Monte-Carlo code for correlated spins" and added a figure 7 in the extended data showing simulation results.

>> The sentence "To our knowledge no Monte Carlo code exists which simultaneously follows the decay from scission of both fragments on an event-by-event basis…" is very confusing, as it would mean ignoring a relatively large body of literature on fission in the past ~15 years that has seen the development of various such codes, e.g., GEF, FIFRELIN, CGMF, FREYA. This is even more confusing since the authors refer to FREYA and FIFRELIN further down in their response. What is actually missing in those codes? If it is about the emission of neutrons and gamma rays before the full acceleration of the fragments, then a much broader discussion of what they authors have implemented is warranted, as technical details (lacking at present) would matter tremendously.

There has clearly been a misunderstanding and obviously our explanation was insufficient. Our code is not intended in any way to compete or be benchmarked against full fission-decay models such as FREYA, GEF, CGMF, FIFRELIN, etc., which we are familiar with. These codes have indeed have taken up to 15 years to develop. The sole problem we wish to solve is to determine what range of slopes we might expect to see experimentally if fragment spins are correlated at scission, i.e. the determination of the location of the blue band in the inset of figure 4. Our code was written for this very precise purpose, which is to propagate spin dispersions in both fragments from scission to yrast, which occur due to emission of neutrons and statistical gamma rays. The code is as complex as is necessary to perform correctly this one unique task. The requirements of the code are (i) complete user control over the spin distribution parameters at scission, (ii) total control over the widths of the spin dispersions due to emission of both neutrons and gamma rays, and (iii) the ability to output the resulting spin distribution observed at yrast when setting conditions on the spin distribution in the partner fragment. In response to the question "what is wrong with other codes?" the answer is nothing, our code does something else. We have now modified the section in supplementary information to better explain this.

>> That the authors developed their own Monte Carlo code is positive though, but it is difficult to judge the quality of the results given the lack of details on what this code does. For instance, it does not treat the statistical gamma decay, which is performed using the relatively recent RAINIER code. What does that mean for the competition between prompt

neutrons and gammas? Are the known neutron data, including the dependence of the average neutron multiplicity (and P(nu)!) on the fragment mass, which would have a direct impact on the dependence of the gamma-ray multiplicity vs fragment mass, well reproduced? Limited but important comparisons between the above-mentioned codes have been performed. The same cannot be said of the present code. Results shown in Fig.7 are reassuring and expected though.

The sole point is that once developed, we could then use our code to perform a sensitivity analysis to answer the question: If fragment spins were correlated at scission what range of slopes would we expect to observe at yrast if we perturb the scission spin and dispersion parameters? These parameters were the mean value and skewness of the distribution at scission and the magnitudes of the neutron and statistical gamma ray dispersions. Perturbations covered a wide range of realistic possibilities for these parameters. Our code does indeed use known nubar and p(nu) distribution, but since experimentally we were looking at spin correlations between 3n and 1n fragment partners, we know that only 3 neutrons (or 1 neutron) will be emitted and retain only these events. It is therefore not a surprise that our results in figure 7 are what might be expected, since we are using dispersions compatible with the known physics of post-scission neutron and gamma decay.

• The simple model of the "snapping of a taut elastic band" introduced by the authors is interesting but obviously very schematic. It is very similar to the "random neck rupture" model introduced by Brosa et al to explain the saw-tooth behavior of the prompt neutron multiplicity as a function of the fragment mass. There, the physical intuition was based on the Rayleigh instabilities experienced by a fluid given a certain surface-to-volume ratio constraint. The physical intuition presented here is less clear as there is no distinction made between the translation kinetic and rotational energies expected from the snapping of this stretched elastic.

> We have added a sentence at the beginning of the discussion section to make the principle elements of the elastic band analogy clear. The analogy is both accessible to the general reader and contains essential ingredients of the problem. Indeed we reference the random neck rupture of Brosa, which describes the neck/band snapping position. We have now also developed a parallel microscopic model (see discussion) which goes beyond the limitations of this analogy, where the intuition is from statistical theory.

>> I indeed appreciate the efforts of the authors to find some classical analogy that can be understood more broadly. Let me however focus on the "microscopic" model. I would first claim that there is very little "microscopic" in this model. Those are phenomenological formulae derived from statistical physics, and they are not obtained from nucleon-nucleon microscopic calculations. There are additional simplifications in this model (e.g., E_x proportional to A_N, where A_N is crudely defined from deviations from A=78 or 130) that make it even more phenomenological in nature. I do agree with the authors that the data are consistent with the expectations from "statistical theory for a post-scission, uncorrelated, spin generation mechanism." I would however refrain from using the term microscopic.

We agree and have replaced the term microscopic with statistical to make this distinction.

The authors offer the idea that fluctuations in the direction of motion of the nascent fragments are what cause the development of two independent torques. This is probable, similar to the idea presented in their reference [9], but the lack of discussion of translational vs rotational energies makes it difficult to analyze this idea further. While the kinetic energy of the fragments was not recorded in the present experiments, some results of TKE vs fragment mass are well known, and could have been used to discuss this interesting question.

> The question of a direct correlation between TKE and Erot is intriguing, especially in the light of the recent results by Chebboubi et al. (Physics Letters B 775 (2017) 190–195), where an anti-correlation between TKE and is shown for 132Sn.
We remark that the relation between TKE and Erot is experimentally challenging to investigate directly as Erot is small compared to TKE (around 2% of TKE), and to our knowledge, a suitable experimental setup does not currently exist, although we are able to extract values of average Erot at the yrast line from our current data. The shape of <KE>(A) is known to be negatively correlated with fragment deformation at scission and is approximately constant for the light fragment, has a local maximum for A~132, and decreases for higher A. Since we observe a spin-sawtooth both for the light and heavy fragment, it seems no direct relationship between (A) and <KE>(A) exists. Erot increases with and decreases with fragment deformation, and an increase in Erot will not necessarily lead to a decrease in TKE, but rather in the excitation energy Ex. In the fission models FREYA and FIFRELIN, Erot cannot influence TKE as Erot is determined after TKE (taken from experiments) is removed. The
same is true for Ref [9], where TKE is taken as the Coulomb energy of the two fragments and related to the compactness of the configuration at scission. However, Ref[9] predicts fragments at scission with the same spin. If the referee had something else in mind that we've missed, then we are happy to apply further changes to the manuscript.

>> My main point is that the proposed model is simplistic and convenient to fit a rather limited set of data. According to this model, the two independent torques in the fragments originate in the snapping of the band, but that's only true if there are deviations from strictly aligned fragments, which is highly probable indeed. If we follow the analogy of the band, the torque is actually created when the band is twisted, and the larger the twist the larger the torques. The authors invoke the uncertainty principle (on fission fragments!) in a qualitative manner to generate some torque. That's all very generic and intuitive but rather non predictive. What would be the predictions from such model for more compact or more elongated fragments, which would strongly correlate with TKE and Erot?

We would like to thank the referee for pointing out this interesting extension to our elastic band analogy. We would expect that compact scission configurations would have higher TKE (with closer centres of charge generating greater Coulomb repulsion), lower TXE and hence lower spin and vice-versa, consistent with Chebboubi et al. For Erot it's not clear. There is a positive correlation between spin and Erot, but also an anti-correlation since larger deformations have higher moments of inertia and hence lower Erot.

• The idea briefly mentioned that "quantum entanglement" between the moving fragments could be witnessed is far-fetched, to say the least, given the exceptional conditions (and isolation from any disturbing environment) that the production of entangled elementary particles require.

> We too think this idea is far-fetched. This is why we wrote “does not appear to apply” in our original submission. We brought this up because the eminent theoretician J. Dobaczewski recently proposed the idea (https://arxiv.org/abs/1910.03924), which was subsequently repeated in this recent review article on nuclear fission (M. Bender et al. J. Phys. G (2020), https://iopscience.iop.org/article/10.1088/1361-6471/abab4f). Our intent was to dampen speculation about something that we consider highly unlikely. However, we have now removed the sentences referring to this idea.

>> Thank you. I don’t think this idea should be taken too seriously, despite its natural appeal.

• Under Consequences, the authors mention the issue of gamma-ray heating in a nuclear reactor as a potential issue that their current results addresses. They also mention “delayed g rays from isomeric states which contribute to the decay heat after reactor shutdown.” This is true but not coming from the fission fragments prior to beta-decay, so it isn’t relevant to the present discussion.

> Here we disagree with the referee. There is a definite contribution to the total decay heat coming from prompt population of isomeric states (~20 nuclei) in the time range relevant for modelling Fukushima-type reactor accidents (seconds, minutes, hours, days). Many of these isomers have strong prompt population (e.g. 133mTe 11/2- 55.4 m, 136mI 6- 47 s, 132mSb 8- 2.8 m, etc.) These isomeric decays are not the main source of decay heat, which mostly comes from beta-decays close to stability, but the population of these isomers nonetheless needs to be understood and decays included in the decay heat. We therefore think they are relevant in the discussion.

>> Those isomers are already taken into account in decay heat calculations, as long as they are present in the decay data libraries. The exact decay paths have a very small impact on the final results, and as I mentioned earlier and as the authors confirm, most of the decay heat will come from beta decays. In more realistic simulations of reactor designs, such “details” should be taken into account indeed. Please see below for my more general comment on fundamental science vs applied technologies.

To carry out the post-neutron emission calculation of the mass weighted averages shown in Fig.4 requires the knowledge of neutron multiplicity per fragment mass, which is well-known in the case of 252Cf (sf) only. What was used for the two other fissioning systems at the 1.9 MeV incident neutron energy for which no measurement exist (to my knowledge).

> The referee is correct. Prompt fission neutron (PFN) multiplicities are very challenging to measure for neutron-induced fission of fertile nuclei. Corrections are made using data from (Oberstedt et al.) where 238U(n,f) PFN is inferred from measurements in the neighbouring fissile isotope 235U(n,f). For 232Th(n,f) the only available information in the literature comes from calculations (A. Tudora et al). The variation of PFN(A) between different systems is rather small, so the introduction of a slight model-dependence to make this small

correction for 232Th(n,f) is justified. We have now revised figure 4 in response to the suggestion of referee 2.

>> Thank you for the clarification, and indeed, I believe this is correct and that such corrections would be small indeed.

What is of course of interest to the reactor physics community is essentially the heat/energy deposited by g rays produced in those fission reactors, which is a step beyond what is discussed here.

> This is true, but to get there requires a gamma-transport calculation with high-precision information on the source-terms from gamma-emitting processes, i.e. gamma-ray spectra and average multiplicities (measurements which are top of the high priority request list for the OECD nuclear energy agency). These are currently impossible to model accurately due to lack of information on fragment spin distributions, which we now provide. We would argue that all physicists, including reactor physicists, would be genuinely interested in a description of the mechanism behind these prompt gamma emissions (analogous to Kepler's laws) rather than just the nuclear data measurements (analogous to Tycho Brahe's cataloging of the night sky).

>> Definitely a cheap shot addressed to reactor physicists… While I was not arguing about the obvious (in my mind) importance of fundamental research vs the more pragmatic and applied engineering R&D (also important!), justifying the former based on its supposed (long-term) benefits to the latter is a delicate proposition, which I often endorse, whenever appropriate. Unfortunately, it is often misused or abused, at the expense of developing long-lasting and trusting relationships between the two. If the authors want to make the present claim, I encourage them to first discuss with nuclear reactor physicists and engineers to make sure they are behind it. A more subtle and appropriate statement would be to emphasize the role of advanced computer simulations of new reactor designs that may eventually benefit from more realistic nuclear data. This is a longer-term prospect, but definitely of importance to the nuclear energy community, but which will require significant developments beyond what is currently available. As for the OECD/NEA HPRL mentioned here, it is unfortunate that it is not updated more regularly and/or more thoroughly. It is a daunting task, and I am not blaming anyone for this state of affairs. I do recognize the need for fundamental science to piggy-back on well-funded shorter-term ventures, but let's not fool anyone in the process.
We apologize if this was what came across. This was not at all our intention. We were simply arguing that we think we have significant and unexpected results on a fundamental mechanism which other scientists would be interested to read about.

• One of the main limitations of the present approach is that only even-even fragments are considered, which in many cases, is a small subset of all fragments produced. The extension of (or at least an attempt to extend) the conclusion presented here to all fragments is lacking.

> The method can be extended to extract average spins since the level schemes of odd nuclei are included in the global fit. If the same procedure can be applied and the ground

state spin subtracted from the result to obtain , then the results appear to follow the curves presented in figure 1 (e.g. 133Sb 4.18(14), 135I 5.75(14), 139Xe 7.01(6), 143Ba 8.1(3)) for the 238U(n,f) reaction. However, we prefer to present only the even-even data which we are sure we can solidly defend.
For many odd nuclei one major problem is that the ground state spin is unknown/tentative (e.g. 135Sb (7/2+), 147La (3/2+), 99Y (5/2+), etc.). Furthermore, if the ground state spin is high (e.g. 7/2) the spin distribution measured at/near yrast may not be representative of the spin distribution at entry because states with spin below the ground state are not properly sampled.
An interesting question which could be addressed by the inclusion of odd-nuclei would be the presence, or absence, of an odd-even staggering effect in average spin coming from an extra microscopic contribution from unpaired protons in two odd-Z fragments (Naik). At present, we are not confident that the MSM has sufficient sensitivity for odd nuclei to measure variations in spin to the sub-hbar levels which might be required to detect such an effect (see our subsequent response on the sensitivity of the technique).

>> I fully agree that the analysis is on more solid footing for the even-even nuclei than even-odd ones. The fact that the latter are still being considered in the analysis is hopefully good, and I'd hope the authors have checked that their results are not biased significantly either way.

We do not observe biases due to the inclusion of only even-even fragments within the sensitivity of the method.

• "The major strength of the MSM is that it uses all the available g-ray intensity and coincidence data (…) and condenses this information into a single number, the average spin after neutron emission." I would not call this a "strength" since it means many details of the decay sequence are lost in the process, and several assumptions have to be made to get to this result. And no information on the distribution of spins can be obtained.

> The referee is correct. We have removed the non-neutral word "strength" and shortened this sentence.

>> Good. It was a very small remark, but thank you for addressing it.

• The conclusion that the Yrast sequence of transitions is the only information needed to obtain an average initial spin is troubling, but could be correct for the simplified case of even-even fragments. This result is presented as the conclusion of a numerical sensitivity study but its interpretation is not pursued further.

> We include quantitative information on this study in the "methods: non-inclusion of weaker transitions" section along with an interpretation/explanation of why this turns out to be the case.

>> This explanation is satisfactory. Thank you.

• The validity of the MSM approach is "demonstrated" through the comparison of fusion-evaporation vs neutron transfer reactions. The spin difference in those cases is massive, and it is unclear how this example demonstrates that the MSM method is sensitive enough to distinguish average spin values between different fragmentations, separated by only a fraction of hbar.

> The referee raises an excellent point and we have now added a new sub-section in methods entitled "Sensitivity of the MSM" where we quantify the sensitivity via two different methods. We also remove the text describing application of the MSM to other reactions to save on space.

>> It is nice to see more details regarding this point. In discussing the unknown ~10% spread of the data points beyond the statistical trend, the authors mention two "local" reasons both related to our limited knowledge of the nuclear structure for some fragments. Those local effects are in fact important when studying trends since they relate to only specific parts of the phase space, as opposed to being a systematic bias or unrecognized but fully correlated error. This effect may not be strong enough to impact the reported saw-tooth shape, but could certainly disrupt it.

Since the trends occur over a range of typically 15 nuclei in the light peak and a further 15 in the heavy peak, occasional local perturbations to these trends are not seen to influence the overall shape of the saw-tooth patterns, as evidenced in figure 1. Correlated sources of uncertainty are explained in the next point.

• The crux of the analysis presented in this paper, and therefore its main conclusions, is obtained through the use of the Radware software package. A very large global fit is performed with "thousands of parameters." No discussion of uncertainties is presented at any point in the publication. Such a global fit should come with specific metrics, or figure of merit, and quantifiable uncertainties associated with those, included strong correlations among the many parameters. The authors only mention the "powerful visual comparisons" and "visual checks" that can be performed. While this is definitely useful, it is certainly impossible to go through all g-g correlation data obtained in this work. The systematic study and reporting of those uncertainties is simply lacking.

> We now realize that we did a poor job explaining the fitting procedures used and the details of the statistical error analysis and apologise for the omission of this important information. We would like to assure the referee that uncertainty analysis is something we have taken very seriously. Some key information on Radware can be found in this article (D.C. Radford, Nucl. Instrum. Meth. A361 297 (1995)). However, to fully address the referees concerns we have completely revised and extended the methods section "Application of the MSM to the fission data", now entitled "Fitting procedures and statistical uncertainties". We have now made explicit every single step of the fitting/analysis procedure, how uncertainties are treated at each stage and report their median values. We show how they propagate through into the final result. Figure 8 in the extended data shows examples of the spin distributions behind each data point and allows visualization of the associated uncertainties on side-feedings which leads to the final results.

>> This added section, including Fig.8, is definitely helpful. It still lacks many quantitative details that would be useful to grasp the full procedure and in particular the treatment of correlations, which seems to be lacking. This is however a complicated topic that would require much more space than can reasonably be used by such a manuscript. A general concern is that most global fitting procedures fail to consider local deviations. A global Chi^2/d.o.f. becomes of little use when studying trends, as seen in various nuclear mass predictions using semi-classical and microscopic approaches.

We have now extended this section to give more methodological details, particularly on the importance of assuring local fit convergence. We have added a new section entitled propagation of error and variance-covariance which provides additional quantitative information, in particular on potential correlated sources of uncertainty. Minor changes to average spin uncertainties occur when taking into account these correlations but these are seen to have no impact on the results or conclusions.

• Under the “Determination of the side-feeding of the first 2+ state,” the authors mention a possible experimental bias undetected in their recent analysis of fission yield anomalies in the 238U(n,f) reaction. More than just the side feeding issue addressed here, other reasons such as biasing due to a high-multiplicity gating on near-spherical nuclei could also cause some misinterpretations. This was covered in a recent paper by Fotiades et al (Phys. Rev. C 99, 024606 (2019)). Similar assumptions are being made in the present paper, although the experimental setup is different and the conclusions probably not impacted as much by those assumptions.

> The thorough work by Fotiades et al., demonstrates clearly the impact and importance of 2+ side-feedings on fission yield measurements using techniques of gamma-ray spectroscopy. Indeed the origin of our previously-published 238U(n,f) yield anomaly is clearly related to the large increase in 2+ side-feeding for 132Sn and its nearest neighbours. These 2+ side-feedings can now be measured directly in our current work due to the much higher resolving power of the ν-Ball spectrometer over our previous setup.
We can demonstrate that the potential biases due to high-multiplicity gating near spherical nuclei are negligible, since the 252Cf fission tag enables us to precisely quantify these effects. Increasing the (offline) minimum trigger condition from $M_\gamma \geq 2$ to $M_\gamma \geq 6$ results in a global increase in average fragment spin of only 1.7%. The gamma-ray multiplicity distributions measured with ν-Ball when gating on different fragments turn out to be remarkably similar (including 132Sn and neighbours). The saw-tooth pattern assures that on average, low gamma multiplicities from one nucleus are compensated by high multiplicities from its partner.

>> Interesting, and somewhat unexpected. I am not sure I understand the number of 1.7% correctly though. If this is a “global increase”, then it drowns the local effect that could be occurring near spherical fragments, and again a global, systematic perturbation would not impact the saw-tooth shape, but local ones would. I believe the authors have done their due diligence here, but this explanation still remains rather opaque to me.

When plotted, the entire saw-tooth pattern is observed to move upwards slightly by 1.7% and there are no local effects where systematically higher or lower values than the average occur in any particular mass region. We expected the increase would be greater, but the fact that it isn't is almost certainly due to our calorimeter covering 75% of the solid angle which weakens the correlation between emitted and detected multiplicities. We therefore conclude that applying the minimum trigger condition of $M\gamma >= 3$ in our experiments does not introduce any bias to our results.

However, assuming that the ground-state feeding is zero for all fragments is certainly wrong. The quantitative impact of such assumptions is difficult to obtain without performing a comprehensive set of calculations that uses all known information about the nuclear structure and decay schemes of the fragments.

> We agree with the referee. Our original assumption of zero direct ground-state feeding is incorrect. As we suggested in our initial submission, an extrapolation of the spin distribution to zero spin is possible. We initially hesitated to do this because we did not wish to be criticized for applying a correction which is certain to amplify the saw-tooth pattern.
To address the referees comment we have completely redone the data analysis and now fitted the spin distribution associated with each data point and used it to extract a value for the 0+ feeding. These values are then used to correct the average spin data presented in figure 1, assuming appropriately large uncertainties on the extrapolations. Figure 8 in the extended data illustrates many examples of the yrast spin distributions and the fits used to make the extrapolation. We have modified the section "Ground state feeding" in the methods section to describe the procedure.

>> That's helpful. I would expect the error bars in Fig.1 to be significantly higher near A~130, based on the results shown in Fig.8, but it might just be the optical illusion of having a relative error applied smaller quantities. However, it also reflects the fact that the authors use the Bethe formula to fit their data, even though the data are really lacking near A~130.

The relative uncertainties are typically a factor of 4-5 higher for A~130 when compared to A~140, but since the average spins around A~130 are smaller, the increase in absolute size is smaller.

Reviewer Reports on the Second Revision:

Referee #3 (Remarks to the Author):

Dear Authors,

I started answering and commenting your responses and additional material point by point, but quickly realized that I was satisfied with most/all of your answers. I appreciate the evident and serious effort you put into addressing all my initial concerns.

I have no further objection or comments (except perhaps a typo on the RAIN(I)ER code in the text; the reference is ok).

Thank you.