

# Angular momentum generation in nuclear fission

J.N. Wilson<sup>1</sup>, D. Thisse<sup>1</sup>, M. Lebois<sup>1</sup>, N. Jovančević<sup>1</sup>, D. Gjestvang<sup>2</sup>, R. Canavan<sup>3,12</sup>, M. Rudigier<sup>3,4</sup>, D. Étasse<sup>5</sup>, R.-B. Gerst<sup>6</sup>, L. Gaudefroy<sup>7</sup>, E. Adamska<sup>8</sup>, P. Adsley<sup>1</sup>, A. Algora<sup>9,29</sup>, M. Babo<sup>1</sup>, K. Belvedere<sup>3</sup>, J. Benito<sup>10</sup>, G. Benzoni<sup>11</sup>, A. Blazhev<sup>6</sup>, A. Boso<sup>12</sup>, S. Bottoni<sup>11,13</sup>, M. Bunce<sup>12</sup>, R. Chakma<sup>1</sup>, N. Cieplicka-Oryńczak<sup>14</sup>, S. Courtin<sup>15,16</sup>, M.L. Cortés<sup>17</sup>, P. Davies<sup>18</sup>, C. Delafosse<sup>1</sup>, M. Fallot<sup>19</sup>, B. Fornal<sup>14</sup>, L. Fraile<sup>10</sup>, A. Gottardo<sup>21</sup>, V. Guadilla<sup>19</sup>, G. Häfner<sup>1,6</sup>, K. Hauschild<sup>1</sup>, M. Heine<sup>15</sup>, C. Henrich<sup>4</sup>, I. Homm<sup>4</sup>, F. Ibrahim<sup>1</sup>, Ł.W. Iskra<sup>11,14</sup>, P. Ivanov<sup>12</sup>, S. Jazwari<sup>3,12</sup>, A. Korgul<sup>8</sup>, P. Koseoglou<sup>4,21</sup>, T. Kröll<sup>4</sup>, T. Kurtukian-Nieto<sup>22</sup>, L. Le Meur<sup>19</sup>, S. Leoni<sup>11,13</sup>, J. Ljungvall<sup>1</sup>, A. Lopez-Martens<sup>1</sup>, R. Lozeva<sup>1</sup>, I. Matea<sup>1</sup>, K. Miernik<sup>8</sup>, J. Nemer<sup>1</sup>, S. Oberstedt<sup>23</sup>, W. Paulsen<sup>2</sup>, M. Piersa<sup>8</sup>, Y. Popovitch<sup>1</sup>, C. Porzio<sup>11,13,24</sup>, L. Qi<sup>1</sup>, D. Ralet<sup>25</sup>, P.H. Regan<sup>3,12</sup>, K. Rezyunkina<sup>26</sup>, V. Sánchez-Tembleque<sup>10</sup>, S. Siem<sup>2</sup>, C. Schmitt<sup>15</sup>, P.-A. Söderström<sup>4,27</sup>, C. Sürder<sup>4</sup>, G. Tocabens<sup>1</sup>, V. Vedia<sup>10</sup>, D. Verney<sup>1</sup>, N. Warr<sup>6</sup>, B. Wasilewska<sup>14</sup>, J. Wiederhold<sup>4</sup>, M. Yavahchova<sup>28</sup>, F. Zeiser<sup>2</sup> and S.Ziliani<sup>11,13</sup>

<sup>1</sup>Université Paris-Saclay, CNRS/IN2P3, IJCLab, 91405 Orsay, France

<sup>2</sup>University of Oslo, Department of Physics, P.O. Box 1048, Blindern, 0316 Oslo, Norway

<sup>3</sup>Department of Physics, University of Surrey, Guildford GU2 7XH, United Kingdom

<sup>4</sup>Technische Universität Darmstadt, Fachbereich Physik, Institut für Kernphysik, 64289 Darmstadt, Germany

<sup>5</sup>LPC Caen, 6 Boulevard Maréchal Juin, 14000 Caen, France

<sup>6</sup>Institut für Kernphysik, Universität zu Köln, 50937 Köln, Germany

<sup>7</sup>CEA/DAM Bruyères-le-Châtel 91297 Arpajon Cedex, France

<sup>8</sup>Faculty of Physics, University of Warsaw, PL 02-093 Warsaw, Poland

<sup>9</sup>IFIC, CSIC-University of Valencia, Valencia, Spain

<sup>10</sup>Grupo de Física Nuclear & IPARCOS, Universidad Complutense de Madrid, CEI Moncloa, 28040 Madrid, Spain

<sup>11</sup>INFN sez. \ Milano, Via Celoria 16, 20133 Milano, Italy

<sup>12</sup>National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom

<sup>13</sup>Dipartimento di Fisica, Università degli Studi di Milano, Via Celoria 16, 20133 Milano, Italy

<sup>14</sup>Institute of Nuclear Physics, Polish Academy of Sciences, PL-31342 Krakow, Poland

<sup>15</sup>IPHC, 23 rue du Loess, 67037 Strasbourg, France

<sup>16</sup>CNRS, UMR7178, 67037 Strasbourg, France

<sup>17</sup>RIKEN Nishina Center, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

<sup>18</sup>School of Physics and Astronomy, University of Manchester, Oxford Road, Manchester M13 9PL, United Kingdom

<sup>19</sup>Subatech, IMT-Atlantique, Université de Nantes, CNRS-IN2P3, F-44307 Nantes, France

<sup>20</sup>INFN Laboratori Nazionali di Legnaro, Viale dell'Università, 2, I-35020 Legnaro, Italy

<sup>21</sup>GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstr. 1, 64291 Darmstadt, Germany

<sup>22</sup>CENBG, UMR5797, Université de Bordeaux, CNRS, F-33170 Gradignan, France

<sup>23</sup>European Commission, Joint Research Centre, Directorate G for Nuclear Safety and Security, Unit G.2, 2440 Geel, Belgium

<sup>24</sup>TRIUMF, 4004 Wesbrook Mall, Vancouver, BC V6T 2A3, Canada

<sup>25</sup>Grand Accélérateur National d'Ions Lourds, Bd Henri Becquerel, 14076 Caen, France

47 <sup>26</sup>*Institute for Nuclear and Radiation Physics, KU Leuven, 3000 Leuven, Belgium*

48 <sup>27</sup>*Extreme Light Infrastructure-Nuclear Physics (ELI-NP)/Horia Hulubei National Institute for*  
49 *Physics and Nuclear Engineering (IFIN-HH), Strada Reactorului 30, 077125 Bucharest-*  
50 *Măgurele, Romania*

51 <sup>28</sup>*Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences, 1784*  
52 *Sofia, Bulgaria*

53 <sup>29</sup>*Institute for Nuclear Research (Atomki), H-4001 Debrecen, Hungary*

54  
55 **When a heavy atomic nucleus fissions, the resulting fragments are observed to emerge spinning<sup>1</sup>;**  
56 **this phenomenon has been an outstanding mystery in nuclear physics for over 40 years<sup>2,3</sup>. The**  
57 **internal generation of around 6-7 units of angular momentum in each fragment is particularly**  
58 **puzzling for systems which start with zero, or almost zero, spin. There are currently no**  
59 **experimental observations which enable decisive discrimination between the many competing**  
60 **theories for the angular momentum generation mechanism<sup>4-12</sup>. Nevertheless, the present**  
61 **consensus is that excitation of collective vibrational modes generate the intrinsic spin before the**  
62 **nucleus splits (pre-scission).**

63 **Here we show that there is no significant correlation between the spins in fragment partners,**  
64 **which leads us to conclude that angular momentum in fission is actually generated *after* the**  
65 **nucleus splits (post-scission). We present comprehensive data showing that average spin is**  
66 **strongly mass dependent, varying in saw-tooth distributions. We observe no significant**  
67 **dependence of fragment spin on the mass or charge of the partner nucleus, confirming the**  
68 **uncorrelated, post-scission nature of the spin mechanism. To explain these observations, we**  
69 **propose that collective motion of nucleons in the ruptured neck of the fissioning system generates**  
70 **two independent torques, analogous to the snapping of an elastic band. A parametrisation based**  
71 **on occupation of angular momentum states according to statistical theory well-describes the full**  
72 **range of experimental data. This new information on the role of spin in nuclear fission is not only**  
73 **important for the fundamental understanding and theoretical description of fission, but also has**  
74 **consequences for the  $\gamma$ -ray heating problem in nuclear reactors<sup>13,14</sup>, for the study of the structure**  
75 **of neutron-rich isotopes<sup>15,16</sup>, and for the synthesis and stability of super-heavy elements<sup>17,18</sup>.**

76  
77 The stability of heavy atomic nuclei is governed by a delicate balance between the  
78 Coulomb repulsion of the protons which attempt to deform the nucleus, the nuclear surface  
79 tension which drives towards spherical configurations, and quantum shell effects which add  
80 extra stability for certain nuclear shapes. Fission occurs when there is a perturbation of this  
81 balance in favour of the Coulomb repulsion. It is an exothermic, dynamical process that begins  
82 as instability in the nuclear shape, which after passing the point of no-return (saddle point)  
83 becomes more and more elongated. The nascent fragments form a neck as they move rapidly  
84 apart, which quickly snaps (scission). Shell effects in the nascent fragments give rise to certain  
85 favoured mass splits, which for low energy fission of actinide nuclei (typically containing  $\sim 240$   
86 nucleons) produces a light fragment of mass  $A \sim 100$  and a heavy fragment of mass  $A \sim 140$ .  
87 Post scission, the decay of each excited fragment is a statistical process. It initially proceeds  
88 through efficient removal of excitation energy via emission of typically 0 - 2 neutrons and 1 - 3  
89 high-energy  $\gamma$  rays. Subsequently, the emission of several more  $\gamma$  rays, which usually carry  
90 away two units of angular momentum each, removes the majority of the angular momentum  
91 and the remaining excitation energy. This prompt de-excitation process ends at the fragment  
92 ground states usually within a few nanoseconds<sup>19</sup>.

93 There are many competing theories of how a fissioning nucleus generates its intrinsic  
94 angular momentum, and where in the above sequence of events it occurs. One class of  
95 explanations proposes that it arises from the excitation of collective vibrational modes such as

96 bending, wriggling, tilting and twisting of the system before it splits (pre-scission). These  
97 theories suggest the vibrations are either initiated by thermal excitations<sup>4-6</sup>, arise from quantum  
98 fluctuations<sup>7,8</sup>, or both together<sup>9</sup>. Post-scission theories suggest that the angular momenta are  
99 generated either from Coulomb forces<sup>10</sup> or from deformed fragments which have coupled  
100 orientations<sup>11,12</sup>. Since the angular momentum is quickly carried away by the  $\gamma$  rays, the  
101 experimental study of the generation mechanism necessarily involves detailed observation of  
102 the prompt  $\gamma$ -ray emission.

103 Experimental attempts to understand the intrinsic spin generation started with low-  
104 resolution detection of prompt fission  $\gamma$ -rays in correlation with fragment mass<sup>20,21</sup>, which  
105 revealed saw-tooth shapes in the  $\gamma$ -ray yields which are strongly related to spin. The major  
106 difficulty was the separation of  $\gamma$  rays emitted from the two fragments, and the existence of  
107 these patterns was called into question in a later experiment where no saw-tooth was  
108 observed<sup>22</sup>. Another experimental approach involves spectroscopy of isomeric (long lived)  
109 excited states found in certain nuclei. Measurements of isomer population are highly sensitive  
110 to small relative changes in spin. However, only a small subset of all the isotopes produced in  
111 fission have such isomeric states and it is difficult to measure trends over a large range in  
112 mass<sup>23</sup>. In this present work, we use a third technique<sup>24</sup> based on high-resolution spectroscopy  
113 which allows both separation of  $\gamma$  rays from the two fragments and the study of trends over a  
114 large mass ranges.

115 To probe intrinsic generation of angular momentum also requires systems with initial  
116 spin of zero or almost zero, namely spontaneous fission or neutron-induced fission. Heavy-ion  
117 or charged-particle-induced fission reactions are unsuitable since they generate high initial  
118 angular momenta<sup>25</sup>, which can obscure the origin of the intrinsic spin.

119 We present unique and extensive experimental data obtained from fission experiments  
120 carried out at the ALTO facility of IJC Laboratory in Orsay with the LICORNE directional,  
121 neutron source<sup>26,27</sup> coupled to the high-performance  $\nu$ -Ball  $\gamma$ -ray spectrometer<sup>28</sup>. We carried out  
122 high-resolution spectroscopy of fast-neutron-induced fission of <sup>232</sup>Th and <sup>238</sup>U, and the  
123 spontaneous fission of <sup>252</sup>Cf with the addition of an ionisation chamber<sup>29</sup>.

124

## 125 **Results**

126 For each of the three systems studied we identified characteristic  $\gamma$ -ray decay patterns of  
127 excited states in around 30 even-even nuclei (with even numbers of both protons and neutrons).  
128 For each even-even fission fragment we extracted the average spin after neutron emission  
129 using a method developed at the University of Manchester<sup>24</sup> which combines all the available  
130  $\gamma$ -ray transition intensity and coincidence information (see methods).

131 Our results (Fig. 1) definitively confirm that fragment spins vary strongly as a function  
132 of fragment mass in saw-tooth distributions, similar to the patterns previously observed in  $\gamma$ -  
133 ray yields<sup>20,21</sup>. We note that a given fragment spin appears to depend only on the fragment  
134 mass, with no significant relationship to the mass of the system which emits it nor the mass or  
135 charge of the partner nucleus with which it emerged. This observation does therefore not  
136 support theoretical explanations based on post-scission Coulomb effects<sup>10</sup>, where a dependence  
137 of spin on the product of the fragment charges,  $Z_1Z_2$ , would be expected.

138 Additionally, large asymmetries in average spin are observed for certain fragment pair  
139 combinations (e.g. <sup>86</sup>Se and <sup>150</sup>Ce from <sup>238</sup>U(n,f)), where the spin of the heavy fragment can be  
140 more than double that of its light partner. The existence of such asymmetries does not support  
141 the post-scission explanation based on coupled orientations of deformed fragments<sup>11,12</sup>, which  
142 explicitly predicts spins of equal magnitudes. Indeed, the existence of such large spin  
143 asymmetries provokes the question of how spin generation could possibly occur pre-scission if

144 the fragments are in contact and participating in a correlated collective motion. In that case  
145 expected fragment spins at scission would be  $+I$  and  $-I$  units. To investigate further, we studied  
146 the correlation between spins of the most strongly populated fragments in the  $^{238}\text{U}(n,f)$   
147 reaction. For a given nucleus,  $\gamma$ -ray transitions of increasing spin were selected from its  
148 partner, constraining the partner population to higher and higher spins. We then examined how  
149 the average spin of the given nucleus evolved in response (Fig. 2). For example, the most  
150 strongly populated partner nucleus of  $^{96}\text{Sr}$  is  $^{140}\text{Xe}$ . By demanding observation of a  $\gamma$  ray  
151 emitted from the lowest  $8^+$  state in  $^{140}\text{Xe}$  we constrain this nucleus to be populated with  
152 average spins higher than 8 units of angular momentum. The corresponding average spin in  
153  $^{96}\text{Sr}$  is deduced by measuring the corresponding coincident  $\gamma$  ray intensities. By varying the  
154 spin conditions and the isotopes studied, we obtain the fragment spin correlations.

155 The observed slopes are clearly consistent with zero, suggesting an uncorrelated, post-  
156 scission spin generation mechanism. The data do not support pre-scission theoretical  
157 explanations<sup>4-9</sup> confirming what was suspected from the large spin asymmetries (Fig. 1). It  
158 appears that each fragment has no knowledge of the spin generated in its partner.

159 This unexpected conclusion may resolve the historical controversy surrounding  
160 previous experimental results<sup>20,21,22</sup>. For fragment spins which are generated independently, the  
161 event-by-event correlations measured in<sup>22</sup> would not be expected to generate a saw-tooth  
162 pattern. Hence this absence of saw-tooth may support rather than contradict our current  
163 findings.

164  
165 **Discussion**  
166 A post-scission, uncorrelated origin of angular momentum suggests that the fragments have  
167 become two separate, independent quantal systems. This can be viewed from both macroscopic  
168 and statistical/single-particle points of view.

169  
170 **Post-scission generation of two independent torques** Macroscopically, we suggest that  
171 fragments acquire their spin in a process analogous to the snapping of an elastic band. A neck  
172 forms between the two emerging fragments which undergoes first a stretching, then a rupture  
173 and finally a relaxation during which the potential energy from the deformed neck (i.e. elastic)  
174 transforms into kinetic energy. For asymmetric fission of the actinide nuclei we assume a  
175 double cluster, with the cores of the nascent fragments lying near doubly-closed shells and the  
176 remaining nucleons from the neck shared between them after rupture (Fig. 3).

177 We suggest that the nucleons from both halves of the ruptured neck drive the  
178 generation of angular momentum in each fragment. The relative sizes of torques will depend  
179 on the number of neck nucleons and thus the precise location of the neck rupture, i.e. the  
180 configuration at scission. Classically, the neck would rupture in the middle at its weakest point.  
181 However, in the subatomic world a gap can appear at any point<sup>30</sup>, with decreasing probabilities  
182 for more extreme partitions. We suggest that how the system arrives at a specific scission  
183 configuration will not have any subsequent impact on the generation of post-scission spin and  
184 that the fragments retain no memory of their formation after scission.

185 At scission the former neck nucleons are located far from the centre of masses of the  
186 newly-born fragments in two very elongated configurations. Such extreme elongations have  
187 large surface energies which provide the restoring forces towards more spherical shapes.  
188 Fluctuations in the aggregate direction of motion of these former neck nucleons generate the  
189 two independent torques. Small angular deviations from the fission axis of the collective  
190 nucleon motion must occur due to Heisenberg's uncertainty principle for spin/orientation of a  
191 system<sup>31</sup>. Uncertainties in the direction of the resulting linear momentum, along the fission axis

192 will result in small perpendicular components that will generate a distribution of angular  
 193 momenta. Angular momenta in both fragments will point in a plane perpendicular to the fission  
 194 axis consistent with previous experiments<sup>1</sup>, although there will be no correlation or constraint  
 195 on their relative orientations. The resulting orbital angular momentum,  $\vec{I}_o$ , of the fragments  
 196 with respect to each other, generated by the components of the motion perpendicular to the  
 197 fission axis, assures the conservation of the total angular momentum  $\vec{I}_1 + \vec{I}_2 + \vec{I}_o = 0$ .

198 The dramatic fragment shape change from elongated to more spherical shapes will also  
 199 generate heat as the surface energy converts into internal excitation energy, setting the stage for  
 200 subsequent evaporation of neutrons. Angular momentum, excitation energy and emitted  
 201 neutron multiplicity will thus be strongly correlated with each other. Indeed, similar saw-tooth  
 202 distributions are known to occur in average neutron multiplicities as a function of fragment  
 203 mass<sup>32,33</sup>.

204  
 205 **Comparison of the variation in average spins to that expected from statistical theory** In  
 206 the statistical/single-particle view, if the newly-formed fragments are independent, then their  
 207 excited states would be expected to have an angular momentum occupation entirely according  
 208 to statistical theory. For an excited nucleus, the probability distribution,  $P(I)$ , of angular  
 209 momenta,  $I$ , was first derived by Hans Bethe<sup>34</sup> and is expected to be:

$$P(I|\sigma^2) = \frac{2I + 1}{2\sigma^2} \exp\left(-\frac{(I + 1/2)^2}{2\sigma^2}\right)$$

211 where  $\sigma$  is known as a spin-cutoff parameter describing the width of this distribution and is  
 212 directly related to the average spin value,  $\langle I \rangle \approx 1.15\sigma$ . From statistical theory (see methods)  
 213 we derive a smooth parametrisation which can be tested for compatibility with our average  
 214 spin data (Fig. 1):

$$\langle I \rangle = cA_N^{1/4} A_F^{7/12}$$

217 where  $c$ , is a constant and the only free fit parameter,  $A_F$  is the fragment mass, and  $A_N$  is the  
 218 mass of neck nucleons. For light and heavy fragments we use values of  $A_N = (A_F - 78)$  and  
 219  $A_N = (A_F - 130)$  near the doubly-magic Ni and Sn shell closures. The derivation presented  
 220 here has limitations and is not intended as a full description, but as an illustration of the idea  
 221 (see methods). An extended theoretical description would involve more complex dependencies  
 222 of the parameters due to structure effects.

224 Six independent fits using the above parametrisation for each light and heavy peak in  
 225 the three different systems were performed. The fitted constants are remarkably similar, with a  
 226 mean of  $c = 0.196$  and standard deviation of  $\sigma_c = 0.009$ , a relative variation of only  $\sim 4\%$   
 227 suggesting that the fragment spins fall on a universal curve. This simple parametrisation thus  
 228 appears to capture the main ingredients of the spin-mass relationship. We conclude that the  
 229 experimentally observed variation in the average spins is thus consistent with what is expected  
 230 from statistical theory for a post-scission, uncorrelated, spin generation mechanism. There may  
 231 be other second order effects (e.g. Coulomb forces) that are not yet accounted for, but these are  
 232 clearly small.

233 A concise suggestion for the mechanism of intrinsic angular momentum generation in  
 234 the light of our new data is as follows, although we recognise that other interpretations may  
 235 also be possible:

236 A fissioning nucleus which starts with zero or near-zero spin undergoes: (i) an  
237 unstoppable shape instability from Coulomb forces; (ii) a neck formation between the two  
238 emerging fragment clusters; (iii) a neck stretching and rupture (scission) with the birth of two  
239 deformed, newly-independent quantal systems; (iv) a shape relaxation of each fragment as  
240 surface potential energy converts to excitation of the internal nucleonic degrees of freedom; (v)  
241 a resulting occupation of different angular momentum states occurring entirely according to  
242 statistical theory for two independent excited nuclei.

243 In the equivalent macroscopic picture, the last two steps can also be seen as: (iv) a  
244 shape relaxation where aggregate collective motions of the nucleons have off-axis components  
245 generating two independent torques; (v) the statistical distributions of torques creates two  
246 independent distributions of spins.

## 247 **Consequences**

248 Understanding the angular momentum generated in fission is important for fundamental  
249 reasons, but also has consequences for other fields. In nuclear energy applications, fragment  
250 spin is related to reactor  $\gamma$ -ray heating effects<sup>13,14</sup>, either through the number of prompt  $\gamma$  rays  
251 that transport it during reactor operation, or the delayed  $\gamma$  rays from isomeric states which  
252 contribute to the decay heat after reactor shutdown. For these reasons many recent  
253 measurements of prompt  $\gamma$ -ray characteristics such as average multiplicity  $M_\gamma$  have been  
254 carried out<sup>35-37</sup>. Currently, only purely empirical connections between these characteristics and  
255 mass of the fissioning system have been made<sup>38</sup>. However, we are now able to understand  
256 better the underlying fundamental relationships if we combine our data with known fragment  
257 yield information (Fig. 4). Here, we manage to relate two independent average quantities, the  
258 mass and the spin, for light, heavy and average fragment masses, and use our parametrisation  
259 to make predictions for other systems.

260 We also note that fission is a production mechanism used to study the structure of  
261 exotic nuclei<sup>15,16</sup>. Thus, understanding spin generation will allow determination of which  
262 excited nuclear states can be accessed.

263 Finally, outside the actinide region fragment-yield distributions evolve due to the  
264 changing influence of shell closures. For example, a transition from asymmetric to symmetric  
265 fission occurs for nuclei beyond  $^{258}\text{Fm}$ <sup>39</sup>. In the newly-discovered region of  $\beta$ -delayed fission<sup>40</sup>  
266 around  $^{180}\text{Hg}$ , the shell effects which drive the configuration at scission are not well  
267 understood. For fission regions which are less well explored, measurements of spin-sensitive  $\gamma$   
268 ray data could yield valuable information on neck formation and the relevant shell closures  
269 involved.

## 270 **Conclusion**

271 A full theoretical description of nuclear fission requires incorporation of the intrinsic angular  
272 momentum generation mechanism. We have presented extensive experimental data on  
273 fragment spins in different systems from which it is now finally possible to discriminate  
274 between the many competing theoretical explanations of this mechanism. We show that  
275 fragment spins are uncorrelated, demonstrating the post-scission nature of the mechanism.  
276 Theoretical explanations based on pre-scission collective vibrations<sup>4-8</sup>, post-scission Coulomb  
277 excitations<sup>10</sup>, or coupling through fragment deformations are not supported by our data<sup>9,11</sup>. A  
278 parametrisation based on the expected occupation of spin states according to statistical theory  
279 well-describes the experimentally observed mass dependence of average spins.  
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370

371 **Fig. 1 | Dependence of average spin on fragment mass.** Average spins extracted for even-  
372 even nuclei produced in fast-neutron induced fission of  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and spontaneous fission of  
373  $^{252}\text{Cf}$  are presented along with statistical uncertainties. Single-parameter fits to the data are  
374 shown in black lines. The fitting parametrisation developed to explain the angular momentum  
375 generation mechanism is presented in the discussion section.

376 **Fig. 2 | Correlation between fragment spins.** Correlations between fragment and partner  
377 spins for the six most strongly populated fragments in the  $^{238}\text{U}(n,f)$  reaction with associated  
378 statistical uncertainties. Weighted linear fits to the data points for each nucleus are shown. The

379 fitted slopes are compared to the expected slopes for the spin mechanisms (a) pre-scission with  
380 correlated spins or (b) post-scission with uncorrelated spins in the inset. The blue band (a) was  
381 determined from Monte-Carlo simulations of the de-correlating effects of the neutrons and  
382 statistical  $\gamma$  rays (see methods and extended data Fig.3).

383  
384 **Fig. 3 | Schematic diagram of post-scission angular momentum generation.** Independent  
385 torques for different scission configurations are shown with neck nucleons displayed in green.  
386 The straight black arrows illustrate sizes and example directions of the linear momentum  
387 vectors which generate the associated angular momenta. The corresponding positions on the  
388 saw-tooth distribution of the resulting average spins are shown on the right.

389 **Fig. 4 | Relationship between average fragment spins and average masses.** Fragment yield-  
390 averaged data with statistical uncertainties are shown. Left Panel: Data are compared to  
391 predictions from the parametrisation for the three fissioning systems, with light peak (L), heavy  
392 peak (H) and average for the system (A) marked. Right panel: The same data are plotted as a  
393 function of the average fragment mass with the black lines showing predictions for other  
394 systems from the parametrisation. Purple diamonds show specific predictions for the major  
395 fissile isotopes,  $^{233,235}\text{U}$ ,  $^{239}\text{Pu}$  and in addition,  $^{245}\text{Cm}$ .

396

## 397 **Methods**

398

399 **Experimental Setup** Samples of  $^{238}\text{U}$  (81 g) and  $^{232}\text{Th}$  (129 g) were irradiated with a pulsed  
400 neutron beam from the LICORNE neutron source (400 ns period) in the centre of the v-Ball  
401 spectrometer for total acquisition times of 216 hours and 450 hours, respectively. The average  
402 neutron energy that provoked fission was 1.9 MeV. Triggerless data from the 184 detectors in  
403 the v-Ball array were written to disk at high data rates of typically between 1 and 3 million  $\gamma$ -  
404 ray hits per second, and processed later offline. Each detected  $\gamma$ -ray energy was associated with  
405 a unique 64-bit time stamp accurate to sub-nanosecond precision, thanks to the state-of-the-art  
406 FASTER digitisation system<sup>41</sup>.  $\gamma$ -ray coincidence events were identified offline with a  
407 minimum trigger condition defined as at least two unsuppressed high-resolution Germanium  
408 (Ge) detectors and at least one other detector module (BGO or LaBr<sub>3</sub>) firing within a short 80  
409 ns time window. These events were subsequently sorted into two- and three-dimensional  
410 histograms for further offline analysis. An additional data set was gathered from the  $^{252}\text{Cf}$   
411 spontaneous fission source inside an ionisation chamber<sup>29</sup> placed in the centre of the v-Ball  
412 array for 52 hours. With this latter setup, one fragment was detected in-flight, while the other  
413 fragment was stopped in the backing of the sample.

414

415 **Data Analysis** Examples of  $\gamma$ -ray coincidence spectra are shown in Extended Data Fig. 1 and  
416 Extended Fig. 2. which support the main findings of the article. The lack of dependence of the  
417  $^{140}\text{Xe}$  intensity pattern on the fissioning system is shown in Extended Data Fig. 1, and the lack  
418 correlation between  $^{140}\text{Xe}$  and  $^{96}\text{Sr}$  fragment intensity patterns are shown in Extended Data Fig.  
419 2.

420

421 The main experimental data on average spins after neutron emission presented in this  
422 article rely on a method which was initially developed at Manchester University in the late  
423 1980's and described fully in ref<sup>24</sup>. It will henceforth be referred to as the Manchester Spin  
Method (MSM).

424 The MSM relies on measuring the relative intensity of every resolvable  $\gamma$ -ray transition  
 425 for a given nucleus populated in the reaction of interest. At each level with spin  $I$ , the intensity  
 426 difference between the observed ingoing and outgoing transitions is computed. This difference  
 427 is defined as the direct side-feeding,  $S$ , of the state. The average spin populated is therefore the  
 428 side-feeding-weighted average of the level spins over all  $n$  levels,  $\langle I \rangle = \sum_{i=1..n} I_i S_i /$   
 429  $\sum_{i=1..n} S_i$ . A further small correction in the result is necessary to account for the angular  
 430 momentum carried away by the statistical transitions, which depends on the reaction and is  
 431 deduced from  $\gamma$ -ray decay models at around 1 extra unit in the case of fission.  
 432 The MSM condenses all the available  $\gamma$ -ray intensity and coincidence data for a given nucleus  
 433 into a single number, the average spin after neutron emission. It is thus a powerful  
 434 experimental tool to study angular momentum effects in nuclear reactions. The method  
 435 measures a cumulative intensity flow through many different excited states, all of which will  
 436 eventually reach the ground state. There is a redundancy in the measured information and the  
 437 method has a low sensitivity to individual  $\gamma$ -ray intensities (i.e. a large perturbation in the  
 438 intensity value of any particular  $\gamma$  ray from the decay pattern has a small impact on the result).  
 439 For example, the inclusion or exclusion of the intensities of transitions from states other than  
 440 those with the lowest energy for a given spin (so-called yrast states) levels in the calculation is  
 441 seen to have very little impact on the result (see the section on “non-inclusion of weak  
 442 transitions”). If the yrast sequence of transitions is observed, then an average spin can be  
 443 extracted from the data.

444 We note that the first experiment where the MSM was applied used only 12 Compton  
 445 suppressed small-volume Ge detectors to study heavy-ion induced fission. In our work, we  
 446 measure neutron-induced fission with a high-performance third-generation  $\gamma$ -ray spectrometer  
 447 with 106 large-volume, Compton Suppressed Ge crystals and state-of-the-art, triggerless signal  
 448 digitisation technology.

450 **Application of the MSM to v-Ball data** In the present work on nuclear fission, we measured  
 451 the average spin in around 30 even-even nuclei in each system (see Extended Data Tables 1, 2  
 452 and 3). Even-even nuclei have relatively simple, well-known, decay schemes and are generally  
 453 much easier to study. Even-odd and odd-odd isotopes often have highly fragmented decay  
 454 patterns with many low-energy transitions, which are difficult to detect. The presence of a  
 455 neutron beam pulsation, or in the case of the spontaneously fissioning  $^{252}\text{Cf}(\text{SF})$ , direct  
 456 detection of one of the fission fragments in the ionisation chamber is crucial for distinguishing  
 457 between  $\gamma$  rays from prompt fission and those from subsequent fragment  $\beta$  decays. This latter  
 458 source of  $\gamma$  rays is usually associated with low multiplicity events ( $M_\gamma \sim 2-3$ ), but for certain  
 459 isotopes can be comparable to that of fission. Emission of  $\gamma$  rays after  $\beta$  decays is uncorrelated  
 460 in time, whereas 95% of prompt fission  $\gamma$  rays are emitted within a few nanoseconds of the  
 461 beam pulse or fission event. Without the beam pulsation,  $\gamma$  rays from  $\beta$ -decay and prompt  
 462 fission events are difficult to discriminate. This can lead to difficulties in extracting fragment  
 463 average spin from intensity measurements, since population of nuclei via both processes  
 464 occurs. The closer to stability the nucleus, the more of a problem this represents.  $^{252}\text{Cf}(\text{SF})$  has  
 465 been extensively studied by spectroscopists over the last twenty years<sup>42,43</sup> but mostly from data  
 466 sets without direct fission fragment detection, where the primary focus has been on extending  
 467 knowledge on the nuclear structure of exotic neutron-rich nuclei. Spin effects in  $^{252}\text{Cf}(\text{SF})$  may  
 468 have been difficult to study without an ability to discriminate  $\gamma$  rays from fission and  $\beta$ -decay.

470  **$\gamma$ -ray coincidence data and efficiency calibrations** Application of the MSM requires  
 471 determination of the  $\gamma$ -ray full energy peak detection efficiency over a wide range of energy

472 100 keV - 5 MeV. Each of the three systems studied ( $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{252}\text{Cf}$ ) has its own unique  
473 efficiency curve due to different target/chamber geometries producing slightly different self-  
474 shielding effects at lower energies. These were determined by combining GEANT IV<sup>44</sup>  
475 simulations of the setup for the highest energy part (2 - 5 MeV), source measurements, and  
476 measurements from the fission coincidence data for the lowest energy part (100 - 500 keV).  
477 For the lowest part of the energy range, self-shielding effects in the massive  $^{232}\text{Th}$  and  $^{238}\text{U}$   
478 targets are particularly important, and are difficult to simulate due to the complex, non-uniform  
479 distribution of fissions within these targets. The drop in efficiency below 200 keV is significant  
480 and is measured from the experimental data by gating on  $\gamma$ -ray yrast cascades in rotational  
481 nuclei (e.g. Ce and Mo) from above and measuring efficiency ratios for the transitions below.  
482 Uncertainties on these efficiencies are included in the data analysis for the measurement of  $\gamma$ -  
483 ray intensities and in the subsequent deduction of average spins after neutron emission.

484  
485 **Fitting procedures** Global fits of many thousands of  $\gamma$ -ray coincidences were performed in  
486 two dimensional (2D)  $\gamma$ - $\gamma$  coincidence matrices using the Radware software package<sup>45</sup>. Two  
487 dimensional analysis is essential to measure 4+ state side-feedings. Since many nuclei share  
488 similar transition energies, a global 2D fitting procedure is needed for accurate measurements  
489 of transition intensities. Odd-even and odd-odd nuclei also need to be included so that all the  
490 possible coincidences can be identified in a particular matrix slice or region. Level scheme  
491 information from the evaluated ENSDF libraries<sup>46</sup> containing level spins, excitation energies,  
492 transition energies and coincidence relationships, are used as the starting point for each  
493 nucleus. Peak width parameters are fixed from a pre-determined width calibration as a function  
494 of energy. The intensities,  $G_k$ , and energies of the observable transitions are then fitted  
495 simultaneously for all nuclei in a global fit with thousands of free parameters. Subsequent local  
496 fits for each nucleus are then performed to check convergence at the local level, with global  
497 parameters fixed and only local parameters free to vary. Global and local fits are then repeated  
498 iteratively until convergence is achieved. At each stage, Radware calculates a  $\chi^2/\text{degree of}$   
499 freedom which is used to verify and assure convergence for each nucleus. Additionally,  
500 Radware also allows for powerful visual comparisons between the fitted  $\gamma$ -ray coincidences  
501 and the experimental spectral data. This facilitates a large number of visual checks to be  
502 performed to assure the level scheme of each fragment is correctly fitted and the local fit has  
503 fully converged. Global fits serve only as second order corrections to fit properly the rare  
504 occurrences that one fragment contains a pair of transitions of similar energies to those in  
505 another fragment.

506 To process the results of the fitting procedure and to extract side-feedings and average  
507 spins for each nucleus, new software has been developed which operates on the fitted intensity  
508 and peak position output from Radware. The side-feeding  $S_i$  of each level is computed from the  
509 sum of all observable transition intensities,  $G_k$ , feeding in and out of each level.

510 The software checks the level scheme transition intensities for self-consistency.  
511 Negative side-feedings are unphysical and if detected may signal a potential problem with the  
512 fitting of transitions feeding in or out of a particular level. Finally, the code computes the  
513 average spin for each nucleus studied by combining the level spins  $I_i$  and the side-feeding  $S_i$   
514 information  $\langle I \rangle = \sum_{i=1..n} I_i S_i / \sum_{i=1..n} S_i$ .

515  
516 **Propagation of uncertainties and variance-covariance** The computed statistical uncertainty  
517 on the intensity of a particular transition is dependent on statistical variations in the number of  
518 counts at coincidence peak positions in two dimensions. A relative uncertainty in the level of  
519 background of 5% is assumed along with a typical relative uncertainty in the detection

520 efficiency of 3%. For transitions below 200 keV the relative uncertainty on the detection  
 521 efficiency rises to 20% due to the significant drop in detection efficiency over this energy  
 522 range.

523 To determine the uncertainties on the extracted average spins  $\langle I \rangle$ , the uncertainties on  
 524 the fitted experimental intensity data are propagated through the MSM. However, the  
 525 intensities,  $G_k$ , and side-feedings,  $S_i$ , are not necessarily independent and correlations may exist  
 526 between these parameters. Therefore, correct mathematical treatment of error propagation  
 527 requires the incorporation of potential correlated sources of uncertainty. Analysis of variances  
 528 and covariances are needed first for the intensities,  $G_k$ , to determine the uncertainties on the  $S_i$   
 529 and then for the side-feedings to determine the error on average spin  $\sigma_{\langle I \rangle}$  in the following way  
 530

$$\sigma_{\langle I \rangle}^2 = \sum_{i=1..n} I_i^2 \sigma^2(S_i) + \sum_{i=1..n} \sum_{j=1..n (i \neq j)} I_i^2 I_j^2 \text{cov}(S_i, S_j)$$

531 where  $\text{cov}(S_i, S_j)$  is the matrix of covariances.  
 532

533 As the v-Ball detector array uses Ge detectors which have an excellent resolution, a  
 534 high detector granularity, and a "low" overall efficiency (~5%), the vast majority of  
 535 covariances between intensity parameters are zero. Within the same level scheme, the off-  
 536 diagonal elements of the variance-covariance matrix are typically ( $< 0.05$ ), hence the  
 537 independence of the  $G_k$ 's can be considered a realistic assumption. However, the same cannot  
 538 be said of the  $S_i$ 's which are computed from intensity differences between neighbouring  
 539 transitions in the scheme. The adjacent side-feedings  $S_i$  are thus strongly correlated with each  
 540 other giving rise to both large negative and positive off-diagonal elements in the corresponding  
 541 covariance matrix ( $\sim |0.4-0.8|$ ). To perform the propagation requires the computation of a  
 542 covariance matrix  $\text{cov}(S_i, S_j)$  for each data point of  $\langle I \rangle$ . This is complex and laborious, and  
 543 given the number of data points, each one derived from a separate level scheme with its own  
 544 unique set of coincidence relationships, this procedure for uncertainty calculation is  
 545 challenging.

546 A more practical method for obtaining good estimates of the statistical errors associated  
 547 with each average  $\langle I \rangle$  is to fit the side-feeding distribution as Extended Data Fig. 4 and use  
 548 the resulting uncertainty on the fitted average of this distribution. Here, there may be some  
 549 small dependence of the uncertainty on the exact form of the fitting function chosen. This  
 550 procedure for uncertainty estimation yields uncertainties comparable in size to the application  
 551 of variance-covariance analysis.

552 Using the example of the  $^{238}\text{U}(n,f)$  coincidence data, the observable intensities vary  
 553 from the strong, e.g.  $^{140}\text{Xe } 4^+ \rightarrow 2^+$  at 3.42(11)% of the total yield, to the very weak  $14^+ \rightarrow 12^+$  in  
 554  $^{150}\text{Ce}$  at 0.024(16)% of the total yield. The median relative statistical uncertainties on transition  
 555 intensities from the global fit is 13%, and for level side-feedings is 24%. This gives rise to a  
 556 typical relative average spin uncertainty of around 5%.

### 557 558 **Sensitivity of the MSM**

559 The level of accuracy, or sensitivity, of the MSM is an important question. To what extent are  
 560 side-feeding distributions measured at or near the yrast line distorted by local quirks of the  
 561 nuclear structure, leading to non-statistical inaccuracies in the average spin measurement for a  
 562 particular nucleus? There are two empirical answers to this question: The first is addressed by  
 563 the sensitivity analysis of the method to the inclusion or exclusion of non-yrast states (r.m.s  
 564 average difference 5.9%). This implies a potential variation in the sensitivity of the method in

565 the range of 0.3-0.6 h due to the degree of incompleteness of the spectroscopic information.

566 A second estimation of the sensitivity, or accuracy of the MSM can be obtained from  
567 analysis of the non-statistical variations of the data points around the fitted trends. The  
568 correlation coefficients obtained from the fits (see Extended Data Table 4) have values of  
569 typically  $R^2=0.85$ , implying that  $\sim 15\%$  of the variation is not accounted for by the fit. The  
570 statistical uncertainties account for an additional  $\sim 5\%$  of the variation (e.g. r.m.s average of  
571 5.5% for the  $^{238}\text{U}(n,f)$  reaction). Hence  $\sim 10\%$  of the variation of the variation remains  
572 unaccounted for. This can originate from three sources: second order physics effects not  
573 included in the smooth parametrisation, local spin miss-assignments/errors in the literature  
574 level scheme information, and local biases due to peculiarities of the local nuclear structure.  
575 The trend is measured over a range of approximately 4-10 h and hence we deduce that in the  
576 worst case, the sensitivity of the method is in the range 0.4 -1.0 h.

577

### 578 **Methods: Corrections applied in the MSM**

579

580 The MSM involves some further small corrections due to possible residual coincidences from  $\beta$   
581 decay, for the side feeding of the first  $2^+$  state, for the presence of isomeric states, and finally  
582 for statistical transitions from the continuum of unresolved non-yrast states. The correction  
583 methods are outlined in the following subsections, followed by a description of how a  
584 transition intensity can be deduced indirectly, if it cannot be obtained directly or accurately  
585 fitted.

586

587 **Beta decay** The experimental conditions were arranged to strongly suppress  $\beta$  decay, achieved  
588 by tagging one fission fragment in the ionisation chamber for  $^{252}\text{Cf}(\text{SF})$  and by pulsation of the  
589 neutron beam with 400 ns period in the cases of  $^{232}\text{Th}(n,f)$  and  $^{238}\text{U}(n,f)$ . Additional corrections  
590 were employed to remove any residual  $\gamma$ -ray coincidences from  $\beta$  decay in the neutron-induced  
591 reaction data by subtracting an uncorrelated background. Background coincidence matrices  
592 were created using a pre-prompt trigger window 200 ns before the beam pulse of exactly the  
593 same size as the prompt window (80 ns). Typically, the correction applied is very small, since  
594 these matrices contained only 1-2% of the total counts of the prompt matrices, yet these  
595 subtractions are potentially important for the fragments closest to stability towards the end of  
596 the  $\beta$ -decay chains in fission (e.g.  $^{98}\text{Zr}$ ,  $^{142}\text{Ba}$ , etc.).  $\gamma$ -ray coincidences from these nuclei will  
597 have larger components produced by this unwanted population process. If the correction is not  
598 applied this could lead to underestimates of the average spin in these particular nuclei due to  
599 the presence of unwanted  $\beta$ -decay population pathways at lower spins.

600

601 **Determination of the side-feeding of the first  $2^+$  state** The side feeding of the first excited  
602 state ( $2^+$  in almost all even-even nuclei) cannot be measured directly from the  $\gamma$ - $\gamma$  coincidences  
603 of a particular nucleus. However, as noted in the original MSM paper<sup>29</sup>, it is possible to  
604 determine this side-feeding by selecting a strong transition in a partner fragment. The intensity  
605 ratio of the transition from the first excited state to the ground state, and the transition(s)  
606 feeding the first excited state can then be measured from the resulting spectrum and uncertainty  
607 determined. This ratio, labelled  $G_{(2/4)}$  and shown in Extended Data, Tables 1 and 2, is always  
608 greater than or equal to unity, since negative side-feedings are unphysical.

609

610 These ratios cannot be determined directly for all of the fragments studied either  
611 because they and their partners are weakly populated, or in some cases the transition energy is  
612 a doublet common to both fragment and partner, or two neighbouring partners. We therefore fit  
the trends of  $G_{(2/4)}$  ratios as a function of fragment mass in the light and heavy peaks for both

613 the  $^{232}\text{Th}$  and  $^{238}\text{U}$ , and use the fitted values with appropriate uncertainties. The  $G_{(2/4)}$  ratios for  
614 the light peak show a gradual trend towards unity at the highest masses. However, the  $G_{(2/4)}$   
615 ratio in the heavy peak is initially high (around 2.5) near the doubly-magic Sn shell closure and  
616 decreases rapidly towards unity with increasing mass. In the most extreme case, the side-  
617 feeding of the  $2^+$  state in  $^{132}\text{Sn}$  populated in the  $^{238}\text{U}(n,f)$  reaction accounts for some 60% of the  
618 total side-feeding intensity. This phenomenon may thus account for some of the observed  
619 anomaly at  $Z=50$  when using the  $\gamma$ -ray coincidence method to determine fission yields<sup>47</sup>.

620 For the  $^{252}\text{Cf}(\text{SF})$  system it is not possible to deduce the  $2^+$  side-feedings from gating on  
621 the partner fragments since the partner fragment decays in-flight, so its transitions are Doppler  
622 broadened. In the case of  $^{252}\text{Cf}(\text{SF})$  we use  $G_{(2/4)}$  ratio values deduced from the fits to the  
623  $^{238}\text{U}(n,f)$  data. The  $^{232}\text{Th}(n,f)$   $G_{(2/4)}$  ratios show similar variations with mass, but we assume the  
624  $^{238}\text{U}(n,f)$  trends provide better estimates. This is preferable to assuming  $2^+$  state side-feeding  
625 values of zero for  $^{252}\text{Cf}(\text{SF})$ , since it allows a better comparison of average spins in all three  
626 systems, but may necessarily introduce some small systematic bias.

627  
628 **Statistical transitions** The statistical side feeding transitions will also carry away a small  
629 quantity of angular momentum. In the original MSM paper calculations were used to estimate  
630 the average number of statistical transitions (2.5) and average angular momentum per transition  
631 (0.4 units)<sup>48</sup>. Here, we use these same values to facilitate comparison of results. These do not  
632 impact the shape of the observed saw-tooth distributions but will instead just shift them  
633 globally up or down in spin.

634  
635 **Isomeric states and delayed transitions** Calculating the average spin for a nucleus with a  
636 strongly populated isomer requires an additional step in the analysis. For isomeric transitions  
637 with lifetimes in the ns -  $\mu\text{s}$  range, the  $\gamma$ -ray decay below the isomer can occur outside the  
638 trigger window and thus the  $\gamma$ -rays and their coincidences with states above the isomer are not  
639 observed, leading to an underestimate of the average spin if no correction is applied. The  
640 correction for  $^{252}\text{Cf}(\text{SF})$  data is very simple, since we can just increase the size of the prompt  
641 window from 100 ns to 4  $\mu\text{s}$ . This results in an increase in the average spin of the most affected  
642 nuclei,  $^{132}\text{Sn}$  and  $^{134}\text{Te}$ , of 12% and 14% respectively. No other nuclei show statistically  
643 significant increases in the deduced average spin for an extended coincidence window.  
644 Applying corrections for isomers in the  $^{232}\text{Th}(n,f)$  and  $^{238}\text{U}(n,f)$  data sets is more difficult. The  
645 prompt window is increased from 80 ns to 400 ns, and the corresponding increase in spin for  
646 these key isomeric nuclei is measured. A further correction is then applied using an  
647 extrapolation to account for the missing isomeric coincidences beyond the 400 ns window.

648 For all three fissioning systems the nucleus  $^{130}\text{Sn}$  presents a unique problem. A  $10^+$ , 1.6  
649  $\mu\text{s}$  isomer decays to the  $7^-$  state through an unobservable 96 keV transition and this  $7^-$  state has  
650 a half-life of 1.7 m. Hence, there is missing intensity for this nucleus. We include  $^{130}\text{Sn}$  in our  
651 data, but acknowledge the existence of a potential systematic error in the calculation of the  
652 average spin for this particular case. However, since the neighbouring  $^{132}\text{Sn}$  also has a similar  
653 high spin isomer, and the inclusion or exclusion of these decays changes the average spin by  
654 only 12%, we assume that the systematic underestimate of average spin for  $^{130}\text{Sn}$  will be  
655 smaller than the statistical uncertainties.

656  
657 **Redundancy and the indirect deduction of intensity information** As mentioned previously,  
658 there is some redundancy in the  $\gamma$ -ray transition intensity since we measure a cumulative  
659 intensity flow. This redundancy can be exploited to recover missing intensity information in  
660 the rare case that it is necessary. A problem that can occur during a two-dimensional

661 coincidence analysis is that occasionally, certain coincidences or transitions can be obscured by  
662 the presence of others if they are too close in energy. This generally presents more of a  
663 difficulty for a small number of weak transitions in nuclei with the lowest production yields.  
664 For example, there is a strong background of random 511 keV  $\gamma$  rays from electron-positron  
665 annihilation. For a nucleus that has a weak transition very close to this energy it is often  
666 impossible to measure its intensity directly due to the large statistical fluctuations present after  
667 subtraction of this dominating background.

668 If such doublets or multiplets are too close in energy to resolve by two-dimensional  
669 peak fitting, information on the intensity of the obscured transition can still be recovered from  
670 the intensity flows into and out its initial and final states. For the general case, the intensity,  $G$ ,  
671 of a  $\gamma$ -ray transition between initial state  $A$  and final state  $B$  can never be smaller than the  
672 intensity balance into state  $A$  and never be larger than the intensity balance out of state  $B$ , since  
673 this would result in negative side-feedings for  $A$  or  $B$ , which is unphysical.

674 If  $G_{Ai}$  and  $G_{Ao}$  are the measured ingoing and outgoing intensities from state  $A$ , and  $G_{Bi}$   
675 and  $G_{Bo}$  those for state  $B$ , then the intensity of the missing transition intensity,  $G_x$ , must obey  
676 the following relation  $(G_{Ai}-G_{Ao}) \geq G_x \geq (G_{Bo}-G_{Bi})$ . The best estimate of the intensity  $G_x$  is thus  
677 the average  $((G_{Ai}-G_{Ao})+(G_{Bo}-G_{Bi}))/2$  of the upper and lower bound and implies the side-  
678 feedings of state  $A$  and state  $B$  are equal. The impact of deducing a  $\gamma$ -ray intensity on the  
679 average spin measured for a particular nucleus is not significant, since the measurement  
680 integrates the intensity information from many transitions and the deduced intensity is usually  
681 a very good estimate of the real intensity.

682

### 683 **Methods: Potential sources of bias**

684

685 The MSM has several sources of potential bias outlined in the following subsections:

686

687 **Level schemes** Since we are observing neutron-rich nuclei far from stability in these  
688 experiments, the level schemes in the literature (ENSDF databases) may have spin assignments  
689 of certain levels which are only tentative, and in some cases incorrect by 1 or 2 spin units. This  
690 may have a small impact on the average spin extracted for a particular nucleus. However, the  
691 main side-feeding branches usually occur at lower spins with the  $2^+$ ,  $4^+$  and  $6^+$  yrast levels  
692 accounting for a large fraction of the total side feeding intensity in most cases. These states are  
693 usually well measured with unequivocal spin assignments. Miss-assignment of the spins of  
694 certain states may either slightly lower or slightly raise the average spin deduced for a  
695 particular nucleus. However, the effect will be local, can occur in either direction, and will be  
696 confined to a particular nucleus. No global systematic effect is expected.

697

698 **Ground state feeding** The direct side-feeding of the ground state is impossible to measure  
699 using  $\gamma$ -ray spectroscopy. However, we can estimate it from an extrapolation of the spin  
700 distribution towards zero. Extended Data Fig. 4 shows how this extrapolation is performed  
701 from fits to the spin distribution associated with each data point. The extracted  $0^+$  feeding is  
702 given an appropriately large relative uncertainty. The  $0^+$  feeding is typically 3-5% of the total  
703 side-feeding intensity in most cases, but increases in the vicinity of closed-shell nuclei (up to  
704 18% in the case of  $^{132}\text{Sn}$ ). The impact of this correction on the average spin values results in a  
705 slight lowering which is smaller than the statistical errors. However, for  $^{132}\text{Sn}$  and its near  
706 neighbours with significant  $0^+$  feeding the average spin values drop more significantly by  
707 typically 0.3 h. Performing this correction has no impact on the conclusions.

708

709 **Non-inclusion of weaker transitions** A potential bias in extraction of average spin could  
710 occur as a function of the fragment yield. Nuclei which are more weakly populated in general,  
711 may have fewer observable transitions and levels available for inclusion in the weighted sum.  
712 However, we conclude that the MSM method is very insensitive with respect to the inclusion  
713 or exclusion of non-yrast levels. Provided transitions from levels in the yrast sequence are  
714 visible, a reliable extraction of average spin can be made. The non-observation of weak  
715 transitions at the top of the yrast sequence has little impact on the final result, since if the  
716 transitions are weak at this point, the side-feedings are also weak and contribute little to the  
717 result. To quantify this potential bias average spins in the 15 most strongly populated nuclei  
718 were recalculated after fitting only the yrast sequences and ignoring the presence of all other  
719 non-yrast states and transitions. The r.m.s. difference between the two sets of values was found  
720 to be 5.9%. The transition rates of statistical side-feeding transitions are orders of magnitude  
721 faster than the intra-yrast cascade transitions and this probably accounts for why the difference  
722 is small. Finally, if the measured average spins for all nuclei studied are plotted against  
723 fragment yields, the two quantities are seen to be almost entirely uncorrelated.

724  
725 **The trigger condition** For the case of the  $^{252}\text{Cf}(\text{SF})$  the trigger condition was an anode signal  
726 of the ionisation chamber corresponding to detection of one fragments in flight, with the other  
727 stopped rapidly in the backing of the sample. This gives a clean, unambiguous signal that a  
728 fission has occurred. For the  $^{232}\text{Th}(\text{n},\text{f})$  and  $^{238}\text{U}(\text{n},\text{f})$  reactions the fission discrimination is less  
729 perfect. While the beam pulsation allows discarding of events which are uncorrelated in time, a  
730 minimum multiplicity condition is also used in the prompt trigger window. This is essential to  
731 discriminate fission from the complex background of other low-multiplicity processes which  
732 also occur during the beam pulse, such as inelastic scattering  $^{238}\text{U}(\text{n},\text{n}')$ ,  
733  $^{27}\text{Al}(\text{n},\text{n}')$ ,  $^{72,73,74,76}\text{Ge}(\text{n},\text{n}')$ ,  $\text{p}(^7\text{Li}, ^7\text{Li})$  Coulomb excitation of the primary beam, and the  
734 intrinsic activity of the targets. The intrinsic activity is a particular problem for the  $^{232}\text{Th}$  target,  
735 as a fraction of its decay also occurs during the prompt beam pulse. Since the majority of  $\gamma$ -  
736 rays detected during the experiment come from these low-multiplicity processes, a minimum  
737 trigger condition of  $M_\gamma \geq 3$  is essential to preferentially select fission events. For the best  
738 discrimination the trigger condition should be placed at even higher multiplicities but we have  
739 deliberately kept it at 3 to minimise any potential trigger biases, even though this results in  
740 larger backgrounds. From the  $^{252}\text{Cf}(\text{SF})$  data it is possible to study the impact of the  
741 multiplicity trigger condition on the results. Raising the minimum trigger condition from  
742 ionisation chamber fission tag from a minimum multiplicity of two to three has no significant  
743 impact on the measured average spins. Effects are, however, observed at higher multiplicity  
744 conditions. A global increase in the average spin for all nuclei of around one spin unit is  
745 observed for an increase of around 4 units in detected multiplicity. This correlation is  
746 completely expected and gives further confidence in the key observation of this paper, namely  
747 the absence of spin correlations between fragment partners. The reason the slope of the  
748 correlation of average spin with detected multiplicity is not larger is mostly due to the  
749 imperfections in the v-Ball calorimeter (68% efficiency for detecting a single  $\gamma$  ray at 1 MeV).  
750 An event of detected multiplicity (or fold) of 4 will thus have significant contributions from  
751 emitted multiplicities of 4, 5, 6, 7 and 8.

752  
753 **Derivation of the spin parametrisation from statistical theory** The expected probability  
754 distribution,  $P(I)$ , of angular momenta,  $I$ , for an excited nucleus following the work of Hans  
755 Bethe<sup>35</sup> is:

$$P(I|\sigma^2) = \frac{2I + 1}{2\sigma^2} \exp\left(-\frac{(I + 1/2)^2}{2\sigma^2}\right)$$

756  
 757 where  $\sigma$  is known as a spin-cutoff parameter describing the breadth of this distribution. In the  
 758 Fermi gas model, the spin cut-off parameter depends directly on the nuclear temperature,  $T$ ,  
 759 and is related to the excitation energy  $E_x$  and level density parameter,  $a$ .  
 760

$$T = \sqrt{\frac{E_x}{a}}$$

761  
 762 In this model, the spin cut-off parameter is then usually defined as the product of the rigid body  
 763 moment of inertia,  $J_{rigid}$ , and the temperature:

$$\sigma^2 = J_{rigid}T$$

764  
 765 where for a spheroidal nucleus

$$J_{rigid} = \frac{2}{5}A_F R^2$$

766  
 767 so  $J_{rigid} \propto A_F^{5/3}$ . Using a level density parameter,  $a$  which is proportional to  $A_F$  the variation in  
 768 the spin cut-off parameter with fragment mass can then be defined in the following way:  
 769

$$\sigma^2 \propto \sqrt{E_x} A_F^{7/6}$$

770  
 771 If we assume that the excitation energy of the fragment is proportional to the mass of the  
 772 nucleons from the ruptured neck (i.e.  $E_x \propto A_N$ ), we obtain this final parametrisation based on  
 773 statistical theory which can be used to fit our average spin data in Fig. 1:  
 774

$$\langle I \rangle = c A_N^{1/4} A_F^{7/12}$$

775 where  $\langle I \rangle \approx 1.15\sigma$ . This smooth parametrisation of the mass dependence with only one free  
 776 parameter can be used as a fitting function, analogous to the smooth fitting of nuclear mass  
 777 variations with the Weissacker formula<sup>49</sup>. An extended theoretical description would also have  
 778 additional local variations in  $E_x(A_F)$ , level density  $a(A_F)$  and  $J_{rigid}(A_F)$  due to structure  
 779 effects. However, the smooth functional dependence of  $\langle I \rangle(A_F)$  captures the major ingredients  
 780 of the variation.

781 **Monte-Carlo code for correlated fragment spins** For the data presented in Fig. 2, a  
 782 dispersion propagation Monte-Carlo code was developed to understand what experimental  
 783 slopes we would expect to see if the intrinsic angular momentum is generated by pre-scission  
 784 mechanisms which produce correlated spins at scission (i.e. the precise width and location of  
 785 case a. in the inset of Fig. 2). The emission of neutrons and statistical  $\gamma$  rays in each fragment  
 786 will have a de-correlating effect on any spin measurements carried out at or near the yrast lines.  
 787 The precise and only purpose of this code is to propagate realistic spin dispersions from  
 788 scission to yrast simultaneously in both fragments due to emission of neutrons and statistical  $\gamma$   
 789 rays. It allows for (i) complete user control over the spin distribution parameters at scission, (ii)

790 total control over the widths of the spin dispersions due to emission of both neutrons and  $\gamma$   
791 rays, and (iii) the ability to output the resulting spin distribution observed at yrast when setting  
792 conditions on the spin distribution in the partner fragment.

793 Dispersions in spin due to neutron emission were taken as random  $\pm 0.5\hbar$  per emitted  
794 neutron. To obtain statistical  $\gamma$  ray spin dispersions, the RAINIER code<sup>50</sup> was used to fully  
795 model the  $\gamma$  decay of several representative spherical and deformed fission fragments. Typical  
796 statistical  $\gamma$  ray spin dispersion distributions with a width of around  $\pm 1.5\hbar$  were then imported  
797 into our Monte-Carlo code.

798 The placement of different gating conditions at yrast on one fragment could then be  
799 simulated and the effect on the resulting spin distribution at yrast in the partner fragment could  
800 be determined (see Extended Data Fig. 3). A simulated experimental relationship between the  
801 gating condition at yrast in one fragment and the “measured” average spin in the other could  
802 then be probed and an “experimental” slope deduced. With these tools, a sensitivity analysis of  
803 the results to the parameters of the initial spin distribution and neutron/ $\gamma$  dispersions could be  
804 performed. The blue band in the inset of Fig. 2 for case a) gives a range of expected slopes  
805 (0.4-0.6) for fully correlated spins at scission for reasonable variations of these parameters. The  
806 conclusion is that statistical emission will slightly weaken any spin correlations present at  
807 scission but will not destroy them.

808 [Note also that similar experimental data exists for 1n partners. These are not shown in  
809 Fig. 2, which would have become much too cluttered. However, the results obtained are  
810 similar, with no significant slopes observed.]

811 An extension of this Monte-Carlo code to include the  $\gamma$ -ray spectrometer granularity and  
812 detection efficiency was also developed. Here, the concern was that demanding observation of  
813 a “high spin” state in one fragment might reduce the overall efficiency of detection due to  
814 potential biases towards higher multiplicity events involving many detectors. Since v-Ball is a  
815 highly granular array (106 Ge separate Ge elements at large distances) this effect on the  
816 expected slopes proved to be completely negligible. However, for the case of very close-  
817 packed detector arrays (e.g. 6 clover detectors placed in cubic geometry) small negative slopes  
818 could arise due to such biases, suggesting an artificial anti-correlation between fragment spins.

819

## 820 **Data Availability Statement**

821 All data from which the conclusions of this paper are drawn are contained within this  
822 manuscript. All other data can be made available on reasonable request. The large quantities of  
823 raw data (approx. 120 Tb) are shared within the nu-ball collaboration on servers at the CNRS-  
824 IN2P3 Centre de Calcul in Lyon (<https://cc.in2p3.fr>). The ALTO facility of IJC laboratory has  
825 a transparent data management policy which complies with the relevant European directives on  
826 open data ([https://ec.europa.eu/digital-single-market/en/european-legislation-reuse-public-](https://ec.europa.eu/digital-single-market/en/european-legislation-reuse-public-sector-information)  
827 [sector-information](https://ec.europa.eu/digital-single-market/en/european-legislation-reuse-public-sector-information)). Raw data from the nu-ball collaboration will be made publicly available  
828 after a period of 5 years.

## 829 **Code Availability Statement**

830 All codes used in the data analysis can be made available on reasonable request.

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856

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886

### 887 **Author contributions**

888

889 JNW participated in the construction of the v-Ball spectrometer, contributed to the  
890 experimental data taking, organised the v-Ball international collaboration, performed the  
891 analysis work presented here and wrote the main body of the paper.

892 DG helped with experimental and theoretical discussions, interpretation of results, manuscript  
893 preparation and resubmissions, calculations, plots and bibliography.

894 DT constructed the spectrometer, calibrated and optimized the spectrometer, kept the  
895 spectrometer running, contributed to the experimental data taking, performed data processing  
896 of the large quantities of triggerless data and helped distribute it to the collaboration.

897 ML organised the v-Ball project, led the construction of the spectrometer, organised the  
898 experimental campaign, kept the spectrometer running, contributed to the experimental data  
899 taking and measured v-Ball performances.

900 MR, NJ, RC, GH, RL and R-BG helped with the cabling of the v-Ball spectrometer, supported  
901 the running of the spectrometer (filling with liquid nitrogen, monitoring detectors, etc),  
902 calibrated and optimised the spectrometer, contributed to the experimental data taking and  
903 performed offline data analysis.

904 DE developed and helped deploy the digital electronics used for the v-Ball DAQ.

905 LG developed and deployed the  $^{252}\text{Cf}$  ionisation chamber.

906 SO, CS, TK, PHR, ABI, NW, SL, BF, AA, MF, LF and others contributed to the theoretical  
907 discussions and interpretation of results.

908 SS helped with organisation, discussions and interpretation, bibliography and manuscript  
909 preparation.

910 FZ carried out fragment decay simulations using the RAINIER code.

911 All listed collaborators helped keep the experiment, the spectrometer and the data acquisition  
912 systems running over the period of 7 weeks during which the data were collected.

913

### 914 **Competing Interests**

915

916 The authors declare that they have no competing financial interests.

917

### 918 **Author Information**

919 Correspondence and requests for materials should be addressed to Jonathan Wilson (email:  
920 [jonathan.wilson@ijclab.in2p3.fr](mailto:jonathan.wilson@ijclab.in2p3.fr)).

921

### 922 **Extended Data**

923

### 924 **Extended Data Figure Legends and Tables**

925

926 **Extended Data Fig 1. |  $\gamma$ -ray coincidence spectra for  $^{140}\text{Xe}$ .** Spectra are gated by the  $2^+ \rightarrow 0^+$   
927 transition for the three different fissioning systems studied in this work. The spins of states  
928 emitting the yrast sequence of transitions are marked. Strong  $\gamma$  rays from the binary partner  
929 fragments are indicated.  $\gamma$  rays from fragment partners in  $^{252}\text{Cf}(\text{SF})$ , such as  $^{112}\text{Ru}$ , were  
930 detected in flight and are thus not visible due to Doppler broadening. The  $^{252}\text{Cf}(\text{SF})$  spectrum  
931 has much fewer counts, but similar experimental sensitivity is achieved due to the elimination  
932 of significant backgrounds by direct detection of the fission fragment in the ionisation chamber  
933 with the  $\gamma$ - $\gamma$  coincidences.

934  
935 **Extended Data Fig. 2 | Coincident  $\gamma$ -ray spectra from the  $^{238}\text{U}(\text{n},\text{f})$  reaction gated on**  
936 **transitions from  $^{140}\text{Xe}$  emitted from states of increasing spin.** The fits to transitions  
937 decaying out of specific states of the partner nucleus  $^{96}\text{Sr}$  are shown in red. The 492 keV  
938 transition from the  $6+$  state in  $^{96}\text{Sr}$  in the 3<sup>rd</sup> panel is deduced from its neighbours rather than  
939 fitted, due to contamination. The intensity pattern is not observed to vary and the average spins  
940 in  $^{96}\text{Sr}$  show no significant changes. The relationships between partner spins for several more  
941 nuclei are shown in Fig. 2.

942 **Extended Data Fig. 3 | Monte Carlo simulations of events with correlated spins at**  
943 **scission.** Placing conditions on the minimum spin at yrast of events in fragment 1 affects the  
944 yrast distributions of event spins in fragment 2.

945  
946 **Extended Data Fig. 4 | Examples of experimental spin distributions for a range of nuclei**  
947 **observed in the  $^{238}\text{U}(\text{n},\text{f})$  reaction.** Statistical uncertainties are shown. To eliminate the odd-  
948 even staggering effect and facilitate easy visualization, side-feedings of odd spins are  
949 redistributed equally between the two neighbouring even spins. The red curves are fits to the  
950 experimental data with one free parameter and are used to extract  $0+$  side-feedings via an  
951 iterative procedure.

952 **Extended Data Table 1:**  $^{232}\text{Th}(\text{n},\text{f})$  average spin data. Fragment yields are taken from the  
953 ENDF.BVII evaluation.<sup>51</sup>

954  
955 **Extended Data Table 2:**  $^{238}\text{U}(\text{n},\text{f})$  average spin data. Fragment yields are taken from the  
956 ENDF.BVII evaluation.<sup>51</sup>

957  
958 **Extended Data Table 3:**  $^{252}\text{Cf}(\text{SF})$  average spin data. Fragment yields are taken from the  
959 ENDF.BVII evaluation.<sup>51</sup>

960  
961 **Extended Data Table 4:** Fit parameters for the light and heavy peak data for the three  
962 fissioning systems shown in Fig. 1. The fitting function is defined as  $\langle I \rangle = cA_N^{1/4}A_F^{7/12}$  with a  
963 single free parameter,  $c$ .

964

965







