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Orlandi, R., Asai, M., Makii, H. et al. (2026) Deformation and Magicity in Heavy Actinides: First Observation of the Ground-State Rotational Band of  $^{252}\text{Fm}$ . Physical Review Letters. 082501. ISSN: 1079-7114

<https://doi.org/10.1103/7gzt-ldn5>

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# Deformation and magicity in heavy actinides: first observation of the ground-state rotational band of $^{252}\text{Fm}$

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(Dated: March 19, 2026)

The ground-state rotational band of  $^{252}\text{Fm}$  ( $Z=100$ ,  $N=152$ ) was investigated using two different experimental setups at the JAEA Tandem Accelerator Laboratory. The  $2^+ \rightarrow 0^+$  and  $4^+ \rightarrow 2^+$  transitions were observed in the  $\alpha$  decay of  $^{256}\text{No}$  via  $\alpha$ - $\gamma$  coincidence spectroscopy. The energies of the  $(6^+) \rightarrow (4^+)$ ,  $(8^+) \rightarrow (6^+)$  and  $(10^+) \rightarrow (8^+)$  transitions were identified in the prompt  $\gamma$ -ray spectrum of  $^{252}\text{Fm}$  produced in the  $^{18}\text{O}+^{249}\text{Cf}$  multi-nucleon transfer reaction. The new data reveal that, compared with neighboring fermium isotopes and  $N=152$  isotones in the region,  $^{252}\text{Fm}$  has the lowest first excited  $2^+$ -state energy, 41.88(16) keV, and the largest kinematic moment of inertia. These properties constitute new evidence for the deformed shell gaps at  $Z=100$  and  $N=152$ , supporting the understanding of  $^{252}\text{Fm}$  as a deformed doubly magic nucleus.

Nuclear structure progress is closely tied to the discovery and prediction of magic numbers, which indicate closed-shell configurations of protons and neutrons (2, 8, 20, 28, 50, 82, and 126 for neutrons) that enhance nuclear stability, and are associated with spherical nuclear shapes. Within two decades of the foundational works by Mayer and Jensen [1],  $Z=114$  and  $N=184$  were proposed as the next proton and neutron magic numbers [2, 3], with modern models suggesting  $Z=114$ , 120, or 126 and  $N=184$  [4, 5]. These shell closures are expected to hinder spontaneous fission in superheavy elements, creating a region of relatively stable nuclei known as the island of stability (IoS), whose exact location remains unknown. The nuclear structure of heavy nuclei is also strongly influenced by “deformed shell closures”, which are energy gaps among nucleon orbits in non-spherical nuclei with large quadrupole deformation ( $\beta_2 \simeq 0.25 - 0.30$ ). These are smaller than spherical shell gaps, and their effects are more localized in the  $(Z, N)$ -plane. However,

they reveal themselves in the ground-state observables, and their impact on superheavy elements can be significant [6]. For example, deformed shell gaps at  $Z=108$  and  $N=162$ , which are manifested by  $Q_\alpha$  values [7–9], enable the existence of nuclei up to near the shores of the IoS [6], and the production of nuclei as heavy as  $^{294}\text{Og}$  ( $Z=118$ ,  $N=176$ ) [10].

In the region near  $A=252$ ,  $Q_\alpha$  values, partial spontaneous-fission half-lives [11–13] and two-nucleon separation energies [14] also show evidence of deformed shell gaps at  $N=152$  and, although less pronounced, at  $Z=100$  [15, 16]. Due to the limited spectroscopic data, the origin and extent of these gaps are not fully understood, and models yield conflicting predictions (*cf.*  $Z=100$ ,  $N=152$  of microscopic-macroscopic models [9, 17] vs.  $Z=96$ , 98, or 104,  $N=150$  and 152 (and occasionally  $N=148$ ) from non-relativistic and relativistic energy density functionals [18–21]). Furthermore, nuclei near  $A=252$  are key benchmarks for nuclear models

of superheavy elements, and can provide a link to high- $j$  orbits in the IoS, such as the neutron  $h_{11/2}$  or the proton  $j_{15/2}$ ,  $i_{13/2}$  and  $f_{5/2}$ : low- $K$  components of such orbits, split by deformation, are lowered in energy and can be found near the Fermi level of deformed nuclei near  $Z=100$  and  $N=152$  [19, 22, 23].

Gamma-ray spectroscopy, and particularly the energy of the first excited  $2^+$  state,  $E(2^+)$ , and the moment of inertia (MoI) of even-even nuclei, can help to probe these gaps. At a deformed gap, in fact, the lower level density at the Fermi surface is believed to hinder pairing correlations, leading to a larger MoI [15, 17, 19] and a lower  $E(2^+)$ . Sudden changes in  $E(2^+)$  and MoI can thus be interpreted as structural transitions driven by the shell gaps. For example, the weakening of the  $N=152$  gap in Pu isotopes was elucidated from the  $\gamma$ -ray spectroscopy of  $^{245,246}\text{Pu}$  [24].

One of the least known even-even isotopes in the  $A \simeq 252$  region, mainly because of low production cross sections in fusion-evaporation reactions with stable beams and targets, is the key nucleus  $^{252}\text{Fm}$  ( $Z=100$ ,  $N=152$ ). Only its first excited state energy, determined from  $\alpha$ -decay energy differences, is currently known ( $42.1 \pm 1.3$  keV) [25]. Here we report for the first time on the measured energies of the lowest five  $\gamma$ -ray transitions of its ground-state rotational band. The  $2^+ \rightarrow 0^+$  and  $4^+ \rightarrow 2^+$   $\gamma$  rays were observed in the  $\alpha$ - $\gamma$  coincidence spectrum of  $^{256}\text{No}$   $\alpha$  decay. The energies of the  $(6^+) \rightarrow (4^+)$ ,  $(8^+) \rightarrow (6^+)$  and  $(10^+) \rightarrow (8^+)$  transitions were determined instead through the in-beam  $\gamma$ -ray spectroscopy of  $^{252}\text{Fm}$ , produced using the  $^{18}\text{O} + ^{249}\text{Cf}$  multi-nucleon transfer (MNT) reaction. Both experiments were carried out at the JAEA Tandem accelerator facility in Tokai, Japan. Since details of the experimental setups have been published elsewhere [25–27], only brief summaries are provided here.

The  $\alpha$  decay of  $^{256}\text{No}$  was studied using a gas-jet cou-

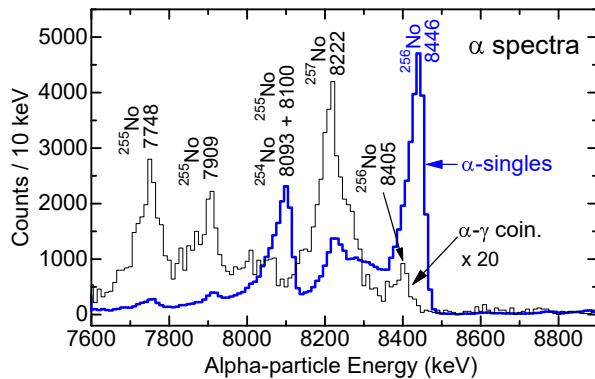


FIG. 1. Alpha-energy spectra of the No isotopes produced in the  $^{12}\text{C} + ^{249,250,251}\text{Cf}$  experiment, using the  $\alpha$ - $\gamma$  coincidence setup. The  $\alpha$ -singles spectrum observed 0–6 seconds after source deposition is shown by the thick solid line. The  $\alpha$  energy spectrum in coincidence with X and  $\gamma$  rays is multiplied by 20.

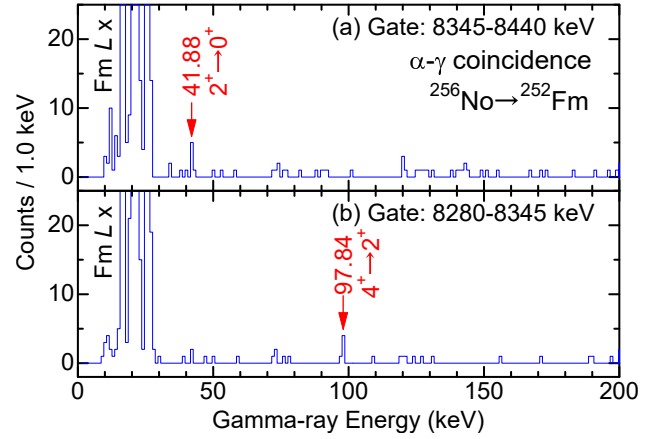


FIG. 2. Gamma-ray energy spectra in coincidence with  $\alpha$  particles, with energy (a) between 8345 and 8440 keV and (b) between 8280 and 8345 keV, corresponding to the 8405 and 8308 keV  $^{256}\text{No}$   $\alpha$  transitions to the  $2^+$  and  $4^+$  states in  $^{252}\text{Fm}$ , respectively. The  $\gamma$ -ray peaks associated with the  $\alpha$  decay of  $^{256}\text{No}$  to  $^{252}\text{Fm}$  are labeled in red.

pled  $\alpha$ - $\gamma$  detection system [26]. In the experiment, a 71.4-MeV, 540-pnA  $^{12}\text{C}$  beam impinged on a  $420 \mu\text{g}/\text{cm}^2$  mixed Cf target made of  $^{249}\text{Cf}$ (63%),  $^{250}\text{Cf}$ (12%) and  $^{251}\text{Cf}$ (25%). The target had a 1.6 mm diameter and a total radioactivity of 4.1 MBq. Reaction products recoiling out of the target were thermalized in He gas, attached onto KCl aerosol clusters, and transported through a 25-m long He-jet capillary to a rotating wheel  $\alpha$ - $\gamma$  detection system, where the products were deposited onto a  $120 \mu\text{g}/\text{cm}^2$ -thick polyethylene tetraphthalate foil. The wheel was periodically rotated to transport the deposit to a detector station equipped with two Si PIN photodiodes ( $28 \times 28 \text{ mm}^2$  active area) and two Ge detectors placed behind the PIN photodiodes. The energy resolution (FWHM) was 50 to 100 keV for  $\alpha$  particles and about 0.85 keV for 60 keV  $\gamma$  rays.

Figure 1 shows the Si spectra obtained in the  $^{12}\text{C} + ^{249,250,251}\text{Cf}$  experiment, where nobelium isotopes were produced mainly via  $^8\text{Be}$  cluster-transfer and, in part, fusion evaporation ( $\alpha\text{xn}$  channel) reactions. The peak at 8446 keV in the  $\alpha$  singles spectrum is attributed to the  $\alpha$  decay of  $^{256}\text{No}$  to the ground state of  $^{252}\text{Fm}$ ; the deduced half-life agrees very well with the literature value of 2.91(5) s. This is also supported by the fact that the 8446-keV peak disappears in the  $\gamma$ -coincident  $\alpha$  spectrum, while the 8405-keV peak (feeding the  $^{252}\text{Fm}$   $2^+$  state) becomes visible.

Figure 2 shows the  $\gamma$ -ray spectra in coincidence with the  $\alpha$  particles in the energy range (a) between 8345 and 8440 keV and (b) between 8280 and 8345 keV, which correspond to the  $\alpha$  decays of  $^{256}\text{No}$  to the  $2^+$  and  $4^+$  states in  $^{252}\text{Fm}$ , respectively. The  $\gamma$ -ray peaks at 41.88(16) keV and 97.84(20) keV in Figs. 2(a) and 2(b), observed for the first time, are attributed to the  $2^+ \rightarrow 0^+$  and  $4^+ \rightarrow 2^+$   $\gamma$ -ray transitions in the ground-state band of  $^{252}\text{Fm}$ . From the energy of these  $\gamma$  rays, the exci-

tation energy of the  $4^+$  state can be determined to be 139.7(3) keV. Taking into account the  $E2$  theoretical total internal conversion coefficients (1657 and 29.3 [28] for the 42- and 98-keV  $E2$  transitions), the  $\gamma$ -ray detection efficiency ( $\sim 35\%$  at both energies), the total detected  $^{256}\text{No}$   $\alpha$  counts (80500), and for  $\alpha$ -branching ratio of 10(2)% (deduced in [25]) to the  $2^+$  state and assuming a branching of  $\sim 0.5\%$  to  $4^+$  state, the coincident  $\gamma$ -ray counts are calculated to be 1.7(3) and  $\sim 4.6$ , respectively. These values are consistent with the observed counts of 6 and 7. The total integral of the Fm L X-ray counts in the 13–27 keV energy range, seen in Fig. 2(a), is 453, in excellent agreement with the calculated counts of 478, expected from the internal conversion of the 41.88-keV transition. The experimental and calculated X-ray spectra, as well as the  $\alpha$ - $\gamma$  coincidence matrix, are included in the Supplementary Material [29].

To observe transitions from higher excited states, a different setup was employed for the in-beam  $\gamma$ -ray spectroscopy of  $^{252}\text{Fm}$ , which was produced via the MNT reactions induced by a 140.4-MeV  $^{18}\text{O}$  beam irradiating a  $104\text{-}\mu\text{g}/\text{cm}^2$   $^{249}\text{Cf}$  target, electrodeposited on a  $270\text{-}\mu\text{g}/\text{cm}^2$  nickel backing foil. In this experiment, depending on the number of neutrons and protons exchanged between colliding nuclei, several isotopes of californium, einsteinium and fermium were populated. The isotopes produced were identified on an event-by-event basis by detecting the complementary light ejectile, using an array of  $\Delta E$ -E silicon telescopes [27, 30, 31]. From the angle and energy of the light ejectiles, using binary-reaction kinematics, the excitation energy of the target-like isotopes was extracted with an uncertainty of approximately 300 keV [27]. An array of 4 coaxial Ge detectors was used to detect the  $\gamma$  rays. New  $K$  isomers in  $^{248}\text{Cf}$  observed in this experiment were presented in a recent publication [27].

The particle-identification spectrum obtained with the

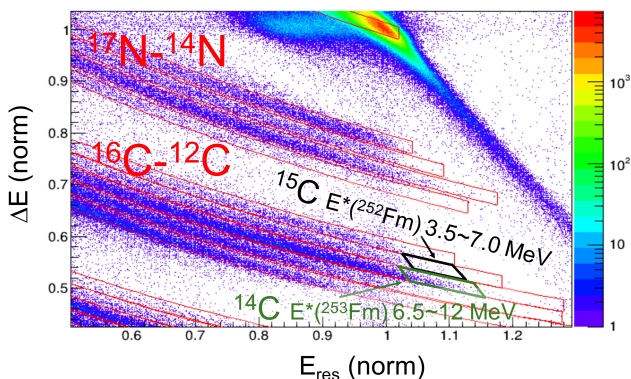


FIG. 3. Typical particle identification plot achieved with the Si array. The energy deposited in one of the  $\Delta E$  detectors is plotted versus the residual energy ( $E_{res}$ ) deposited on the E detector. The data are normalized to the energy of the  $^{18}\text{O}$  elastic-scattering peak for each combination of  $\Delta E$  and  $E$  strip. The  $^{15}\text{C}$  and  $^{14}\text{C}$  gates used to study  $^{252}\text{Fm}$  are also drawn. See text for details.

Si array is shown in Fig. 3. Ejectiles are well separated and identified.  $^{252}\text{Fm}$  events were selected using two different gates, shown in Fig. 3. The first gate selects events in which  $^{252}\text{Fm}$  was directly produced via the  $^{249}\text{Cf}(^{18}\text{O}, ^{15}\text{C})^{252}\text{Fm}$  ( $Q=-17.749(5)$  MeV [32]) reaction, with an excitation energy below the neutron separation energy,  $S_{1n}=7.214(15)$  MeV [32]. The alternative gate selects  $^{14}\text{C}$  events from the  $^{249}\text{Cf}(^{18}\text{O}, ^{14}\text{C})^{253}\text{Fm}$  reaction, which correspond to an excitation energy of  $^{253}\text{Fm}$  above  $S_{1n}$  (5.542(5) MeV) but below  $S_{2n}$  (12.756(14) MeV) [32], which allows for the evaporation of one neutron, leading to excited  $^{252}\text{Fm}$ .

TABLE I. Energy (keV), assigned transition and number of net counts for the different  $^{252}\text{Fm}$   $\gamma$ -ray transitions highlighted in Figs. 4(a), (b) and (c).

| Gate                          | Energy (keV) | Transition                 | Net counts |
|-------------------------------|--------------|----------------------------|------------|
| $^{15}\text{C}$               | 153(1)       | $(6^+) \rightarrow (4^+)$  | 9(5)       |
| $^{15}\text{C}$               | 206(1)       | $(8^+) \rightarrow (6^+)$  | 18(6)      |
| $^{15}\text{C}$               | 257(1)       | $(10^+) \rightarrow (8^+)$ | 10(5)      |
| $^{14}\text{C}$               | 153(1)       | $(6^+) \rightarrow (4^+)$  | 45(10)     |
| $^{14}\text{C}$               | 206(1)       | $(8^+) \rightarrow (6^+)$  | 11(7)      |
| $^{15}\text{C}+^{14}\text{C}$ | 153(1)       | $(6^+) \rightarrow (4^+)$  | 48(10)     |
| $^{15}\text{C}+^{14}\text{C}$ | 206(1)       | $(8^+) \rightarrow (6^+)$  | 30(15)     |
| $^{15}\text{C}+^{14}\text{C}$ | 257(1)       | $(10^+) \rightarrow (8^+)$ | 14(9)      |

The spectra obtained using the  $^{15}\text{C}$  and  $^{14}\text{C}$  gates are presented in Figs. 4 (a) and (b), and their sum in Fig. 4 (c). Due to the high selectivity of the  $^{15}\text{C}$  gate, the peaks appearing in Fig. 4 (a) correspond to transitions in  $^{252}\text{Fm}$ . Peaks with at least 5 counts are found at 101(1), 137(1), 153(1), 186(1), 196(1), 206(1), 230(1), 257(1), 289(1) and 486(1) keV. The amount of collected data does not suffice to place them in the level scheme using double gating. Some of these transitions are likely to belong to excited rotational bands, similar to those observed for example in  $^{250}\text{Fm}$  [33]. We note that the 153- and 206-keV transitions can also clearly be seen in the  $^{14}\text{C}$ -gated spectrum (vertical lines are drawn to guide the eye). From systematics, from the energies of the  $2^+ \rightarrow 0^+$ , and  $4^+ \rightarrow 2^+$   $\gamma$  rays determined in our  $\alpha$ - $\gamma$  experiment, and from the fact that they are among the strongest peaks in the  $^{15}\text{C}$ -gated spectrum, we believe that the 153-, 206-, and 257-keV lines belong to the ground-state rotational band, and we tentatively assign them to the  $6^+ \rightarrow 4^+$ ,  $8^+ \rightarrow 6^+$  and  $10^+ \rightarrow 8^+$  transitions. Their intensities are summarized in Table I. As in our previous study of  $^{248}\text{Cf}$  [27], the Fm K X-ray peaks (121 and 115 keV) in Fig. 4 have intensities comparable to the observed  $\gamma$  rays. The 42- and 98-keV transitions cannot be seen due to internal conversion. In MNT reactions, the average excitation energy  $E^*$  of the compound nucleus increases with the number of nucleons exchanged between beam and target [34]. This is why the number of events with  $E^*(^{252}\text{Fm}) < 10$  MeV is only  $\sim 5\%$  of all the events in which  $^{15}\text{C}$  was detected, compared to nearly 90% with  $E^*(^{248}\text{Cf}) < 10$  MeV for the  $^{19}\text{O}$  channel [27].

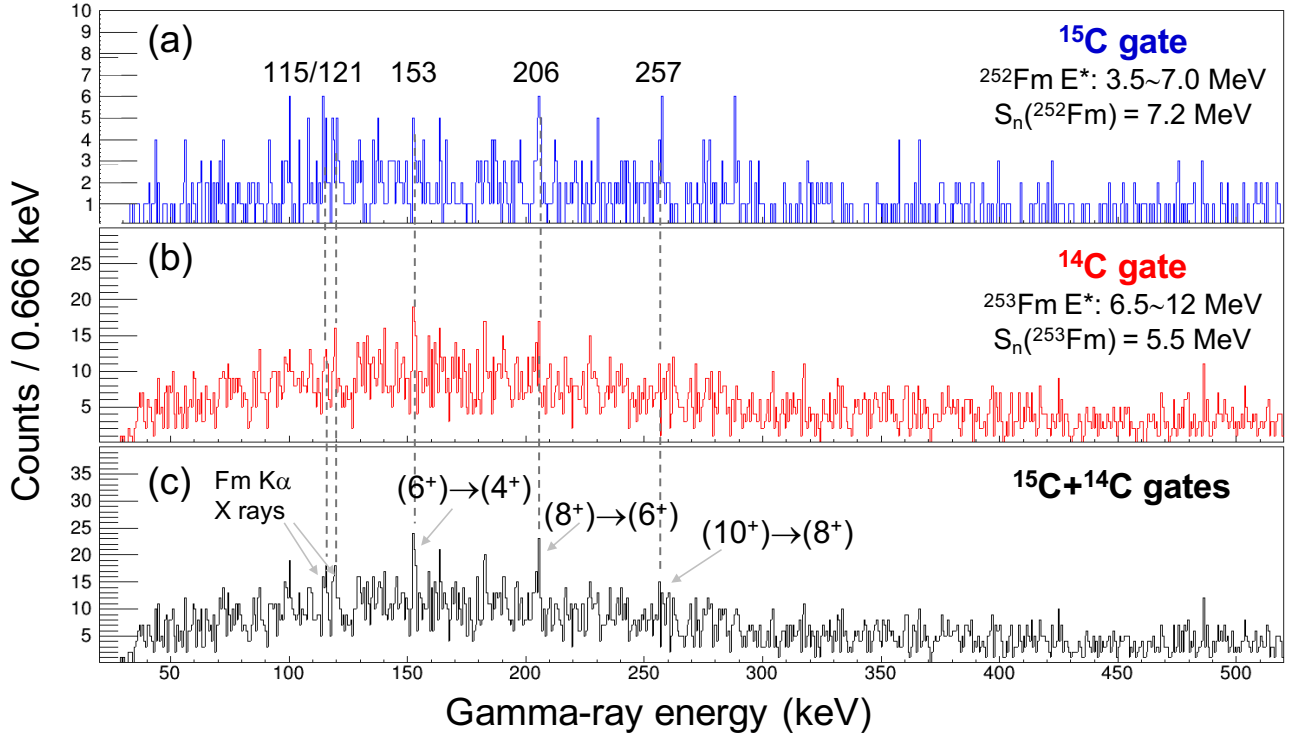


FIG. 4. Gated  $\gamma$ -ray spectra coincident with events associated in  $^{252}\text{Fm}$ . Spectrum (a) was produced by gating on  $^{15}\text{C}$  events, for a corresponding  $^{252}\text{Fm}$  excitation energy between 3.5 and 7.0 MeV, while spectrum (b) by gating on  $^{14}\text{C}$  events, for a  $^{253}\text{Fm}$  excitation energy between 6.5 and 12.0 MeV. Spectrum (c) is the sum of (a) and (b).

Despite the limited number of counts, the observation of the same  $\gamma$  rays using independent  $^{15}\text{C}$  and  $^{14}\text{C}$  gates strongly supports that they originate from  $^{252}\text{Fm}$ .

The results obtained in our experiments can be used to discuss the proton and neutron deformed shell gaps at  $Z=100$  and  $N=152$ . The  $E(2^+)$  value measured using  $\alpha$ - $\gamma$  spectroscopy is plotted in Fig. 5 together with the energies of the first excited  $2^+$  state of even-even isotopes from curium to rutherfordium [35]. The current measurement reveals that  $^{252}\text{Fm}$  has the lowest  $E(2^+)$  both among fermium isotopes and  $N=152$  isotones. The minimum among fermium isotopes is consistent with a  $N=152$  deformed shell gap. Furthermore, the comparison of  $E(2^+)$  values with other  $N=152$  isotones is supportive evidence for a  $Z=100$  deformed gap. This could not be established before due to the 1.3-keV uncertainty on our previous value obtained using  $\alpha$ -decay energies [25]. For Cf, No and Rf isotopes, from the known  $E(2^+)$  values alone the minimum cannot yet be established. The current lowest values are at  $N=150$  for Cf and  $N=152$  for No and Rf isotopes.

Using the five  $\gamma$ -ray transitions of  $^{252}\text{Fm}$ , we calculated the experimental kinematic moment of inertia (MoI)  $\mathcal{J}^{(1)} = \hbar^2(2I - 1)/E_\gamma(I)$ , where  $I$  is the spin of the initial state. In Fig. 6, the kinematic MoI of  $^{252}\text{Fm}$  is plotted together with those of even-even (a) fermium isotopes and (b)  $N=152$  isotones [35], and fitted using the Harris parameterization [36].  $^{252}\text{Fm}$  exhibits the largest

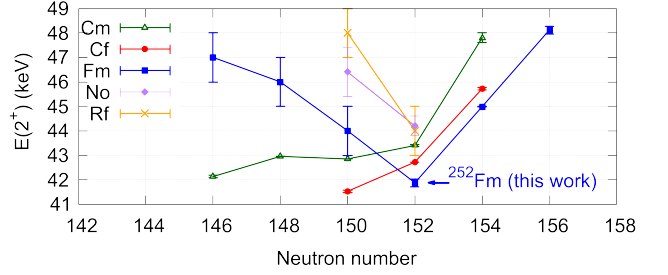


FIG. 5. Energy of the first excited  $2^+$  state in even-even isotopes of curium, californium, fermium, nobelium and rutherfordium, plotted as a function of neutron number. With the exception of  $^{252}\text{Fm}$ , experimental data are from [35].

kinematic MoI both among fermium isotopes and  $N=152$  isotones, supporting the deformed shell gaps at  $N=152$  and  $Z=100$ .

The second derivative of the binding energy as a function of proton (or neutron) number,  $\delta_{2p}$  ( $\delta_{2n}$ ) is another quantity that can reflect the presence of shell gaps (see Supplementary Material [29]). The peak of experimental  $\delta_{2p}$  is at  $Z=100$  for  $N=152, 154$  isotones, but at  $Z=98$  in the  $N=150$  isotones [29], and this is possibly linked to the fact that the MoI of  $^{248}\text{Cf}$  is  $\sim 1\%$  larger than that of  $^{252}\text{Fm}$  [37]. Note that the  $\delta_{2p}$  and  $\delta_{2n}$  values (frequently labelled in the literature as two-proton and two-neutron shell gaps) cannot be taken as the direct measure of these

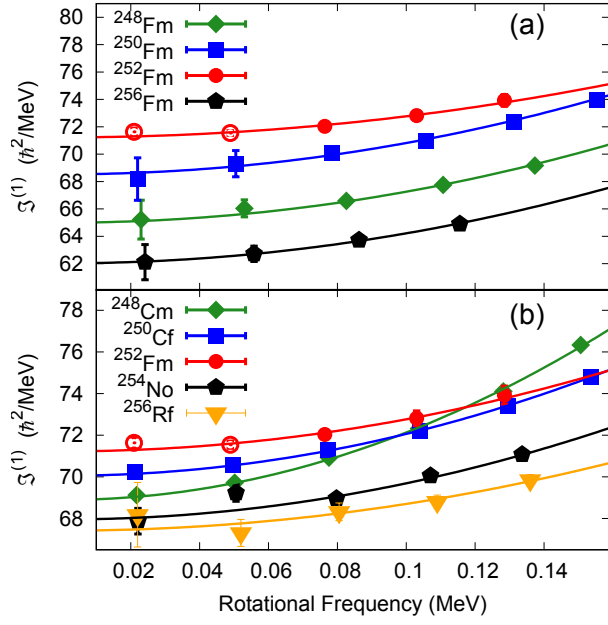


FIG. 6. Kinematic moment of inertia (symbols) of even-even (a) fermium isotopes and (b)  $N = 152$  isotones. The lines are quadratic fits from Harris parameterization. For  $^{252}\text{Fm}$ , open (full) symbols represent values obtained by  $\alpha$ - $\gamma$  spectroscopy (MNT reactions).

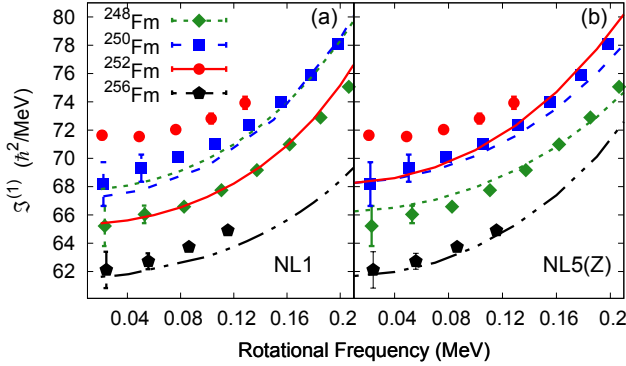


FIG. 7. Experimental (symbols) and calculated (lines, color-matched to symbols) moments of inertia of even-even fermium isotopes. See text for details.

gaps since they are affected not only by the size of the gaps but also by pairing and deformation changes (*cf.* Appendix of Ref. [38]). Thus, in the case of relatively modest  $\delta_{2p}$  and  $\delta_{2n}$  values (as in the case of actinides and superheavy nuclei) it is important to supplement the identification of deformed shell gaps by fingerprints (such as maxima of the moments of inertia) which test the shell structure in a different way.

Theoretical interpretation of the rotational properties of nuclei in this region has been carried out in the past in

different theoretical frameworks (see, for example, Refs. [21, 39]). Fig. 7(a) shows the results of cranked relativistic Hartree-Bogoliubov (CRHB+LN) calculations with the NL1 functional and approximate particle number projection by means of the Lipkin-Nogami (LN) method of Ref. [39]. The experimental kinematic MoI of  $^{252}\text{Fm}$  is substantially underestimated, while that of  $^{248}\text{Fm}$  is overestimated. On the other hand, the data in  $^{250,256}\text{Fm}$  are reproduced rather well. To improve the description of MoIs in this region, the following steps have been undertaken as compared with Ref. [39]: (i) proton and neutron pairing strengths were optimized individually and isospin dependence of neutron pairing suggested in Refs. [40, 41] has been taken into account and (ii) the new NL5(Z) functional globally fitted in Ref. [42] has been used in the calculations. Fig. 7(b) shows that MoI of the  $^{248,250,256}\text{Fm}$  nuclei are well described in this framework. There is also substantial improvement in the description of  $^{252}\text{Fm}$ , but the calculated MoI is still smaller than the experimental one. This is because even in the new NL5(Z) functional the size of the  $N = 152$  deformed shell gap is underestimated.

In conclusion, we observed for the first time the lowest five  $\gamma$ -ray transitions of the ground-state rotational band in  $^{252}\text{Fm}$ , and improved the precision on the energy of the first excited  $2^+$  state. The comparison of our results with the  $E(2^+)$  and MoIs of even-even fermium isotopes and  $N=152$  isotones serves as new evidence of both the  $N=152$  and the  $Z=100$  deformed shell gaps in  $^{252}\text{Fm}$ , supporting the image of a deformed doubly magic  $^{252}\text{Fm}$ . This work also provides useful benchmarks to theoretical models on heavy and superheavy elements, and indicates the need for their further improvement.

*Acknowledgments* — The authors express their gratitude to the JAEA Tandem accelerator crew for providing stable  $^{12}\text{C}$  and  $^{18}\text{O}$  beams and for their continued support. We thank H. Kudo and T. Mitsugashira for providing the mixed Cf material which was chemically purified at International Research Center for Nuclear Materials Science, Institute of Materials Research (IMR), Tohoku University. This work was partly supported by the REIMEI research grant of ASRC, JAEA. R.O. acknowledges partial support of JSPS KAKENHI Grant Number 25K07344. E.I. acknowledges the support by the International Joint Research Promotion Program of The University of Osaka, and JSPS KAKENHI Grant Number JP 17H02893. T.T.P. acknowledges support from JAEA as a special research student. A.N.A. acknowledges support from UK STFC. This material is also based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics under Award No. DESC0013037. We thank Y. Utsuno for fruitful discussions.

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