





Spontaneous fission instability of the neutron-deficient No and Rf isotopes: The new isotope ^{249}No

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In the heaviest elements, the instability of atomic nuclei against spontaneous fission leads to ever shorter nuclear half-lives. Upon falling below a timescale of 10^{-14} s, the border of existence of isotopes is crossed because this is the timescale on which the formation of atomic shells occurs. Analysis of the experimental data on the spontaneous fission half-lives of Rf isotopes in relation with their expected single-particle orbitals hint at a potentially abrupt decrease in half-lives of unknown neutron-deficient Rf isotopes with neutron numbers <149 , which suggests that the isotopic border is already almost reached. However, this conjecture, which cannot be explained within the current knowledge, was directly related to uncertainty in the experimental data on ^{253}Rf . We revisited the decay of ^{253}Rf and identified two fission activities, which are attributed to decays of the two different states with half-lives of $12.8^{+7.0}_{-3.4}$ ms and 44^{+17}_{-10} μs . In addition, hitherto unknown α decay in ^{253}Rf , which is followed by α decay of the new isotope ^{249}No with a half-life of 15^{+74}_{-7} ms, was observed. Based on our new data, no abrupt decreases in half-lives of the neutron-deficient No and Rf isotopes are expected, which is in line with theoretical predictions. Fission half-lives of the two different states in ^{253}Rf are benchmark cases for the theoretical description of the single-particle orbital influence on the fission process.

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Electrons and the nucleus, which they surround, constitute an atom. The electrons occupy quantum orbitals, i.e., shells, which form on the timescale of $\approx 10^{-14}$ s. Thus, an existence of chemical elements, i.e., of their isotopes, occurs only for nuclei with half-lives $\gtrsim 10^{-14}$ s [1]. Meanwhile, nuclear systems can exist for much shorter timescales ($\ll 10^{-14}$ s) and their radioactive decays carry information about the coexistence of the strong and the electromagnetic forces at their extreme. A prime region to study such effects is that of the heaviest nuclei whose instability is primarily defined by the fission process [2–6].

In a classical representation of the atomic nucleus as a structureless charged nuclear liquid drop [3,4] that consists of Z protons and N neutrons, heavy nuclei with $Z \gtrsim 100$ would be unstable against fission on a timescale of $\leq 10^{-14}$ s. However, nucleons occupy quantified discrete orbitals and effect fission in “collective” and in individual ways. Collective effects appear on the potential-energy surface of a nucleus all along its shape evolution towards the scission point at which the nucleus splits into two primary fragments. As a consequence, the potential-energy barrier of the nucleus is strongly effected by the shell structure (e.g., showing a double-humped shape [5,7]), which results in an increased stability against fission. An understanding of such nuclear properties led to the prediction of the existence of superheavy nuclei (SHN) with $Z \gtrsim 100$ and with half-lives much longer than 10^{-14} s [8,9].

Presently, the heaviest nuclei with proton or neutron numbers up to $Z = 118$ or $N = 177$ are known with half-lives being in the range of 10^{-6} – 10^5 s [2]. These show that the limit of the existence of superheavy atomic species has not yet been reached. Accordingly, the search for SHN with larger Z and N , and with half-lives exceeding the “isotopic” border is an active topic in both physics and chemistry [10–15].

An effect of a single nucleon occupying the last orbital in an odd- A nucleus on the fission process is evident in its significantly longer half-life when compared with those of the neighboring even-even ones in which all nucleons are paired and thus the total spin is zero [6]. In fact, this effect is quantitatively less understood than the above-mentioned collective one for which the theoretical description is better established. Theories qualitatively explain the effect of the single-particle (SP) configuration on fission [16,17]; however, their predictive power for the experimental half-lives of odd- A and odd-odd nuclei still needs to be improved.

The problem is complicated by the absence of a direct experimental measure of SP-effects in odd- A nuclei. Empirically, to express the effect of the SP configuration on fission, a hindrance factor (F_H) is used:

$$F_H(K^\pi, A) = \frac{T_{\text{sf}}(K^\pi, A)}{[T_{\text{sf}}(0^+, A-1)T_{\text{sf}}(0^+, A+1)]^{1/2}}, \quad (1)$$

where K and π are the projection of spin onto the symmetry axis and the parity of the odd- A nucleus, respectively. T_{sf} is the experimental partial spontaneous fission (SF) half-life of the ground states in the odd- A nuclei and in the even-even nuclei.

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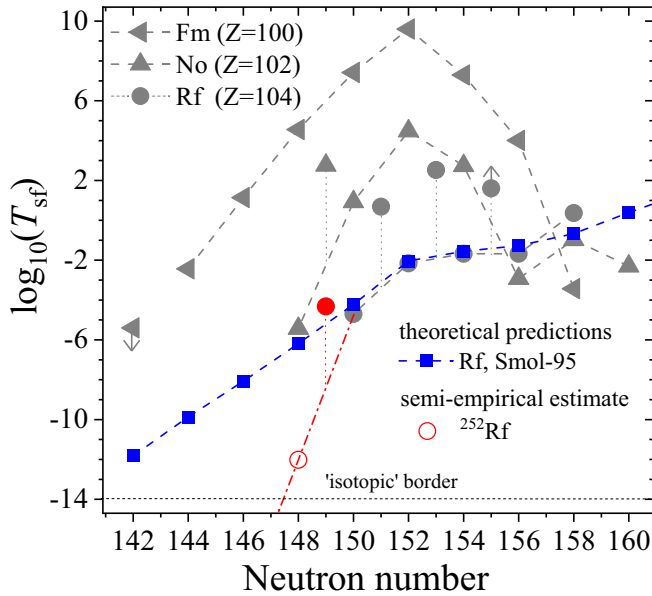


FIG. 1. Spontaneous fission half-lives of Fm, No, and Rf isotopes [6]. Theoretically predicted half-lives for Rf isotopes (Smol-95, [19]) are shown by rectangles. Dashed lines connect even-even isotopes. Vertical dotted lines show the fission hindrance factors for odd- A nuclei. The dash-dotted line crossing to the isotopic border shows an abruptly falling tendency in half-lives of neutron-deficient Rf isotopes based on an empirically estimated half-life of 1 ps for ^{252}Rf . This estimate is based on a $48 \mu\text{s}$ half-life of ^{253}Rf [6] and an assumed $F_H = 10^4$. See text for details.

The F_H s deduced from known T_{sf} are in the range of 10^2 – 10^{10} and show a dependence on K^π , Z , and A [6]. Currently, the exact empirical relation between the F_H and K^π is uncertain, but the same SP levels in the neighboring isotopes and isotones show F_H values agreeing within a factor of 10^2 . It should be noted that F_H calculated by Eq. (1) strongly depends on the properties of the neighboring even-even nuclei whereas T_{sf} are strictly due to the fission path of the respective nucleus on its potential-energy surface. Such an artificial effect on F_H can be seen in the case of ^{257}Fm , where the largest $F_H = 6.7 \times 10^9$ seemingly overestimates the effect of the corresponding SP configuration because of the short T_{sf} of ^{258}Fm caused by the change in the fission path compared with ^{256}Fm [6].

F_H , in fact, is not very valuable for theory whereas the direct experimental T_{sf} is a credible quantity. Experimental T_{sf} values of Fm, No, and Rf isotopes are shown in Fig. 1. The longest T_{sf} in the even-even nuclei are observed at $N = 152$, which is thought to originate from an enhanced shell gap in these deformed nuclei. However, such a feature is barely visible in Rf isotopes. This has been explained by the disappearance of the outer fission barrier leading to a single-humped barrier in Rf [8,18]. Since only the single-humped barrier exists in neutron-deficient ($N < 152$) Rf isotopes, their decreasing trend in T_{sf} is predicted to occur without sudden changes in the slope in the $N < 152$ isotopic sequence (see Fig. 1) [19–21], which reflects a smoothly decreasing trend of calculated fission barrier heights [19,22,23].

Meanwhile, the above-expected trend of T_{sf} can be examined in the hitherto unknown ^{252}Rf ($N = 148$) for which T_{sf} can be estimated by taking the known T_{sf} for ^{254}Rf ($23 \mu\text{s}$ [24]) and ^{253}Rf ($48 \mu\text{s}$ [25]) and using Eq. (1). As mentioned above, the main uncertainty in such an estimation is the F_H , which relates to a particular SP configuration. Theoretically the SP configuration of the ground state of ^{253}Rf (^{253g}Rf) is predicted to be either $9/2^-$ [734] [26] or $7/2^+$ [624] [27,28]. The latter, which is the ground state in the lighter $N = 149$ isotones [29], results in $F_H = 10^5$ and 9.8×10^4 in ^{251}No and ^{245}Cm , respectively. The $9/2^-$ [734] state features $F_H = 3.5 \times 10^6$ and 8.2×10^3 in ^{249}Cf and ^{255}Rf , respectively. Accordingly, if the $48 \mu\text{s}$ fission belongs to ^{253g}Rf , which has such a high- K SP configuration, then one can roughly estimate its F_H to be $\approx 10^4$. A half-life for ^{252}Rf is then estimated as $T_{\text{sf}} = 1 \text{ ps}$, i.e., 10^{-12} s . This is a very small value that leads to an abrupt fall in T_{sf} compared with theory [19–21] and also compared with the systematic trend established by ^{254}Rf and ^{256}Rf (see Fig. 1). Moreover, these results reveal that ^{250}Rf would already be beyond the “isotopic” border of the element Rf, thus excluding an experimental expansion of the neutron-deficient region of Rf isotopes towards the proton drip line. This signature for an abrupt fall still remains if the above-mentioned experimental F_H uncertainty of 10^2 for the assumed value of 10^4 is used. At the same time, a sudden fall cannot be explained by a change in the single-humped barrier shape. Meanwhile, Fig. 1 shows a similarly rapid decrease in T_{sf} also in the lighter $N = 148$ isotone ^{250}No compared with ^{252}No , which, however, can be interpreted as a sign for the transition from the double-humped barrier to the single-humped barrier [21].

To shed light on these problems, we have investigated the decay of ^{253}Rf at the gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) at GSI, Darmstadt, Germany.

In this Letter, we report the identification of fissions from two different states of ^{253}Rf . Measured partial fission half-lives of both these states represent a new benchmark case for the theoretical description of the single-particle effect on the fission process. We also report a hitherto unknown α decay of ^{253}Rf and the discovery of the new isotope ^{249}No .

A pulsed (5-ms-long pulses, 50 s^{-1} repetition rate) $^{50}\text{Ti}^{12+}$ beam was accelerated by the Universal Linear Accelerator UNILAC. The beam energy in the middle of the 0.6-mg/cm^2 -thick ^{204}PbS target (isotopic composition: 99.94% ^{204}Pb ; 0.04% ^{206}Pb ; 0.01% ^{207}Pb ; 0.01% ^{208}Pb) was 234.3 MeV, which corresponds to an excitation energy of 18.5 MeV of the compound nucleus $^{254}\text{Rf}^*$, at which a maximum cross section of $\approx 0.3 \text{ nb}$ was expected for the $1n$ evaporation channel [30]. Four targets were mounted on a wheel which rotates synchronously with the beam macro structure [31].

For separation and collection of evaporation residues (ERs) in the focal-plane detector, TASCA was operated with helium gas at 0.8 mbar pressure and with a magnetic rigidity ($B\rho$) of 2.14 Tm [32,33]. The efficiency of TASCA to guide ERs to the implantation detector was estimated to be 60% [32,34,35]. We used a double-sided silicon detector with 144 vertical (X) and 48 horizontal (Y) strips on the front and back sides, respectively. Energy resolutions (FWHM) of both the X and Y strips were about 40 keV for the 5.8 MeV α particles from the

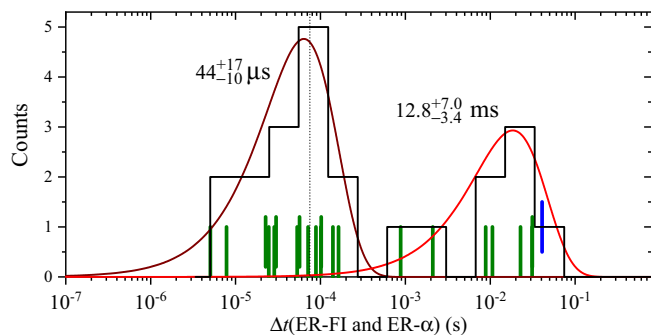


FIG. 2. Shown are the individual correlation times of fission (an offset value of 0.2 counts is applied to separate the close in time events) and α (an offset value of 0.5 counts is applied to highlight the one α count) events attributed to ^{253}Rf . The time distribution of all events is also shown. Radioactive decay curves calculated according to Ref. [39] are shown. For details see text.

^{244}Cm external source. Energy calibrations were made using α decays of nuclei produced in the $^{48}\text{Ca} + ^{176}\text{Yb}$ reaction. Signals from the X and Y strips were pre-amplified with different gains to provide two energy branches up to about 20 and 200 MeV. All pre-amplifier signals were digitized by 100-MHz-sampling FEBEX4 analog-to-digital converters [36,37]. In 70% of the data, the shape of each signal was stored in an 80- μs -long trace and in 30% with a 60- μs -long trace [38]. The average beam intensity on the targets was $\approx 4 \times 10^{12}$ particles per second, which resulted in an average counting rate of about 60 Hz.

We identified 12 spatially correlated events, each consisting of ER and fission (FI) with correlation times (Δt) exceeding $\approx 80 \mu\text{s}$. In addition, nine ER traces containing the second signal corresponding to the FI were observed, thus, showing the ER–FI with $\Delta t = 0.1\text{--}80 \mu\text{s}$. Individual Δt values of these 21 ER–FI events are shown in Fig. 2 together with the deduced time distribution. The events are best separated into two groups with different half-lives [39]. Fourteen events with shorter Δt result in a half-life of $44^{+17}_{-10} \mu\text{s}$, which agrees with $48 \mu\text{s}$ reported for ^{253}Rf in Ref. [25]. Meanwhile, the seven FI events with longer Δt values result in an average of $\Delta t = 15^{+10}_{-5} \text{ms}$. This is similar to the $T_{\text{sf}} = 6.2 \text{ms}$ fission of ^{256}Rf [6], which could be produced in the $^{50}\text{Ti} + ^{207}\text{Pb}$ reaction. We expected to observe ≈ 0.02 events of ^{256}Rf based on the cross section of $\approx 3 \text{nb}$ [30,38] and the 0.01% of ^{207}Pb in the target. Therefore, these seven FI events can also be attributed to ^{253}Rf but to a different state than the one with $44^{+17}_{-10} \mu\text{s}$. It should be noted that, in the original ^{253}Rf -discovery experiment [25], a similar 11 ms fission activity was also observed; however, it was not excluded to originate from ^{256}Rf because of an uncertainty in the amount of impurity of ^{207}Pb .

In addition, we observed six events in the energy range of 8–10 MeV detected during beam-off periods. Three of them were found to form the α -decay chain shown in Fig. 3. The properties of the last α are in fine agreement with the known α decay of ^{245}Fm [40]. The Q -values of 9.31(3) and 9.21(3) MeV corresponding to the first and second α , respec-

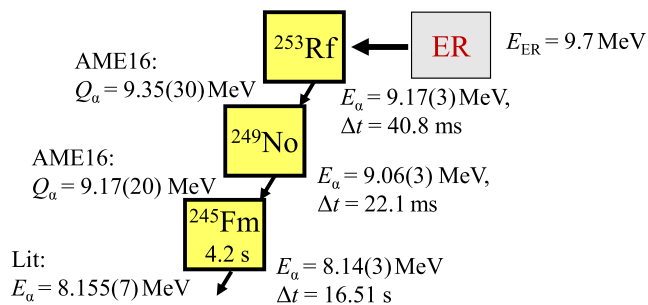


FIG. 3. Measured α -decay chain attributed to ^{253}Rf . Experimental energies of implantation signals (ER), α particles, and correlation times Δt are given together with predicted Q_α values (AME16: from Ref. [41]). The known energy for ^{245}Fm (Lit:) is taken from Ref. [40]. Thick-frame boxes mark beam-off events.

tively, are in agreement with the evaluated ones for ^{253}Rf and ^{249}No (taken from Ref. [41]). Moreover, $\Delta t = 40.8 \text{ms}$ for the first α is similar to the above-mentioned $\Delta t = 15^{+10}_{-5} \text{ms}$ of the seven FI events. We therefore attribute the α -decay chain and the seven FI events to originate from the same state with $T_{1/2} = 12.8^{+7.0}_{-3.4} \text{ms}$ in ^{253}Rf .

The total cross section calculated from all 22 events attributed to ^{253}Rf was $0.27^{+0.07}_{-0.06} \text{nb}$, which agrees with $0.30^{+0.08}_{-0.07} \text{nb}$ from Ref. [25].

Finally, the inspection of all traces of ER, α , and FI events revealed one case of an ER–electron–FI sequence, which may point to the existence of another isomeric state in ^{253}Rf (see Supplemental Material [42] and below).

For the fission and α -decaying state in ^{253}Rf a partial α -decay half-life (T_α) of $\approx 100 \text{ms}$ was deduced with its $\approx 12.5\%$ α branching. The state populated in the new isotope ^{249}No decays by α -particle emission with an energy of 9.06(3) MeV and a half-life of 15^{+74}_{-7}ms . The ratios between the measured T_α and the one calculated by the semi-empirical expressions given in Refs. [43,44] for T_α as a function of the measured Q_α are 1.5 (102 ms/ 66 ms) and 0.5 (15 ms/ 28 ms) for ^{253}Rf and ^{249}No , respectively. These values indicate favored α transitions. Such cascades of favored α transitions occur in ^{251}No via ^{247}Fm to ^{243}Cf ([45], see Fig. 4) from ground and isomeric states with $7/2^+$ [624] and $1/2^+$ [631] configurations, respectively. Accordingly, the presently identified states in ^{253}Rf are assigned to have the same SP levels as in the lighter ^{251}No isotone, and the proposed tentative decay schemes are shown in Fig. 4. We attribute $7/2^+$ [624] and $1/2^+$ [631] for the 12.8 ms and 44 μs states, respectively. These assignments are based on theoretical predictions [16,17,46], which show that fission from a low- K state occurs faster than from a high- K state in an odd- A nucleus.

Comparison of the measured Q values with the evaluated Q values suggest that the observed α decays to be ground-state-to-ground-state transitions or transitions involving low-lying excited states. The ground-state SP levels in the lighter $N = 149$ isotones are $7/2^+$ [624]; thus, this SP level could also be the ground state of ^{253}Rf . However, the $1/2^+$ [631] orbital, which is located at excitation energies of 384 and 356 keV in ^{243}Pu and ^{245}Cm , respectively, and was not found below

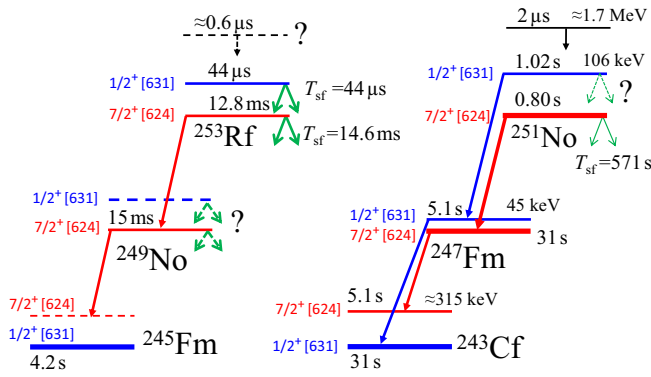


FIG. 4. Suggested decay schemes for the presently observed Rf and No isotopes are shown together with those for the lighter No and Fm isotones (only a part of known levels together with their favored α transitions are shown) [45]. All single-particle configurations are tentatively assigned. Decays from nuclear levels marked by dashed lines were not observed or were not unambiguously identified. Half-lives of levels are given together with energies for excited states where known. The unknown fission branches and electromagnetic transition are marked by question marks. For details see text and Supplemental Material [42].

≈ 700 keV in ^{247}Cf and ^{249}Fm [29], suddenly appears as a low-lying isomeric state in ^{251}No . Thus, it appears possible that $1/2^+[631]$ may become the ground state in ^{253}Rf . Therefore, with the present data we cannot make unambiguous assignments on the ordering of these two levels and on the ground state. Nevertheless, based on the ground states of all known $N = 149$ even- Z isotones down to ^{243}Pu [29], we consider a decay scheme in which the $7/2^+[624]$ orbital remains the ground state as more likely. The possible existence of a second high-lying isomeric state in ^{253}Rf similar to the one in ^{251}No shown in Fig. 4 is discussed in Supplemental Material [42].

In the case of ^{249}No , the systematics of the lighter $N = 147$ isotones [47] suggest the $7/2^+[624]$ orbital to be ^{249g}No .

Now, by using $T_{\text{sf}} = 14.6$ ms for the $7/2^+[624]$ state we evaluate $T_{\text{sf}}(^{252}\text{Rf})$ with the above-assumed $F_H = 10^4$. This results in $T_{\text{sf}}(^{252}\text{Rf}) = 93$ ns, which is in line with the theoretical value of 650 ns [19], which excludes an abrupt fall in T_{sf} of neutron-deficient Rf isotopes (see Fig. 1). Nevertheless, this kind of analysis of T_{sf} of even-even and odd- A nuclei by involving the empirically assumed F_H seems still to be informative once a SP level is known for the latter ones. For instance, by taking $T_{\text{sf}} = 15$ ms as a lower limit for the α -decaying $7/2^+[624]$ state in ^{249}No , $T_{\text{sf}}(^{250}\text{No}) = 3.8(3) \mu\text{s}$ [48] and $F_H = 10^4$, one gets $T_{\text{sf}}(^{248}\text{No}) > 0.5 \mu\text{s}$. This points to a smooth decrease in T_{sf} of No isotopes with $N < 148$,

which is in line with the attributed single-humped barrier [21] and similar to that in Rf. In fact, as a consequence of the absence of an abrupt fall in T_{sf} for even-even nuclei and of the SP-effect on fission, the α -decay branch becomes observable in ^{253}Rf and also in ^{249}No . We note that this region of No and Rf isotopes, which is known to be dominated by SF has not been expanded over the last two decades.

Finally, by taking the ratio between the half-lives of ^{253m}Rf and ^{253g}Rf , we get a difference in F_H s, $\Delta(F_H)$, due to the $7/2^+[624]$ and the $1/2^+[631]$ orbitals, which results in $\Delta K = 3$. Our extracted $\Delta(F_H) \approx 332$ is “free” of any assumptions because they are, e.g., made in Eq. (1) and reveal a difference in retardations of fission processes due to the SP configurations. This finding is greatly valuable for the theoretical description of the influence of the SP configuration on fission because fissions occur from the same quantum system, i.e., ^{253}Rf around similarly low excitation energies. Experimentally, it opens up a new perspective to investigate the effects of the SP configuration on fission by finding similarly decaying low-lying states in other odd- A nuclei, where different SP configurations and ΔK are involved (e.g., ^{243}Fm [40,49], ^{245}Md [50], and ^{247}Md [6]). In such a way one can get more insights into still-scarcely-known cases of fission of odd-odd nuclei and of high- K isomeric states in even-even nuclei [6,24,46,48,50,51]. Potential first candidates are ^{249}No and ^{251}No (see Fig. 4). Based on the same SP configuration, one could estimate $T_{\text{sf}}(^{251m}\text{No}) \approx 1.7$ s for the $1/2^+[631]$ state based on its $T_{\text{sf}}(^{251g}\text{No}) = 571$ s of the $7/2^+[624]$ state and the above $\Delta(F_H) \approx 332$. This estimate does not exclude direct fission with a branching of up to 60% from the α -decaying ^{251m}No , which has a 1.02 s half-life [42].

In conclusion, we identified two fission activities in ^{253}Rf with half-lives of $12.8_{-3.4}^{+7.0}$ ms and $44_{-10}^{+17} \mu\text{s}$. A hitherto unknown α -decay chain from ^{253}Rf and the new isotope ^{249}No , which has a half-life of 15_{-7}^{+74} ms, were discovered. No abrupt falls in half-lives of the more neutron-deficient No and Rf isotopes are expected, which promises further expansion of this region towards the proton drip line. The difference in the fission half-lives of two states in ^{253}Rf is a new experimental approach to examine fission hindrance. This finding together with other similar cases which should be explored in the future will provide benchmark cases for the theoretical description of the influence of single-particle orbitals on the fission process.

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