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Search for Electron-Capture Delayed Fission in the New Isotope ^{244}Md

J. Khuyagbaatar,^{1,2,*} H. M. Albers,² M. Block^{1b},^{1,2,3} H. Brand,² R. A. Cantemir,² A. Di Nitto^{1b},^{3,†}
 Ch. E. Düllmann^{1c},^{1,2,3} M. Götz,^{1,2,3} S. Götz,^{1,2,3} F. P. Heßberger,^{1,2} E. Jäger,² B. Kindler,² J. V. Kratz,³
 J. Krier,² N. Kurz,² B. Lommel^{1b},² L. Lens,^{2,3} A. Mistry,² B. Schausten,² J. Uusitalo,⁴ and A. Yakushev^{1b},²

¹Helmholtz Institute Mainz, 55099 Mainz, Germany

²GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany

³Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany

⁴University of Jyväskylä, 40351 Jyväskylä, Finland



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The electron-capture decay followed by a prompt fission process was searched for in the hitherto unknown most neutron-deficient Md isotope with mass number 244. Alpha decay with α -particle energies of 8.73–8.86 MeV and with a half-life of $0.30^{+0.19}_{-0.09}$ s was assigned to ^{244}Md . No fission event with a similar half-life potentially originating from spontaneous fissioning of the short-lived electron-capture decay daughter ^{244}Fm was observed, which results in an upper limit of 0.14 for the electron-capture branching of ^{244}Md . Two groups of fission events with half-lives of $0.9^{+0.6}_{-0.3}$ ms and 5^{+3}_{-2} ms were observed. The $0.9^{+0.6}_{-0.3}$ ms activity was assigned to originate from the decay of ^{245}Md . The origin of eight fission events resulting in a half-life of 5^{+3}_{-2} ms could not be unambiguously identified within the present data while the possible explanation has to invoke previously unseen physics cases.

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Atomic nuclei with extreme numbers of protons and neutrons far away from the beta-stability line are one of the main objects to study the interplay of two fundamental interactions of nature, namely the repulsive electromagnetic and the attractive nuclear forces [1,2]. However, such nuclei do not exist in nature, thus they have to be artificially produced in nuclear reactions [1,3]. Of special interest are the heavy nuclei. According to theoretical models that describe an atomic nucleus as a charged liquid drop, they are unstable against fission due to the dominance of the “disruptive” Coulomb force over the attractive nuclear force (see recent review articles [2–4] and references therein). However, thanks to nuclear shell structure, fission in the heaviest nuclei is retarded [2–4]. Once the fission probability is reduced, the stability of nuclei is determined by other radioactive decay modes such as α -particle emission and β decay (β^\pm and electron capture, EC).

The heaviest known nuclei with proton numbers $Z > 105$, thus, have been identified mostly by their α -decay chains, while their instability against β decay was so far investigated only scarcely [3,5–7]. Despite some speculation [8], such a decay path has not been identified conclusively yet in nuclei with $Z > 105$. Recently, it has been predicted that β decay of odd-odd superheavy nuclei (SHN) may be followed by fission of an excited even-even daughter SHN with a much larger probability than in the known cases for heavy nuclei up to Md ($Z = 101$). This so-called β /EC-delayed fission (β DF/ECDF) was suggested to become one of the main decay modes of β -decaying SHN with probabilities that strongly depend on both height

and shape of the fission barrier [9]. Accordingly, the presence of β DF in SHN can be considered as a benchmark for examining the structure of the fission-barrier in SHN.

The known ECDF cases for nuclei with largest Z are ^{246m}Md [10] and ^{250}Md [11]. Their relatively high ECDF probability has been explained in Ref. [9] as being due to a negligible outer barrier influence. In this context, one of the next steps for examining the predictions given in Ref. [9] is the study of hitherto unknown ^{244}Md , for which an ECDF probability of about 20% has been predicted, if EC decay takes place.

In the present work, we report results from the $^{50}\text{Ti} + ^{197}\text{Au}$ fusion-evaporation reaction in which the new isotope ^{244}Md was identified and decay properties of ^{245}Md , ^{241}Es , and ^{240}Es were confirmed.

The experiment was performed at the gas-filled trans-actinide separator and chemistry apparatus (TASCA) at GSI, Darmstadt [12]. A pulsed (4–5 ms long pulses, 5 or 50/s repetition rate) $^{50}\text{Ti}^{12+}$ beam with an average pulse intensity of $\approx 10^{11}$ ions per pulse was accelerated by the universal linear accelerator UNILAC and bombarded a rotating ^{197}Au target [13] with an average thickness of about 0.63 mg/cm^2 . Two different beam energies resulting in center-of-target energies of 239.8 and 231.5 MeV were used [14], leading to compound nucleus excitation energies of $E^* = 32.7$ and 26.2 MeV at which the productions of ^{244}Md and ^{245}Md , respectively, are expected [15].

TASCA was filled with helium gas at 0.8 mbar pressure and the magnetic rigidity was set at 2.05 Tm [16,17]. A double-sided silicon strip detector (DSSD) comprising 144

vertical (X) and 48 horizontal (Y) strips on the front and back sides, respectively, was used to detect implanting ERs and their subsequent decays. The efficiency for implanting ERs into the DSSD was estimated to be 60% [16,18]. Energy calibrations were performed using α decays of nuclei produced in the $^{48}\text{Ca} + ^{176}\text{Yb}$ reaction and energy resolutions (FWHM) of both X and Y strips of the DSSD were about 40 keV for 5.8 MeV- α particles.

In the present experiment, all signals from the preamplifiers were processed with fast digital electronics, which replaced the previous combined analog and digital electronics [19–24]. Signals from the X and Y strips were amplified with different gains to provide two energy branches up to about 20 and 200 MeV. The signals were digitized by 100 MHz-sampling FEBEX4 14-bit ADCs developed by the GSI experiment electronics department [19,25]. The shape of each signal was stored in a 30 μs -long trace. A multiwire proportional counter (MWPC) was mounted in front of the implantation detector. Any event with an energy in the range of $E = 6$ –20 MeV having a coincident MWPC signal was considered as implantation signal of an ER.

Spatial- (in the same X/Y strips) and time- (<30 s) correlated ER-like and α -like events were searched and the results are shown in Fig. 1(a). Three and ten events with $E = 8.6$ –8.9 MeV were found at $E^* = 26.2$ and 32.7 MeV, respectively.

The energies of the three events observed at $E^* = 26.2$ MeV, at which the $2n$ channel cross section of the $^{50}\text{Ti} + ^{197}\text{Au}$ reaction is expected to be larger than at

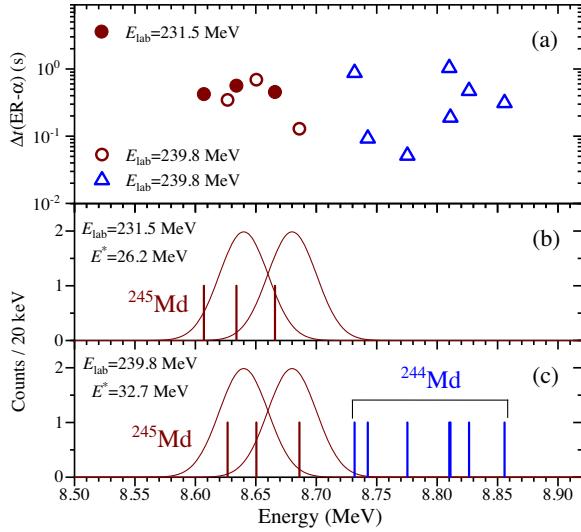


FIG. 1. Correlation times (Δt) of the α events to the preceding implantation (ER) signals shown as function of their energy (a). The events assigned to decays of ^{244}Md and ^{245}Md are marked by triangles and dots, respectively. The events observed at $E^* = 26.2$ and 32.7 MeV are marked by full and open symbols and their energies are shown in (b) and in (c), respectively. Gaussian peaks correspond to expected α -decay energies of the known ^{245}Md [26]. See text for details.

$E^* = 32.7$ MeV, are in agreement with the known energies of 8.64(2) and 8.68(2) MeV for ^{245}Md [26]. In the event-by-event analysis, we found a total of seven α -decay chains ending with α decays of ^{241}Es with $E = 8.12(2)$ MeV [27]. One such chain is shown in Fig. 2. Detailed data on all decay chains detected in the present work are given in Ref. [27].

The energies of the ten events observed at $E^* = 32.7$ MeV are shown in Fig. 1(c). They reveal three events within the energy range of ^{245}Md and the appearance of seven α events with higher energies.

Time distributions of all events assigned to ^{245}Md [27] and to its daughter ^{241}Es are shown in Figs. 3(a) and 3(b), respectively. The half-lives of $T_{1/2} = 0.33^{+0.15}_{-0.08}$ s and $4.3^{+2.4}_{-1.2}$ s, which are extracted according to Ref. [31], confirm the values of $0.35^{+0.23}_{-0.16}$ s and 8^{+6}_{-4} s for ^{245}Md and ^{241}Es , respectively, as reported from an experiment using the $^{40}\text{Ar} + ^{209}\text{Bi}$ reaction [26]. Cross sections of $\sigma = 59^{+31}_{-23}$ pb and 28^{+26}_{-17} pb for ^{245}Md were deduced for the present $^{50}\text{Ti} + ^{197}\text{Au}$ reaction at $E^* = 26.2$ and 32.7 MeV, respectively.

Seven further α events with similar Δt but with $E = 8.73$ –8.86 MeV were attributed to originate from ^{244}Md produced in the $3n$ channel of the fusion-evaporation reaction of $^{50}\text{Ti} + ^{197}\text{Au}$. From the time distribution of these events $T_{1/2} = 0.30^{+0.19}_{-0.09}$ s was deduced as shown in Fig. 3(c). To confirm the identification, α -decay chains stemming from ^{244}Md , which includes the known ^{240}Es with an α -decay branch of 70%, were searched (see Ref. [27]). One of these chains is shown in Fig. 2 and

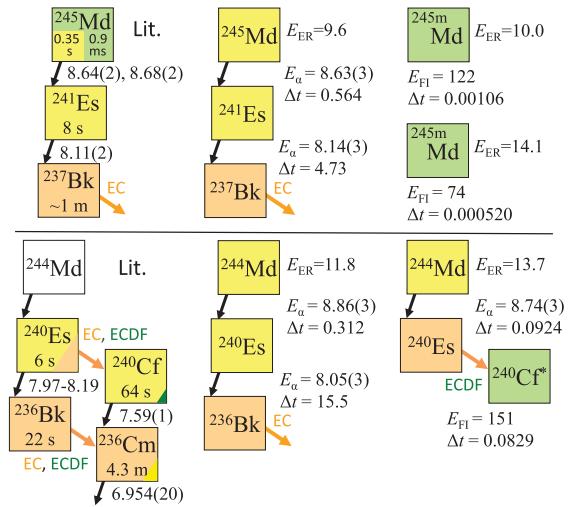


FIG. 2. The known properties (α -decay energies are given in MeV) of the nuclei that could potentially be produced in the radioactive decay (α and electron capture) of ^{245}Md (Lit. [26]) and ^{244}Md (Lit. [28–30]) are shown on the left side. Examples of observed decay chains assigned to ^{245}Md and ^{244}Md at $E^* = 26.2$ and 32.7 MeV, respectively, are shown. Energies of ER, α and fission events are given in MeV and correlation times in s.

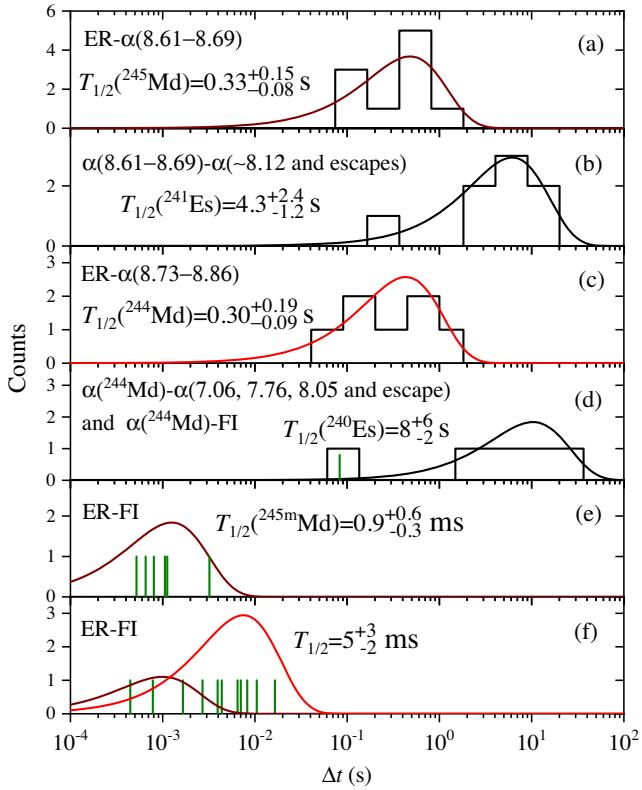


FIG. 3. Correlation time (Δt) distributions of the α events assigned to decays of ^{245}Md (a), ^{241}Es (b), ^{244}Md (c), and ^{240}Es (including the one fission event, for which its Δt is shown) (d). Correlation sequences, energy ranges of the α events, extracted half-lives are given in each plot. Correlation times of fission events observed at $E^* = 26.2$ and 32.7 MeV are shown in (e) and in (f), respectively. Curves correspond to the deduced half-lives according to Ref. [31]. See text for details.

its properties agree with the expected decay scheme of ^{244}Md [28–30]. Three more ER- α chains were followed by α events with $E = 7.06, 7.76$, and 0.67 MeV after $2.99, 27.4$, and 5.6 s, respectively [27]. The latter one was assigned to an α particle escaping the DSSD in backward direction, thus only leaving a partial energy signal. The secondary events are attributed to ^{240}Es , which emits α particles within the wide energy range of $7.97\text{--}8.19$ MeV [27,28].

We note that energies of 7.06 and 7.76 MeV are significantly lower than the known energy range of $7.97\text{--}8.19$ MeV for the α particles from ^{240}Es [27,28], for which we cannot give a definite answer based on the present data. It may happen that their energies were not fully detected, or they could originate from so far unknown α transitions in ^{240}Es or ^{240}Cf [27]. Their assignment to ^{240}Es is further supported by the observation of one additional 6.89 MeV beam-off α -like event detected 106 s after the 7.76 MeV- α , which can be interpreted to originate from the decay of ^{236}Cm (see Fig. 2 and [27]). In fact, in one of the α -decay chains assigned to ^{245}Md , the

energy of the α member belonging to ^{241}Es was 7.45 MeV, which could be a similar case as for the above events.

Another α event of 8.74 MeV assigned to ^{244}Md was followed by a fission (FI) event detected 83 ms later [27]. This event is shown in Fig. 2 and is attributed to originate from ECDF of ^{240}Es , for which a branching of $\approx 5\%$ is known [27,28]. Finally, from the Δt of the four α events and one FI event following the α decay of ^{244}Md , $T_{1/2} = 8^{+6}_{-2}$ s (see Fig. 3) was deduced, which is similar to the $T_{1/2} = 6(2)$ s of ^{240}Es and thus agrees with and confirms the findings in Ref. [28]. These results confirm the assignment of ^{244}Md .

In addition, 6 and 11 FI events correlated with ER-like signals with energies of $8\text{--}14$ MeV within a short time were observed at $E^* = 26.2$ and 32.7 MeV, respectively [27]. The time distribution of the six events at $E^* = 26.2$ MeV shown in Fig. 3(e) results in $T_{1/2} = 0.9^{+0.6}_{-0.3}$ ms, which is again in agreement with the findings in Ref. [26]. Based on the known decay properties of ^{245}Md [4] and ^{247}Md [4,10], these events were attributed to originate from an isomeric $1/2^-$ [521] state in ^{245}Md . Two of six ER-FI events are shown in Fig. 2. These six fission events result in $\sigma = 41^{+24}_{-17}$ pb.

The time distribution of the eleven events at $E^* = 32.7$ MeV indicates the presence of an activity with longer $T_{1/2}$ than ^{245m}Md . However, correlation times of these events are too short to be compatible with $T_{1/2}$ of the α -decaying state of ^{244}Md with $T_{1/2} \approx 0.30$ s. The non-observation of fission events with a half-life similar to ^{244}Md reveals that we did not observe the EC decay, which would be source for the ECDF process in ^{244}Md . An upper limit of 0.14 can be deduced for the EC-decay branching in ^{244}Md based on the observed seven α -decay chains.

Among the 11 FI events, we expect to detect up to three FI events with 0.9 ms half-life based on the present and known data [26] for $^{245,245m}\text{Md}$. Accordingly, the three FI events with shortest Δt corresponding to $T_{1/2} = 0.7^{+1.0}_{-0.3}$ ms and produced with $\sigma = 22^{+20}_{-13}$ pb are attributed to originate from ^{245m}Md [see Fig. 3(f)]. The remaining eight FI events with $T_{1/2} = 5^{+3}_{-2}$ ms, as shown in Fig. 3(f), have a different origin. We exclude an origin in actinide fission isomers because their production is negligibly low in transfer channels of $^{50}\text{Ti} + ^{197}\text{Au}$. The used Au target material was of high chemical purity, which excludes fusion or transfer reactions with impurities in the target as source.

Let us consider their potential association with ^{244}Fm (i) or ^{244}Md (ii).

(i) The isotope ^{244}Fm , which decays by spontaneous fission with $T_{1/2} = 3.12$ ms [30], is in fact, the only isotope, which would well explain the origin of the eight FI events [27]. Hence, one could potentially attribute them to the decay of ^{244}Fm produced in the $p2n$ channel. They result in $\sigma = 59^{+29}_{-21}$ pb, which is comparable with the 65^{+34}_{-25} pb deduced from the α events assigned to ^{244}Md .

However, in this region of heavy nuclei, p_{xn} channels are evidently populated alongside the xn ones, but with cross sections often a factor of 10 or more smaller than those of the xn ones [30,32–34]. Such a σ value for the p_{2n} channel comparable to the xn ones is presently unknown for syntheses of heavy nuclei with $Z \gtrsim 96$ [30,33,34] while it is known to occur with high probabilities in the region of neutron-deficient nuclei with $Z < 96$ [21,35,36]. Nevertheless, if such an unexpectedly high p_{2n} cross section occurs in the present study, this would indeed be an intriguing observation, which would substantially strengthen the motivation for syntheses of SHN in p_{xn} channels, as suggested in Ref. [32].

(ii) These eight ER-FI events could also originate from ^{244}Md . In this case, fission would occur from a different state than the α -decaying one with $T_{1/2} \approx 0.30$ s. However, such a short-lived fission from an odd-odd nucleus, in which fission is known to be strongly retarded compared to even-even cases, has never been observed previously [5,6]. Compared to even-odd and odd-even nuclei, where the hindrance factor for fission (the ratio between experimental and calculated unhindered half-lives [4]) is mainly determined by the property of a single unpaired nucleon [4], in the odd-odd cases, most low-lying states are characterized by a coupled single-proton and single-neutron configuration. However, the magnitude of fission hindrance in nuclei with odd numbers of both nucleon types is still unknown, as well as its dependence on the total spin and Nilsson orbital quantum numbers. Various semiempirical estimates exist and mostly predict that the hindrance is at least similar to, or higher than for neighboring isotopes or isotones [4]. In this regard, a hindrance factor of about 10^4 has been attributed to ^{243}Fm ($N = 143$ isotope of ^{244}Md) due to its unpaired single neutron. On the other hand, hindrance factors of $>10^4$ have been evaluated for the fissions of ^{247}Md and ^{247m}Md ($Z = 101$ isotope). Thus, for low-lying states in ^{244}Md one can assume that the spontaneous fission partial $T_{1/2}$ should have a gain relative to its unhindered $T_{1/2}$ by a factor of $>10^4$. Unhindered $T_{1/2}$ is typically estimated as the geometric mean of the half-lives of neighboring even-even nuclei, i.e., $^{244,246}\text{No}$ and $^{242,244}\text{Fm}$ by which the effect of the fission barrier can be isolated. However, experimental data are only known for the latter isotopes, thus the partial fission $T_{1/2}$ of ^{244}Md can only be compared with the Fm isotopes. By taking ^{244}Fm , which is the even-even isobar of ^{244}Md , one can argue that fission from ^{244}Md has to have a partial $T_{1/2}$ much ($>10^4$ times) longer than $3.12(8)$ ms [30]. However, this will be revised once also the $T_{1/2} < 4$ μs of ^{242}Fm is considered. In this case, 5_{-2}^{+3} ms fission from ^{244}Md will be $>10^3$ times longer than the fission in ^{242}Fm , which would point to a large hindrance. Accordingly, these comparisons lead to a still incomplete picture of fission from the odd-odd ^{244}Md . Still, fission from ^{244}Md is not excluded to occur with a half-life of ≈ 5 ms. If this was the case, then such a short

$T_{1/2}$ should be a result of effects from the neutron-proton configuration and fission barrier.

To conclude, the origin of the eight ER-FI events with 5_{-2}^{+3} ms cannot unambiguously be attributed to one particular of the above scenarios (i) and (ii). More interestingly, for neither scenario experimental evidence has been observed to date. Thus, each scenario would lead to an intriguing physics case. At the same time, both scenarios might contribute partially to the observed ER-FI events, leading to a third hypothetical scenario (iii). It could be that ^{244}Md was produced as a product of the $2n$ channel, but in a state that partially decays into ^{244}Fm via EC/ β^+ (hereafter: EC) decay with a short $T_{1/2}$ ($<5_{-2}^{+3}$ ms). However, such a short $T_{1/2}$ has never been observed for EC decay [6], which is a process governed by the weak interaction. Keeping in mind that the scenario (iii) is hypothetical, nevertheless, additional analyses searching for the presence of an isomeric state in ^{244}Md were carried out.

An isomeric state is retarded to decay via electromagnetic transitions, thus, it deexcites predominantly through emission of a conversion electron (CE). Recently, it has been shown that CEs from the decay of short-lived isomeric states can be efficiently detected in the digital trace of the preceding signal [23,24,37]. Since in the present experiment the shape of each signal was stored, we inspected the 30 μs -long traces of the seven ER signals followed by α decays of ^{244}Md , checking for the presence of a low-energy signal associated with the decay of an isomeric state. In one case, we found a low-energy signal, which can be attributed to the detection of a conversion electron [see Fig. 4(a)]. This signal was detected 20 μs after the ER, which gives an

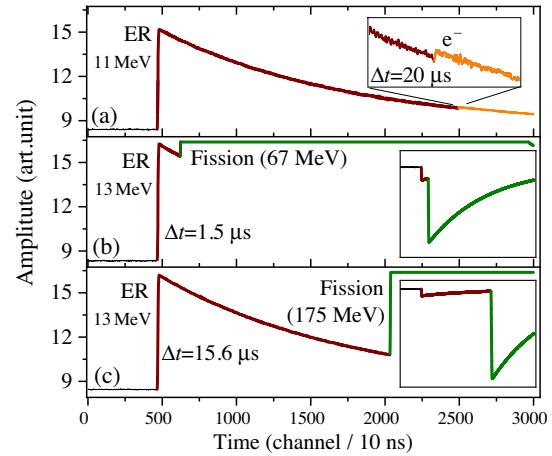


FIG. 4. The X-strip trace of the ER correlated with an α event with an energy of 8.81 MeV (assigned to ^{244}Md), in which the low-energetic signal (a close look is given in the inset) was observed (a). X-strip traces of ER-like events, in which the FI-event signal (energies are given) were detected are shown in (b) and (c). In the insets, the Y-strip traces, in which the FI-event signals are stored with full shapes are shown. Time differences between two signals are given. See text for details.

estimate for the $T_{1/2}$ of a potential isomeric state. Despite that one event is insufficient to make a definite conclusion on its assignment, this observation indicated the existence of a short-lived isomeric state in ^{244}Md . Upon decay of this isomeric state in ^{244}Md by EC, a delayed fission resulting in ECDF with a probability of $\approx 20\%$ could also take place as predicted in Ref. [9].

If such a short-lived EC decay occurred in ^{244}Md , then we might observe its ECDF branch by detecting ER-FI events with a similarly short correlation time, i.e., $< 30 \mu\text{s}$. Two ER-like events with an additional FI-like signal in their traces were found and are shown in Figs. 4(b) and 4(c). The energies of the implantation signals agree well with the average energy of ERs of ^{244}Md and ^{245}Md [27]. These FI events occur 1.5 and 15.6 μs after the ER signals. Overall, these two events and the one ER with a conversion electron lead to $T_{1/2} \approx 9 \mu\text{s}$ for the decay of an isomeric state in ^{244}Md . The EC branching of this isomeric state calculated from the numbers of observed α -decay events of ^{244}Md and fissions from ^{244}Fm would be $\approx 44\%$. In the EC decay of this isomeric state, ECDF would occur with a probability of $\approx 20\%$ based on the observed numbers of two short- and eight long-correlated ER-FI events assigned to ^{244}Fm . This result would be in fine agreement with the prediction from Ref. [9]. However, despite these surprising outcomes of additional analyses, the short $T_{1/2}$ for an EC decay of only about 9 μs , which is more than about 10^6 orders of magnitude shorter than the typical partial EC-decay half-lives of known neutron-deficient nuclei, scenario (iii) remains a hypothesis.

In conclusion, we synthesized the new isotope ^{244}Md in the $^{50}\text{Ti} + ^{197}\text{Au}$ reaction and observed its α decay with α -particle energies in the range of 8.73–8.86 MeV and with a half-life of $0.30^{+0.19}_{-0.09} \text{ s}$. We did not observe EC decay of ^{244}Md for which an upper limit of 0.14 was given. In addition, we detected eight short-lived fission events. Their origin has been discussed by involving three different possible scenarios, where each one leads to a previously unseen physics case.

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*J.Khuyagbaatar@gsi.de

[†]Present address: Università degli Studi di Napoli “Federico II,” 80126 Naples, Italy

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