Rapid Communications | Editors' Suggestion

Stability of the heaviest elements: K isomer in ^{250}No

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Decay spectroscopy of ^{250}No has been performed using digital electronics and pulse-shape analysis of the fast nuclear decays for the first time. Previous studies of ^{250}No reported two distinct fission decay lifetimes, related to the direct fission of the ground state and to the decay of an isomeric state but without the possibility to determine if the isomeric state decayed directly via fission or via internal electromagnetic transitions to the ground state. The data obtained in the current experiment allowed the puzzle to finally be resolved, attributing the shorter half-life of $t_{1/2} = 3.8 \pm 0.3$ μs to the ground state and the longer half-life $t_{1/2} = 34.9^{+3.9}_{-3.2}$ μs to the decay of an isomeric state. ^{250}No becomes, thus, one of a very few examples of very heavy nuclei with an isomeric state living considerably longer than its ground state. This phenomenon has important consequences for the nuclear-structure models aiming to determine the borders of the island of stability of superheavy elements.

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In recent decades, there has been important progress in the synthesis and investigation of superheavy elements despite the difficulty of such studies due to extremely low production cross sections. Recent studies of high-spin structures in transfermium isotopes ($Z > 100$) have demonstrated that the enhanced stability of heaviest nuclei may also be influenced by the phenomenon of K isomerism [1–3]. K isomers occur when a nuclear state is formed with a large value of K, the projection of the total angular momentum along the nuclear symmetry axis. In even-even nuclei, breaking of a pair of nucleons can lead to the creation of a high-K configuration, whose decay is strongly hindered according to selection rules, resulting in formation of a metastable state. Although K isomerism is a common feature in the transfermium region [4–7], only a few cases have been observed in which their half-lives are longer than that of the ground state. Those cases, indeed, are of special interest since they point to significant enhancement of stability against the fission despite the high excitation energy ($\approx$1 MeV) and the low fission barrier ($\approx$6 MeV).

Very few cases have been reported in heavy nuclei: ^{270}Ds [8] with an isomeric state half-life of $6.0^{+8.2}_{-2.2}$ ms compared to 100^{+140}_{-40} μs of the ground state, ^{256}Es with an isomeric state half-life of 7.6 h (25 min for the ground state) [9], and ^{254}Rf [10] with an isomeric state with a half-life of 247 ± 73 μs and a ground-state half-life of 23.2 ± 1.1 μs. This inversion of stability demonstrates the significant role that K isomerism can play in studies of superheavy nuclei [2]. The possibility that K isomers could be more stable (have longer lifetimes) in superheavy elements has been recently discussed qualitatively in terms of the shape of the fission barriers using configuration constraints in calculations of the potential energies and fission barriers but without inclusion of dynamical effects or the calculation of lifetimes [5]. Theoretical predictions of fission probabilities (and lifetimes)

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for high-K multiquasiparticle states in superheavy nuclei are still challenging, which is partially due to the paucity of experimental data.

An interesting case of possible inversion of stability due to the presence of an isomeric state has been found in ^{250}No. Two distinct short-lived activities in ^{250}No were first observed in a study performed in Dubna in 2003 by Belozerov *et al.* [11]. The longer of them, however, was tentatively attributed to ^{249}No, although the authors did not rule out the possibility that these events come from the deexcitation of an isomeric state in ^{250}No. A few years later, a study carried out at Argonne National Laboratory using the fragment mass analyzer demonstrated unambiguously that these two different lifetimes are, indeed, related to the decay of ^{250}No, namely, to the direct fission of the ground state and to the decay of an isomeric state [12]. The shorter half-life ($t_{1/2} = 3.7^{+1.1}_{-0.8}$ μs) was assigned to the ground state whereas the longer half-life ($t_{1/2} = 43^{+22}_{-15}$ μs) was attributed to the decay of a $K^{\pi} = 6^{+}$ isomeric state. This assignment was made based solely on the relative cross section of the two different decays. The sensitivity of the experiment was, however, insufficient to discriminate between two possible scenarios: the longer-lived activity could either result from direct fission of the excited level or from fission of the ground state that is delayed by K-forbidden electromagnetic transitions from the isomer to the ground-state band. A solution of this problem was not possible due to experimental limitations related to dead time and the ability to observe low-energy electromagnetic transitions. Therefore, only a lower limit for the partial fission half-life of the isomer could be determined, i.e., >43 μs. Tentative spin and parity assignments for the isomeric state were obtained based on multiquasiparticle blocking calculations [12].

The fission properties of ^{250}No have recently been also investigated at the separator SHELS at Dubna [13]. In this experiment, the half-lives, the total kinetic energies of fission fragments, and the neutron multiplicities were measured. The measured half-lives of the two fission activities were $t_{1/2} = 5.1 \pm 0.3$ and $t_{1/2} = 36 \pm 3$ μs, in good agreement with previous results.

In the present Rapid Communication, we report on detailed results of decay spectroscopy of the isomeric state in ^{250}No with the motivation of addressing the question of which decay scenario is correct: direct fission of the isomeric state or decay via electromagnetic transitions to the ground state.

A novel approach enabled separation of the low-energy signals, corresponding to the emission of conversion electrons from preceding evaporation residue implantation and subsequent fission events. Using pulse-shape analysis in conjunction with a data-acquisition system incorporating modern digital electronics, proper identification of signals with arrival times differing by less than 10 μs was possible. Consequently, the electromagnetic decay branch has been observed for the first time, leading to an unambiguous assignment of the longer-lived activity to an isomeric state. The present Rapid Communication, demonstrates the gain that can be achieved in heavy-element studies by using digital electronics, which only very recently has been successfully implemented in this type of experiment [10,14–16].

The experiment was carried out at the Accelerator Laboratory of the Department of Physics, University of Jyväskylä. The ^{250}No nuclei were produced using the ^{204}Pb (^{48}Ca, $2n$)^{250}No reaction. Highly enriched ($>99.9\%$ purity) targets of ^{204}Pb were used, mounted on a rotating wheel to withstand the beam intensity of 50–100 pnA.

The ^{204}PbS target had an average thickness of 440 μg/cm^2 and was evaporated on a carbon layer of 40 μg/cm^2 and covered with a carbon layer of 10 μg/cm^2. The study was performed with a beam energy in the middle of the target estimated to be 218.0 ± 1.0 MeV. The evaporation residues (ERs) recoiling out of the target were separated from the primary beam, beam- and targetlike products by the gas-filled separator RITU [17,18], according to their magnetic rigidity, and implanted onto the focal plane spectrometer GREAT [19]. The focal plane detection system consisted of a multiwire proportional counter (MWPC), which provided the energy loss (ΔE) and the time of flight (TOF) between the MWPC and the double-sided silicon strip detectors (DSSDs) where the separated reaction products were implanted. The implantation detector was composed of two detectors of 300 μm-thick DSSDs placed side by side. Each of the DSSDs had a size of 60×40 mm and was segmented with a strip pitch of 1 mm. The X side of each detector was equipped with analog electronics, and the gain was optimized for high-energy particles, i.e., fission fragments and target recoils. The Y side was read out by digital electronics, namely, Lyrtech's VHS-ADC (high-speed signal processing system) cards. Thus, allowing direct digitization of the preamplifier signals at a sampling rate of 100 MHz and 14-bit resolution with corresponding waveforms of maximum length 10 μs (traces), enabling signals from different energy deposition processes to be separated. Signals separated by more than 10 μs were recorded in different events. For the Y side, the energies of the particles detected by the DSSD were extracted by applying the moving window deconvolution algorithm [20]. A high digital gain was selected, optimized for low-energy events, such as conversion electrons or α particles, leading to a saturation of signals from fission fragments.

The DSSDs were surrounded by 28 silicon PIN diodes in a box configuration on the upstream side. The DSSDs and the PIN detectors were calibrated with a triple-α source containing ^{239}Pu, ^{241}Am, and ^{244}Cm, and the Y strips were, in addition, calibrated using a ^{133}Ba electron source for the electron energy calibration. A large-volume germanium Clover and two EUROGAM germanium Clover [21] detectors were installed around the DSSD detectors in order to detect γ rays coming from the deexcitation of the ^{250}No nucleus. In addition, a 15-mm-thick double-sided Planar germanium strip detector with a 120×60-mm active area and a strip pitch of 5 mm was placed directly behind the DSSD inside the same vacuum chamber to detect low-energy γ rays. The germanium detectors were calibrated with ^{133}Ba and ^{152}Eu sources. All germanium and PIN detectors were also instrumented with digital Lyrtech VHS-ADC cards. The triggerless total data readout data-acquisition system was used [22] whereby the energies and times of all events in the various detectors are collected and time stamped to an accuracy of 10 ns. Data analysis was carried out using the GRAIN software package [23].

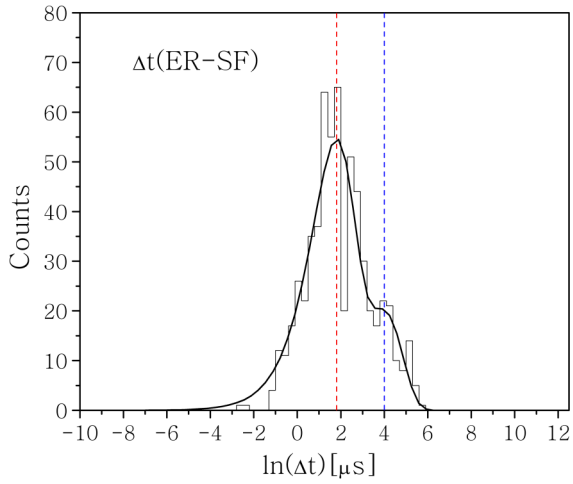

FIG. 1. Time distribution of SF events following the ER implantation. Data are fitted with a two-component decay curve (solid line). The mean values of each component are underlined by the dashed colored lines, red and blue, respectively. The maximum search time was 300 μs.

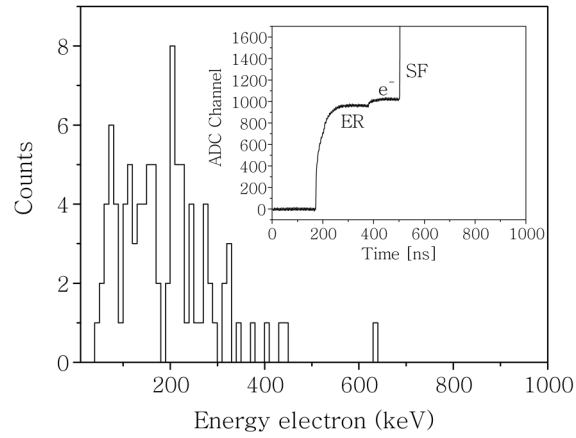

FIG. 2. Electron energy spectrum from ER-e$^-$SF correlations. The inset shows a representative wave form for a ER-e$^-$SF decay sequence.

The identification of the ER was performed by demanding a ΔE signal in the MWPC detector in coincidence with an energy signal in the DSSD implantation detector. Coincidences between the TOF signals and the total-energy signals, measured in the DSSD, were used to discriminate against scattered beam and targetlike reaction products. The events where implantation of an ER was followed by a fission event (SF) observed in the same pixel, up to a maximum correlation time of 300 μs, were assigned to ^{250}No nuclei. During the beam time of 112 h, 613 such events were observed. Among them were 370 events where evaporation residues were found in the same trace (within 10 μs) as the fission event, which illustrates the importance of using digital electronics for this Rapid Communication. The remaining events were detected in separated events for which the trace and pulse-shape analysis is not required. A fission event appears as a saturation in the trace signals due to the digital gain chosen on the Y side whereas for the X side an energy of greater than 50 MeV was demanded.

In order to deduce the half-life of the ^{250}No decay, the time differences between the implantation of an ER and a subsequent SF event were analyzed and are presented in Fig. 1. The time distribution was used to determine the half-lives using the method suggested by Schmidt [24]. The solid line in Fig. 1 is the result of fitting the data with two components and represents the time distribution of fission events correlated with ^{250}No evaporation residues, clearly showing the occurrence of two activities with different half-lives. The peak position determines the lifetime via the $\ln(\tau)$ relation. The half-life corresponding to the main peak is equal to 3.8 ± 0.3, in good agreement with values reported by Peterson *et al.* [12] and Svirikhin *et al.* [13]. The shoulder on the right side, highlighted by the blue dashed line, indicates larger correlation times.

In order to distinguish between the two possible decay scenarios, electromagnetic transitions were searched for. K-isomeric states known in this region of nuclei typically deexcite via several rapid internal transitions with accompanying emission of conversion electrons and γ rays, which is followed some time later by the decay of the ground state. Consequently, the so-called calorimetric method was applied [25] whereby events in which, at least, one electron (i.e., a low-energy signal) was observed between the implantation of ^{250}No and a subsequent fission event in the same pixel.

A total of 97 such events were observed. The resulting total electron energy spectrum together with a typical ER-e$^-$-SF (evaporation residue-electron-fission) trace is shown in Fig. 2. One can see that the three kinds of events can be easily distinguished due to their very different amplitude scales: ER (with an energy of 8–20 MeV), conversion electrons (with an energy up to 700 keV), and fission fragments (with an energy >50 MeV) which corresponds to saturation of the trace as described earlier. Thus, we can unambiguously show experimentally that the isomeric state decays to the ground state via electromagnetic transitions followed by the ground-state fission.

The half-life of the isomeric state was deduced by fitting the logarithmic decay-time distribution of conversion electrons correlated with an ER detected at the focal plane and requiring correlation with an SF of ^{250}No, see Fig. 3(a). A half-life of $t_{1/2} = 34.9^{+3.9}_{-3.2}$ μs has been extracted. For comparison, the time distribution of ER-SF correlations, provided an electron was detected in between ER implantation and SF, is also shown in Fig. 3(b) for the same events. As seen from Fig. 3, both fission events and electron events have relatively long and similar half-lives. This implies that the decay of the ground state by SF must have a short half-life compared to the isomeric state so that the delay due to decay of the ground state does not change the time distribution significantly. Hence, the long-lived component in Fig. 1 is unambiguously assigned to the fission decay of the ground state delayed by the electromagnetic decay of the isomer, whereas the short component can be related to the ground-state decay of ^{250}No.

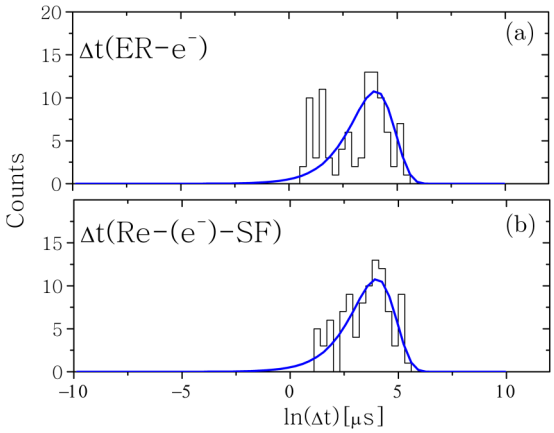

FIG. 3. (a) Evaporation residue-electron decay-time difference spectra in correlations with a fission event following the electron. The solid blue line corresponds to the fit using a one-component decay curve. The maximum searching time was $\Delta t = 300$ μs. (b) Decay-time distributions for the ER-SF correlation requiring an electron burst in the DSSD.

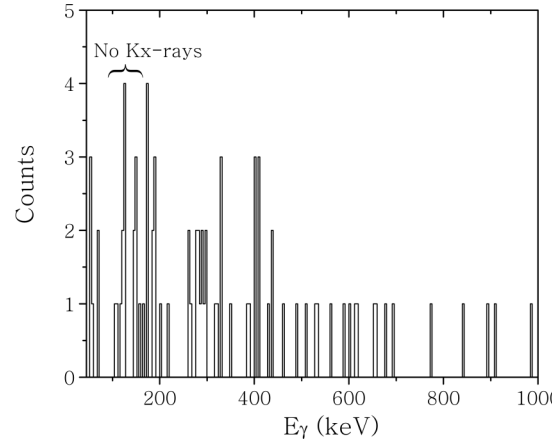

FIG. 4. γ-ray spectrum in coincidence with emitted electrons following the deexcitation decay of the isomer from the Planar and Clover germanium detectors.

In order to estimate the production rate for the ground state and for the isomeric state, considering that the detection efficiency for electrons may not be 100%, only the total ER-SF events were considered. By integrating the two distributions shown in Fig. 1, an isomer population ratio relative to the ground state of about 41(13)% was obtained. This value is consistent with the known population of isomeric states in this mass region (e.g., ^{252,254}No [26,27]), which are typically about 10%–30%.

The total production cross section for ^{250}No was estimated considering the total number of ERs followed by fission events. The following factors were taken into account: RITU transmission 33(5)% and DSSD coverage 83(5)%. The uncertainty comes mainly from the beam dose estimate obtained by averaging the number of incoming ions on the focal plane and the uncertainty in RITU transmission. The uncertainty in the excitation energy take into account the uncertainty of the beam energy from the cyclotron (0.5%) and the energy lost in the target. The resulting production cross section is $\sigma = 32(8)$ nb at a compound nucleus excitation energy in the middle of the target of 24.2 ± 1.5 MeV.

Gamma rays associated with the deexcitation of the long-lived isomeric state have also been investigated. The delayed γ rays following the deexcitation of isomeric states were detected in the germanium detectors surrounding the focal plane, i.e., Planar and Clover germanium detectors. A coincidence with the conversion electrons from the decay of the isomeric state was demanded using the conditions $\Delta t(\mathrm{ER} - e^-) <$ 300 μs and $Ee^- < 700$ keV. The resulting γ-ray spectrum is presented in Fig. 4. Two peaks at $\approx$127 and $\approx$143 keV are observed, consistent with characteristic K_α and K_β x rays in nobelium. Clusters of γ rays at 250–300 and at 400 keV as well as a few counts at an energy above 800 keV are present, indicating possible transitions from the isomeric state towards the ground state. Although the statistics is clearly not sufficient to firmly establish the decay path, the high-energy γ-ray transitions could correspond to deexcitation of a sideband to the ground-state rotational band, in line with the systematics of this mass region. Indeed, sidebands are commonly observed in $N = 150$, 152 isotones (e.g., ^{254}No, ^{252}No, ^{250}Fm, etc. [6,26,28,29]) as well as in other $N = 148$ isotones. For instance, a recent study of ^{244}Cm underlines the presence of a sideband with spin and parity $K^\pi = 2^-$ and a bandhead at 933.6 keV, interpreted as an octupole vibrational band [30]. Clearly, more detailed investigations should be performed in order to unambiguously outline the decay path of the isomeric state in ^{250}No and assign its spin and parity. Nevertheless, a rough estimate of the excitation energy of the isomeric state can be obtained by summing the γ-ray energies together with those of the conversion electrons measured in prompt coincidences, suggesting that the isomer may be located at $\approx$1.2 MeV.

Albeit only a hypothesis, such a value is in agreement with the results of extensive calculations performed by Delaroche *et al.* [31] using Hartree-Fock-Bogoliubov with two quasiparticle blockings and recently by Liu *et al.* [2] using a configuration-constrained potential-energy surface (PES).

A summary of experimental [32–35] and theoretical information [31] for the isomeric states in $N = 148$ isotones is shown in Fig. 5. The calculations suggest that a $K^\pi = 6^+$ state with a two-quasi-neutron $(5/2^+[622] \otimes 7/2^+[624])$ configuration is the most likely candidate for a two-quasiparticle isomer with an excitation energy of 1.01 MeV, corroborated by Ref. [2] which predicts an excitation energy of 0.83 MeV. Similar assignments have been performed in other $N = 148$ isotones. The predicted energies of the alternative $K^\pi = 7^-$ configuration are much higher than the experimental observations.

In order to search for a possible α branch of the ground-state decay of ^{250}No, which has not been observed in previous experiments, correlations of an ER implantation with a subsequent signal in the same pixel, corresponding to an energy of 5–10 MeV and a time difference less than 1 ms, were searched for. Two α events were found with energies of 9490 and 9270 keV with decay times of 142 and 105 μs,

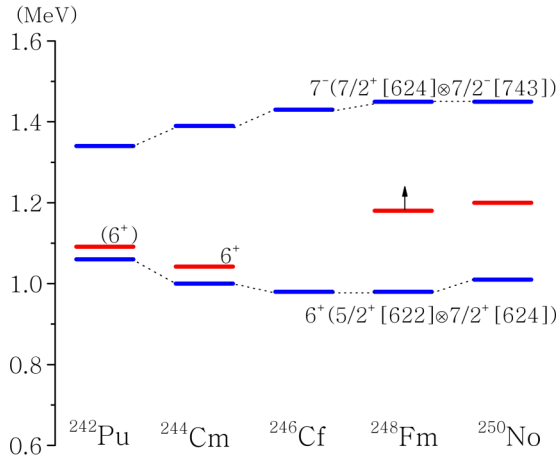

FIG. 5. The red lines: experimental isomeric state energies (MeV) for $N = 148$ isotones [32–35]. The blue lines: theoretical calculation of Delaroche *et al.* [31].

respectively. It is interesting to note that a possible origin of these decays could be associated with the long-lived isomeric state. Indeed, based on experimental and extrapolated mass excesses, an α decay energy of 8.8 MeV is expected for the ground-state decay of ^{250}No [36]; therefore, the somewhat higher α energies are potentially a signature of α decay from the isomeric state. However, with present data, we cannot make a definite conclusion.

To summarize, a new internal transition branch has been measured for the first time stemming from the isomeric state in ^{250}No, which decays towards the ground state with a half-life of $t_{1/2} = 34.9^{+3.9}_{-3.2}$ μs followed by the ground-state fission with a half-life of 3.8 ± 0.3 μs. The longer half-life of the isomeric state compared to the ground state suggests that there is substantial fission hindrance due to the isomeric state. The fission from the isomeric state in ^{250}No is hindered by a factor of more then 19 compared to the ground state [29]. The hindrance has been calculated on the basis of the partial SF half-lives for the isomer and the ground state. A lower limit of 50% has been estimated for the electromagnetic branch, assuming 100% emission and detection of an electron in the decay path to the ground state. Configuration-constrained PES calculations [5] have been performed for ^{250}No and other heavy nuclei (^{254}No, ^{256}Fm) with an emphasis on the relative fission probabilities of high-K isomers and their respective ground states. These calculations predicted a significant increase in the K-isomer fission barrier (6.89 MeV, compared to the corresponding ground-state fission barrier of 5.45 MeV) and estimated a half-life reduction of about five orders of magnitude per MeV of barrier height. Our new experimental result supports these calculations.

To provide more insight regarding the possibility of increased barrier heights for multiquasiparticle excitation, new experimental data are needed. The investigation of ^{248}No where the fission barrier may disappear due to the dramatic decrease in the fission barriers with the neutron number would also be of great interest. From the systematic of the half-lives for the even-even isotopes in the $N = 152$ region, it appears that the nobelium nuclei present the sharpest drop on either side of $N = 152$, which appears to indicate that their stability gain from this deformed shell closure is the most significant. This is remarkable since none of the existing calculations of fission barriers in this region [37–40] predict such a dramatic change. Moreover, these nuclei around $N = 152$ region may also gain additional stability due to the presence of isomeric states.

Further systematic studies of the half-lives of even-even nuclei in the region, and, in particular, their behavior as a function of neutron number, may improve our understanding of the fission process and, more specifically, of the stability against fission of normally deformed high-K isomers. In this context, they provide guidance in the search for the heaviest elements where multiquasiparticle high-K isomers may also lead to enhanced stability.

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