Decay and Fission Hindrance of Two- and Four-Quasiparticle K Isomers in ^{254}Rf

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Phys. Rev. Lett. **115**, 132502 — Published 24 September 2015
DOI: 10.1103/PhysRevLett.115.132502

Decay and fission hindrance of two- and four-quasiparticle K isomers in ^{254}Rf

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(Dated: August 11, 2015)

Two isomers decaying by electromagnetic transitions with half-lives of 4.7(1.1) μs and 247(73) μs have been discovered in the heavy ^{254}Rf nucleus. The observation of the shorter-lived isomer was made possible by a novel application of a digital data acquisition system. The isomers were interpreted as the K^{π}=8$^-$, ν^2(7/2$^+$[624], 9/2$^-$[734]) two-quasineutron and the K^{π}=16$^+$, 8$^-\nu^2$(7/2$^+$[624], 9/2$^-$[734])$\otimes$8$^-\pi^2$(7/2$^-$[514], 9/2$^+$[624]) four-quasiparticle configurations, respectively. Surprisingly, the lifetime of the two-quasiparticle isomer is more than four orders of magnitude shorter than what has been observed for analogous isomers in the lighter $N = 150$ isotones. The four-quasiparticle isomer is longer lived than the ^{254}Rf ground state which decays exclusively by SF with a half-life of 23.2(1.1) μs. The absence of sizable fission branches from either of the isomers implies unprecedented fission hindrance relative to the ground state.

What is the maximum number of protons and neutrons that can be held in a nucleus? The key to answering this fundamental question lies in the interplay between the attractive nucleon-nucleon interactions and the repulsive Coulomb force that acts between protons. According to the simplistic liquid-drop model, nuclei with $Z > 100$ should undergo fission instantaneously. However, shell corrections, which reflect the quantum nature of a nucleus, increase the barrier against fission and the so-called super-heavy nuclei (SHN), with many more protons, can be produced in fusion reactions and live long enough to be detected in a laboratory.

Recently, the nuclear landscape was extended to $Z = 118$ [1], approaching a predicted island of relative stability centered around new spherical proton and neutron shell closures, where the shell effects are significantly enhanced. The exact values of the magic numbers are still elusive (see Ref. [2] and references therein), which provides an impetus for further experimental and theoretical studies. The island of stability is connected to the mainland of stable nuclei by a shoal, which is centered around the deformed subshell closures at $Z = 100$, $N = 152$ and $Z = 108$, $N = 162$, consisting of nuclei that gain stability as a result of prolate deformation. In deformed, axially-symmetric nuclei, the projection of the angular momentum of individual nucleons onto the symmetry axis, Ω, is a good quantum number. When the neutron or proton Fermi surface (or both) is near orbitals characterized by large Ω, multi-quasiparticle states with high K value, where $K=\sum_i \Omega_i$ is a sum over all unpaired nucleons, can be formed at relatively low excitation energies. These states are often long lived, since depopulating transitions proceed between levels with significantly different K values [3]. The observation of these so-called K isomers and elucidation of their properties can provide valuable information on single-particle excitation energies and on residual nucleon-nucleon interactions, such as pairing and spin-dependent interactions between unpaired nucleons, which are an essential input for understanding the structure of SHN.

Spontaneous fission (SF) is an important decay mode of SHN. Fission is also critical during the formation of SHN in fusion reactions as it competes with neutron evaporation during the cooling process. K isomers provide the means to study the role of pairing and K conservation during the fission process, which are key elements for the accurate description of fission lifetimes and dynamics. Theoretical predictions for SF decay probabilities from K isomers using a one-dimensional approach and the WKB approximation concluded that the SF rate could be orders of magnitude slower than that from ground states due to higher and wider fission barriers [4, 5]. By incorporating a dynamical treatment of pairing, SF of high-K states was found to depend critically on dynamically-induced superfluidity in the tunneling process [6]. Recently, the possibility that K isomers could be more stable in SHN has been discussed in terms of the shape of the fission barriers when using configuration constraints in calculations of the potential energies and fission barriers, but without inclusion of dynamical effects or the calculation of lifetimes [7, 8]. State-of-the-art nuclear density functional theory calculations have underscored the importance of the interplay between pairing and shape parameters during the fission process [9]. Information on fission from high-K isomers is essential for testing emerging models of SF. Such data, however, remain very scarce [10].

Following the early discovery of two-quasiparticle (2-qp) isomers in ^{250}Fm ($Z = 100$) and ^{254}No ($Z = 102$) [11], many spectroscopic studies have been carried out for trans-fermium nuclei in recent years. Two-quasiparticle K isomers were observed in several even-even, $N = 150$ isotones, from ^{244}Pu ($Z = 94$) to ^{252}No ($Z = 102$) [11–15], and were associated with the K^π=8$^-$, two-quasineutron ν^2(7/2$^+$[624],9/2$^-$[734]) configuration. This assignment was supported by the properties of rotational bands feeding the isomers in ^{250}Fm [13] and ^{252}No [16]. In all cases, the isomers decay to the members of the K^π=2$^-$ octupole band and to the 8$^+$ member of the ground-state band. The ^{254}Rf nucleus is the heaviest known $N = 150$ isotone, but no information is available about excited structures in this nucleus. Its ground state disintegrates by SF with a relatively short half-life, but the published values are discrepant: $T_{1/2}$=500(200) μs [17], 23(3) μs [18], and 29.6($^{+0.7}_{-0.6}$) μs [19].

In this Letter, we report on the discovery of two isomers in ^{254}Rf using a novel approach involving a pulse-shape analysis in conjunction with a digital data acquisition system. Two experiments were carried out in which ^{254}Rf nuclei were produced via the ^{206}Pb(^{50}Ti, 2n) fusion-evaporation reaction and studied using fission tagging. In the first measurement, ^{50}Ti ions at 242.5 MeV impinged onto a 0.5-mg/cm^2, 99.948%-enriched ^{206}Pb target at the Argonne National Laboratory ATLAS facility. The targets were mounted on a rotating wheel to withstand a beam current between 100 and 200 pnA. Recoiling residues were dispersed according to their mass-to-charge state ratio by the Fragment Mass Analyzer (FMA) [20] and, after passing through a parallel-grid avalanche counter, were implanted into a 100 μm-thick, 64×64 mm^2, 160×160 strip double-sided silicon strip detector (DSSD) at the focal plane. SF events from Rf isotopes with mass A = 254-256 were unambiguously identified by implementing spatial and temporal correlations between implanted residues and high-energy (>100 MeV) decay events in the DSSD. Signals from the DSSD were digitized using 100 MHz, 14-bit digitizers [21], enabling readout of waveforms spanning 10 μs, starting 1.5μs before the leading edge of the signal. This advancement in instrumentation has an especially large impact in studies of this type, where low-energy internal conversion electrons from high-K isomeric states are expected with lifetimes as short as a few μs. A total of 28 fission events corresponding to A=254 associated with implanted ^{254}Rf nuclei were observed. In four of the ^{254}Rf fission events, a small signal corresponding to a burst of internal conversion electrons following an isomeric decay was observed as a pile-up on the ^{254}Rf implant waveform. Decay times and energies of electrons occurring within 10μs after implantation were extracted through detailed analysis of the waveform. An additional fifth electron was observed at a longer decay time of 515 μs after implantation, with the subsequent fission occurring 38 μs later. These events were interpreted as evidence for the existence of two isomers in ^{254}Rf.

A follow-up experiment was carried out at the Lawrence Berkeley National Laboratory using the high-efficiency Berkeley Gas-filled Separator (BGS) [22] to charcterize the two isomers. Ions of ^{50}Ti, accelerated to 244 MeV by the 88-Inch Cyclotron, impinged on a 0.5-mg/cm^2 target of ^{206}Pb, mounted on a rotating wheel for 132 hours with an average intensity of about 275 pnA. Recoiling evaporation residues were separated from other reaction products and unreacted beam ions by the BGS, before passing through a multi-wire proportional counter. Three 1-mm-thick, 32×32 strip, 64×64 mm^2 DSSDs, equipped with logarithmic preamplifiers, in a corner-cube configuration at the focal plane of the BGS served as implantation detectors for reaction products, combining to provide a total of 3×32×32=3072 pixels. Signals from all detectors were digitized as for the FMA experiment described above. However, the digitizer firmware was redesigned in order to better match experimental conditions [23]. In addition, γ rays emitted at the focal plane were measured by three clover HPGe detectors, positioned ∼4 mm behind each of the DSSDs.

Seven hundred twenty three SF events from ^{254}Rf nuclei were cleanly identified by selecting implant events in the DSSD with an energy of 6-17 MeV that were immediately followed by a fission event in the same pixel. The time difference between these implant events and the subsequent fission can be seen in Fig. 1. The exponential fit, corrected for unobserved feeding from a short-lived isomer discussed below (see Fig. 1), resulted in a half-life of $T_{1/2}$ = 23.2(1.1) μs for the ground state of ^{254}Rf, in good agreement with the 23(3) μs value reported in [18], but shorter than 29.6$^{+0.7}_{-0.6}$ μs measured in [19].

A characteristic signature of isomeric decays in ^{254}Rf is the emission of several internal conversion electrons followed by the ground-state fission. Consequently, a search for isomers was carried out by selecting chains of events wherein at least one electron burst was observed in-between implantation and subsequent ^{254}Rf fission in the same DSSD pixel, using approach described

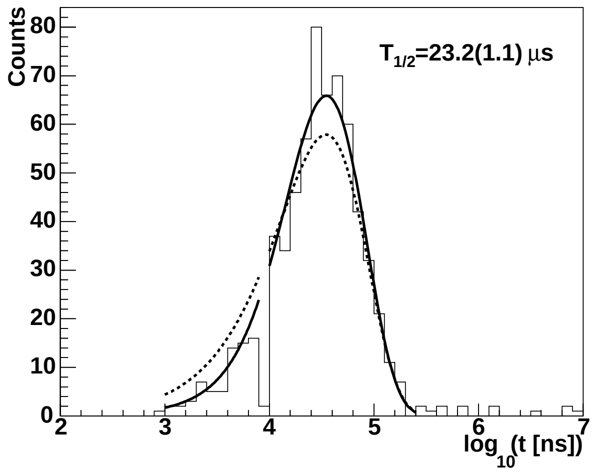

FIG. 1. The decay time distribution for the ^{254}Rf ground-state fission events. The dashed line corresponds to the exponential decay fit. The solid line corresponds to the fit which takes into account undetected feeding from a short-lived isomer (the half-life of the isomer was fixed at 4.7 μs). The latter fit indicates that about 15±5% of the fission events were delayed by the undetected isomer decays. The gap in the fitted function represents the dead time at the end of each event.

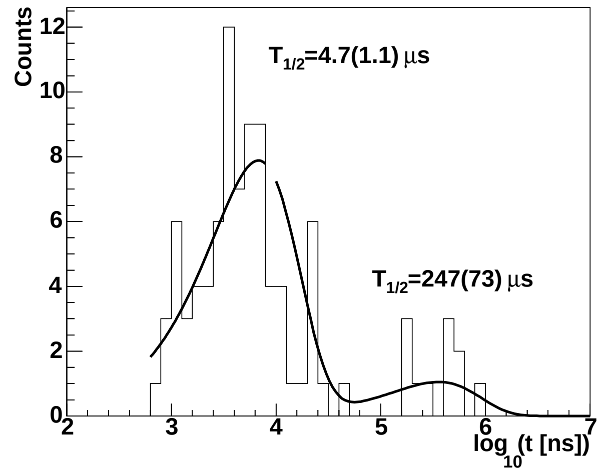

FIG. 2. Decay-time distributions for the electrons emitted from both isomers in ^{254}Rf. The solid line corresponds to the two-component exponential decay fit.

in Ref. [24]. The waveform of one such implant-electron-fission chain is shown as inset in Fig. 3(a). The time difference between the implant and the detection of the first electron signal for these event chains can be seen in Fig. 2, where two distinct groups are visible, implying the presence of two isomeric states. Eighty-two electrons were associated with a shorter-lived isomer with a half-life of 4.7(1.1) μs, while 11 electrons were associated with a longer-lived isomer with $T_{1/2}$ = 247(73) μs. In addition, there are seven event chains wherein two successive electron bursts were detected inbetween the implantation and the ^{254}Rf ground-state fission; the waveform of one such electron-electron event is presented in inset in Fig. 3(b). For these events, the measured decay times of the first and second generation electrons are consistent with the longer-lived isomer feeding the shorter-lived one. Based on the number of observed ground-state fission events and isomeric electron events, isomer population ratios relative to the ground state of about 25% (after correcting for unobserved shorter-lived isomer decays) and about 2% were deduced for the shorter- and longer-lived isomers, respectively. Given the typical isomer ratio values of 10-30% [13–15] for 2-qp isomers in neighboring nuclei and a value of ∼4% reported for the 4-qp isomer in ^{254}No [25], one may conclude that the isomers in ^{254}Rf are most likely associated with 2- and 4-qp excitations. Figures 3(a) and (b) contain the energy distributions for electrons associated with the shorter-lived and the longer-lived isomer, respectively. Both spectra extend to about 450 keV, i.e., 100 keV higher compared to the 2-qp isomers in the lighter $N = 150$ isotones. This supports the 4-qp nature of the longer-lived isomer. If both isomers were of 2-qp character, one would expect the energy difference between them to be less than ∼200 keV. Gamma rays detected in prompt coincidence with electrons emitted following the decay of the 2-qp isomer are shown in Fig. 3(c). An 893-keV line with five counts is clearly visible in this spectrum. There is also a cluster of three counts at 853 keV and two counts at 829 keV, indicating two other possible transitions. Similar, high-energy γ rays connecting the octupole band with the ground-state band are known to follow the isomer decays in the lighter $N = 150$ isotones [13–15]. The sum of the 893-keV γ-ray energy and the 450-keV maximum electron energy indicate that the 2-qp isomer is located at about 1350 keV or higher. Multi-quasiparticle, pairing-blocking calculations were carried out using the approach outlined in Ref. [26]. The main difference was that the single-particle energies for orbitals near the proton and neutron Fermi surfaces were adjusted to reproduce the known one-quasiparticle states in neighboring, odd$-A$ nuclei. In addition, the effect of the residual nucleon-nucleon interactions was taken into account, using the approach discussed in Ref. [27]. The calculations aimed at predicting the ordering of the multi-quasiparticle states rather than their exact excitation energies. The predicted lowest-lying 2- and 4-qp states in ^{254}Rf are presented in Fig. 4. The calculations suggest that the K^π=8$^-$, ν^2(7/2$^+$[624],9/2$^-$[734]) configuration is the most likely candidate for the 2-qp isomer, similarly to the assignments made in the lighter $N = 150$ isotones. The alternative K^π=8$^-$, π^2(7/2$^-$[514],9/2$^+$[624]) and the K^π=5$^-$, π^2(1/2$^-$[521],9/2$^+$[624]) configurations are predicted higher in energy. The 4-qp

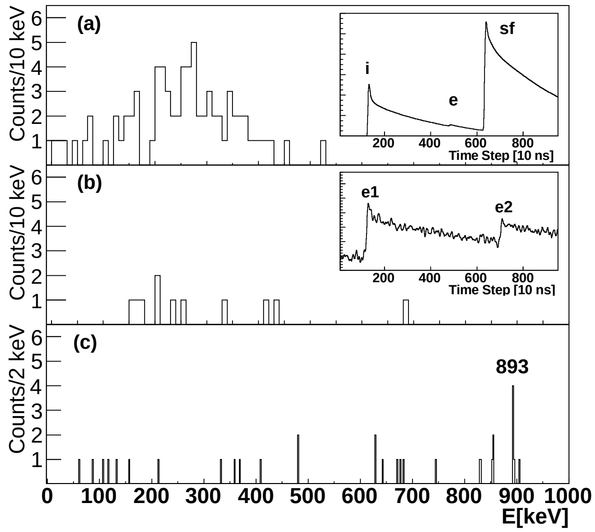

FIG. 3. Energy spectra for electrons emitted from (a) the 2-qp and (b) the 4-qp isomer, and (c) spectrum of γ rays coincident with electrons emitted following the decay of the 2-qp isomer. Insets in panel (a) and (b) contain waveforms for implant-electron-fission (i-e-sf) and electron-electron (e1-e2) decay sequences, respctively.

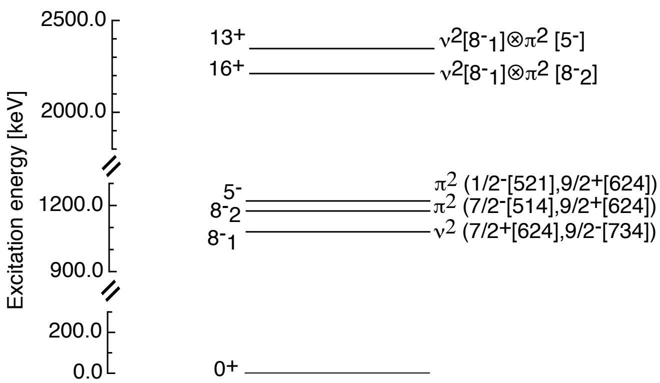

FIG. 4. Two- and four-quasiparticle states in ^{254}Rf predicted using the multi-quasiparticle calculations.

isomer is most likely associated with the K^π=16$^+$, ν^2(7/2$^+$[624],9/2$^-$[734])$\otimes\pi^2$(7/2$^-$[514],9/2$^+$[624]) configuration. The same lowest 2-qp and 4-qp configurations were predicted recently in Ref. [28]. Interestingly, the measured lifetime of the 2-qp isomer in ^{254}Rf is 4-5 orders of magnitude shorter when compared with the two-qp isomer half-lives of 1.92(5) s and 109(6) ms observed in the lighter $N = 150$ isotones ^{250}Fm [13] and ^{252}No [14, 15], respectively. One possible explanation could be that a different, lower-K configuration is associated with the isomer in ^{254}Rf, such as K^π=5$^-$, π^2(1/2$^-$[521],9/2$^+$[624]), which would lead to a shorter lifetime since depopulating transitions would be less K forbidden. This scenario was proposed to explain an unexpectedly short lifetime of the lowest isomer in ^{256}Rf [29]. However, the K^π=5$^-$ state is predicted to lie ∼150 keV higher in energy (see Fig. 4). In order to bring the K^π=5$^-$ state down in energy, an unanticipated, sudden deformation change in ^{254}Rf would be needed that leads to a different ordering of the proton single-particle levels. Also, the electron and γ-ray spectra associated with the 2-qp isomer are quite different from spectra obtained for the lowest isomer in ^{256}Rf [30, 31].

In the case of the K^π=8$^-$, ν^2(7/2$^+$[624],9/2$^-$[734]) assignment, several factors may contribute to the much shorter lifetime. Firstly, the observed decrease in hindrance of the M1 decay branch to the 7$^-$ member of the octupole band with increasing atomic number in the $N = 150$ isotones [10], extrapolated to ^{254}Rf, could account for more than an order of magnitude decrease in the lifetime of the isomer when compared to ^{252}No. The preffered decay of the isomer through the octupole band is also consistent with the weak population of the 8$^+$ level of the ground-state band, as manifested by the absence of the 8$^+\to$6$^+$ ground-state band transition around 250 keV in the γ-ray spectrum in Fig. 3(c). Secondly, the I^π=8$^-$ member of the K^π=2$^-$ octupole band could be located very close in energy to the K^π=8$^-$ isomer, leading to accidental mixing and thus shorter lifetime, as recently observed in ^{174}Lu [32]. The K^π=8$^-$ isomer is located approximately 25 keV above the I^π=7$^-$ member of the octupole band in the neighboring $N = 150$ isotones ^{250}Fm [13] and ^{252}No [14, 15]. The γ-ray transitions observed in the present experiment have energies similar to those of the transitions connecting the octupole and the ground-state bands in ^{250}Fm and ^{252}No, implying that the properties and relative positions of the two bands could be very similar in ^{254}Rf. On the other hand, the observed maximum electron energy in ^{254}Rf is about 100 keV higher compared to ^{250}Fm and ^{252}No, suggesting that the K^π=8$^-$ isomer in ^{254}Rf is located in close proximity to the I^π=8$^-$ octupole band member, which is expected to be ∼95 keV above the 7$^-$ level. Higher energies of de-exciting transitions would also result in the shorter isomer lifetime.

No definitive evidence was found for a fission branch for either of the two isomers. In the case of the 2-qp isomer, potential fission events are obscured by ground-state fission. There is no visible excess of fission events between 1 and 9 μs in the ground-state decaytime spectrum in Fig. 1. Ten counts represent one standard-deviation departure in this time interval, which corresponds to an upper limit for the fission branch from the 2-qp isomer of about 10%. Six fission events with decay times consistent with the half-life of the 4-qp isomer were observed. However, these could also be associated with ground-state fission events following decays of the 4-qp isomer which escaped detection. If all six fission events originated from the isomeric state, the upper limit for the fission branch from this level would be 40%. Consequently, the lower limits for the partial fission half-life are $T_{1/2}(SF)>50$ μs and 600 μs, corresponding to a fission hindrance of $HF = T^{iso}_{1/2}(SF)/T^{gs}_{1/2}(SF) >2$ and >25, for the two- and 4-qp isomers, respectively. The ^{250}No nucleus is the only other known case where fission from a K isomer is hindered with respect to ground-state fission [33]. This provides evidence that K isomers in SHN could live significantly longer than their ground states.

In conclusion, two isomers were discovered in ^{254}Rf and were interpreted as the two-quasineutron K^π=8$^-$, ν^2(7/2$^+$[624], 9/2$^-$[734]) and K^π=16$^+$, 8$^-\nu^2$(7/2$^+$[624], 9/2$^-$[734])$\otimes$8$^-\pi^2$(7/2$^-$[514],9/2$^+$[624]) 4-qp configurations consistent with their decay pattern and in agreement with the multi-quasiparticle calculations. Both isomers have very unexpected properties. The half-life of the 2-qp isomer is 4-5 orders of magnitude shorter than that of equivalent isomers in lighter $N = 150$ isotones. More detailed spectroscopic information is required to pin down the exact reason for this abrupt change. The 4-qp isomer lives longer than the ground state. Despite the fact that the ^{254}Rf ground state rapidly undergoes fission, no such branch was found for either of the two isomers, implying significant fission hindrance in the isomeric states. This mechanism could lead to longer apparent lifetimes of the heaviest existing nuclei above $Z = 118$ possibly extending the nuclear landscape that can be accessed experimentally to even heavier elements.

This material is based upon work supported by the US Department of Energy, Office of Science, Office of Nuclear Physics, under contract number DE-AC02-06CH11357, DE-FG02-94ER408341 and DE-FG02-94ER40848 and by NSF grant number PHY-1203100. This research used resources of ANL's ATLAS facility, which is a DOE Office of Science User Facility.

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