

In-beam study of ^{254}No

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Abstract. Excited states of the $Z = 102$ nuclide ^{254}No have been studied in the reaction $^{208}\text{Pb}(^{48}\text{Ca},2n)$ by means of in-beam γ -ray spectroscopy in combination with recoil gating and recoil decay tagging. A Ge detector array, consisting of four clover detectors, and a gas-filled separator were used. Six γ -ray lines were observed and associated with E2 transitions in the ground state band of ^{254}No , the highest-lying of these being the $16^+ \rightarrow 14^+$ transition. Based on global systematics and the extrapolated 2_1^+ excitation energy, the value $\beta_2 = 0.27 \pm 0.03$ was extracted for the quadrupole deformation. An improved value for the half-life of ^{254}No , $T_{1/2} = (48 \pm 3)$ s, was determined.

PACS. 27.90.+b $220 \leq A < 250$ Properties of nuclei; nuclear energy levels – 21.10.Hw Spin, parity, and isobaric spin – 23.20.Lv Gamma transitions and level energies

1 Introduction

Efforts aimed at the synthesis of new elements have, following a steady progress in detection techniques, led to production of elements up to proton number 112 [1]. The elements with $Z = 107$ –112 were produced using cold fusion reactions [2] where Pb or Bi targets were bombarded with ions of Cr–Zn so that the 1n evaporation channel was observed. To separate the new nuclei from unwanted particles, the fast and efficient method of in-flight separation with the on line velocity filter SHIP [3] at GSI, Darmstadt, was used.

The ultimate goal of the study of the heaviest elements is not only to find their limit of existence but also to reach the long-predicted island of spherical super heavy nuclei [4]. The main result from recent progress in this field is the discovery that the shell stabilized island is not separated from the peninsula of known nuclei by the sea of fission as originally expected. Rather, a region of enhanced stability

against fission due to a deformed shell closure at $Z = 108$ and $N = 162$ has been found. Hence, the main decay mode for isotopes of the heaviest known elements is α decay.

The most important component in the deformation of the isotopes of the heaviest known elements is the quadrupole deformation. It is predicted to have a relatively constant value of $\beta_2 \sim 0.24$ in a large region on the chart of nuclei centered around ^{256}No [5, 6]. The relative importance of higher order terms depends on the nuclide. The shell stabilization of the strongly bound nucleus ^{270}Hs ($Z = 108$), for example, has been suggested to originate from a negative hexadecapole (β_4) deformation in the ground state [5]. It is of interest that some HFB [7] and HF+BCS [8] calculations and mean-field models [9] predict a larger deformation $\beta_2 \sim 0.28$ – 0.30 than the above mentioned Nilsson-Strutinsky type of calculations.

Two of the most important goals of further studies in the region of the heaviest elements are experimental establishment of their deformation and deeper insight into their production mechanism in heavy-ion-induced fusion reactions. Measurement of the excitation energies of the ground state bands of even-even nuclei provides valuable information on the deformation. Determination of feeding

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distributions gives indications concerning the limitation on angular momentum values leading to the formation of evaporation residues.

Recent progress in the identification of γ rays from weakly produced nuclei has led to a rapid increase in knowledge about the structure of very neutron-deficient nuclei [10]. The Recoil Decay Tagging (RDT) method [11, 12] has made it possible to perform in-beam γ -ray experiments on nuclei which have production cross sections on the order of $1 \mu\text{b}$ or even less [13]. The cross sections for neutron evaporation exit channels in fusion reactions with stable targets decrease rapidly when the proton number of the compound system increases. For example, the cross section for the reaction $^{208}\text{Pb}(^{40}\text{Ar}, 3\text{n})^{245}\text{Fm}$ has been measured to be (18 ± 5) nb [14]. The corresponding production rate is too small for in-beam studies using presently available techniques. However, the use of two doubly magic nuclei as reaction partners in the cold fusion reaction $^{208}\text{Pb}(^{48}\text{Ca}, 2\text{n})^{254}\text{No}$ leads to the anomalously high production cross section of $\sim 2 \mu\text{b}$ [15] and provides a unique opportunity for an in-beam experiment on a trans-fermium nuclide.

Very recently, the RDT method was used in conjunction with this reaction in two laboratories to detect γ rays from the $Z = 102$ nucleus ^{254}No . At Argonne National Laboratory (ANL), five E2 transitions in the ground state band were observed [16] using the Gammasphere Ge detector array and the Fragment Mass Analyzer (FMA). In the present work, performed at the Department of Physics, University of Jyväskylä (JYFL), this result has been confirmed and the ground state band has been extended by one transition. In the above mentioned experiments, the predicted ground state deformation of ^{254}No was also confirmed.

2 Expected properties of excited states of nuclides around ^{254}No

The quadrupole deformation of even-even nuclei in the vicinity of ^{254}No is such that their excited states are characterised by rotational bands with first-excited 2^+ energies of around 45 keV [17]. The 2_1^+ excitation energy has been measured for $^{248,254,256}\text{Fm}$ on the basis of α - or β decay studies [17]. Until recently, no data were known for No isotopes but there is an estimate of (50 ± 7) keV for the 2_1^+ energy of ^{256}Rf from α decay of ^{260}Sg [18]. On theoretical grounds, no sudden changes in structure are expected for ^{254}No . There is, however, a deformed neutron subshell closure ($N = 152$) at ^{254}No which has a dramatic effect on spontaneous fission half-lives [4]. All in all, it is important to determine the magnitude of the deformation experimentally and compare with theoretical predictions.

In recent calculations using the macroscopic-microscopic approach [19], the ground state moments of inertia of very heavy even-even nuclei were deduced. This allowed the determination of the excitation energy of the first rotational 2^+ state, E_{2^+} . Two minima were obtained for E_{2^+} , one (41.7 keV) for ^{254}No and the other (40.0 keV) for

^{270}Hs . These are related to minima of the neutron pairing-energy gap Δ_n at $N = 152$ and $N = 162$, respectively, since a small pairing correlation corresponds to a large moment of inertia [19].

One should note that the low energies of the few lowest-lying transitions in the ground state bands lead to high internal conversion coefficients [20], making their observation in an in-beam γ -ray study very difficult.

Finally, one should consider the possibility of the existence of K-isomers, since for these nuclei the proton and neutron Fermi surfaces lie in the vicinity of high- Ω orbitals. There is experimental evidence, with no isomeric transitions observed, for the existence of an isomeric state with a half-life of (280 ± 40) ms in ^{254}No [21]. This may be a $K^\pi = 8^-$ two-quasi-particle state [21]. The isomeric ratio, σ_m/σ_g in the reaction $^{12}\text{C} + ^{249}\text{Cf}$ was measured to be about 0.4.

3 Experimental details

The ^{48}Ca beam was produced using the ECR ion source and accelerated with the JYFL $K = 130$ MeV cyclotron. Self-supporting stationary targets of enriched ($\sim 99\%$) ^{208}Pb with thicknesses varying between 250 and 700 $\mu\text{g}/\text{cm}^2$ were used. The total irradiation time was 350 h. The beam intensity, which was limited by the maximum allowed counting rate of the Ge detectors surrounding the target (10 kHz), was 10 pA ($6 \times 10^{10} \text{ s}^{-1}$). The cyclotron beam was pulsed (3 ms on and 2 ms off) during the cross section determination (see below) while in the γ -ray study no pulsing was used. The consumption of the enriched ^{48}Ca material was 0.11 mg/h.

Fusion products were separated from the primary beam and unwanted reaction products by the RITU gas-filled recoil separator [22]. The pressure of the He filling gas was 1 mbar. The He gas was separated from the beam line vacuum by a carbon window of 20 mm diameter and 50 $\mu\text{g}/\text{cm}^2$ thickness. The cooling effect of the gas allowed the use of stationary targets; tests revealed that the targets still survived at a beam current of about 50 pA. The total efficiency for observing fusion products in the focal plane detector was estimated to be 25 %.

Prompt γ rays from the target were observed with the SARI array (Segmented Array at RITU). It consisted of four clover detectors positioned at 50 degrees relative to the beam direction. Three of the detectors were segmented. The detectors were operated without Compton suppression shields. The photo-peak efficiency of the array was 1.7 % at a target-to-detector distance of 15 cm. The detectors were operated in the add-back mode. In addition, a set-up of three mini-orange (MO) type electron spectrometers around the target was used [23]. Due to an intense low energy delta electron background the transmission window of the MO spectrometers was centered at around 700 keV to observe possible transitions from vibrational bands to the ground state band, and therefore was outside the region of the first two transitions of the rotational band in ^{254}No . The remaining background of low energy delta electrons was further suppressed by the

use of 11 μm thick Mylar foils between the target and the electron detectors.

The residues were implanted into a position sensitive 16-strip Si detector (PSSD) measuring 80 mm (horizontal) \times 35 mm (vertical) at the focal plane of RITU. Here the position, energy and time information for the fusion products and any subsequent α decays was recorded. For γ rays detected in the SARI array, energy and time information was recorded when in coincidence with signals in the PSSD. Energy and position signals from the PSSD were processed through two different amplifier branches. The low-gain energy range (recoil branch) was 10–200 MeV and the high-gain range (α branch) 0–12.5 MeV.

Germanium detectors were positioned adjacent to the focal plane PSSD in order to detect isomeric or delayed γ rays. Details of this part of the experimental setup will be discussed in a forthcoming publication.

To determine the optimum bombarding energy to be used in the experiment, the excitation function for the production of ^{254}No was measured using a target of 500 $\mu\text{g}/\text{cm}^2$ thickness and a pulsed cyclotron beam. The yield of ^{254}No α particles was measured at different bombarding energies, using Ni degrader foils to adjust the beam energy. The maximum yield was observed at a bombarding energy of 216 MeV, as measured halfway through the target. The cyclotron settings were then changed so that an energy of 216 MeV was obtained at target midpoint after passing the beam through the 50 $\mu\text{g}/\text{cm}^2$ carbon beam window. The yield of ^{254}No α particles was once again measured and found to be consistent with the maximum production rate observed. No beam energy adjustments were made when target thicknesses differing from 500 $\mu\text{g}/\text{cm}^2$ were used. This led to slightly less than optimum production rates for some targets. The observed rate of ^{254}No α particles in the PSSD and the cross section of (3.4 ± 0.3) μb [15] obtained by radiochemical methods allow us to extract the value of (15 ± 4) % for the total efficiency of the separator system. This is significantly less than the value of 25 % which is based on both experimental results and calculations. Taking this value, however, the production rate observed in the present work is in good agreement with the cross section values of ~ 2 μb measured using the velocity filter SHIP at GSI [15] and the VASSILISSA separator at Dubna [24].

4 Results

Gamma rays were assigned to ^{254}No using the recoil decay tagging (RDT) method [11,12]. Fusion products observed in the focal plane Si detector were identified by correlating them with their subsequent α decay events. Only those prompt γ rays detected in coincidence with the wanted fusion products were then accepted. For ^{254}No , the ground state to ground state decay α particles have an energy of (8093 ± 14) keV, and the α decay branching ratio is (90 ± 4) % [17]. The other observed decay modes are electron capture $((10 \pm 4)$ %) and spontaneous fission (0.25 %) [25]. Approximately 15–20 % of the α decays of ^{254}No are expected to feed the first excited 2^+ level at about 45 keV in

^{250}Fm . Since the decaying nuclei are imbedded in the Si detector, the energy of conversion electrons and other radiation depopulating the 2^+ level adds to the energy signal corresponding to the ground state to 2^+ state α particle transition [26]. As a consequence, only one α peak with an energy of 8093 keV is observed. In addition, there is about 45 % probability that the α particle escapes from the Si detector leaving only part of its energy in the crystal.

The previously measured half-life of ^{254}No , (55 ± 5) s [27] is rather long for RDT studies and sets strict limits on the count rate of evaporation residue-like and α particle-like events to avoid excessive accidental correlations. Figure 1a shows part of the recoil- γ TAC gated particle energy spectrum measured in the low-gain energy range. The energy window used in the correlation search for evaporation residues, corresponding to a nominal range (no pulse height defect correction) of 26–40 MeV, is shown in the figure. The average count rate of events classified as possible fusion products as determined by the energy condition only was 0.36 s^{-1} . The rate of real fusion products in the focal plane is determined mainly by the production of ^{254}No since charged particle evaporation is suppressed by the high Coulomb barrier and since the 1n and 2n excitation functions are narrow.

Figure 1b shows the singles particle energy spectrum in the high-gain energy range. The three main α peaks in the spectrum belong to ^{254}No and its decay products ^{250}Fm (half-life 30 min) and ^{246}Cf (36 h). The following procedure was followed to generate the spectrum in Fig. 1b. For each of the 16 individual strips, a plot of total energy vs vertical position was generated for the 8.09 MeV ^{254}No peak. Fits were then made to correct the energy signal for position dependence, and the resulting 16 individual α particle energy spectra were gain matched. The α particle energy calibration was based on the energies of ground state to ground state transitions from ^{246}Cf (6.750 MeV), ^{250}Fm (7.43 MeV) and ^{254}No . The full-width-at-half-maximum of the ^{254}No α peak was 24 keV.

For the recoil- α correlation analysis, α particle-like events in the energy range 8060–8120 keV were accepted. The average count rate within this window was 0.023 s^{-1} . In addition to ^{254}No α particles, primary beam particles scattered down to low energies are within the window. It is known from previous work using Pb targets at recoil separators (see e.g. [28]) that one of the primary multi-nucleon transfer product background activities is ^{213}Rn with an α particle energy of 8.09 MeV i.e. similar to that of ^{254}No . The short half-life of ^{213}Rn , 25 ms, and significantly different recoil kinetic energy make it easy to discriminate against this activity. No γ rays identified as belonging to ^{213}Rn were observed in the present work.

The number of accidental evaporation residue – α particle correlations depends on the effective granularity of the focal plane detector. The vertical position resolution was measured to be better than 0.5 mm (FWHM) on the basis of observed correlations. The nominal number of detector elements is thus rather large, on the order of 1000. The width of the vertical position distribution of true correlated recoil- α pairs was measured to be

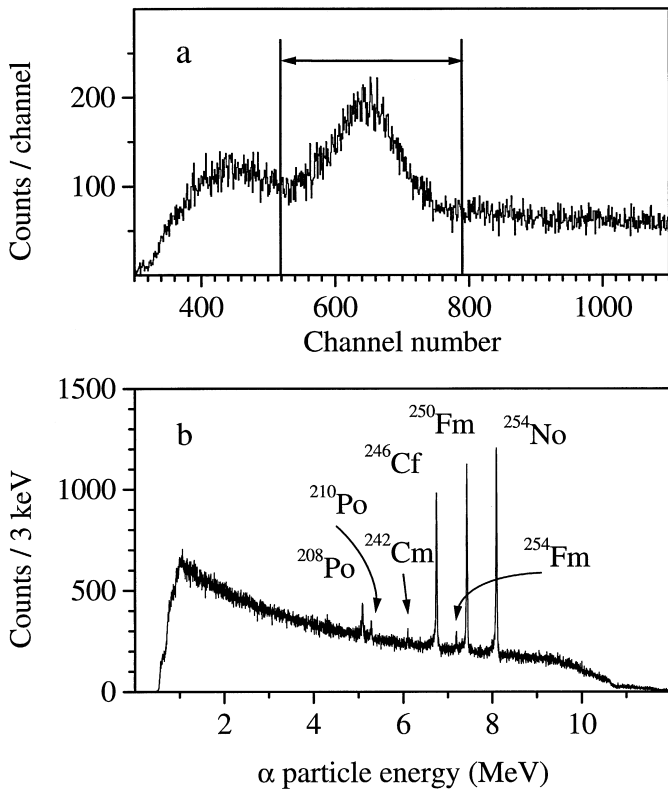


Fig. 1. (a) Particle energy spectrum measured in the focal plane PSSD in the low-gain range. A 100 ns wide gate has been used for the γ -recoil TAC to reduce the amount of background. The arrow marks the energy window used in determining the ^{254}No half-life and in the correlation search to generate the γ -ray spectra. (b) Particle energy spectrum in the high-gain (α) range. In addition to α peaks from ^{254}No and its decay products, peaks from the long-lived α activities $^{208,210}\text{Po}$ produced in an earlier experiment are visible in the spectrum. The presence of ^{254}Fm is due to the electron capture decays of ^{254}No and ^{254}Md

13 mm (FWHM). However, the background events which fall within the energy window for fusion products are almost evenly distributed over the detector surface. The effective granularity is thus roughly equal to half the nominal one, or ~ 500 . Taking into account the above mentioned average count rates of evaporation residue-like and α particle-like events and the granularity, one can estimate that the average time difference for accidental nucleus- α correlations is on the order of 1500 s.

In Fig. 2 we show the observed time difference distribution of evaporation residue – α particle pairs generated using the above energy conditions. The maximum allowed time and vertical position differences were 2500 s and ± 0.4 mm, respectively. One can see two components in the distribution [29]. There is a long-lived accidental component resulting from the following two cases: (i) the α particle-like event was a scattered beam particle, (ii) the nucleus from which the α particle was emitted was outside the position window or was lost due to dead time. The apparent half-life of this artificial component, ~ 1000 s, corresponds

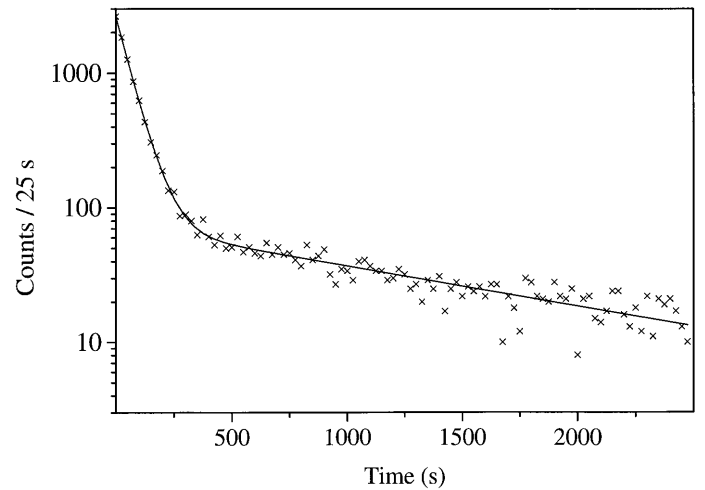


Fig. 2. The measured decay curve of ^{254}No . See text for the explanation of the two components

well with the above given average random time difference of about 1500 s. The apparent life time of the physical component corresponding to real nucleus- α correlations is distorted [29] according to the formula

$$\lambda_{obs} = \lambda + r$$

where λ_{obs} is the observed decay constant, λ is the real decay constant and r is the sum of the counting rates of acceptable recoil events and α events. By applying this correction to the present case a half-life of (48 ± 3) s is determined for ^{254}No . This is in slight disagreement with the previously published value of (55 ± 5) s [27]. Part of the discrepancy can be traced back to the time calibration used in [27] which was based on the half-life of ^{214}Ra . Taking into account the presently accepted value for this half-life, the corrected ^{254}No half-life from [27] is (52 ± 5) s. We conclude that the 8.10 MeV α activity observed in the present experiment is ^{254}No .

In Fig. 3a we show the γ -ray spectrum in coincidence with fusion products identified as ^{254}No nuclei on the basis of recoil- α correlations. The maximum search time used was 200 s. In addition, a 100 ns wide gate was placed in the γ -recoil TAC spectrum for acceptable fusion products. In Fig. 3b a spectrum is shown which consists of γ rays observed in coincidence with recoil events falling within the energy and γ -recoil TAC windows discussed above. The same set of γ -peaks is observed in both spectra. The peaks in the spectrum of Fig. 3b, generated using the recoil gating method, contain roughly three times as many counts as the RDT spectrum in Fig. 3a. This is in agreement with the estimated number of escape α particles which do not contribute to the RDT spectrum and the losses due to dead time of the data acquisition system.

In addition to No X-rays, γ -ray transitions having energies of 158.9, 214.1, 267.2, 318.2, 366.5, and 414.0 keV were observed and assigned to originate from ^{254}No . The γ -ray energies have an uncertainty of 0.3 keV except for the 414 keV line which has an uncertainty of 1.0 keV. The first five of these transitions were also observed in

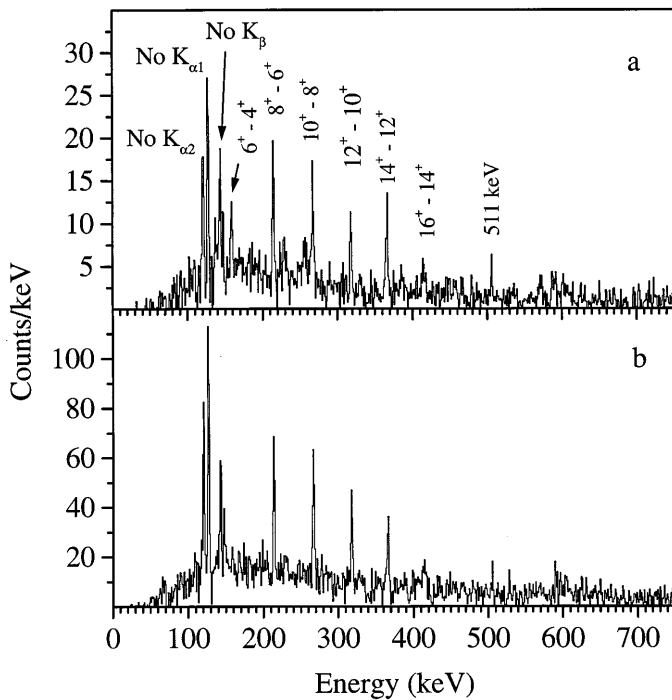


Fig. 3. (a) Spectrum of γ rays in coincidence with ^{254}No evaporation residues. (b) Spectrum of recoil-gated γ rays. The intensity of the 159 keV $6^+ \rightarrow 4^+$ transition in (b) is reduced due to a peak of similar energy in the subtracted background spectrum

the ANL experiment [16], and assigned to an E2 cascade in the ground state band of ^{254}No up to a 14^+ state. The assignment of the 366 keV $14^+ \rightarrow 12^+$ transition was tentative. Our data which contain more counts, are in accordance with these assignments. The six transitions can be explained most naturally by assuming that they form an E2 cascade in the ground state band of ^{254}No as follows. The kinematic moment of inertia $I^{(1)}$ can be expressed in terms of the Harris parameters I_0 and I_1 and the rotational frequency ω as

$$I^{(1)} = I_0 + I_1\omega^2.$$

A very good fit with a reduced $\chi^2 = 1.2$ results when the lowest energy transition of 158.9 keV is assigned as the $6^+ \rightarrow 4^+$ transition and so on up to the $16^+ \rightarrow 14^+$ transition with an energy of 414 keV (see also [16]). The values of the Harris parameters are $I_0 = 68.2 \text{ } \hbar^2/\text{MeV}$ and $I_1 = 162.4 \text{ } \hbar^4/\text{MeV}^3$ representing a moment of inertia which increases slightly with the rotational frequency. In Fig. 4 we show the proposed ground state band level scheme for ^{254}No based on our results.

The statistics were not sufficient for obtaining $\gamma\gamma$ -coincidences. However, it is of interest to note that the Gammasphere data clearly indicate that the 414 keV transition is in coincidence with other transitions in the ground state band [16].

The analysis of the conversion electron data and the focal plane Ge detector data is in progress.

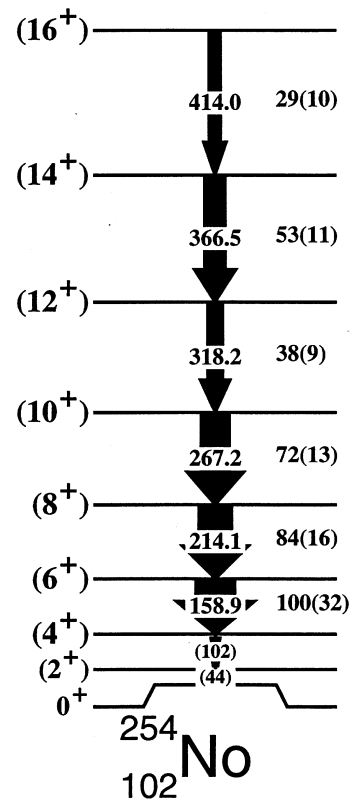


Fig. 4. The proposed ground state band level scheme of ^{254}No . The intensities are based on the RDT spectrum and include the calculated contribution from internal conversion for E2 transitions and have been normalised to 100 for the $6^+ \rightarrow 4^+$ transition

5 Discussion

We did not measure $B(E2)$ values from which the quadrupole deformation of ^{254}No could be determined. It is, however, possible to extract the ground state deformation parameter β_2 from the extrapolated energy of the 2_1^+ state using global systematics [30,31]. The extracted values of the Harris parameters can be used to estimate values of $(44.2 \pm 0.4) \text{ keV}$ and $(102.0 \pm 0.3) \text{ keV}$ for the energies of the $2^+ \rightarrow 0^+$ and the $4^+ \rightarrow 2^+$ transitions, respectively. The $2^+ \rightarrow 0^+$ transition energy yields a deformation parameter β_2 of 0.27 ± 0.03 . This result is in good agreement with values calculated using the macroscopic-microscopic method. Typical examples are $\beta_2 = 0.251$ [5], 0.252 [32], and 0.246 [6]. There is a difference between the treatment of pairing in [5] and [32], but the β_2 values do not differ for ^{254}No . The Harris parameters can also be used for deducing the expected transition energy for the $18^+ \rightarrow 16^+$ transition, 457 keV.

The nucleus ^{254}No survives fission at least up to spin $16\hbar$. The 16^+ state of the rotational band has an excitation energy of 1885 keV. In the present experiment the excitation energy of the compound nucleus ^{256}No , 21 MeV, was higher by $\sim 2.5 \text{ MeV}$ than in the ANL experiment [16] in which the multiplicity distribution suggested that residues were formed with spin values up to $\sim 18\hbar$. The

observed band structure and the regular decrease of the γ -ray intensities show no indication of the fission barrier causing a band termination or strong deviations from the rotational pattern.

There is no indication of a backbend in our data. Cranked Shell Model calculations were performed for ^{252}No and ^{254}No in order to ascertain whether quasi-particle alignment effects may be expected for these nuclei. The deformed Woods-Saxon potential [33] was used with "universal" parametrization [34]. Calculated deformation parameters for ^{252}No and ^{254}No were taken from [6]. The resulting quasi-nucleon Routhian plots for ^{254}No indicate strong neutron interactions at $\hbar\omega = 0.25$ MeV (corresponding to a transition energy of $E_\gamma = 500$ keV). For ^{252}No , neutron interactions are predicted at the lower frequency of 0.22 MeV ($E_\gamma = 440$ keV). These alignment effects should be evident as a backbend or upbend in a plot of spin vs transition energy for the ground state bands. Unfortunately, the highest-lying observed transition for ^{254}No in the present work is below 500 keV. An additional experiment on ^{252}No may provide a better opportunity to observe alignment effects in these nuclei. Such a study would also partly answer the question whether the 2_1^+ energy has a local minimum at $N = 152$ [5, 19].

There are statistically significant γ -peaks in the energy range ~ 570 – 600 keV in the recoil decay tagged spectrum (Fig. 3a) which can also be seen in the Gammasphere data. These transitions may be the links between the ground state band and excited vibrational bands. Such bands have been observed in several nuclei in this region, e.g. ^{256}Fm [35].

In addition to the study of even-even nuclei, it is important to learn about the single-particle states in the future. Two difficulties are then encountered. The cross sections to produce odd-mass isotopes in the vicinity of ^{254}No are at most on the order of a few hundred nb [15], and only part of the γ -ray cascades are expected to contribute to the main quasi-particle band. In the neutron case, deformed sub-shells are predicted at $N = 152$ and $N = 162$, separated by a region with a very high single-particle level density. Consequently, the $1/2^+$ ground state of ^{255}No is for the moment only tentatively established. In the proton case, a deformed sub-shell is predicted at $Z = 100$ [5, 32], followed by the orbitals $1/2^-$ [521], $7/2^-$ [514], and $9/2^+$ [624], but so far no spin assignment has been made for the ^{255}Lr ground state. The reasonably high cross section for the production of ^{255}Lr in the reaction $^{209}\text{Bi}(^{48}\text{Ca}, 2n)$, ~ 400 nb [15], with a unique α decay energy in this region and a relatively short half-life (~ 22 s) render this nuclide the most attractive goal for future studies.

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