ON THE NUCLEAR STRUCTURE OF ^{229}Ra

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ABSTRACT

The nucleus ^{229}Ra has been studied via β^- -decay of ^{229}Fr , the half-life of which was measured to be (50.2 ± 0.4) s. A level scheme with seven excited levels has been constructed. In the Nilsson classification the ground state is assumed to be $5/2^+[633\downarrow]$, levels at 142.8 keV and 213.0 keV are described as $1/2^+[631\downarrow]$ and $3/2^+[631\uparrow]$ and levels at 137.6 keV and 479.2 keV as $3/2^-[761\uparrow]$ and $1/2^-[501\downarrow]$. The level structure of ^{229}Ra is similar to the structure in the lighter Ra-isotopes, which have been described as octupole deformed nuclei.

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1 Introduction

The nucleus ^{229}Ra is situated at the border of a mass region where experimental and theoretical evidence for the existence of reflection-asymmetric nuclear shapes has been found [1, 2]. Important signatures of the octupole deformation [2, 3] are close-lying states of equal spin and opposite parity which are connected by fast E1 transitions and ‘anomalous’ decoupling parameters for $K = 1/2$ bands. Such evidence has been found for nuclei around mass number 225 and the present work is part of an investigation to explore how far in mass number the region of octupole deformation extends.

Previously, only the ground state properties of the ^{229}Ra nucleus were known [4]; its ground state nuclear spin and dipole and quadrupole moments had been determined from collinear laser spectroscopy [5] to be $I = 5/2$, $\mu_I = 0.51$ n.m. and $Q_s = 2.9$ b, the latter numbers given with 5% and 15% estimated errors, respectively. The aim of the present study on the β^- -decay of ^{229}Fr was to provide information about the excited states of ^{229}Ra in order to shed light on the nuclear structure of ^{229}Ra .

For the ^{229}Fr parent nucleus the half-life had been previously determined [6, 7] to be (48 ± 2) s while no information was available on its ground state properties. This is an illustration of the scarcity of data for nuclei in the octupole-quadrupole transition region and is a strong motivation for experimental investigations around mass number 229.

2 Experimental procedure and results

Francium isotopes were produced in a UC_2 target by spallation reactions induced by 600 MeV protons from the CERN synchro-cyclotron. After surface ionization the nuclide ^{229}Fr was obtained as a mass-separated beam from the ISOLDE-2 on-line separator [8]. The activity was deposited on the aluminium-coated Mylar tape of a tape transport system working in a start-stop mode to collect ^{229}Fr -sources and carry them in front of the detector system. The system consisted of a mini-orange type electron spectrometer and two high-purity Ge gamma detectors permitting γe^- and $\gamma\gamma$ coincidences to be recorded simultaneously. The physical arrangement of the apparatus was the same as in ref. [9].

The detector used in the electron spectrometer was a 300 mm² Si(Li) crystal with a thickness of 2 mm, cooled to liquid-nitrogen temperature by means of a copper cold finger. An absorber block placed between the source and the detector shielded the latter from direct X-rays and γ -rays. A set of permanent magnets positioned around the spectrometer axis deflected electrons from the source to the detector. The magnet configuration was chosen to give an approximately uniform efficiency ($\sim 0.5\%$) over a broad range of energies [9].

The vacuum chamber of the electron spectrometer had a thin Kapton window to minimize absorption of low-energy photons which were detected by the γ -detectors. These detectors had active volumes of 68 cm³ and 88 cm³, typical resolutions (FWHM) of 0.8 keV at 122 keV and 1.8 keV at 1.3 MeV and the smaller one had a thin entrance window to enable detection of low-energy photons. Energy and efficiency calibrations for these detectors were performed with ^{133}Ba , ^{152}Eu , ^{207}Bi and ^{241}Am sources in addition to a commercially obtained, absolutely-calibrated reference source of different radionuclides.

Coincident events between any pair of the three detectors were selected by standard hardware circuitry and the information from the time-to-amplitude and analog-to-digital converters was stored on magnetic tape in event-recorded mode. Collection time was usually 40s and measuring time 50s, but for one measuring period a collection time of 50s and a measuring time of 100s were used to enhance the daughter products activities in the sources.

The decay of the ^{229}Fr activity as well as the growth and decay of the daughter activities were also observed in a multi-spectrum analysis mode using cycles of 50s collection time followed by 10×10 s measuring time. Some examples of the decay-curves are displayed in fig. 1. From the error weighted average of the 14 most prominent lines in the γ spectrum, which includes some of the Ra KX-rays, a half-life of $T_{1/2}=(50.2\pm 0.4)\text{s}$ for ^{229}Fr was obtained. This is in good agreement with the previous [7] value of $(48\pm 2)\text{s}$.

2.1 Results of the γ -ray measurements

An example of a singles γ -spectrum is displayed in fig. 2. The spectrum shows a number of lines from the background activities of ^{152}Eu and ^{232}Th as well as lines following β^- decay of the daughter nucleus ^{229}Ra .

The stronger γ -lines corresponding to transitions in ^{229}Ra were identified from direct measurements of their half-lives. Otherwise, the identification of γ -transitions in ^{229}Ra is based on comparisons to spectra with longer measuring times (which have enhanced daughter product activity) and on observed coincidences with established lines. Energies and intensities of the γ -lines assigned to the decay of ^{229}Fr are listed in table 1.

Only the strongest transitions could be studied in the coincidence measurements due to the limited number of events acquired in the experiment. In particular the low energy transitions easily escape detection due to their large conversion and the γ -coincident e^- -spectra do neither have high enough statistics nor the necessary resolution to remedy the problem. Examples of the γe^- coincidence spectra are shown in fig. 4. A summary of the coincidence studies is given in table 2.

Figure 1: Decay curves of some Ra KX-rays and some strong γ -lines assigned to the ^{229}Fr decay. The statistical errors are mostly smaller than the size of the data points.

Figure 2: Singles γ -spectrum following ^{229}Fr decay

Table 1: Energies and intensities of γ -rays in ^{229}Ra

Transition energy E ΔE (keV)		Relative intensity I ΔI		Dominant multipolarity	Placement in level scheme
26.2	0.1	9	2	M1	(168.9→142.8)
44.3	0.1	5	1	E2	(213.0→168.9)
61.6	0.1	17	2	M1	
65.6	0.1	66	2	M1	
96.1	0.1	15	1		
107.0	0.1	37	2	M1	
137.6	0.1	138	3	E1	137.6→0.0
142.8	0.1	577	6	E2	142.8→0.0
168.9	0.1	52	2	M1	168.9→0.0
213.0	0.1	39	2	M1	213.0→0.0
217.7	0.1	9	2	M1	
266.1	0.1	126	3	E1	479.2→213.0
274.2	0.1	9	2	M1	
305.6	0.1	8	2		518.5→213.0
310.3	0.1	$\cong 1000$		E1	479.2→168.9
336.4	0.1	664	8	E1	479.2→142.8
341.7	0.1	15	2	M1	479.2→137.6
349.6	0.1	200	4	E1	518.5→168.9
352.8	0.1	10	2		569.5→213.0
358.9	0.1	38	2		
372.9	0.1	13	6		
375.7	0.1	95	4	(M1)	518.5→142.8
399.0	0.1	8	2		
404.3	0.1	8	2		
413.7	0.1	9	2		
428.4	0.1	26	3	M1	565.9→137.6
453.1	0.2	9	4		
455.9	0.1	12	3	(E1)	
484.4	0.1	10	2		
486.2	0.1	10	2		
491.4	0.4	8	2		
522.7	0.1	14	4		
525.8	0.2	14	4		
563.1	0.1	19	3		
569.8	0.1	28	3		
573.9	0.2	10	4		
587.4	0.1	15	3		
589.8	0.2	16	3		
598.6	0.3	7	3		
607.1	0.3	3	2		
611.3	0.3	10	4		
615.1	0.2	11	3		
626.3	0.1	15	3		
757.2	0.1	16	4		
1256.0	1.0	114	30		(1398.8→142.8)

Table 2: Results of $\gamma\gamma$ -coincidence studies in ^{229}Ra

Gated transition (keV)	Energies (keV) of coincident γ -transitions
61.6	<u>310.3</u>
65.6	<u>310.3</u> , <u>336.4</u>
107.0	<u>310.3</u>
137.6	341.7, 404.3, <u>428.4</u>
142.8	(266.1), <u>310.3</u> , <u>336.4</u> , 349.6, 358.9, 375.7, 1256.0
168.9	<u>310.3</u>
213.0	266.1
266.1	142.8, 213.0
310.3	61.6, <u>65.6</u> , 107.0, <u>142.8</u> , 168.9
336.4	65.6, (107.0), <u>142.8</u>
349.6	65.6, (107.0), <u>142.8</u>
358.9	142.8
375.7	<u>142.8</u>
428.4	<u>137.6</u>
1256.0	142.8

Strong lines in the coincidence spectra are underlined
 Parentheses indicate weak lines

2.2 Internal conversion coefficients

An electron singles spectrum corresponding to a total measuring time of ~ 1 hour is shown in fig. 3. The experimental resolution is ~ 2.6 keV (FWHM). Simultaneously, singles γ -ray spectra were recorded in order to determine the internal conversion coefficients. Areas of the lines were obtained from standard peak fitting routines and the efficiency corrections were the same as in ref. [9]. The resulting values for the conversion coefficients are listed

Figure 3: A singles electron spectrum from the decay of ^{229}Fr to ^{229}Ra

in table 3 with their statistical errors. The dominant multiplicities given in table 3 were obtained by comparison to theoretical values for E1, E2 and M1 transitions calculated by Rösler et al. [10]. For the strong 137.6 keV γ -line no conversion lines were detected above background in the electron spectra. The transition was therefore given an obvious E1 assignment.

3 Experimental results and decay scheme

The ground state spin of ^{229}Fr has not been experimentally determined so far, while the spin sequence 5/2, 3/2, 3/2, 1/2 has been measured [11] for the ground states of $^{221,223,225,227}\text{Fr}$. Both the ground state spin and the magnetic dipole moments for these nuclei are described [2] by the reflection-asymmetric rotor model. The main factor for the level ordering in the model is the quadrupole deformation parameter which increases with

Table 3: Experimental conversion coefficients for transitions in ^{229}Ra

Transition energy (keV)	Conversion shell					Dominant multi- polarity
	K	L_{1+2}	L_3	M	N	
26.2				<160(36)		M1
44.3		141(35)	<65(16)	<71(2)		E2
61.6		17(2)	<28(4)	2.5(4)		M1
65.7		<6.9(1)	0.32(5)	1.7(1)		M1
107.0		1.64(8)	0.15(6)	0.40(15)		M1
142.8	<0.6(1)	0.66(1)	0.37(5)	0.27(1)	0.09(1)	E2
168.9	2.7(2)	0.34(4)	0.07(3)	0.12(3)	0.09(3)	M1
213.0	1.15(12)	0.26(4)		0.09(2)		M1
217.7	0.5(2)					M1
266.1	0.034(12)					E1
274.2	0.6(3)					M1
310.3	0.020(2)	<0.0005(1)				E1
336.4	0.017(2)	0.005(2)				E1
341.7	0.25(9)					M1
349.6	0.036(7)	<0.009(1)				E1
352.8	<1.4(3)					
358.9	0.05(4)					
375.7	<0.08(2)	0.03(1)		<0.035(14)		(M1)
404.3	0.6(2)					
428.4	0.14(4)	0.08(2)				M1
455.9	0.026(11)					(E1)

neutron number. Unless the quadrupole moment of ^{229}Fr should be significantly smaller than for ^{227}Fr and the octupole deformation significantly larger, both events being rather unlikely, the ground state spins of the two are expected to be the same. Because of the reflection asymmetry the parity could be of either sign, but the main component is likely to be the $1/2^+[400\uparrow]$ Nilsson state. Based on this assumption the direct population of the ^{229}Ra ground state is expected to be very weak and the most prominent β^- -transition would be to the state with the largest $1/2^-[501\downarrow]$ Nilsson component.

Figure 4: Coincident e^- spectra gated on the 310.3 and 336.4 keV γ transitions.

3.1 Level scheme of ^{229}Ra

The ground state spin of ^{229}Ra has been previously measured [5] to be $5/2$ and in the construction of the level scheme a $5/2^+$ assignment to the ground state has been assumed in accordance with the model prediction [2]. Also the theoretical magnetic dipole and electric quadrupole moments of $0.63\mu_N$ and 2.88 b agree reasonably well with the measured values [5] of $(0.51\pm 0.03)\mu_N$ and for the quadrupole moment of $(2.9\pm 0.4)\text{ b}$ and [12] $(3.1\pm 0.2)\text{ b}$. In addition the spin and parity for the ground state of the isotone ^{231}Th is [13, 14, 15] also $5/2^+$. The parities given in the level scheme (fig. 5) are relative to the assumed positive parity for the ground state.

The ground state level: The lowest level observed in the experiment is assumed to be the ground state. It is fed by the four transitions 137.6 keV, 142.8 keV, 168.9 and

Figure 5: Level scheme of ^{229}Ra with energies in keV

213.0 keV which are positioned via coincidence spectra. As discussed above it is assumed to have a $5/2^+$ assignment.

The level at 137.6 keV: Because the 137.6 keV transition has only been observed in coincidence with transitions of higher energies and of considerably smaller intensities, it is assumed to connect a level at 137.6 keV with the ground state. The E1 character of the transition assigns the opposite parity of the ground state and a spin of $3/2$, $5/2$ or $7/2$ to the level. The strong direct feeding of this level in the β^- -decay of ^{229}Fr virtually excludes the two highest of these spins, however. A $3/2^-$ assignment is therefore given to the 137.6 keV level with a high degree of confidence.

The level at 142.8 keV: Taking into account the conversion factor, which is calculated to be 2.17 and is experimentally in the range 1.4 – 2.0, the 142.8 keV E2 transition is the strongest in the spectrum. It is coincident with a number of other strong transitions with higher energies and is assumed to decay directly to the ground state, thereby defining a level at 142.8 keV. The spin of this level is limited to $1/2$ or $3/2$ by the feeding of the E1 transition from the $1/2^-$ state at 479.2 keV and the seemingly pure E2 character of the 142.8 keV transition to the ground state makes a $1/2^+$ assignment to this level most likely.

The level at 168.9 keV: From the coincidences, the 168.9 keV M1 transition and the 310.3 keV E1 transition define this level. The highly converted 26.2 keV M1 transition is assumed to connect this level to the 142.8 keV level thus establishing the intensity balance. From the assumed spins of the ground state and 479.2 keV levels, the 168.9 keV level is assigned $3/2^+$.

The level at 213.0 keV: This level is defined by the coincident pair of transitions of energy 213.0 keV and 266.1 keV and of M1 and E1 multipolarity. The latter is also coincident with the 142.8 keV transition and is therefore the transition feeding the 213.0 keV level. From the $1/2$ spin of the 479.2 keV level and $5/2^+$ assignment of the ground state, the 213.0 keV level is found to have $3/2^+$.

To account for a slight discrepancy in the intensity balance, the 44.3 keV E2 transition is assumed to depopulate the 213.0 keV level. Its placement in the scheme is based on energy fit only, however, as it was too weak to be observed in the coincidence spectra.

The level at 479.2 keV: Three strong E1 transitions at 266.1 keV, 310.3 keV and 336.4 keV depopulate this level and determine its parity to be the opposite of that for the ground state. From energy considerations the weaker 341.7 keV M1 transition is assumed to connect the 479.2 keV level with the 137.6 keV level. No γ -rays are found to populate the 479.2 keV level implying it to have, by far, the strongest direct population in the β^- decay of ^{229}Fr . As already discussed, the ground state of ^{229}Fr is assumed to have spin $1/2^+$, and therefore the 479.2 keV level is given a $1/2^-$ assignment. In the β^- -decay of the isotope ^{231}Ac , which also has spin $1/2^+$, 93% of the decays [14] proceed to a $1/2^-$ level at 554 keV in ^{231}Th , similar to the present case.

The level at 518.5 keV: The relatively strong 349.6 keV E1 and 375.7 keV (M1) transitions and from energy fit also the 305.6 keV transition, depopulate the 518.5 keV level. The E1 character of the 349.6 keV transition assigns negative parity to the level, in contradiction to the (M1) assignment of the 375.7 keV transition. We have not been able to resolve this problem on basis of our data, but the upper limit of 0.009 for the L_1+L_2 conversion coefficient of the 349.6 keV transition is only in agreement with E1 multipolarity. A negative parity is therefore assumed with the most probable assignment being $1/2^{(-)}$ or a $3/2^{(-)}$.

As for the 479.2 keV level no γ -ray feeding is found and therefore a relatively strong direct population in β^- decay is observed. In the β^- -decay of the isotope ^{231}Ac the population pattern for the 554.4 keV and 593.5 keV levels [14] in ^{231}Th is similar to the 479.2 keV and 518.5 keV levels in the present case. In analogy with ^{231}Th we tentatively assign $3/2^{(-)}$ to the 518.5 keV level.

If the 479.2 keV and 518.5 keV levels are related a 39.3 keV transition might be

expected, which would escape detection in the γ -spectrum because of the strong X-ray line at 39.9 keV from the ^{152}Eu background. Some strength, however, is found which could be due to a 39.3M line in the electron spectra coincident with the 310.3 and 336.4 keV γ -gates.

The level at 565.9 keV: This level is defined by the 428.4 keV M1 transition to the 137.6 keV level, which restricts the level spin to $1/2$, $3/2$ or $5/2$ with negative parity. Unfortunately, the 352.8 keV line was too weak to be recorded in the coincidence spectra, but energy considerations make this transition a candidate for the $565.9 \rightarrow 213.0$ keV transition.

The level at 1398.8 keV: The coincidence of the 142.8 keV and 1256.0 keV γ -transitions defines this level of which the parity and spin remain undetermined. It is included in the list mainly because the 1256.0 keV transition is relatively strong (table 1) and therefore indicates strong β^- -feeding also to higher-lying levels in ^{229}Ra .

Other levels: Because of low statistics in the coincidence spectra the 61.6, 65.7 and 107.0 keV transitions could not be placed in the level scheme with a high degree of confidence. Of these, the 65.7 keV transition is probably defining a level at 544.8 keV with spin $1/2^-$ or $3/2^-$. Its decay to the 479.2 keV level would make it an alternative to the 518.5 keV level as the $3/2^-$ member of the $K^\pi = 1/2^-$ band but would require a larger rotational parameter for ^{229}Ra than for the isotone ^{231}Th .

The present data suggest a level at 107.0 keV with the 61.6 keV transition feeding it from the 168.9 keV level. The ordering is determined from the coincidence of the 107.0 keV transition with the 310.3, 336.4 and 349.6 keV transitions while the 61.6 keV transition is only coincident with the first one. The 61.6 and 107.0 keV transitions have about the same total intensity; there would be no significant β -feeding to a level at 107.0 keV.

4 Discussion

It is generally believed that ^{229}Ra is at the borderline of or outside of the mass region where static octupole deformation occurs, but in a recent reference [3] it is suggested that ^{229}Ra is in fact octupole shaped. The main reason for this is the disappearance of the reversed odd-even staggering [5] of the mean square charge radius for ^{229}Ra . For the isotone ^{231}Th it was concluded in ref. [15] that no evidence for an octupole deformation was found, while the authors of ref. [3] state that the positive sign for the decoupling parameter of the $[631\frac{1}{2}]$ band is a sign of a dynamical octupole effect.

When the levels observed in ^{229}Ra are compared to the corresponding levels in the isotopes $^{225,227}\text{Ra}$ and the isotone ^{231}Th (see fig. 6), no sudden changes are seen in the excitations, with a possible exception for the lowest negative parity state.

In particular the energy difference between the $1/2$ and $3/2$ members of the $[631\frac{1}{2}]$ -like rotational band decreases with increasing mass number in the Ra-isotopes and is the same for the isotones ^{229}Ra and ^{231}Th . Although the $5/2$ member of the band was not populated in the present experiment, the decoupling parameter may be assumed to be very nearly the same for the two latter nuclei, i. e. positive. For mass numbers ≥ 233 the decoupling parameter is negative [3] as expected for a purely quadrupole deformed nucleus. Even the levels of ^{229}Ra therefore seem to be influenced by the octupole driving force and the spin $1/2$ states at 142.8 and 479.2 keV may possibly be considered as a parity doublet with a somewhat large energy splitting.

A low-lying $3/2^-$ state has so far escaped detection in ^{231}Th and in the present experiment no low-lying $5/2^-$ state has been found. Whether this is due to a different position of the Fermi level or a possible difference in the influence of octupole forces cannot be judged from the present data, but the energy of the $3/2^-$ state follows the same smooth trend through the Ra isotopes as the $1/2^+$ state.

Figure 6: Comparison of levels in ^{229}Ra with selected levels in the isotopes $^{225,227}\text{Ra}$ and in the isotone ^{231}Th

5 Summary and conclusions

In the present study the half-life of ^{229}Fr has been measured to be $(50.2 \pm 0.4)\text{s}$ and for the first time a level scheme for ^{229}Ra has been constructed. The levels observed seem closely related both to levels in the octupole deformed nucleus ^{225}Ra and to levels in the isotone ^{231}Th . The decoupling parameter of the $[631\downarrow]$ -like Nilsson band seems to be positive in ^{229}Ra as in the other Ra-isotopes and in ^{231}Th , which may be taken as evidence for dynamic and/or static octupole deformations in these nuclei. More extensive level schemes and measurements of life-times in ^{229}Ra are required to draw firm conclusions on its type of deformation, but effects of the octupole force seem to be present also in this nucleus.

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