PHYSICAL REVIEW C VOLUME 52, NUMBER 1 JULY 1995

^{223}Ra nuclear spectroscopy in ^{14}C cluster radioactivity

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(Received 26 January 1995)

The energy spectrum of ^{14}C ions emitted from ^{223}Ra sources implanted at ISOLDE has been measured with the spectrometer SOLENO. The highest statistics (899 events) and the best energy resolution (90 keV) obtained so far in cluster decay allow a real spectroscopic study. Hindrance factors for transitions to the ground state and first excited state in ^{209}Pb, and limits for the next three low-lying states, have been determined. The data can be explained by a parent wave function containing mainly an $i_{11/2}$ spherical component.

PACS number(s): 23.70.+j, 27.90.+b

Since the discovery of ^{14}C spontaneous emission from ^{223}Ra [1], confirming earlier theoretical predictions [2], a large number of C, O, F, Ne, Mg, and Si radioactivities have been detected. The measured half-lives [3, 4] are in good agreement with predicted values within the analytical superasymmetric fission (ASAF) model, improved since 1980 (see [5, 6] and references therein). Various models, extending either a fission theory or the traditional α-decay one ([7, 8] and others mentioned in [6]), have also contributed to the understanding of the decay mechanism.

A new generation of experiments started with the discovery [9] of a fine structure in the energy spectrum of ^{14}C ions emitted by ^{223}Ra. The fine structure results [9–11] obtained with the magnetic spectrometer SOLENO raised the question of using such experiments to determine spectroscopic factors defined by the nuclear structure models. The controversy on the population of the $15/2^-$ state of ^{209}Pb in ^{14}C decay of ^{223}Ra, either in measurements [10, 11] or in theoretical interpretations [12, 13], reinforced the need of an immediate answer. In the present paper we describe a new measurement of the fine structure in ^{14}C emission from ^{223}Ra with a high precision, yielding meaningful spectroscopic factors. Our choice for this decay relies on additional experimental and theoretical grounds. The highest probability of ^{14}C emission from odd-mass nuclei occurs in ^{223}Ra, allowing the use of a selective magnetic spectrometer. Theoretically, it gives the opportunity to study the nuclear structure of the deformed parent, because the quasispherical daughter, ^{209}Pb, possesses well-known low-lying excited levels.

The high-quality source, requested by the fine-structure experiments, was made by implanting at ISOLDE ^{223}Ra ions into a C catcher. The previously used chemically separated radioactive sources had a severe drawback in measuring spectroscopic factors: there was a low-energy tailing which made it difficult to measure small size peaks which fell on the tail of larger ones. Moreover, spurious peaks appeared on the low-energy side of pronounced peaks. The ^{223}Ra source was produced at the ISOLDE mass-separator GPS on line to the 1 GeV proton beam of the PS-Booster synchrotron. A ThC [14] target (55 g/cm^2 thick) was bombarded with a beam of up to 2.4×10^{13} protons/pulse at an average repetition period of 2.4 s. The strong ^{223}Ra source was made by implantation, during two days, of the ^{223}Ra + ^{223}Fr singly charged ions of 60 keV, on a vitreous carbon $13\times15\times2$ mm catcher plate. The beam spot of 2 mm width was moved on the surface of the catcher by means of the steering quadrupoles, in order to avoid sputtering losses. Nine equidistant implantation locations were chosen: one on the center and eight on the circumference of a circle of radius 2 mm. A second source of much lower activity (1/1000), was produced in similar conditions for calibration. A diaphragm was placed on each source, with an aperture of ϕ=8 mm which corresponded to the implantation area. After tranportation to Orsay, a control measurement was performed by arranging a source and a Si detector in direct view of each other inside a vacuum chamber at a large distance ($\approx$2 m). Also, a diaphragm of small diameter (ϕ=5.5 mm) was placed on the detector surface, so that a moderate α-counting rate was obtained. The activity of the strong ^{223}Ra source was found to be 211 MBq (5.70 mCi) at the end of the irradiation and the contamination by ^{224}Ra was less than 1/1000. First measurements with SOLENO showed a strong emanation of Rn from the source. A 1000 Å of Al was then deposited on its surface by evaporation; it was sufficient to stop the emanation of Rn without altering the detection energy resolution. As a bonus, the deposit of Al together with the implantation depth in

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0556-2813/95/52(1)/267(4)/$06.00

carbon catcher of $\approx$300 Å represent a total thickness of $\approx 33\ \mu\mathrm{g/cm^2}$ to be crossed by ^{14}C ions emerging from the source, enough to attain the charge state equilibrium [15]. According to the standard setup of the spectrometer, the source and the Si detector (of 450 mm^2 with a ϕ=22 mm diaphragm) were placed on the axis of symmetry at the opposite extremities. We have used a detector with an energy resolution of 15 keV for the α ray of ^{241}Am. A good vacuum ($\approx 10^{-7}$ Torr) was maintained during the measurements.

The control of the spectrometer was established by measuring the transmission curve. It is the effective solid angle Ω at the entrance versus the parameter $y = B\rho/I$, where $B\rho$ is the magnetic rigidity of the ions and I the electric current in SOLENO. This curve was obtained by counting, for several successive values of the current I, the number of 5.717 MeV α particles from the low-activity calibrated ^{223}Ra source. The resulting curve is identical to the one previously obtained [11]. For ^{14}C measurements, the electric current of the spectrometer was set to focus on the detector the fully stripped ^{14}C^{6+} ions from the source. As the energy of these ions is different when they feed the ground state of ^{209}Pb or its excited states, two values of the current (278 and 281 A) were alternately used. Each of them focuses ^{14}C ions corresponding to the ground state of ^{209}Pb and its four low-lying excited states on top of the relatively flat transmission curve. The goal in the choice of the current values was that the location of the $15/2^-$ state for I = 278 A would coincide with the location of the $11/2^+$ state for the I = 281 A value. In this way a straightforward comparison between the populations of the two states is possible.

Data were acquired for two weeks. At the beginning, there was a total number of $N = 2.24\times10^{14}$ nuclei of ^{223}Ra present in the source. A number of $0.4511\times N$ nuclei decayed during the measurement for the current of SOLENO set to I = 278 A value and a number of $0.1324\times N$ for the current set to the I = 281 A value. The counting rate of $\approx$60 c/s was due to α radiation emitted from Rn and its daughters. The neutral atoms of this gas, emanated by the source, diffuse towards the detector. Advantage was taken to use the detected α particles to monitor the detector gain variation in time. In the off-line analysis of the event-by-event data, the gain corresponding to each detected ^{14}C ion was monitored by the channel position of the 6.623 MeV α ray of ^{211}Bi. The present results and the previously obtained ones [9] are plotted in Fig. 1. The energy calibration as extrapolated from known α rays was used to recognize the approximate location of the ^{14}C clusters. The energy resolution (full width at half maximum) in ^{14}C detection is 90 keV to be compared to the value of 250 keV in [9]. The peaks have no tail towards low energies and there is no background. A total number of 899 ^{14}C were detected: 130 events for the transition to the ground state of ^{209}Pb, and 768 events for that leading to the first excited state. No event was detected at the location of the second or third excited states. One solitary event has fallen at the position of the $1/2^+$ state. In comparison with the first experiment on the fine structure [9] in ^{14}C radioactivity (see Fig. 1) the number of events is now more than twice as many. The agreement in the population of the ground state (g.s.) and first excited state is clear, but all the events falling at higher excited states in [9] are obviously due to low-energy tailing in the source. The peak found in [10] at the location of the $15/2^-$ state is spurious, probably originating in the inhomogeneity of the source.

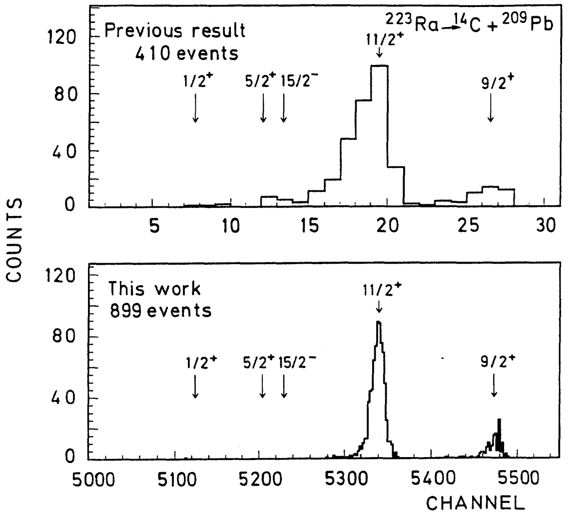

FIG. 1. ^{14}C kinetic energy spectrum obtained in the present work (bottom), compared to the previous [9] one (top). Note the considerable improvement of the resolution. The positions of excited states of the daughter nucleus ^{209}Pb are shown with arrows. The vertical scales are in counts per channel.

Much investigation has been done to identify the origin of the solitary event in the present experiment. It could not be a ^{14}C ion emitted from the ^{224}Ra contained in the source, since it does not lay at the corresponding peak position (channel 5250). The possibility that it would originate from α multiple pileup was examined. We have analyzed the α spectrum cumulated during the whole experiment (Fig. 2). The energy position of the event

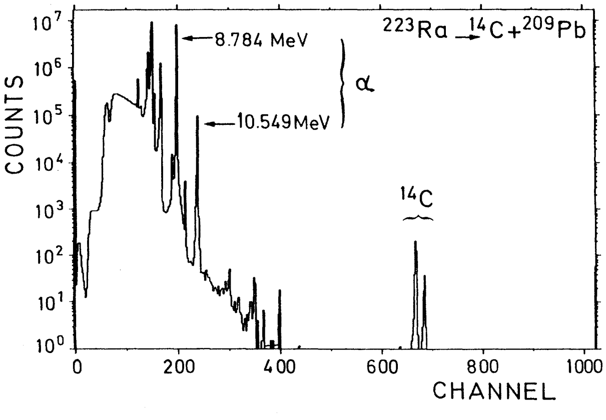

FIG. 2. α particle and ^{14}C energy spectra, cumulated during the whole experiment. The 8.784 and 10.549 α lines are emitted by ^{212}Po, which is a member of the ^{224}Ra decay chain.

restricts its origin to a triple pileup among two α of 8.784 MeV and one long-range α of 10.549 MeV, emitted by ^{212}Po. By taking into account the counting balance in random coincidences, such a triple pileup event is 10^5–10^6 times less probable. Hence this origin is to be excluded. Another possibility is that its lower energy is due to a degradation along the path from the source to detector. Finally, only a new experiment with higher statistics can tell us whether it corresponds or not to the $1/2^+$ excited state in ^{209}Pb.

In Table I we report the number of ^{14}C ions detected in each state and for each of the two current sets of SOLENO, together with the deduced partial half-lives, T, hindrance factors, HF, and spectroscopic factors S_{l_M}.

In a Geiger-Nuttal systematics (see Ref. [11]), we plotted $\log_{10}T$ vs $Q^{-1/2}$ for known ^{14}C emissions from even-even Ra isotopes with $A = 222$, 224, and 226. From the corresponding points we get by least-square fit a straight line. On the same diagram, we plotted the points representing the transitions from ^{223}Ra to the low-lying excited states of ^{209}Pb, listed in Table I. The vertical deviations from the straight line represent the hindrance factors given in Table I in the column "sys". The transition to the ground state (HF=380) is strongly hindered, while the one to the first excited state $11/2^+$ (HF=2) is not, in agreement with our previous results [11]. For transitions to the second and third excited states $15/2^-$ and $5/2^+$, only lower limits (89 and 43, respectively) were determined. The solitary event, if attributed to the transition toward $1/2^+$ excited state of ^{209}Pb, would be characterized by HF ≥ 4. The total branching ratio relative to α decay found in the present work, $(8.9\pm0.4)10^{-10}$, is about 20% higher than in all previous measurements. The relative intensities of transitions to the ground state and to the first excited state are 18% and 82%, respectively. The half-life of ^{233}Ra for ^{14}C decay is $T=10^{(15.04\pm0.02)}$ s. This shorter value could be explained by better ^{14}C collection efficiency. Indeed, the very small low-energy tailing of the present measurement proves the absence of "lost events" by energy degradation in the source.

Large hindrance factors, due to differences in the intrinsic configurations of the parent and daughter, have been measured both in α decay [16], and spontaneous fission [17] of odd-mass or odd-odd nuclei. There are different definitions, some of them relating model-dependent parameters (e.g., the reduced width). A better expression, HF $= T/T_{e-e}$, uses measurable quantities, i.e., T, the measured (or calculated) half-life, and T_{e-e}, the corresponding value for a hypothetical even-even equivalent, estimated from systematics, as above, or from a model. A transition is favored if HF $\simeq 1$, and it is hindered if HF ≥ 5. Within the ASAF model one can calculate T_{e-e} [18] for various angular momenta, l, determined from the total angular momentum and parity conservation. By ignoring l, we get from the experimental T and the ASAF T_{e-e} the hindrance factors given in Table I under the heading $l=0$. The next column shows smaller values, due to the influence of the maximum angular momentum l_M. The half-life $T = \ln 2/\lambda$ (where the decay constant $\lambda = \nu S P$) in principle could be calculated by taking into account the nuclear structure, which should dictate the magnitude of the preformation probability, S, of the emitted cluster into the parent nucleus. The two other model-dependent quantities are ν (frequency of assaults on the barrier) and P (external potential barrier penetrability). The model dependent "experimental spectroscopic factors" $S_l = \lambda_{\rm exp}/(\nu P_l)_{\rm mod} = S_{e-e}/{\rm HF}_l$, have been determined by using the ASAF model. By removing the contribution of the angular momentum, the remaining quantity mainly reflects the nuclear structure properties. In a fission theory the hindrance is the consequence of a larger barrier due to the *"specialization energy"* [19, 20]. In a many-body theory of α decay [21] the preformation probability is determined by an overlap integral of the parent, daughter, and emitted particle wave functions. Such a calculation has not been performed so far for cluster decay modes. One has instead made [13] a qualitative interpretation based on the numerical value of the square of the overlap integral between the $\Omega = 3/2$ Fermi level orbitals for $N = 135$ and the most important $\Omega = 3/2$ spherical orbitals $i_{11/2}$, $g_{7/2}$, $j_{15/2}$, and $g_{9/2}$. The overlap integral was obtained low (4%), high (64%), and low (7%) for the transitions toward g.s., first, and second excited states, respectively. The results of the present experiment are in fair agreement with this interpretation. Nevertheless, the full calculation reproducing the measured HF is still missing.

In conclusion, our goal to give fine-structure results in ^{14}C radioactivity with an ultimate quality was achieved. Several "records" were set: the maximum number of ^{14}C (899) detected, the best energy resolution (90 keV), and no background. We have shown that the background and the spurious peaks in the energy spectrum of previous experiments is given by the bad quality of the chemically

TABLE I. Experimental results for ^{14}C transitions from the $3/2^+$ ground state of ^{223}Ra to various final states of ^{209}Pb (excitation energy, E^*, spin and parity, J^π). T is the half-life, HF is the hindrance factor, determined by using T_{e-e} values from systematics (sys) and ASAF model with zero (l=0) or maximum (l_M) angular momentum, and S_{l_M} is the spectroscopic factor.

Final states E^*(MeV) J^π	Events[a]	$\log_{10}T(s)$	HF sys	HF l=0	HF l_M	$\log_{10}S_{l_M}$
0.000 $9/2^+$	92+38	15.79±0.05	380	269	86	-12.72
0.779 $11/2^+$	570+198	15.13±0.02	2.2	1.7	0.5	-10.68
1.423 $15/2^-$	0+0	> 18.08	>89	>72	>6.1	<-12.00
1.567 $5/2^+$	0+0	> 18.09	>43	>37	>21.3	<-12.39
2.032 $1/2^+$	0+1	≥ 18.06	≥4	≥3.5	≥3.0	≤-11.60

[a] At a SOLENO current $I = 278$ A, and $I = 281$ A, respectively.

prepared source. Concerning spectroscopic interest, ^{14}C radioactivity gives original results to test nuclear models. The interpretation of Ref. [12] lead to the wrong conclusion that the transition towards $15/2^-$ is favored. Our present result shows that it is hindered, in agreement with [13]. An eventual theoretical interest concerning the $1/2^+$ excited state of ^{209}Pb could provide motivation for a new experiment with a more intense source.

It is a pleasure to thank Gary Blanpied for a careful reading of the manuscript, and J.F. Berger and M. Girod for encouragement and helpful discussions. We are also grateful to the crew of the PS-Booster and of ISOLDE at CERN, to the crew of the Orsay tandem, in particular to D. Sznajderman, and to the health service staff. A MESR grant has been provided to one of us (D.N.P.).

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