Exotic Nuclear Decay of ^{223}Ra by Emission of ^{14}C Nuclei

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The exotic nuclear decay of ^{223}Ra by emission of ^{14}C nuclei has been investigated by use of an intense radioactive ^{227}Ac source and a magnetic spectrometer with a large solid angle. After a run of 5 d, a group of eleven events was observed at the expected location of ^{14}C in a ΔE-E telescope calibrated with a ^{14}C beam. A branching ratio of $(5.5 \pm 2.0) \times 10^{-10}$ was measured for the emission of ^{14}C nuclei relative to α particles from ^{223}Ra in agreement with the previously reported ratio of $(8.5 \pm 2.5) \times 10^{-10}$.

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Nearly a century after Becquerel, Rose and Jones (RJ) from the University of Oxford have recently reported a novel mode of radioactive decay[1] which was completely unexpected. They claim that the ^{223}Ra nucleus (known as a simple α emitter with a half-life of 11.2 days[2]) occasionally decays by emission of a ^{14}C fragment. The fact that among the nuclei belonging to a natural radioactive chain one may eliminate a substantial amount of charge and mass by the emission of a fragment heavier than an α particle was a surprising discovery. It was therefore highly desirable to confirm this first result by an independent measurement carried out, if possible, with greatly improved experimental techniques.

In the experimental arrangement of RJ, a radioactive source of ^{227}Ac ($t_{1/2} = 21$ yr),[2] with which the lower members of the series (including ^{223}Ra) are in secular equilibrium, was used. The source was placed directly in front of a ΔE-E solid-state (Si) telescope subtending a solid angle of $\frac{1}{3}$ sr, leading to a counting rate of 400 counts/sec. After a run of *189 d,* a group of *eleven events,* with a total energy of about 30 MeV, was observed. On the basis of this result, RJ concluded that these events were due to the emission of ^{14}C from ^{223}Ra with a branching ratio relative to the α particles of $(8.5 \pm 2.5) \times 10^{-10}$.

The aim of our measurement was to reduce the collection time greatly and to select unambiguously the rare decay mode from the high flux of α particles by means of a magnetic spectrometer. Furthermore, we used the same experimental arrangement and ^{12}C, ^{14}C, and ^{16}O particle beams to confirm independently the nature and the energy of the observed events.

Two ^{227}Ac radioactive sources of very different strengths were prepared in our radiochemistry laboratory. Starting with the same original material, the sources were deposited on a Pt disk with an active diameter of 8 mm and had identical geometries. In order to prevent material sputtering, they were covered by thin carbon plus Formvar foils (20–30 μg/cm^2 thick). The strengths of the two sources were measured before and after the experiment using a Si detector with good energy resolution (25 keV) and a geometrical arrangement which allows a reproducible and precise measurement of the solid angle subtended by the detector.

The results for ^{227}Th and ^{223}Ra are given in Table I. Here we stress that if this "intense" source were used with the experimental arrangement of Ref. 1, the α-counting rate would reach $\sim 0.9 \times 10^6$ counts/sec.

Such an "intense" radioactive source must be used with a magnetic spectrometer since this device is able to remove the high flux of α particles and thus to select rare decay modes. The emitted heavy fragments could be unambiguously identified with a standard ΔE-E telescope placed at the focal plane of the spectrometer. To take full advantage of the source strength one needs a magnetic spectrometer with a very large solid angle ($\sim \frac{1}{10}$ sr). Fortunately, the superconducting solenoidal spectrometer SOLENO,[3] with a solid angle of $\frac{1}{20}$ to $\frac{1}{10}$ sr, was available at the Orsay MP tandem accelerator. The transmission curve $\Omega = f(B\rho)$ or solid angle Ω versus magnetic rigidity $B\rho$ has a bell shape with a width at half maximum of $\pm 4\%$ in momentum.

The experimental geometry adopted for this particular investigation is schematically displayed in

TABLE I. Activities of the first three members of the ^{227}Ac radioactive sources used in this work.

	^{227}Ac	^{227}Th	^{223}Ra[a]
"Weak"	3.3 μCi	3.3 μCi	2.80 μCi
"Intense"	247 μCi	247 μCi	210 μCi

[a]The source deposits (^{227}Ac+^{227}Th) had been made about one month before the experiment and therefore secular equilibrium was not yet reached at the time of the experiment.

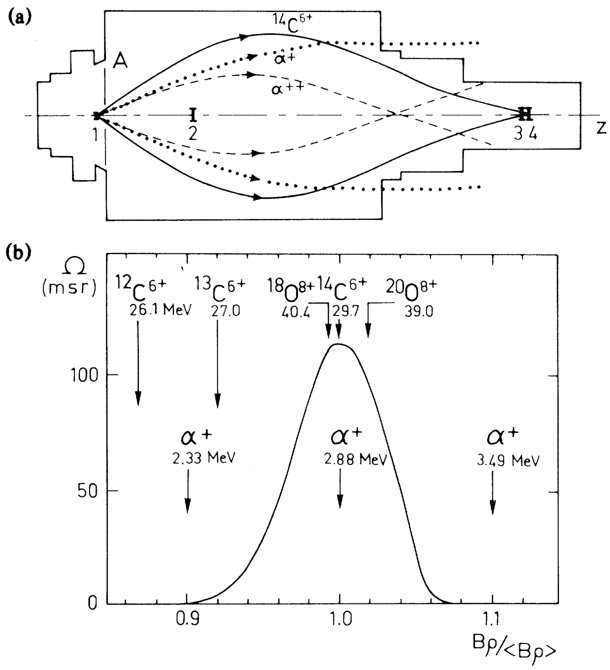

FIG. 1. (a) Schematic representation of the experimental setup used during this work. 1, source location. A, entrance aperature (limiting angle $\pm 20°$). 2, intermediate baffle subtending an angle of $\pm 4°$. Such obturator prevents α particles emitted along the z axis from reaching the ΔE-E telescope. 3,4, ΔE-E telescope location. The setting of SOLENO in this figure corresponds to the focus of $^{14}C^{6+}$ ions of 29.7 MeV (solid lines). (b) Transmission curve of the spectrometer SOLENO. Solid angle Ω vs the ratio of the magnetic rigidity of $B\rho$ of the particles to the central magnetic rigidity $\langle B\rho \rangle = 0.49$ T m. This transmission curve corresponds to the situation illustrated in (a). The other possible rare decay modes are shown with respect to this transmission curve.

Fig. 1(a). The source was mounted inside the spectrometer. In the image plane located at 1.53 m from the source, a Si solid-state ΔE-E telescope was used to identify the atomic number and to measure the total energy of the emitted particles. The ΔE detector was 7.5 μm thick with an active area of 200 mm^2 whereas the E counter was 200 μm thick with an area of 300 mm^2. Both detectors were made in our laboratory. A preliminary calibration was made using ^{241}Am and ThC α particles of 5.48, 6.05, 6.09, and 8.78 MeV, respectively. The thresholds in the ΔE and E counters were set to 0.5 and 1.5 MeV, respectively.

A series of runs were carried out.

First the "weak" ^{227}Ac source was employed to measure the solid angle and the transmission curve of SOLENO. The current was set in order to focus the α particles from ^{227}Th (6.038-, 5.978-MeV doublet) and the resulting counting rate was compared to the one obtained for identical α rays in a detector of known solid angle. The deduced $\Omega = f(B\rho)$ transmission curve is shown in Fig. 1(b). The maximum value of the solid angle is equal to 0.115 ± 0.002 sr.

In a second step, a run of 5 d was made in order to have a measure of possible background events. The experiment was carried out without the ^{227}Ac source and with the current set equal to zero in SOLENO. A similar run was repeated at the end of the experiment. The resulting background spectra will be discussed later.

The search for the emission of heavy fragments was carried out in the following manner: The current of SOLENO was preset to focus selectively the expected $^{14}C^{6+}$ of 29.7 MeV (in the laboratory frame and taking into account the energy loss in the source and in the carbon foil) on the ΔE-E telescope [see Fig. 1(a)]. The 6+ charge state is the most probable one.[4]

For this current setting ($I = 285$ A, $B = 2.43$ T), the spectrometer has its maximum solid angle [see Fig. 1(b)]. The high flux of $^4He^{++}$ ions emitted by the intense ^{227}Ac source are then focused well before the detector because of their lower magnetic rigidity and the corresponding solid angle is equal to zero. The $^4He^{++}$ ions have much higher $B\rho$ values, their focus occurs well beyond the ΔE-E location, and again they are not seen by the counter [see Fig. 1(a)]. Only the degraded $^4He^+$ ions, whose energies are between 2.33 and 3.49 MeV, are within the transmission curve of SOLENO [see Fig. 1(b)]. These particles either are stopped in the 7.5-μm Si counter, or do not give a coincident event because of the lower E threshold.

We have calculated the positions of the other possible exotic nuclear decays suggested by Gamow-factor ratios[1] and by the energies available for the decay and compared these to the transmission curve of SOLENO in Fig. 1(b). Note that the possible rare decay mode $^{227}Ac \rightarrow {}^{20}O + {}^{207}Tl$ was not selected by RJ whereas in our calculations of Gamow-factor ratios, the deduced value is comparable to the one obtained for $^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$.

From Fig. 1(b) it is obvious that only $^{18}O^{8+}$, $^{14}C^{6+}$, and $^{20}O^{8+}$ ions are transmitted with a large solid angle (≈ 100 msr) whereas the $^{12,13}C^{6+}$ ions are not transmitted using the adopted current setting.

Standard fast-slow coincidence techniques were employed to generate a correlated ΔE-E event. The coincidence rate was about 2–4 counts/sec. In addition, the internal clock of the T-1600 SOLAR computer was used to determine the arrival time T of each coincident event. The event $(E, \Delta E, T)$ was stored on magnetic tape.

The full bidimensional spectrum obtained after a run of 5 d is shown in Fig. 2(a). In the lower left-hand corner of the figure one sees the contour diagrams associated with α particles of 6.55, 7.39, and 6.62 MeV originating from ^{219}Ra, ^{215}Po, and ^{211}Bi decays,[2] respectively. The ^{219}Rn, as a gas, escapes from the ^{227}Ac source and may travel inside the vacuum until it reaches the ΔE detector. Its decays and the ones corresponding to its daughters (^{215}Po and ^{211}Bi) are therefore detected by the telescope.

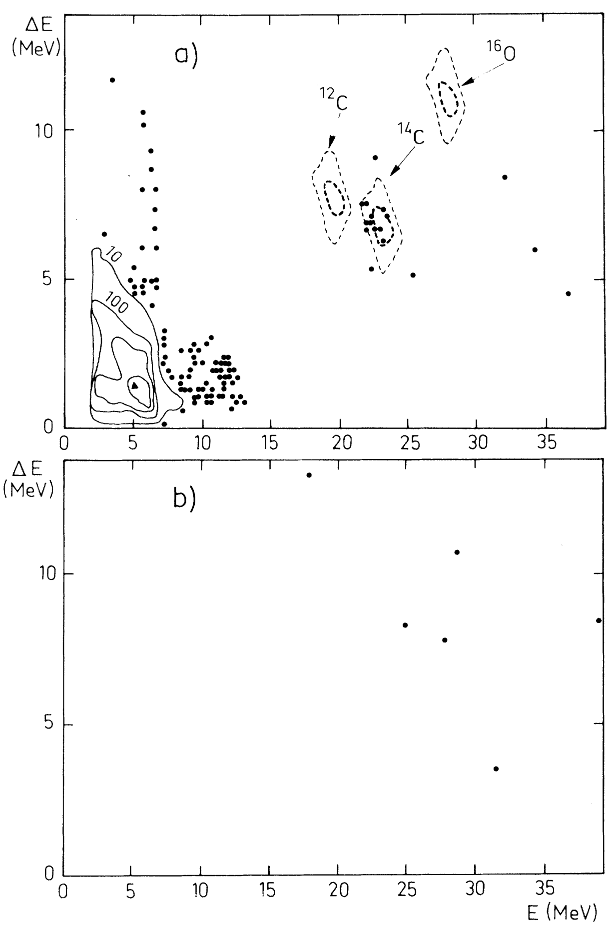

FIG. 2. (a) Full ΔE-E spectrum obtained after a run of 5 d with the intense ^{227}Ac source and a current setting corresponding to Fig. 1(a). The group of eleven events in the middle of the figure correspond to the emitted ^{14}C nuclei from ^{223}Ra. The dashed contours indicate the locations of elastically scattered ^{12}C, ^{14}C, and ^{16}O particles (see text). (b) ΔE-E spectrum obtained after a run of 5 d without the intense ^{227}Ac source and with current set to zero in the spectrometer. Six events randomly distributed are observed in this search for "background" events.

A small number of random coincidences are located along the $\Delta E(\sim 1$ MeV) and E(5–6.5 MeV) lines associated with these α peaks.

In the middle of the ΔE-E plane and well isolated from the α particles a group of *eleven events* was observed. Their energy losses in the ΔE counter (6.8 ± 0.7 MeV) are in very good agreement with that expected for carbon ions in a 7.5-μm Si detector. The measured total energy is equal to 29.4 ± 1.2 MeV and is also very close to the value of 29.7 MeV predicted from Q-value considerations and energy losses.

An additional six events, randomly distributed, were also observed. In Fig. 2(b) the ΔE-E spectrum, obtained during a similar data taking period of 5 d without source and current in the spectrometer, is displayed. Six events were also detected during this search for possible background counts. It appears that they are due to a slight contamination of the ΔE counter by fission products from ^{252}Cf. Such a source was in fact used during preliminary runs to test the response of the ΔE detector to heavy particles. It is very gratifying that the background spectrum explains quite well the previously unidentified six events present in Fig. 2(a).

To confirm independently that the observed events correspond to a ^{14}C decay, we have performed elastic-scattering experiments using energy-analyzed ^{12}C, ^{14}C, and ^{16}O particle beams delivered by the Orsay MP tandem accelerator. The beams were incident upon a ^{12}C self-supporting target of 20 μg/cm^2. The $^{12}C^{6+}$, $^{14}C^{6+}$, and $^{16}O^{8+}$ ions elastically scattered at a laboratory angle of $8.3 \pm 0.2°$ were focused onto the same ΔE-E telescope by the magnetic spectrometer SOLENO. The incident energy of each beam was chosen in order to obtain scattered $^{12}C^{6+}$, $^{14}C^{6+}$, and $^{16}O^{8+}$ ions with an energy in the laboratory exactly equal to that expected for the corresponding heavy fragment, namely, 26.1 MeV for ^{12}C, 29.7 MeV for ^{14}C, and 39.0 MeV for ^{16}O [simulating a 39-MeV ^{20}O fragment: see Fig. 1(b)]. The contour limits at half and at $\frac{1}{10}$ maximum for ^{12}C, ^{14}C, and ^{16}O are shown as dashed lines in the ΔE-E plot of Fig. 2(a).

Clearly the location of the observed group of eleven events obtained before this calibration agrees quite well with the response of our telescope to $^{14}C^{6+}$ ions of 29.7 MeV. Both ^{12}C and ^{16}O contours are far from the observed events.

On the basis of these results, the observed heavy fragments correspond unambiguously to ^{14}C nuclei with a total energy which is in very good agreement with that expected from the decay of ^{223}Ra into $^{14}C + ^{209}Pb$. We would like to stress that both the magnetic rigidity selection of SOLENO and the independent calibration using elastically scattered ^{12}C, ^{14}C, and ^{16}O nuclei rule out the possibility that the emitted fragments are ^{12}C, ^{13}C, ^{18}O, and ^{20}O nuclei. This identification is obtained without referring to any Gamow-factor ratios or preformation-probability considerations, whereas the authors of Ref. 1 rely heavily on these arguments to exclude all the other possible decay modes. Moreover, from the total-energy measurement we conclude that these ^{14}C fragments are the decay products of ^{223}Ra nuclei since the other possible ^{14}C emitters, namely, ^{227}Th and ^{219}Rn, have total energies in the laboratory frame of 27.6 and 26.2 MeV, respectively. These values are outside the error bars of our measured value of 29.4 $\pm$ 1.2 MeV.

From the determination of the ^{223}Ra activity, the value of the solid angle, and the percentage of 6+ charge state for a carbon ion of 29.7 MeV, we have deduced a branching ratio for the emission of ^{14}C nuclei relative to α particles from ^{223}Ra of $(5.5 \pm 2.0) \times 10^{-10}$, a value which is in agreement with the one deduced from the experiment of RJ, viz., $(8.5 \pm 2.5) \times 10^{-10}$.

Summarizing our results, we conclude that the exotic nuclear decay of ^{223}Ra by emission of ^{14}C has been confirmed unambiguously. The magnetic superconducting spectrometer SOLENO used together with intense radioactive sources appears to be an ideal tool for further investigation of similar rare decay modes. We feel that branching ratios of one or two orders of magnitude lower than the one measured in this experiment can be reached with stronger sources.

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