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New measurement of exotic decay of ^{225}Ac by ^{14}C emission

A. Guglielmetti[1,a], R. Bonetti[1], G. Ardisson[2], V. Barci[2], T. Giles[3], M. Hussonnois[4], J.F. Le Du[4], C. Le Naour[4], V.L. Mikheev[5], A.L. Pasinetti[6], H.L. Ravn[3], S.P. Tretyakova[5], and D. Trubert[4]

[1] Istituto di Fisica Generale Applicata dell'Università di Milano and INFN, Sezione di Milano, Via Celoria 16, I-20133, Milano, Italy
[2] Laboratoire de Radiochimie, Université de Nice Sophia-Antipolis 06108 Nice Cédex, France
[3] CERN, EP Division, Geneva, Switzerland
[4] Institut de Physique Nucléaire, BP N° 1 91406 Orsay Cédex, France
[5] Joint Institute for Nuclear Research, Dubna, 141980, Moscow Region, Russia
[6] Dipartimento di Fisica dell'Università di Milano and INFN, Sezione di Milano, Via Celoria 16, I-20133, Milano, Italy

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Abstract. The branching ratio of ^{225}Ac decay by emission of ^{14}C was remeasured under improved experimental conditions by using a radioactive source produced at the ISOLDE mass-separator at CERN and a nuclear track detector technique. The result, $B = \lambda_{^{14}\mathrm{C}}/\lambda_\alpha = (4.5\pm1.4)10^{-12}$, is consistent with the anomalously high value obtained in the 1993 experiment, thus confirming the importance of nuclear-structure effects in this exotic decay.

PACS. 23.70.+j Heavy-particle decay

1 Introduction

One of the most important achievements in the yet ultradecennal research on heavy cluster radioactivity was the discovery of the sensitivity of its partial half-life to the microscopic properties of the mother-daughter nuclei. This resulted from many evidences like, *e.g.*, the fine structure in the energy spectrum of ^{14}C clusters in the well-known ^{223}Ra decay, the hindered decay of odd-A emitters like ^{221}Fr, ^{221}Ra, ^{223}Ra, ^{231}Pa, ^{233}U and others, the anomalously high/low values of two-clusters branching ratios such as ^{24}Ne/^{28}Mg, ^{23}F/^{24}Ne and others [1].

Within such a framework, ^{225}Ac was a special case since while being an odd-Z nucleus its partial decay rate as well as its branching ratio relative to α decay do not seem to exhibit any special hindrance like the one of other odd-A exotic emitters. The situation was summarized in ref. [2] in the case of hindrance factors for ^{14}C emitters. This quantity, borrowed from α decay, measures the degree according to which an odd-A transition is slower in comparison with an even-even one having the same barrier penetrability. While typical hindrance factors, $HF = \frac{\gamma^2(A+1)+\gamma^2(A-1)}{2\gamma^2(A)}$, where $\gamma^2(A)$ is the reduced width of the cluster emitter of mass number A, are in the range 10–100, the one of the $^{225}\mathrm{Ac} \rightarrow {}^{14}\mathrm{C}$ decay measured in 1993 [3] is practically unitary.

The fact that such a case seems surprisingly to behave like an even-even one has been variously justified [3,4] by using arguments based on the microscopic structure of ^{225}Ac.

In order to have a firmer basis for any theoretical interpretation, it would be important to have further experimental data in order, as a first instance, to confirm the 1993 result and possibly to achieve a deeper insight on this rather interesting exotic decay.

Moreover, an important motivation for a renewed study of this decay is the disagreement existing between our 1993 experiment [3], which gave a branching ratio $B(^{14}\mathrm{C}/\alpha) = (6.0 \pm 1.3) \times 10^{-12}$, and the upper limit obtained in 1986 by the Berkeley group, $B < 4.0 \times 10^{-13}$ [5]. Clearly, a confirmation of the latter result and a disproval of ours would imply a considerably higher hindrance, $HF > 15$, therefore perfectly on line with other results on odd-A exotic emitters [2].

We, therefore, prepared a new, strong ^{225}Ac source to be used in two independent experiments:

i) an high efficiency but low-energy resolution experiment to remeasure the integrated decay rate of the $^{225}\mathrm{Ac} \rightarrow {}^{14}\mathrm{C} + {}^{211}\mathrm{Bi}$ decay;
ii) an high-resolution experiment to study the energy spectrum of the emitted ^{14}C clusters.

[a] e-mail: alessandra.guglielmetti@mi.infn.it

2 Source preparation

Like in the previous experiment, the ^{225}Ac source, $T_{1/2} = 10.0$ d, was obtained by taking profit of the intense, mass-separated 60 keV ^{225}(Fr + Ra) beam produced at ISOLDE, CERN. Here, such nuclides are obtained by means of spallation reactions induced by the 1 GeV proton beam of the PS-Booster on a thick ThC_2 target. The nuclides of interest were subsequently transferred by diffusion to a surface ionization source from which Fr and Ra beams were extracted, accelerated and magnetically analysed in the General Purpose Separator of ISOLDE. Finally the ^{225}(Fr + Ra) mass ions were sent in a collection chamber and implanted onto a 2×2 cm^2 vitreous carbon catcher (Carbon Lorraine, France).

A major difference with respect to the 1993 experiment was the increase of the proton beam energy from 600 MeV, given by the then available CERN Synchrocyclotron, to 1 GeV, the present PS-Booster energy. In order to compensate for the decrease of production of nuclides close to the stability line such as ^{225}Fr and ^{225}Ra with increasing primary beam energy, a dedicated, 87 g/cm^2 thick ThC_2 target had to be developed, tested and used for the first time in this collection. With this target an exceptionally high secondary beam intensity was obtained. It reached values as high as 8×10^9 atoms per second with a primary proton beam of the order of 3.5 μA. In the course of 4.8 d irradiation time, about 2×10^{15} ^{225}(Ra + Fr) atoms were collected. The ^{225}Ac source was subsequently obtained by β decay of the implanted ^{225}Fr ($T_{1/2} = 4.0$ min) and ^{225}Ra ($T_{1/2} = 14.9$ d) atoms.

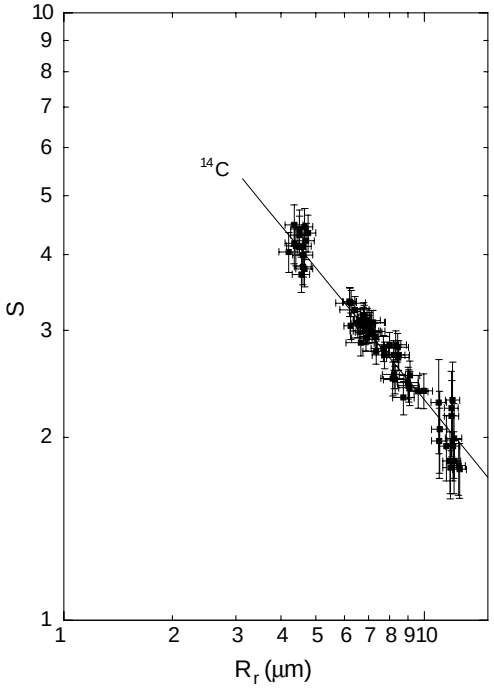

Fig. 1. Comparison of the sensitivity $S = v_t/v_g$ measured for the detected events at different points along the particle trajectory and indicated with small black squares with accelerator calibration curve. R_r is the particle residual range which represents the portion of the total length of the track which, at a certain etching stage, needs still to be developed. The curve is a linear interpolation of experimental values obtained with the 18 MeV ^{12}C beam and shifted for ^{14}C according to the relation $S(^{14}C) = S(^{12}C) \times 14/12$ at the same value of the residual range.

3 The track detectors experiment

In order to carefully remeasure the energy-integrated half-life for ^{14}C emission, we resorted, as in previous works [1, 3], to nuclear track detectors because of their efficiency and selectivity.

We therefore placed the ^{225}Ac source in the center of an aluminum hemisphere, 19.5 cm diameter, whose inner surface was covered with 2.5×2.5 cm^2 barium-phosphate glass plates of the BP-1 type (Schott Glass Technologies, USA) acting as nuclear track detectors. It is well known [1] that, in order to allow a precise particle identification, these detectors need an equally precise calibration, usually done by means of accelerator beams of known energy and ion type. The aim of such a procedure is to establish an empirical relationship between the detector sensitivity, $S = v_t/v_g$ (where v_t and v_g are the etching velocities along the track and in the bulk material, respectively) and the particle residual range [1].

Although we largely used and calibrated this type of detector in the past [6], for this repetition experiment we wanted to use a calibration specifically performed for a glass of the same batch used for the experiment. This was achieved by irradiating a few glass samples by means of 18 MeV ^{12}C beam of the XTU Tandem of Laboratori Nazionali di Legnaro, Italy.

Moreover, in order to make the detector scanning easier and the track parameter measurements more precise, an important step towards particle identification, we decided to increase the signal-to-noise ratio by diminishing the alpha fluence with respect to previous experiment, from $5.9 \times 10^{11} \alpha$/cm^2 to $1.2 \times 10^{11} \alpha$/cm^2. A further improvement was the decrease of the plastic-foil absorber thickness placed in front of the detectors from 19 to 16.9 μm, thus allowing a decrease in the relative error in the track length measurement. The degraded ^{14}C energy comes out to be 18.3 MeV, therefore the same as the one of the beam used in the accelerator calibration.

The irradiation started 73 d after source production and lasted 260 h. After irradiation, the absorbers were removed and the glass plates etched in a 50% stirred HBF_4 solution for 48 h at 65 °C, together with the calibration sample. Detector scanning was done by means of an automated scanning system, based on an image analyzer (Elbek, Siegen, Germany) coupled to an optical microscope and a CCD camera [1], optimized by using the accelerator-produced events in the calibration sample. As a consequence of the experimental improvements described above, the overall scanning efficiency turned out to be 100% , instead of the previously obtained 90%.

We found 14 events in 86.2 cm^2. Figure 1 shows the result of the comparison with the ^{14}C calibration curve, obtained by shifting the one for ^{12}C according to the relation $S(^{14}\mathrm{C}) = S(^{12}\mathrm{C}) \times 14/12$ at the same value of the residual range [7]. The agreement is astonishing. A further improvement was that particle identification was here achieved on the basis of the extraction of the couples (R_r, S) in three different points along the particle trajectories instead of only one like in previous experiment.

From the known source intensity and irradiation time, the branching ratio for ^{14}C decay of ^{225}Ac comes out to be $B = (4.5 \pm 1.4)10^{-12}$ and the corresponding partial half-life $T_{1/2} = (1.9 \pm 0.6)10^{17}$ s. This result was obtained by subtracting from the 14 tracks 2 due to the known ^{14}C decay of ^{221}Fr [2] present in equilibrium within the source. The quoted error is determined from statistics (about 30%), from ^{221}Fr background subtraction (2.5%), from detection efficiency (5%) and from source activity (5%).

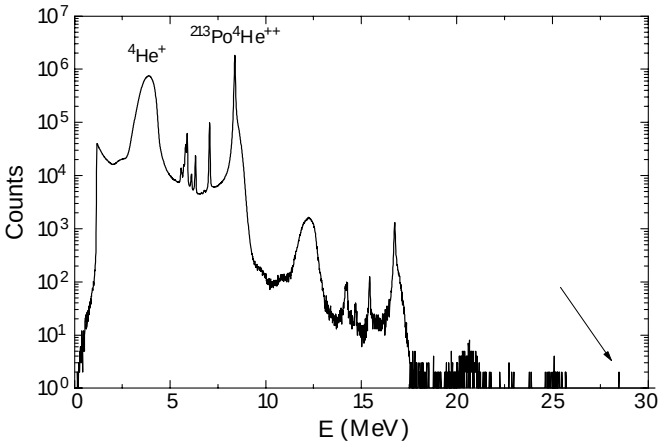

Fig. 2. A typical energy spectrum obtained with the ^{225}Ac source at SOLENO, showing the structures discussed in the text. The one event at ≈ 28.5 MeV is shown with an arrow.

4 The high-resolution experiment

One of the goals of the experiment was to find out which level of the ^{211}Bi residual nuclide is preferentially fed by the ^{14}C decay of ^{225}Ac. As discussed in refs. [3] and [4], two hypothesis have been put forward to explain its unexpectedly high decay rate, in analogy with α decay, in terms of favoured transition either to the ground state or to the first excited one of ^{211}Bi. Calculations performed by means of the cluster model of ref. [8] under both hypothesis are compatible with the experimental data due to the relatively low-excitation energy, $E^* = 404$ keV, of the $J^\pi = 7/2^-$ first excited state of ^{211}Bi, which reduces the barrier penetrability of only a factor 6 relatively to the ground-to-ground-state transition. While theoretical arguments based on the structure of the ^{225}Ac mother nucleus could justify both hypothesis [3,4], only an experimental result could solve such ambiguity, thus possibly confirming the use of ^{14}C radioactivity as a spectroscopic tool after the pioneering experiments with ^{223}Ra decay [4].

Experiments such as the one previously discussed and that of ref. [3] were performed with nuclear track detectors, which, while maximizing the detection efficiency do not have an energy resolution high enough to resolve the ground from the first excited-state decays.

We therefore planned to use a Si detector placed at the focal point of the superconducting solenoidal spectrometer SOLENO of IPN-Orsay. Such a device has been widely used in previous experiments on ^{14}C decay of ^{222,223,224}Ra [9]. In particular, it has allowed to establish the existence of a fine structure in the energy spectrum of ^{14}C clusters [10] which exhibits striking analogies with those of α decays from odd-A parents. The basic idea is to use the selective features of the spectrometer to reject the high flux of α-particles, thus avoiding unwanted phenomena such as pile-up and radiation damage, while keeping the good energy resolution typical of Si detectors.

The radioactive source was therefore moved to Orsay, measured by γ spectroscopy to have an activity $A = 4.44 \times 10^8$ Bq 19 d after the end of CERN collection, and placed inside SOLENO, whose magnetic field was optimized for transmitting 28.6 MeV, 5^+ ^{14}C ions. An ion-implanted Si detector having a surface of 600 mm^2 and a thickness of 100 μm was used.

With such an high source intensity, by assuming a collection efficiency of 200 msr [11] and a branching ratio $B = 6 \times 10^{-12}$ [3], we calculated that the expected number of ^{14}C events was of the order of 1 per day.

Unfortunately, the 8.5 d long experiment turned out to be difficult due to:

1) ^{4}He^{+} particles of degraded energy centered at about 4 MeV emitted by the source, having the same magnetic rigidity as the one of 28.6 MeV ^{14}C and therefore being completely transmitted within the transmission curve. Although the implantation depth of the Isolde beam and the source thickness (0.02 μm) are negligibly small, such a degradation results mainly from multiple collisions within the spectrometer.
2) An important transmission of 8.375 MeV ^{213}Po ^{4}He^{++} particles even in the tail of the transmission curve, in spite of the choice of focussing ^{14}C^{5+} instead of the more abundant ^{14}C^{6+}.

The consequences of the above were not only a radiation damage of the detector but also multiple pile-up events which unfortunately fell also in the energy region between 28 and 29 MeV where ^{14}C events were expected. For example, pure pile-up of three 8.375 MeV ^{4}He^{++} and one 3.9 MeV ^{4}He^{+}, not rejected by using the pile-up rejection circuitry, gives an event at 29 MeV. It is therefore impossible to distinguish between such event and those corresponding to ^{14}C populating either the ground or the first excited state of the daughter nucleus. The situation is shown in fig. 2 where one clearly sees the intense peak of 8.375 MeV ^{213}Po ^{4}He^{++} particles, the ^{4}He^{+} bump centered at 4 MeV, their multiple pile-up peaks and one event in the region of interest.

5 Discussion and conclusions

The positive result obtained with track detectors is clearly consistent with the 1993 one [3]. A weighted average gives $B = (5.3 \pm 1.0)10^{-12}$ and $T_{1/2} = (1.6 \pm 0.3)10^{17}$ s.

Such a consistency is particularly meaningful not only in the light of the experimental improvements related to the track detector technique described above, but also and especially to the source quality. Indeed, a major problem of sources produced with a mass separator is the one of possible contaminants of adjacent mass number. This problem was already known, taken into account and discussed in ref. [3]. Of the two ^{14}C emitters adjacent to ^{225}Ac, it was found that ^{226}Ra would give no contribution because of its by far too long half-life in comparison with the one of ^{225}Ac. On the other hand, ^{224}Ra could be a problem, its half-life being of the order of the one of ^{225}Ac (3.66 d) and its B-value for ^{14}C decay being about two orders of magnitude higher [9]. In ref. [3] it was proposed to solve this problem by allowing a waiting time between source collection and track detectors irradiation long enough (about 7 half-lives) to decrease a possible ^{224}Ra contaminant to negligible levels. By using the same idea, in the present experiment we increased the waiting time to 20 half-lives. The fact that we obtained consistent results proves that the decay we measured has nothing to do with ^{224}Ra decay.

This result therefore confirms the interest of ^{14}C decay of ^{225}Ac from the point of view of nuclear structure. However, it also suggests that an experimental identification of the favoured transition, a crucial information to test the proposed theoretical interpretations [3,4], is a very difficult task indeed. When compared with a similar case largely investigated, ^{14}C decay of ^{223}Ra, we see that such a difficulty is due to the 3 orders-of-magnitude smaller branching ratio which, even with a highly selective device like the magnetic spectrometer used in the present work, produces a too unfavourable signal-to-noise ratio. On the other hand, using of a more selective device would unavoidably imply a smaller collection efficiency, thereby worsening an already critical situation.

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