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EMISSION OF EXOTIC CLUSTERS BY NUCLEI AND DISCOVERY OF A FINE STRUCTURE IN THE ^{14}C DECAY OF ^{223}Ra

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EMISSION OF EXOTIC CLUSTERS BY NUCLEI AND DISCOVERY OF A FINE STRUCTURE IN THE ^{14}C DECAY OF ^{223}RA

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ABSTRACT

This paper is intended as a broad, mainly experimental, survey of the recent field of exotic cluster radioactivity in heavy nuclei. The first part summarizes the development of the field since the first experimental finding in 1984, insisting on ^{14}C emission, and giving a schematic status of the corresponding models. The second part describes in detail the 1989 discovery, in Orsay, of a fine structure of the ^{14}C decay of ^{223}Ra and the search for a similar effect in even-even neighboring nuclei ^{222}Ra and ^{224}Ra. A possible qualitative interpretation of the "hindrance" of the transition to the ground state of ^{209}Pb is proposed.

1. Introduction

It is a pleasure for me to be here to commemorate the 70th birthday of Ray Sheline. I first met him here, in Tallahassee, when I was a young post-doc. During this year 1962 (the year of the Cuban crisis) we worked on the structure of rare earth nuclei using the (d,p) reaction and published together in Phys. Rev. a paper largely cited later. This was the beginning of a long standing link, both personal and scientific.

The choice of my subject for this workshop has been made partly because it has relied heavily on radiochemistry, but also because Ray himself, during a seminar on octupole shapes he was giving at Orsay, casually attracted our attention on the paper just published in Nature by Rose and Jones[1] and dealing with ^{14}C emission.

2. Experimental Study of the Emission of Exotic Clusters

2.1 The Emission of ^{14}C by ^{223}Ra

Summarizing very much, Rose and Jones from the Oxford University, had "simply" put an E x Δ E telescope in front of a ^{223}Ra source (in fact an ^{227}Ac source having a 21 years half life, with which ^{223}Ra (11.2 days) was in secular equilibrium). The source-to-detector distance was large enough (solid angle $\Omega \approx 1/3$ sr) to reduce the very high counting rate from α radioactivity down to an acceptable level. After 189 days of counting they had obtained a group of 11 events with a total energy of about 30 MeV, considered as corresponding to a ^{14}C emission with a branching ratio, relative to α particles, R = (8.5 ± 2.5) 10^{-10}...close to 1 in a billion ! Such an unexpected result(*)was puzzling and it appeared clearly desirable to carefully check, and hopefully confirm, the results of the long and difficult experiment of Rose and Jones.

We had indeed in Orsay a quite unique possibility to do an experimental verification, rapidly and in a rather different way. The goal of our measurement was, using a strong source, to greatly reduce the collection time, and to select unambiguously the rare decay mode from the high flux of α particles by means of a magnetic spectrometer. We also anticipated the important advantage to be able to use various accelerated particle beams to identify precisely the emitted fragments.

- Our radiochemistry laboratory was able, using old and intense radioactive solutions, to prepare an ^{227}Ac source about 300 times stronger than the one used in Oxford.
- A superconducting solenoidal spectrometer, "SOLENO", recently installed at the Orsay MP tandem accelerator, permitted to select with a large solid angle ($\Omega \approx 115$ msr) the desired fragments ($^{14}C^{6+}$, the 6^+ charge state being the most probable one : ≈ 65 %), while very efficiently rejecting the enormous flux of α particles emitted by the source in this solid angle ($\approx 10^6$/sec !). After this magnetic selection the heavy particles were detected and identified by an E x ΔE telescope placed at the focal plane of the spectrometer (see Fig. 1). Both the ΔE (7.5 μm, 200 mm^2) and the E (200 μm, 300 mm^2) Si detectors were produced in our laboratory.
- Beams of ^{12}C, ^{14}C and ^{16}O from our tandem were elastically scattered from a thin C target (the final energy after scattering being in each case the one expected for the emission of such fragments), analyzed by SOLENO, and used to calibrate precisely the telescope and the associated electronics.

In summary (details can be found in Ref.3), after a run of only 5 days, 11 events were observed and clearly attributed to a ^{14}C emission (see Fig. 2). The branching ratio, determined as R = (5.5 ± 2.) 10^{-10}, is in agreement with the value given in Ref.1. It could therefore be safely concluded, after the pioneering work of Rose and Jones[1] and our clear confirmation[3] that - nearly a century after Becquerel - a novel mode of radioactive decay, by emission of ^{14}C, had been discovered.

Several other groups in the world[4-6] also confirmed rapidly the phenomenon, using different techniques : a simple E x ΔE telescope[4] as Rose and Jones, solid state track detectors[5] (the Ra isotopes being produced by a Berkeley group at CERN and separated by the Isolde on - line separator), and a split-pole magnetic spectrometer[6]. This last experiment permitted a clear determination of the mass (A = 14) of the exotic cluster.

2.2 *Search for other ^{14}C Emitters and other Exotic Clusters*

It was a logical continuation, first to extend the search to find other ^{14}C emitters, but also to look for the possibility of emission of heavier exotic clusters.

New ^{14}C emitters have indeed been found : first ^{222}Ra and ^{224}Ra[5] by the Berkeley group (using as above Isolde and track detectors), the result for ^{222}Ra being later confirmed[7] at Orsay with SOLENO ; then ^{226}Ra[7], first at Orsay with SOLENO, and

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(*) Although at the time we were not aware of this fact, we discovered later that the phenomenon of cluster emission had been predicted[2]

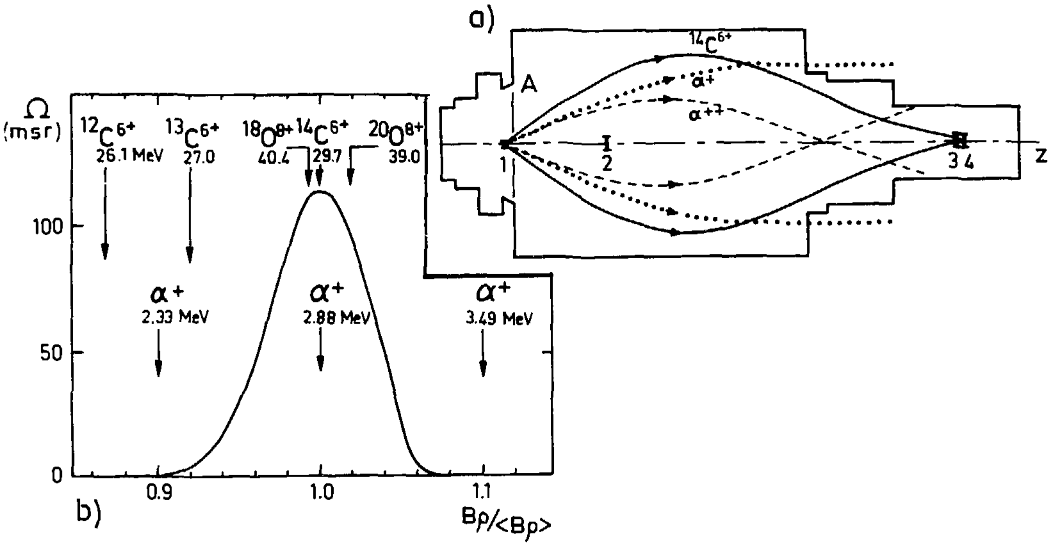

Fig.1(a) Schematic representation of the experimental setup used during our first study[3] of ^{223}Ra exotic emissions. 1, source location A entrance aperture (limiting angle ± 20°). 2, intermediate baffle subtending an angle of ± 4°. Such obturator prevents α particles emitted along the z axis from 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 Bρ of the particles to the central magnetic rigidity $<B\rho> = 0.49$ Tm. This curve corresponds to the situation illustrated in (a). The other possible rare decay modes are shown with respect to this transmission curve.

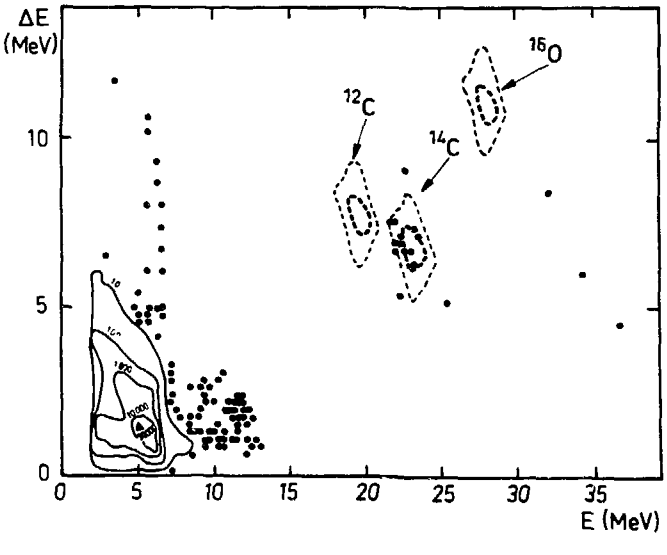

Fig. 2. ΔE-E spectrum obtained after a run of 5 d with the intense ^{227}Ac source and a current setting corresponding to Fig.1 (a). The group of eleven events in the middle of the figure correspond to the ^{14}C nuclei emitted from ^{223}Ra. The dashed contours indicate the locations of elastically scattered ^{12}C, ^{14}C, and ^{16}O particles (see text).

confirmed with improved statistics by a collaboration of the Berkeley and Orsay groups. It should be pointed out that radiochemistry played a key role in the measurements of the Orsay group : ^{227}Ac and ^{230}U sources have been used to generate ^{223}Ra and ^{222}Ra respectively. The ^{230}U itself was obtained from ^{230}Pa produced by irradiation of Th with 34 MeV protons at the Orleans cyclotron. The general characteristics of the sources - obtained after quite complicated multistep radiochemichal separations and purifications - are given in Table 1.

Emission of heavier clusters have also been found[7,8] in several nuclei : first ^{24}Ne by $^{232-233}$U, ^{230}Th and ^{231}Pa, then ^{28}Mg (and Ne) by ^{234}U. Upper limits have also been obtained for the emission of heavier clusters, for example for the emission of ^{34}Si by ^{241}Am. More details and recent results can be found in review papers (see Ref.9, 10 and 11).

Table 1.General characteristics of the sources used at Orsay

Source	Radioactive deposit	Deposit thickness	Deposit diameter	Source activity
^{223}Ra	^{227}Ac	–	8 mm	210 μCi (7.9 MBq.) in ^{223}Ra
^{222}Ra	^{230}U	–	10 mm	600 μCi (22.5 MBq.) in ^{222}Ra
^{226}Ra	$S0_4Ra$	1.8 mg/cm^2	16 mm	2.5 mCi (94 MBq.) in ^{226}Ra

2.3 Summary of the Experimental Results

In this section I will try to extract and outline only the most striking gross results from the impressive amount of data collected since 1984 (see Ref. 10 and 11 for more details). To illustrate these data, the order of magnitude of some measured R values and partial half-lifes for emission of ^{14}C and ^{24}Ne are shown in Tables 2 and 3.

- It is clear that the first case studied, the emission of ^{14}C by ^{223}Ra, was the most favorable one for the R value...
- The branching ratios R for Ne (or Mg)emission are, generally speaking, at least two orders of magnitude smaller than those for ^{14}C.
- When looking at the half lifes of the exotic emissions, it is clear that a minimum is observed, both for C and Ne, when the final nucleus is ^{208}Pb. More generally, the emissions observed always lead to nuclei close to ^{208}Pb.
- An odd-even effect - similar to the one commonly observed for α-decay but much larger - has been reported[7] : the exotic fragment emission rate from an odd-even emitter appears as hindered as compared to the one measured for an even-even emitter. The effect is particularly evident in the Kurie and Knopf diagrams shown in Fig. 3.

3. Theory of Exotic Emissions

As already indicated in section 2.1 -even if we did not notice it at the time - indeed theory predicted[2] beforehand (since 1980 and using different models) the possibility of exotic cluster emission, intermediate between α-decay and fission. Schematically the process can be described alternatively by : i) cluster models, where the cluster has first to be formed at the surface, then to be emitted by tunneling through the Coulomb barrier (two-step process) as in the traditional theory of α-decay ; ii) a very special fission process, extending the fission theory to very asymmetric situations. In any case, it is clear that the emission is - at least partly - governed by energy considerations. In particular it is allowed only when the Q value (energy released in the decay) is positive and one feels intuitively that the rate should increase with Q.

Table 2. Emission of ^{14}C

Emitter	^{221}Ra	^{222}Ra	^{223}Ra	^{224}Ra	^{226}Ra
R	$\lesssim 10^{-13}$	$\approx 3.10^{-10}$	$\approx 8.10^{-10}$	$\approx 4.10^{-11}$	$\approx 3.10^{-11}$
Log. $T_{1/2}$ (sec)	> 14.4	≈ 11	≈ 15.2	≈ 15.9	≈ 21.3
Final nucleus	^{207}Pb	^{208}Pb	^{209}Pb	^{210}Pb	^{212}Pb

Table 3. Emission of ^{24}Ne.

Emitter	^{230}Th	^{231}Pa	^{232}U	^{233}U	^{234}U
R	$\approx 6.10^{-13}$	$\approx 4.10^{-12}$	$\approx 2.10^{-12}$	$\approx 8.10^{-13}$	$\approx 7.10^{-13}$
Log $T_{1/2}$ (sec)	≈ 24.6	≈ 23.4	$\approx 21.$	≈ 24.8	$\approx 25.$
Final nucleus	^{206}Hg	^{207}Tℓ	^{208}Pb	^{209}Pb	^{210}Pb

3.1 Cluster Models

In the classical model of α-decay, the rate of emission is dominated by the probability for the preformed cluster to cross the Coulomb barrier, which itself depends on a quantity known as the Gamow factor, strongly related to the Q value. Although in the case of heavy clusters the preformation probability is completely unknown, the Gamow factors have been used (assuming a preformation probability of 1) to get an idea of the relative branching ratios. As an example, Table 4 shows (in columns 3 and 4) such calculations for the different possible emissions from ^{227}Ac and its decay chain. It is clear that the emission of ^{14}C by ^{223}Ra is strongly favored, in agreement with the experiments summarized in section 2.1. However, no absolute value of R or $T_{1/2}$ can be obtained in this way without :

a) either an ad-hoc prescription,permitting to "mock-up" the trend of the experimental data, as in the quite successful Square Well model (SqW ; Ref. 11), where the preformation probability is arbitrarily taken as 1., the potential is of the "square well + Coulomb" type, the radius and "assault frequency" being treated as arbitrary parameters, fixed once for all to get the best possible agreement with a large set of experimental data.

b) or a true calculation of the preformation probability, or spectroscopic factor S_a : probability *that the final channel*, with a cluster "a" *and a final nucleus* "A-a", is *already* preformed in the parent nucleus. Such calculations have been performed recently[12] in a microscopic way, for clusters up to ^{16}O, in even-even parent nuclei. The results have been used to derive semi-empirical expressions : $S_a = S_\alpha^{(a-1)/3}$, the α spectroscopic factor S_α being taken ≈ 2 times larger for an even-even parent (S_α (even-even)) than for an odd-A parent (S_α (odd-A)). Quite good results have been obtained using these expressions and a potential that fits heavy ions scattering.

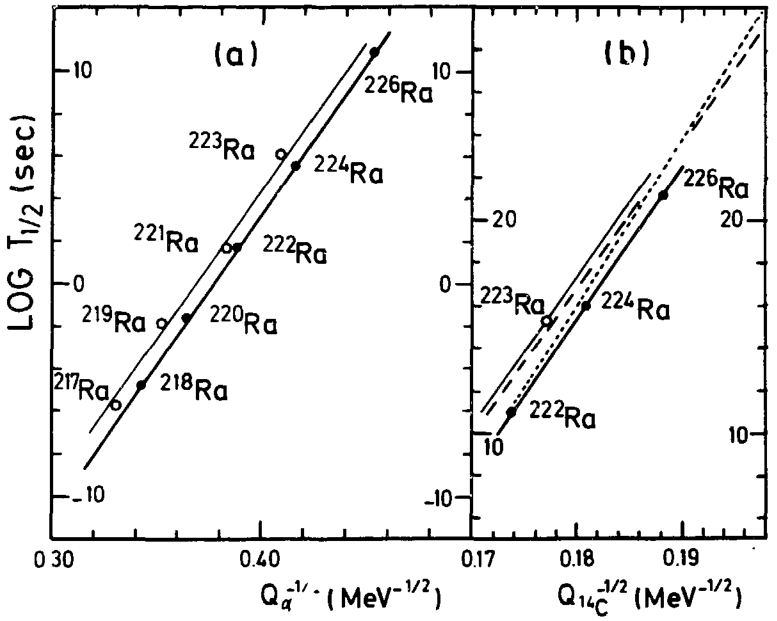

Fig.3. Diagrams of Kurie and Knopf, where the decimal logarithm of the half-life is plotted versus $Q^{-1/2}$, Q being the Q-value of the decay. In (a), the diagram for α-decays of the Ra isotopes. In (b), the diagram of ^{14}C decays of the Ra isotopes. In (b) dashed lines are calculation results of Poenaru et al.[13] and dotted lines are those of Shi and Swiatecki[14].

3.2 Superasymmetric Fission Models

In these models, the potential energy is - like in the fission theory - calculated as a function of the deformation parameter, and used later in computing the emission probability by a quantum mechanical tunneling effect. The deformation parameter is the distance between the separating fragments, assumed to be both spherical. This leads, using some approximations, to an analytical expression depending on a parameter, fixed by a fit : to a large number of experimental data for α-decay, and to the experimental value of R for the ^{14}C emission by ^{223}Ra. Results from such calculations by Poenaru et al.[13] are shown in the last column of Table 4. Another similar model, using the proximity potential, has been developped by Shi and Swiatecki[14], with comparable results.

The half lifes calculated with these fission models are compared to experimental results in Fig.3. A detailed comparison, for the ^{14}C emission by the Ra isotopes.

Table 4. The most probable fragment emissions from ^{227}Ac and its decay chain members (from Ref. 10).

Decays	Q – Values (MeV)	Gamow factor ratio (fragment/α) r_o = 1.15 fm	r_o = 1.20 fm	Branching ratio (fragment/α) (Ref. 13)
$^{227}Ac \rightarrow ^{15}N + ^{212}Pb$	33.30	2.8x10^{-10}	5.6x10^{-9}	4.0x10^{-16}
$^{227}Th \rightarrow ^{19}O + ^{208}Pb$	44.15	4.5x10^{-11}	2.2x10^{-9}	6.3x10^{-17}
$^{227}Th \rightarrow ^{14}C + ^{213}Po$	29.45	5.3x10^{-12}	9.2x10^{-11}	4.0x10^{-16}
$^{227}Th \rightarrow ^{18}O + ^{209}Pb$	44.21	2.1x10^{-10}	9.8x10^{-9}	5.0x10^{-16}
$^{223}Ra \rightarrow ^{12}C + ^{211}Pb$	27.73	6.9x10^{-12}	8.9x10^{-11}	4.0x10^{-15}
$^{223}Ra \rightarrow ^{13}C + ^{210}Pb$	28.85	1.3x10^{-10}	1.9x10^{-9}	4.0x10^{-14}
$^{223}Ra \rightarrow ^{14}C + ^{209}Pb$	31.84	1.2x10^{-5}	1.7x10^{-4}	3.2x10^{-9}
$^{219}Rn \rightarrow ^{14}C + ^{205}Hg$	28.11	2.6x10^{-16}	4.3x10^{-15}	6.3x10^{-21}

including also both the recent exotic cluster preformation calculation by Blendowske and Walliser[12] and the simpler SqW model[11], given in Table 5, shows a reasonable overall agreement.

A more complete and detailed review of existing models can be found in Ref. 10 and 11.

Table 5. Comparison of experimental and theoretical half lifes for ^{14}C emission by the Ra isotopes. ($T_{1/2}$ is expressed in seconds).

	Emitter	^{221}Ra	^{222}Ra	^{223}Ra	^{224}Ra	^{226}Ra
Exp.	(Log. $T_{1/2})_{Exp.}$	> 14.4	≈ 11	≈ 15.2	≈ 15.9	≈ 21.3
Fission models	(Log. $T_{1/2})_{Th.}$					
	Ref. 13	14.3	11.2	15.2	15.9	21.0
	Ref. 14	14.8	11.6	15.7	16.8	22.2
Cluster models	(Log. $T_{1/2})_{Th.}$					
	Ref. 12	14.2	11.8	15.1	16.2	21.1
	Ref. 11	14.1	11.2	15.0	16.0	21.0

4. Fine Structure of Exotic Emissions

4.1 Discovery of a Fine Structure in the Emission of ^{14}C by ^{223}Ra

Due to the strong dependence, discussed in section 3, of the Gamow factor on the Q value, it has been generally considered as quite evident that the decay by exotic cluster emission was always proceeding exclusively by a direct transition to the ground state of the final nucleus. This predominance of the ground state decay - also implicit in the existing models - did not seem so obvious to us and it was decided in 1988 to start a new experimental program to address (and hopefully answer) the question of a possible population of excited levels.

The best test case, as far as the rate of emission was concerned, was naturally the ^{14}C emission by ^{223}Ra...but it was quite demanding on the energy resolution and statistics, the separation between ground state and first excited state of ^{209}Pb being only 779 keV. In our first experiments on ^{14}C emission (section 2.1) the energy resolution, due to detectors,source thickness and statistics, was of the order of 2 MeV (F.W.H.M).

- In order to get improved statistics in a reasonable amount of time, our radiochemists managed to prepare a stronger ^{223}Ra source : starting from an old supply of 150 mg of ^{231}Pa (existing in our laboratory and not treated since 25 years), 70 μg of ^{227}Ac were first recovered. It was not possible to use it directly because the source thickness would have impaired the resolution. Therefore, after about two weeks, the in-grown ^{227}Th was extracted and separated from it. The thin Th source (120 ng of matter), was produced by electrolytic deposition on Pt (for more details, see Ref. 15). The characteristics of the source are given in Table 6.

Table 6. Sources for fine structure studies

Source	Radioactive deposit	Deposit size	Source activity	Method used
^{223}Ra	^{227}Th	0 = 10 mm	1.8 mCi (66 MBq) in ^{223}Ra	Radiochemistry
^{222}Ra	^{230}U	0 = 8 mm	2.3 mCi (85 MBq) in ^{222}Ra	
^{224}Ra	^{224}Ra	$\lesssim$ 4 x 8 mm^2	96 mCi (3550 MBq) in^{224}Ra	Isolde implantation
^{223}Ra	^{223}Ra	15 x 6 mm^2	13 mCi (480 MBq) in^{223}Ra	

- It was decided - because several experiments had already conclusively shown that the emitted cluster was ^{14}C - to use, instead of a telescope of limited size and energy resolution, a simple surface barrier detector of large surface (450 mm^2) built for this purpose and specially selected for good energy resolution with heavy ions (110 keV F.W.H.M with a ^{14}C beam). The increase in size permitted to use SOLENO with a larger solid angle of Ω = 200 msr.

The rate of acquisition was increased in this way by more than one order of magnitude and it was possible to observe 410 ^{14}C events in an 11 days run (to be compared to 11 events in our first measurement[3]). The enormous flux of $\approx 10^7$ α/sec emitted from the source towards SOLENO was very efficiently rejected and did not impair at all the resolution of the experiment. Naturally, a careful energy calibration of all the system, using a ^{14}C beam from our tandem, elastically scattered from a thin Au target, had been performed, as well as background measurements, before the experiment.

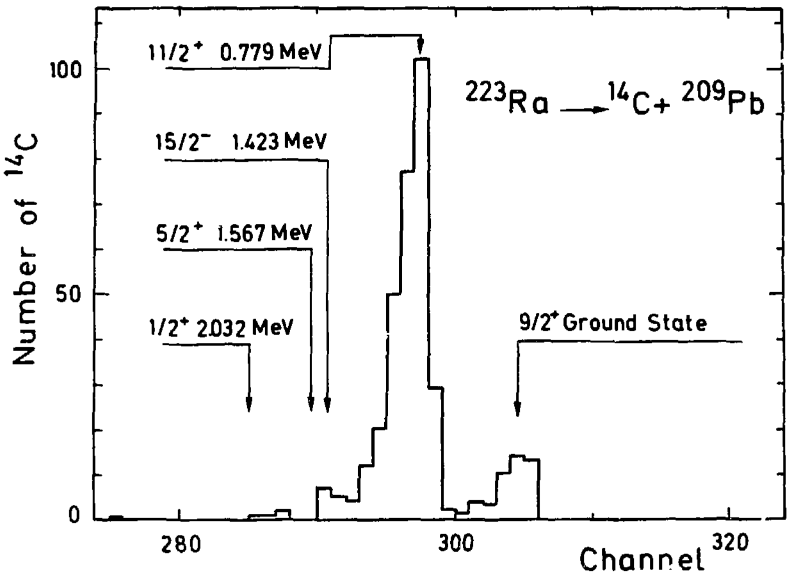

Fig. 4. The histogram is the energy spectrum of ^{14}C particles emitted by ^{223}Ra. The arrows indicate the positions of the ground state and of the first excited states of ^{209}Pb, obtained in the energy calibration with a ^{14}C beam. This spectrum is for a sensitivity range of 4 MeV in excitation energy, corresponding to 50 % transmission of SOLENO.

The energy spectrum observed[16] is shown in Fig. 4. It exhibits clearly two peaks at the positions expected for the ground state and first excited state of ^{209}Pb, and some evidence of population of higher-lying levels, in particular the second excited level at 1423 keV. The energy resolution is 240 keV (F.W.H.M). This spectrum has been obtained by simply adding, without correction, several partial spectra (measured with different currents in SOLENO in order to change the position of the peaks on the transmission curve[16]). After due corrections of efficiencies, it is possible to extract the relative probabilities of population of the levels of ^{209}Pb :

g.s keV : 15 % (± 3)
779 keV : 81 % (± 6)
(1423 keV) : 4 % (± 2)

To summarize : 60 years after the discovery by Rosenblum[17] of the fine structure of the α-emission, a very similar structure is discovered by the Orsay group in the emission of ^{14}C by ^{223}Ra. Strikingly, the dominant decay does not populate the ground state but the first excited state.

4.2 Search for Fine Structure in the Even-Even Emitters

After the above discovery, it seemed important to check the possibility of existence of a fine structure, also for the even-even neighbors ^{222}Ra and ^{224}Ra.

^{222}Ra was studied first, because of its favorable branching ratio ($R \approx 3.10^{-10}$). The measurements had to be performed with a source of ^{230}U constantly generating the short-lived ^{222}Ra ($T_{1/2} \approx$ 37.5 s) as a member of the $^{230}U \rightarrow {}^{226}Th \rightarrow {}^{222}Ra$ family. The ^{230}U itself was prepared and purified from ^{230}Pa, as described briefly in section 2.2 (see Ref. 15 and 18 for details), and electroplated on Pt. Using a very strong source (see Table 6), careful measurements[18] with SOLENO permitted to observe 210 ^{14}C events in 16 days, all but 3 being grouped in a single peak. at the position expected for the ground state of ^{208}Pb (see Fig. 5). The branching ratio is determined as $R = (2.31 \pm 0.31)\ 10^{-10}$, in reasonable agreement with preceding measurements[5,7] and the relative population of the 3^- first excited state of ^{208}Pb, at 2614 keV, is estimated as $< 10^{-2}$. Indeed very simple calculations[11,13,14], based on Gamow factor estimates, predicted an attenuation of the rate by two orders of magnitude for an increase of one MeV in excitation energy, therefore a very small relative population of $\approx 10^{-5}$ for the 3^- level in the present case. This, however, was not considered as a good reason for not studying ^{222}Ra...because the same calculations predicted also a small relative population of $\approx$ 2 % for the first excited level in the decay of ^{223}Ra !

The ^{14}C decay of ^{224}Ra, although its branching ratio is low ($R \approx 4.10^{-11}$), is - due the low energy of 795 keV of its first excited state - an attractive candidate to possibly observe a fine structure : Gamow factor estimates predict the relative population of this state to be $\approx$ 2 % of the ground state one...

^{224}Ra has been studied using very strong (96 mCi, see Table 6) and thin sources, prepared in a new way. They were obtained by implanting mass separated Ra beams from the on-line separator Isolde (CERN) into a vitreous C catcher. The Ra beams were produced by bombarding a thick Thorium carbide target with the 600 MeV proton beam (2.8 μA) of the CERN S.C. Measurements with this source proved more difficult than the preceding ones : i) due to the proximity of the $^{14}C^{6+}$ group corresponding to the ground state of ^{224}Ra and of α particles from the source, the charge state 5^+(instead of 6^+) had to be used at the price of roughly a factor of two reduction in the counting rate ; ii) due to the mode of preparation and to the strength of the source, an Aℓ coating had to be evaporated on it, to reduce the emanation of Radon and to suppress the sputtering of Ra, both giving rise to an α background. An improvement in the pumping still reduced the Radon problem. (for more details, see Ref. 19).

The measurements[19], with SOLENO and a 450 mm^2 Si detector,permitted to record 149 ^{14}C events in about 6 days. All these events are found in a peak (150 keV F.W.H.M) at the position expected for the ground state of ^{210}Pb. No count is observed in the region of excited states (see Fig. 6). An improved value of the branching ratio is determined ($R = (6.5 \pm 1)\ 10^{-11}$) and the relative population of the first excited state is estimated $< 10^{-2}$.

During the ^{224}Ra experiment presently described, a strong (13 mCi) source of ^{223}Ra was also prepared at Isolde and studied at SOLENO, permitting to record 700 ^{14}C events. The results are in reasonable agreement with preceding ones ; unfortunately the peaks, although narrow (150 keV F.W.H.M), have a low energy tail making difficult the observation of weakly populated excited levels above the first excited state of ^{209}Pb.

4.3 Summary of the Experimental Results on the Fine Structure

The main results obtained in our experimental study of the fine structure of the ^{14}C emissions by Ra isotopes are summarized in Table 7. Hindrance factors (H.F).

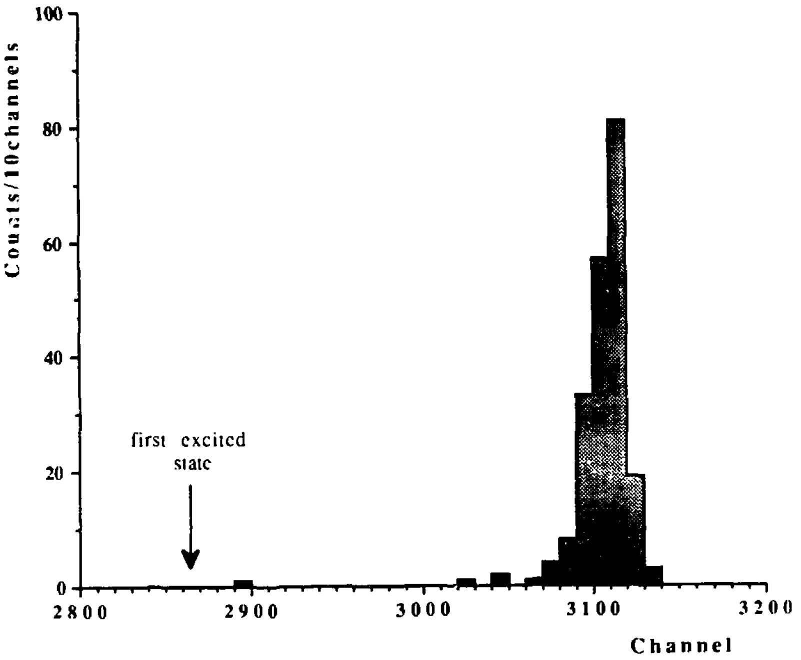

Fig. 5. Total spectrum of the 210 ^{14}C events recorded from the source of ^{222}Ra. The position of the ^{14}C group expected to feed the first excited state of ^{208}Pb is indicated by an arrow.

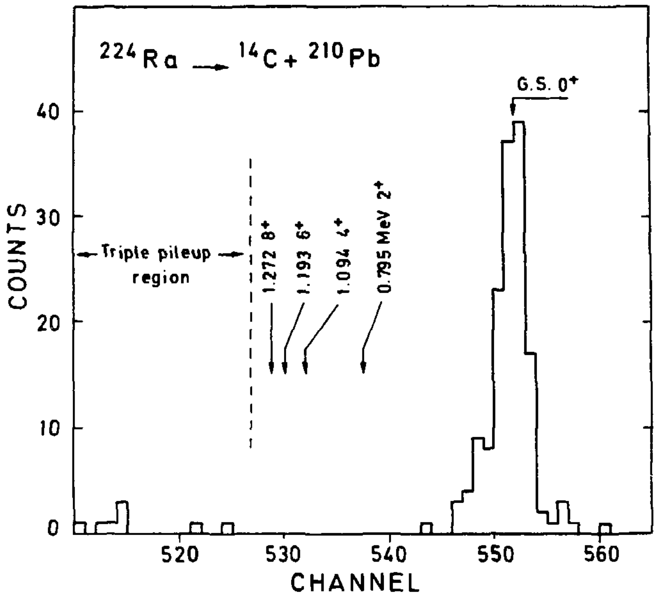

Fig. 6. Total spectrum of the 149 ^{14}C events recorded from the source of ^{224}Ra. The position of the ^{14}C groups expected to feed the ground and some low-lying excited states of ^{210}Pb are indicated by arrows.

which have long been used to characterize the degree of "hindrance" of the different branches in the α decay, are now used for the same purpose for the ^{14}C decay. There are slightly different ways to define these factors : i) They can be defined as the ratio $T_{1/2}(\mathrm{exp.})/T_{1/2}(\mathrm{th.})$, where the theoretical half-life is calculated in the framework of a model fitting the even-even ground states (the SqW model[11] for example, in column 6 of the Table) ; ii) Strictly experimentally, they can be determined, on a Kurie and Knopf diagram (like Fig. 7) as the vertical distances between the points of interest and the straight line connecting the points corresponding to the even-even ground states. Such values are given in colum 7 of Table 7.

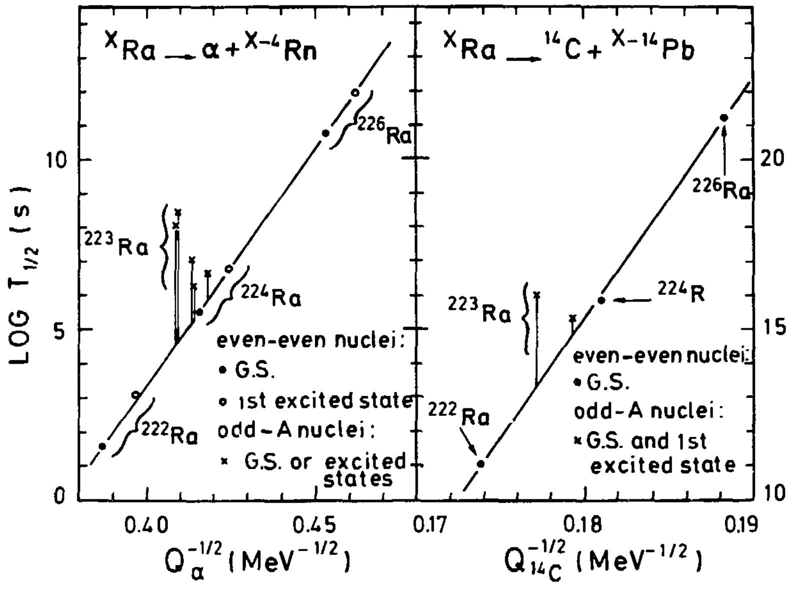

Fig. 7. Kurie and Knopf diagram for α and ^{14}C emissions of Ra isotopes, similar to Fig. 3, but taking into acount the existence of a fine structure.

It should be pointed out that the R value for the ground state of ^{223}Ra is now determined as $R = (1.0 \pm 0.2)\ 10^{-10}$, instead of $6.0.10^{-10}$, value previously adopted for normalization in several predictional models.

5. Interpretation of the Fine Structure

The fission models, indeed, do not contain any information concerning the structure of either the cluster or the heavy nuclei. Their only present possibility could be to predict the *half-life variation with excitation energy, assuming the structure plays no* role.

In principle, cluster models could do better in this case. The results of the SqW model (see Table 7) show an agreement with experimental data which is good for the even-even ground states and reasonable (within a factor of 3) for the excited levels of ^{223}Ra. A recent paper by Blendowske et al.[20] reports similar agreement using S_α (even-even) in all cases. In both models the hindrance factor for the ground state of

Table 7. Main results of the experimental study of the fine structure of the ^{14}C emission by Ra isotopes. ($T_{1/2}$ is expressed in seconds).

Emitter	Level of final nucleus	E_{excit} (keV)	Relative exp. population	Log.$T_{1/2}$ Exp.	Log.$T_{1/2}$ Th[11]	H.F	R	Exp. Ref.
^{223}Ra	g.s	0	15	16.015	13.3	600	$(1 \pm 0.2).10^{-10}$	16, 18, 19
	1st excited	779	81	15.28	14.9	3.0	$(5.2 \pm 0.2).10^{-10}$	16, 18, 19
	2nd excited	1423	4	16.6	16.2	3.0	$\approx (0.26).10^{-10}$	16, 18, 19
^{222}Ra	g.s	0	100	11.2	11.2	1	$(2.31 \pm 0.31).10^{-10}$	18
	1st excited	2614	< 1	> 13.3	16.2	$> 10^{-3}$	$\leq 2.10^{-12}$	18
^{224}Ra	g.s	0	100	15.68	16.0	1	$(6.5 \pm 1).10^{-11}$	19
	1st excited	795	< 1	> 17.89	17.6	> 2	$< 4.10^{-13}$	19

^{209}Pb remains unexplained, this transition being called an "hindered transition". The situation is directly comparable to the one known in the α-decay of odd-A nuclei, in which the decay to the daughter ground state is hindered and at least one transition to an excited state has a low hindrance factor.

A qualitative explanation of the hindrance factors for the ^{14}C decay - very low ($\approx$ 3.) for the first two excited levels of ^{209}Pb at 779 ($J^\pi = 11/2^+$) and 1423 keV ($J^\pi = 15/2^-$), but very high ($\approx$ 600) for the ground state ($J^\pi = 9/2^+$) - has been imagined in 1989, immediately after the experimental discovery of the fine structure, and suggested in different papers[21]. It will be briefly sketched below.

By analogy with the α-decay of odd-A nuclei[22], it is assumed that the preformation probability for the ^{14}C cluster is about the same in ^{223}Ra than in its neighbors ^{222}Ra and ^{224}Ra. The behavior of the odd-A nucleus decay - as compared to the decay of its even-even neighbors - should then be mainly governed by the change in the configuration of the odd-nucleon, more precisely by the overlap between the initial and final nucleon configuration : "favored" transitions are observed when the overlap is large, "hindered" ones when it is small. In our case :

- The leading configurations of the last neutron for the lowest levels of the quasi-spherical ^{209}Pb are well known :

g.s	$9/2^+$	$(2g_{9/2})^1$
779	$11/2^+$	$(1i_{11/2})^1$
1423	$15/2^-$	$(1j_{15/2})^1$

- The spin of the ground state of ^{223}Ra has been measured as J = 3/2 and a recent investigation of the structure of this nucleus by Sheline et al.[23] has concluded that the experimental data are well reproduced by assuming (in addition to a quadrupole deformation $\beta_2 \approx 0.13$) an octupole deformation $\beta_3 \approx 0.1$. This stable octupole deformation implies the existence of a so-called "parity doublet"

$$K^\pi = 3/2^\pm(3/2^+[631] \otimes 3/2^-[761]) \qquad (1)$$

describing the lowest two bands and in particular the ground state.

The odd neutron can be seen as, loosely speaking, "sharing its time" between the two Nilsson orbitals, which arise respectively from the high spin $1i_{11/2}$ and $1j_{15/2}$ shell model orbitals. Therefore, the wave function of the odd neutron in the ground state of ^{223}Ra indeed certainly contains sizable configuration admixtures of the $1i_{11/2}$ and $1j_{15/2}$ shell model orbitals, giving rise to a large overlap and allowing "favored" decays to the corresponding first two excited levels of ^{209}Pb. This is not true for the $2g_{9/2}$ shell model orbital, leading to the observed "hindrance" of the ground state transition. Such an explanation parallels - in its spirit - the one given by Leander and Chen[24] for the ^{223}Ra → ^{219}Rn α-decay.

Ray Sheline and Ragnarsson[25], clearly not aware of the works just described[21], followed later a parallel way. Moreover they performed a calculation[25], in the framework of the reflection - asymmetric rotor model, showing that indeed the ground state of ^{223}Ra has a very large content of the shell-model orbital $1i_{11/2}$ and a very low content of the shell-model orbital $2g_{9/2}$, in agreement with the qualitative interpretation[21] of the hindrance factors for the first excited state and for the ground state of ^{209}Pb. However, this calculation presents two difficulties : i) The calculated energy separation of the two 3/2 band heads is much larger (more than three times)than experimentally found. ii) The ^{223}Ra ground state content of shell-model orbital $1j_{15/2}$ is very small.

Either these two difficulties are linked and show that the calculation has to be improved...or there is a true contradiction between the low $1j_{15/2}$ content theoretically found and the sizeable strength experimentally observed[16] for the transition feeding the 1423 keV ($J^{\pi} = 15/2^{-}$) level of ^{209}Pb. All these points will have to be elucidated.

6. Summary and Discussion

After the pioneering experimental work of Rose and Jones[1] and the quick confirmations[3-6] from all around the world, the existence of a ^{14}C emission by ^{223}Ra has been clearly established. In the next few years other ^{14}C emitters[5,7] and other exotic clusters[7-9], such as Ne, Mg and Si, have been observed. An important new step forward has been the discovery at Orsay of the existence of a fine structure[16] in the ^{14}C decay of ^{223}Ra and the extension of the study to even-even neighbors[18,19].

I feel it is worth to stress here experimental similarities between α-decay and ^{14}C emission : i) There is an odd-even effect in both (see Fig. 3)...indeed related to the fine structure of the odd-A nucleus. ii) The fine structure observed in the ^{14}C decay of ^{223}Ra is similar to the one observed in the α-decay (see Fig. 7) : in both cases the hindrance factor for the ground state of the final nucleus is larger than the hindrance factors for the excited states.

Theoretically, the possibility of exotic emissions has indeed been anticipated[2] before any experiment on this subject was performed. Roughly speaking, the models recently used to describe exotic emissions can be divided into two classes : the "cluster" models, where a preformation is assumed[11,12], and the "fission" models, extending the fission theory to very asymmetric situations[13,14]. A new type of model, describing the exotic cluster as a "soliton" has recently been proposed[26].

Both the "cluster" and the "fission" models have demonstrated their ability to reproduce reasonably the total branching ratios R and half-lifes $T_{1/2}$. This is striking if we remember that these quantities vary enormously with the mass of the cluster and the Q value.

The discovery of the existence of a "fine structure" in the ^{14}C decay has clearly demonstrated the important role played by the nuclear structure in the distribution of the total strength among the levels of the final nucleus, thus favoring the cluster models. Up-to-now the models are able to reproduce reasonably (within a factor of three) the observed results for the even-even emitters and for the decay of the odd-A ^{223}Ra to the excited levels of ^{209}Pb, but only qualitative interpretations (based on more or less precise estimates of the overlap of the initial and final wave functions of the odd neutron) have been proposed[21,25] for the large "hindrance" of the transition to the ground state of ^{209}Pb.

Clearly, an improvement of the cluster models would be welcome...On the other hand, an important point must be clearly confirmed : the existence and intensity of the ^{14}C branch feeding the second excited state of ^{209}Pb in the decay of ^{223}Ra. A high resolution experiment, using the techniques described in Ref. 19 and devised to precisely address this problem, is planned at Orsay in 1992.

It is a pleasure for me to point out and stress the important and decisive part played in the Orsay work reviewed above by Dr. E. Hourani and Dr. M. Hussonnois. I want to acknowledge their help in preparing this review, particularly for a careful and critical reading of the text.

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