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DISCOVERY OF THE RADIOACTIVE DECAY OF ^{223}Ra BY ^{14}C EMISSION AND EXPERIMENTS WITH THE MAGNETIC SPECTROMETER SOLENO

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I. INTRODUCTION

The radioactive decay of ^{223}Ra by ^{14}C nuclei emission was discovered in 1984 by Rose and Jones from the University of Oxford[1]. Essentially, these authors found that ^{223}Ra, in its ground state, which is well known for a long time to be an α-emitter, has a parallel decay by ^{14}C emission with a branching ratio relative to α particles of $(8.5 \pm 2.5) \times 10^{-10}$.

Within a few months following the discovery of Rose and Jones, several groups in different laboratories used various techniques to confirm the discovery[2-5]. The search for other cases led soon to the discovery of the radioactive decay by ^{14}C emission of other isotopes of Ra, i.e. ^{222}Ra[4], ^{224}Ra[4] and ^{226}Ra[6-7]. Evidence of the general character of the phenomenon was obtained through the discovery of the radioactive decay by ^{24}Ne emission of several isotopes, i.e. ^{232}U[8], ^{231}Pa[9], ^{233}U[10] and ^{230}Th[11]. The theoretical predictions of these new radioactivities were published[12] in advance (1980) by A. Sandulescu, D.N. Poenaru and M. Ivascu from Bucharest and W. Greiner from Frankfurt. These authors predicted the existence of radioactivities intermediate between α emission and fission. They showed that ^{14}C should be the most probable emitted ion from ^{222,224}Ra. On the other hand, Rose and Jones did not refer to these predictions neither in the choice of ^{223}Ra as a candidate to be a fragment emitter nor in the explanation of their results. Fortunately, the above cited theoreticians (Poenaru et al.[13-15]), who had derived an analytical superasymmetric fission model, normalized this model to the result of Rose and Jones and provided thereafter realistic predictions which, then, efficiently oriented the following investigators. Another superasymmetric fission model was, also published by Shi and Swiatecki[16)].

Three techniques have been used in the experiments achieved so far :

1) An E x ΔE telescope placed in direct view of a radioactive source. It was the technique used in the pioneering experiment of Rose and Jones[1] and in the subsequent similar experiment of Alexandrov et al.[3]. This technique was rapidly overtaken by the two other more selective techniques.

2) A magnetic spectrometer selecting ^{14}C ions among the high flux of α particles emitted by the same source and directing them toward a detector. A group at Orsay used a spectrometer called SOLENO[17] to confirm[2] the decay of ^{223}Ra into ^{14}C discovered by Rose and Jones and to give evidence[6] of the decay of ^{226}Ra into ^{14}C. A group at Argonne used an Enge Split-Pole spectrometer to measure the mass number of the ^{14}C nuclei emitted by ^{223}Ra[5]. This technique is powerful in the measurement of low branching ratios and of physical characteristics of the fragment like its mass and charge numbers and its kinetic energy.

3) A track recording foil[18], which is sensitive to ions with $Z > 2$. This technique was used by a group from Berkeley in the discovery of the ^{14}C emission from ^{222}Ra[4] and ^{224}Ra[4] and of ^{24}Ne emission from ^{232}U[7]. It was also used by a group at Dubna in the discovery of Ne emission from ^{233}U[10], ^{231}Pa[9] and ^{230}Th[11]. This technique seems to have the capability to measure in the future the smallest branching ratios.

The aim of our present chapter is to review : (i) the experiment of Rose and Jones[1] together with the subsequent one of Alexandrov et al.[3] performed with the same technique and (ii) the experiments[2,6] performed by the group at Orsay on ^{14}C radioactivity of radium isotopes with the magnetic spectrometer SOLENO.

Our review comprises the presentation and comments of the original results and arguments taken in their historical context.

II. EXPERIMENTS WITH THE PIONEERING ExΔE TELESCOPE TECHNIQUE

A. Historical and basic considerations

The existence of radioactivities of heavy nuclei by emission of intermediate mass fragments like C, Ne, Mg,... is a subject which preoccupied several authors in the 1980's. The most engaged authors with regard to this subject were, the members of the group,[12] who gave it a basis on theoretical grounds in their review papers of 1980.

Between 1980 and 1984, besides Rose and Jones[1] who discovered the ^{14}C radioactivity from ^{223}Ra, there was the group of Alexandrov et al.[3] who searched for ^{14}C radioactivities from radium isotopes but without success until the discovery of Rose and Jones. Both groups used traditional tools of quantitative predictions, i.e. the Q-value and Gamow factor considerations. Both had, also, the same detection technique consisting of a Si surface barrier detector telescope ExΔE placed in direct view of a radioactive source.

In order to define the Q-value and the Gamow factor let us consider a heavy nucleus characterized by its charge and mass numbers Z and A and by its mass value M. The decay of this nucleus into a fragment (Z_1, A_1, M_1) and a residual nucleus (Z_2, A_2, M_2) has a Q-value defined as follows :

$$Q = M - M_1 - M_2 \tag{1}$$

When the Q-value for an assumed decay is positive this decay is, energetically, possible and the kinetic energy of the fragment which could be emitted is determined by : $E_1 = Q \times A_2/A$. If one, using a table of masses, calculated the Q-values for decays from a heavy nucleus into a fragment and a residual nucleus, he could find positive Q-values for a large number of parent nuclei and different combinations of fragments and residual nuclei. The highest Q-values one could encounter are those corresponding to decays where the residual nucleus is the doubly magic nucleus ^{208}Pb or a neighboring nucleus.

The traditional Gamow factor for a given decay is associated to the idea that the fragment is preformed inside the parent nucleus and has to cross the coulomb barrier at the surface during its exit. It is the penetration factor by tunneling of the fragment with an energy equal to the Q-value and across the Coulomb potential between the fragment and the residual nucleus. The expression of the Gamow factor in the WKB approximation is :

$$G = \exp\left(-2\int_{r_a}^{r_b} dr(2\mu(U(r)-Q)/\hbar^2)^{1/2}\right) \qquad (2)$$

where $U(r) = Z_1Z_2e^2/r$, $r_a = r_o(A_1^{1/3} + A_2^{1/3})$, r_b is defined by $U(r_b) = Q$ and μ is the reduced mass.

In table 1, there are listed the decays into fragments for some nuclei. For each nucleus, the decays into fragments which are reported are those, among its possible decays, having the highest Gamow factor ratios of fragment to α-particle. Decays of ^{227}Ac and its daughters were taken from ref. 1 and the others from ref. 3.

First, it can be noticed the high sensitivity of the Gamow factor ratios to the variations of r_o. When r_o varies by 0.05 fm, the ratio varies by about one order of magnitude. This fact lowers the confidence in the absolute values of the Gamow factor ratios since the quantity r_o is not determined with a high accuracy.

Then, if, for a given r_o, one compares the values of the Gamow factor ratios of two decays, a large variation is observed. However such a variation is not necessarily meaningful with respect to decay probabilities since a decay probability is equal to a Gamow factor multiplied by a fragment preformation factor which is a quantity completely unknown. It seems that the use of Gamow factor considerations should be restricted to comparisons between two close decays, e.g., two decays from the same parent nucleus or at most from two neighboring nuclei.

B. Experimental procedure

The experimental set up consisted of a ExΔE telescope place in direct view of an ^{227}Ac source. According to the well known property of such a telescope, i.e. discrimination in Z, events corresponding to C ions and α particles should fall on distinct lines in a ExΔE plot. The main experimental conditions used by Rose and Jones[1] and Alexandrov et al.[3] to measure the ^{14}C radioactivity of ^{223}Ra are given in table 2. In fact, both groups had very similar conditions and met, consequently, the same problems.

A source of ^{227}Ac, in which the daughter ^{223}Ra is in secular equilibrium, was chosen because its relatively long half-life of 22 years was convenient for measurements of several months.

The telescope was composed of thin Si detectors of large area. A detector ΔE of a few μm was required in order to allow ^{14}C ions of $\approx$ 30 MeV to cross it and to reach the E detector with a significant part of their energy. Solid angles subtended by the telescopes were as large as a fraction of one steradian.

Despite a short resolving time of 50-100 ns between the ΔE and E signals, multiple pile-up due to the huge rate of α particles generated events extending towards the location of C in the ExΔE plot. Another trouble originating from the high counting rate was the continuous deterioration of the detectors.

C. Results with comments

The results of the experiment of Rose and Jones[1] are presented in the ExΔE plot of figure 1. A group of 11 events is seen inside the dotted lines delimiting the location of the C and at the position of the arrow indicating the energy of ^{14}C nuclei expected to be emitted by ^{223}Ra. The lines and the arrow are an extrapolation of the calibration of the telescope with an α source of ^{241}Am (5.48 MeV). The fact that the group of events falls at the location of ^{14}C expected from ^{223}Ra and does not coïncide with the location of any C isotope expected from ^{227}Ac and its daughters present in the source was in principle a rigourous proof for the decay of ^{223}Ra into ^{14}C. However, the authors had another argument based on preformation probability for the attribution of the observed events to ^{14}C nuclei emitted from ^{223}Ra. The main point in the argument was to compare the preformation probabilities of ^{12}C and ^{14}C inside ^{223}Ra, the ^{12}C as an α-like particle being a priori more likely to be preformed. Taking into account a branching ratio of 8.5×10^{-10} relative to α emission corresponding to the observed events and the Gamow factors of table 1, they deduced a preformation probability of 7×10^{-5} - 4×10^{-7} for ^{14}C and 0.1-10 for ^{12}C relative to that of α emissions. The value obtained for ^{12}C is clearly unacceptable, a fact which excludes the ^{12}C as being the origin of the events.

When the result of Rose and Jones was published, scientists took it with caution. Indeed, several points in their work were questionable mainly : (i) the validity of the preformation considerations, (ii) the accuracy of an extrapolation from 5.5 MeV to 30 MeV in the calibration in energy of the telescope (iii) and the reliability of a very long measurement under multiple pile-up effects and a continuous deterioration of the detectors.

Alexandrov et al.[3] obtained thereafter a result much like that of Rose and Jones (see table 2). They obtained 7 events falling in the ExΔE plot at the location of the expected ^{14}C which was established by a calibration with C and O beams from the cyclotron of Kurchatov.

As soon as the result of Rose and Jones was published a group at Orsay confirmed unambiguously their result with a measurement using a magnetic spectrometer to reject the high flux of α particles and to focus the $^{14}C^{6+}$ ions on a ExΔE telescope.

III. EXPERIMENTS WITH THE MAGNETIC SPECTROMETER SOLENO

A magnetic spectrometer is interesting in ^{14}C radioactivity measurements with its selectivity in magnetic rigidity which allows one to select ^{14}C ions from the very high flux of α particles and to focus them on a detection system. This property, apart from the fact that it protects the detectors against damage by α particles, allows one to use stronger sources and then to measure branching ratios of ^{14}C relative to α particles which are lower than the case of a detection system placed in direct view of a source. The only limit in source intensity originates from the source thickness which should not degrade too much the kinetic energies of the emitted fragments.

The performance of a magnetic spectrometer in ^{14}C radioactivity measurements is higher when its solid angle is larger, at least as far as branching ratio measurements are concerned.

Many spectrometers were certainly considered for ^{14}C radioactivity measurements, but only two led to diffused publications : the superconducting solenoidal coil SOLENO[17] of 100 msr at Orsay and the well-known type ENGE Split-Pole spectrometer at Argonne.

With SOLENO two experiments were performed. The first one confirmed[2] the ^{14}C radioactivity of ^{223}Ra discovered by Rose and Jones. The second one confirmed[6] the ^{14}C radioactivity of ^{222}Ra measured in track detectors by Price et al.[4] and gave evidence of the ^{14}C radioactivity of ^{226}Ra. As to the experiment with the split-pole at Argonne, it measured the mass number of ^{14}C ions emitted by ^{223}Ra[5].

A. Experimental procedure

1. *General characteristics of SOLENO*

This spectrometer is mainly a solenoidal superconducting coil inside a cryostat surrounded by an iron shield and envelopping a cylindrical vacuum chamber. It has a perfect azimutal symmetry. Designed to operate at zero degree with respect to the beam of the Tandem MP of Orsay, it has a solid angle of 100 msr. These characteristics were exploited to detect light particles emitted around zero degree in (^{3}He,$\alpha\gamma$) reaction and to collect the products of fusion-evaporation reactions induced by heavy ions at Tandem energies.

In ^{14}C radioactivity measurements the spectrometer is disconnected from the beam pipe and the radioactive source and the detection system consisting of a ExΔE telescope are placed in the vacuum chamber on the symmetry axis (Fig. 2.a).

After the cryostat is filled with liquid helium, the electric current is set to focus $^{14}C^{6+}$ ions on the telescope. For this setting the doubly charged α particles, α^{++}, are focussed well before the telescope and the singly charged α particles, α^{+}, are focussed well behind the telescope, so both of them do not impinge on the telescope (Fig. 2.a).

2. *Transmission curve of SOLENO*

The focussing efficiency of SOLENO is described by its transmission curve which is the plot of the effective solid angle Ω against the magnetic rigidity of the focussed ion[17](Fig. 2.b). Such a curve is established under each precise geometry used, i.e. for definite diameters and relative distances of the source and the detectors. It is obtained with an α line of a calibrated source by recording the number of α's counted in the telescope versus the electric current in SOLENO. Actually, for each experiment with a given source, an associated transmission curve was established using a thin source made of the same material and of equal diameter. The thin source was calibrated in intensity in a separate measurement using a solid-state detector of known area and placed at a determined distance.

Finally, for each experiment, the associated transmission curve served to determine the electric current setting to focus ^{14}C ions and to estimate the effective solid angle of collection for the focussed ions as follows :

- Let I_α be the current corresponding to the maximum of the transmission curve Ω_{max} obtained with an α-line of energy E_α, the current setting which focus $^{14}C^{6+}$ ions of energy E_C is :

$$I_C = \left(\frac{\sqrt{14E_C}}{6} \Big/ \frac{\sqrt{14E_\alpha}}{2}\right) \times I_\alpha \qquad (3)$$

- For a thin source, the ^{14}C ions emitted have a sharp energy. Their representative point on the transmission curve falls at the maximum, giving an effective solid angle equal to Ω_{max}. For a thick source, ^{14}C ions, e.g. $^{14}C^{6+}$, have a finite spectrum in magnetic rigidity. The effective solid angle is then an average value weighted by this spectrum.

3. *General characteristics of the sources*

The general characteristics of the sources used at Orsay are given in table 3. Sources of ^{227}Ac and ^{230}U were taken to generate ^{223}Ra and ^{222}Ra as daughters. The ^{230}U was obtained by irradiating a thorium target with 34 MeV protons from the cyclotron of Orléans. The source of ^{227}Ac was 300 times stronger than the one used by Rose and Jones. The diameters of the sources were chosen smaller than 16 mm because the collection efficiency of SOLENO from the outer part becomes low. Only the ^{226}Ra source was thick consequently to its strong activity and its long half-life of 1600 years. A problem common to all of the three sources was the existence of an isotope of Rn as a member of their decay chains. Radon, which is a gas, emanates from the source and diffuses across its window. Then it travels inside the vacuum chamber and activates with its daughters the environment of the telescope.

4. *Detection set up*

The telescope consisted of silicon surface barrier detectors made at Orsay. The ΔE detector was 9 µm thick with an area of 200 mm² whereas the E detector was 200 µm thick with an area of 300 mm². A standard electronic set up delivered three parameters : the energies E and ΔE and the time T as given at the output of a TAC circuit (time-to-amplitude couverter) between the two channels. An energy range of 70 MeV was covered in E and of 50 MeV in ΔE. The three parameters E, ΔE and T (with E, ΔE correlated or not) were recorded on magnetic tape in event-by-event mode and checked on line during the experiment. The thresholds on E and ΔE were adjusted for each source to limit the counting rate of the stored events to a few counts per second.

B. Confirmation of the discovery of Rose and Jones

The experiment using ^{227}Ac source was planned with the purpose to overcome the difficulties encountered in the experiment of Rose and Jones which were, mainly, a very high counting rate of α particles producing multiple pile-up with the deterioration of the detectors and an indirect calibration in energy of the telescope. Indeed, these difficulties were completelyremoved. After establishing the transmission curve, a run of 5 days was achieved, in which the counting rate was very low, i.e. a few counts per second. It was followed by a run of 5 days without source and current in SOLENO, which gave an estimate of the background. Finally a calibration of the telescope with beams from the Tandem was carried out. For the calibration, the source was taken out and the spectrometer was connected to the beam pipe. An elastic scattering of the beam on a self-supporting target of ^{12}C, 20 µg/cm² thick, was performed. Scattered ions within the range of θ of 8.1° to 8.5° were focused with the spectrometer on the telescope.

In figure 2.b, is shown the transmission curve. When a current setting of 285 A focuses $^{14}C^{6+}$ ions of 29.7 MeV expected from ^{223}Ra, the corresponding position of these ions comes at the maximum of the transmission curve whereas $^{12}C^{6+}$ of 26.1 MeV and $^{13}C^{6+}$ of 27.0 MeV expected from ^{223}Ra fall outside the transmission curve. The $^{18}O^{8+}$ of 40.4 MeV and $^{20}O^{8+}$ of 39.0 MeV expected to be emitted from ^{227}Th and ^{227}Ac respectively are inside the transmission curve, but these ions are separable by their energy measured in the telescope which is very different from that of ^{14}C nuclei emitted by ^{223}Ra. The α-particle of 5-7 MeV emitted by ^{227}Ac and its daughters fall far outside the transmission curve : to the left when they are doubly charged and to the right when they are singly charged. Only singly charged α^{+} degraded in energy to 3 MeV fall within the transmission curve, but the number of such particles is only a few per second for a source as thin as the ^{227}Ac source which was used.

In figure 3.a, are shown the results of the measurement with the source of ^{227}Ac. Each dot represents an event. At low energy E, where there are a large number of events, countour lines were plotted. Three sets of dashed lines are the result of the calibration. They represent the contour diagrams (at a half and a tenth of the maximum) of the responses of the telescope to ^{12}C et ^{14}C beams having the energies of ^{12}C and ^{14}C nuclei expected from ^{223}Ra and to ^{16}O beam having the energy of ^{18}O and ^{20}O expected from ^{227}Th

and ^{227}Ac. Clearly, there are three types of events in figure 3.a :

- At low energy E up to 13 MeV a large number of events is seen originating from α particles emitted by the radon and its daughters deposited at the region of the telescope.

- At high energy E, apart from a group of 11 events located at $E \approx 23$ MeV, there are six events dispersed over the plane E×ΔE. Obviously, six similar events were found in the background measurement of figure 3.b. The analysis revealed that this type of events was due to a slight contamination with a ^{252}Cf source during a previous test of the telescope.

- The group of 11 events corresponds to a total energy $E+\Delta E = 29.4$ MeV very close to the value 29.7 MeV expected for ^{14}C nuclei emitted by ^{223}Ra. Compared to the results of the calibration, the location of this group coincide with that corresponding to ^{14}C from ^{223}Ra and is very distinct from the location of ^{12}C originating in ^{223}Ra or of ^{20}O and ^{18}O from ^{227}Ac and ^{227}Th respectively. Therefore, the group of events was identified with ^{14}C nuclei emitted by ^{223}Ra, a fact which confirmed the result of Rose and Jones.

It was checked that among all possible decays into fragments of ^{227}Ac and its daughters, apart from the decay of ^{223}Ra into ^{14}C, no one could give fragments fulfilling the three stringent following conditions :

- A total energy of 29.7 MeV
- A location in the E×ΔE plane compatible with that of the group of the observed events.
- A favorable position on the transmission curve of SOLENO.

So, the measurement with SOLENO confirmed unambiguously the decay of ^{223}Ra into ^{14}C, without referring to Gamow factors or emission probabilities.

The branching ratio (^{14}C/α) in the decay of ^{223}Ra was deduced from the number N_C of observed events as follows :

$$\text{B.R.} = N_C/N_\alpha \times 4\pi/\Delta\Omega \times 1/\Sigma \qquad (4)$$

where N_α is the total number of ^{223}Ra nuclei having decayed by α emission during the measurement, ΔΩ is the effective solid angle of SOLENO (0.115 ± 0.002 sr) and Σ the percentage of $^{14}C^{6+}$ in the charge state distribution.

A value of $(5.5 \pm 2.0) \times 10^{-10}$ for the branching ratio was deduced which is in good agreement with the value obtained by Rose and Jones.

C. Confirmation of the decay of ^{222}Ra by ^{14}C emission and evidence for the decay of ^{226}Ra by ^{14}C emission

As soon as the result of Rose and Jones was published[1] and its confirmation by the group of Orsay was known[2], Poenaru et al.[14] published the realistic predictions of their analytical superasymmetric fission model, followed by Shi and Swiatecki[16] who gave in another formulation an equivalent model. In fact, the predictions of these models oriented efficiently the investigators in the choice of the sources to study and so contributed very much in the rapid obtention of the following results with track-recording foils : the decay of ^{222}Ra[4] and ^{224}Ra[4] by ^{14}C emission and of ^{232}U[8] and ^{231}Pa[9] by ^{24}Ne emission.

All these experimental and theoretical findings led, since that moment, to the present understanding of the phenomenons i.e. : a large number of heavy nuclei are fragment emitters and each of them could emit simultaneously several types of fragments with different branching ratios relative to α emission.

Within this context, the second experiment with the spectrometer SOLENO was planned. New detectors were made at Orsay free of contamination and intense sources were prepared in order to reach branching ratios as low as 10^{-12} relative to α emission. The objectives of the experiment were to completement the study of radium isotopes and to search for a new type of radioactivity, i.e. the decay of ^{241}Am into ^{34}Si. The first objective, as it will be seen, was successfully achieved by confirming the ^{14}C radioactivity of ^{222}Ra already measured by Price et al. in track-recording foil and by giving evidence for the ^{14}C radioactivity of ^{226}Ra. The second objective gave only an upper limit for the radioactive decay of ^{241}Am into ^{34}Si.

The experiment began with a measurement of the background (no source and no current in SOLENO) of 5 days, which was repeated at the end. No events were observed in the E$\times\Delta$E plot.

With the 6.556 MeV α line of ^{222}Ra, two transmission curves were made with two weak sources of ^{230}U of two different diameters, 10 mm and 16 mm, which were the two values of diameters used for the strong sources.

A measurement of 3.6 d was performed with the ^{230}U source. The current of SOLENO was set to focus ^{14}C^{6+} ions of 30.48 MeV.

Then a measurement of 9.3 d was carried out with a ^{226}Ra source. Because the source was relatively thick, the current of SOLENO was set to focus ^{14}C^{5+} (most probable charge state at the present energy) expected from ^{226}Ra and presumed to be slowed down by half the thickness of the source.

Finally, a measurement of 8.7 d was devoted to the source of ^{241}Am. The current of SOLENO was set to focus ^{34}Si of 80.93 MeV expected from ^{241}Am.

These measurements were followed by the precise calibration in energy of the telescope with a ^{14}C beam from the Tandem. Elastic scattering of the ^{14}C beam on a Au target of 50 μg/cm^2 was used. The telescope was mounted in front of a window of the reaction chamber at 30° from the zero degree of the beam. Several chosen incident energies of the beam gave scattered ions of the same energies as those expected from the sources. A channeling effect in the detector ΔE was observed in the spectra.

In figure 4, the transmission curve corresponding to the geometry used with the strong source of ^{230}U is shown. This source being thin, the position corresponding to the focussing in the measurement is the maximum solid angle equal to 98 msr. It can be seen, also, that the doubly charged α^{++} in the range of 10-13.3 MeV and the singly charged α^{+} in the range of 2.5-3.3 MeV fall inside the transmission curve. However both of these intervals of energy do not correspond to real values in α emissions from a ^{230}U source.

The bidimensionnal plot in figure 5 is the plot of the results of the measurement with ^{230}U source. Two groups of events are seen. The group at low energy originates from α particles. The other group consisting of 10 events coincide with the location of ^{14}C from ^{222}Ra, given by the dashed line, established in the calibration with ^{14}C beam. It has a mean total energy $E+\Delta E$ agreeing to within 0.4 MeV with the value of 30.5 expected for ^{14}C nuclei emitted from ^{222}Ra. One event is separated from the group under the channeling effect in the ΔE detector. The group of 10 events was identified with ^{14}C nuclei emitted by ^{222}Ra. Taking into account the number of α par ticles emitted by ^{222}Ra throughout the measurement,the solid angle of SOLENO and the percentage (0.65) of $^{14}C^{6+}$ ions in the charge state distribution,a $^{14}C/\alpha$ branching ratio for ^{222}Ra of $(3.1\pm1.0) \times 10^{-10}$ free of systematic error was deduced. It is in good agreement with the result of Price et al.[4]

In ^{226}Ra measurement, the important thickness of the source was the origin of a low-energy tailing in the energy spectrum both of the α-particles emitted by the source and of the ^{14}C nuclei expected to be emitted from ^{226}Ra. The tailing in the spectrum of α-particles increased considerably the number of α^{+} in the interval of 2.6 to 3.4 MeV falling within the transmission curve of SOLENO (fig. 6). Because a large part of these transmitted α-particles was stopping in the ΔE detector, the counting rate of this detector increased reaching about 7000 c/s. On the other hand, the expected tailing of the ^{14}C nuclei was calculated from that measured for a particles and was used to deduce the momentum distribution of the $^{14}C^{5+}$ shown as a dashed line in fig. 6. The product of this spectrum by the transmission curve gave an average solid angle of 36 msr.

In figure 7, are presented the results of the measurement. The single E-type events and the single ΔE-type events shown in the figure originate from α-radiation. Double and triple pile-up are seen in ΔE spectrum. In the E spectrum peak (a) is due to parasitic noise which was observed during the background measurment, whereas peak (b) and the continuum originate from ^{222}Rn and its daughters. In the ExΔE plot, there is one group at low energy originating from α-particles and six dispersed events which were labelled from 1 to 6. The location of ^{14}C ions, is given in a continuous line (central location) and in two dashed lines (contour limits at 1/10 of the central location). The two events labelled 5 and 6 cannot be identified with ^{14}C nuclei due both to their location in the ExΔE plane and their corresponding positions in the transmission curve figure (fig. 6). On the contrary, they could be explained as random coincidences. As to the events labelled 1-4, they cannot be explained as due to random coincidences, but they fall inside the ^{14}C location in figure 7 and inside the transmission curve. Therefore, these events were assigned as ^{14}C nuclei. On the other hand, the most probable decays into fragments of ^{226}Ra and its daughters, according to the predictions of the superasymmetric models of refs. 14 and 16, were considered. It appeared that the decay of ^{226}Ra into ^{14}C not only has the highest fragment/α branching ratio but also is the only decay which gives the energies of the event labelled 1-4. So, the four events were identified with ^{14}C nuclei emitted by ^{226}Ra. The ^{14}C/α branching ratio deduced from the four events was $(3.2 \pm 1.6) \times 10^{-11}$.

The decay of ^{226}Ra into ^{14}C was confirmed[7] afterwards with track-recording foils in a collaboration between the groups of Orsay and Berkeley. A branching ratio of $(2.9 \pm 1.0) \times 10^{-11}$ has been found, in excellent agreement with the measurement with SOLENO.

The measurement with the source of ^{241}Am gave no events in the E×ΔE spectrum. An upper limit of 3 x 10^{-12} for ^{34}Si/α branching ratio from ^{241}Am was set. In addition, since no event has been observed, this measurement with strong source and current in SOLENO has been considered as an ideal background measurement confirming the background measurment performed without source and current in SOLENO.

IV. SUMMARY

The measured branching ratios ^{14}C/α of radium isotopes are reported in table 4 together with the values calculated in the superasymmetric fission model. The agreement between measured and calculated values is within an order of magnitude. The agreement can be considered relatively good since the model is quite simple and general.

These results measured or confirmed with the spectrometer SOLENO contributed very much to constitute the systematics of ^{14}C emission from radium isotopes. Indeed, when upper limits were, thereafter, determined precisely for ^{14}C emission from ^{221}Ra and ^{225}Ra, the plot of the ratio of the measured to calculated branching ratios for the whole series (from ^{221}Ra to ^{226}Ra) showed a clear even-odd effect in the parent nucleus[7]. Hindrance effects for even-odd parents relative to even-even ones as large as two orders of magnitude in branching ratios were found. On the other hand, the systematic study of radium isotopes allowed to study the influence of the deformation of the nuclei on the ^{14}C emission[19,20]. It was concluded[20] that there was neither striking improvement nor visible deterioration brought in the agreement between measured and calculated branching ratios when the calculation takes into account deformation of the nuclei.

V. CONCLUSION

The discovery of the decay of ^{223}Ra into ^{14}C by Rose and Jones was unambiguously confirmed by the group at Orsay using the magnetic spectrometer SOLENO. In addition, the group at Orsay actively participated in building the systematics of results for ^{14}C/α branching ratios measurements of the radium isotopes.

The mutual excellent agreement in branching ratio measurements among the three techniques, ExΔE telescope, the spectrometer SOLENO and the track detector, reflected, finally the reliability and accuracy of each of them.

The capability of the spectrometer SOLENO in branching ratio measurement is limited by the tolerable thickness of the source which should not exceed, for instance, 1mg/cm^2. Nevertheless with such a thickness and for a half-life of the source shorter than 10 years, a measurement of 10 days can reach branching ratios as low as 10^{-13}.

However, it is the capability to measure with SOLENO physical quantities which is at present under study at Orsay. The precise measurement of the energy of the fragments should answer the question whether or not excited states in the daughter are populated in the newly discovered radioactivities.

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Decays into fragments	Q-Value (MeV)	Gamow factor for decay / Gamow factor for α-emission		Ref.
		$r_0 = 1.15$	$r_0 = 1.2$	
$^{227}\mathrm{Ac} \rightarrow {}^{15}\mathrm{N} + {}^{212}\mathrm{Pb}$	33.3	2.8×10^{-10}	5.6×10^{-9}	1
$^{227}\mathrm{Th} \rightarrow {}^{18}\mathrm{O} + {}^{209}\mathrm{Pb}$	44.2	2.2×10^{-10}	1.1×10^{-8}	1
$^{223}\mathrm{Ra} \rightarrow {}^{12}\mathrm{C} + {}^{211}\mathrm{Pb}$	27.7	6.7×10^{-12}	8.5×10^{-11}	1
$\rightarrow {}^{13}\mathrm{C} + {}^{210}\mathrm{Pb}$	28.8	1.1×10^{-10}	1.7×10^{-9}	1
$\rightarrow {}^{14}\mathrm{C} + {}^{209}\mathrm{Pb}$	31.8	1.2×10^{-5}	1.7×10^{-4}	1
$\rightarrow {}^{15}\mathrm{C} + {}^{208}\mathrm{Pb}$	29.1	5.3×10^{-12}	1.0×10^{-10}	1
$^{233}\mathrm{U} \rightarrow {}^{24}\mathrm{Ne} + {}^{209}\mathrm{Pb}$	60.5	-	4.52	3
$\rightarrow {}^{25}\mathrm{Ne} + {}^{208}\mathrm{Pb}$	60.8	-	5.68	3
$^{236}\mathrm{Pu} \rightarrow {}^{28}\mathrm{Mg} + {}^{208}\mathrm{Pb}$	79.7	-	1.20	3
$^{240}\mathrm{Cm} \rightarrow {}^{32}\mathrm{Si} + {}^{208}\mathrm{Pb}$	97.6	-	1.32	3

Table 1 - Gamow factors for some decays into fragments normalized to Gamow factors for the decays into α from the same parent nuclei.

Experimental conditions	Experiment of Rose and Jones[1]	Experiment of Alexandrov et al.[3]
Source	^{227}Ac of 3.3 µCi	^{227}Ac of 85 µCi
Detector ΔE	8.2 µm x 200 mm²	16 µm
Detector E	300 mm²	500 µm
Solid angle ΔΩ	1/3 sr	1/10 sr
Counting rate in α	4000 c/s	25 000 c/s
Measurement time	189 d	30 d
Number of ^{14}C nuclei detected	11	7
^{14}C/α branching ratio	$(8.5\pm2.5) \times 10^{-10}$	$(7.6\pm3.0) \times 10^{-10}$

Table 2 - Experimental conditions of the measurements performed with a telescope E x ΔE placed in direct view of the radioactive source.

Source	Radioactive deposit	Thickness of the deposit	Diameter of the deposit	Activity of the source
^{223}Ra	^{227}Ac	-	8 mm	210 µCi in ^{223}Ra
^{222}Ra	^{230}U	-	10 mm	600 µCi in ^{222}Ra
^{226}Ra	SO_4Ra	1.8 mg/cm²	16 mm	2.5 mCi in ^{226}Ra
^{241}Am	Am_2O_3	1.1 mg/cm²	16 mm	7 mCi in ^{241}Am

Table 3 - General characteristics of the sources used at Orsay.

Decay	Q (MeV)	^{14}C/α measured branching ratios	Ref.	^{14}C/α calculated B.R.	
				Ref. 14-15	Ref. 16
^{222}Ra → ^{14}C+^{208}Pb	33.05	$(3.7\pm0.5)\times10^{-10}$	4	1.0×10^{-11}	1.7×10^{-9}
		$(3.1\pm1.0)\times10^{-10}$	6		
^{223}Ra → ^{14}C+^{209}Pb	31.85	$(8.5\pm2.5)\times10^{-10}$	1	3.2×10^{-9}	6.9×10^{-9}
		$(5.5\pm2.0)\times10^{-10}$	2		
		$(7.6\pm3.0)\times10^{-10}$	3		
		$(6.1\pm0.8)\times10^{-10}$	5		
		$(4.7\pm1.3)\times10^{-10}$	4		
^{224}Ra → ^{14}C+^{210}Pb	30.54	$(4.3\pm1.1)\times10^{-11}$	4	1.6×10^{-12}	6.2×10^{-11}
^{226}Ra ^{14}C+^{212}Pb	28.21	$(3.2\pm1.6)\times10^{-11}$	6	2.0×10^{-12}	3.1×10^{-11}
		$(2.9\pm1.0)\times10^{-11}$	7		

Table 4 - Radioactive decays into ^{14}C of radium isotopes with their measured and calculated branching ratios relative to α particles.

FIGURE CAPTIONS

Fig. 1 - Results of the measurement of Rose and Jones obtained with an E x ΔE telescope in direct view of an ^{227}Ac source. The arrows and the dotted lines are the results of a calibration of the telescope with an α-source : the arrows indicate the energies of ^{12}C and ^{14}C nuclei which could be emitted by ^{223}Ra and the dotted lines delimit the location of the C isotopes in the E x ΔE plane. The dots represent the observed events which were identifed with ^{14}C nuclei emitted by ^{223}Ra. The lower cross is a quadruple pileup from α-particles and the upper cross is an event which was recorded under a thunderstorm. (From ref. 1, used by permission of MACMILLAN JOURNALS LTD).

Fig. 2 - Description of the magnetic spectrometer SOLENO of Orsay used in ^{14}C radioactivity measurements.

(a) Experimental set up inside the vacuum chamber of the spectrometer : A - entrance aperture (±20°), 1 - source location, 2 - obturator (±4°) preventing the source and the telescope to be in direct view with each other, 3,4 - telescope E x ΔE. There are presented the paths of focussed $^{14}C^{6+}$ ions and of unfocussed α^+ and α^{++} particles.

(b) Transmission curve of SOLENO : effective solid angle subtended by SOLENO with respect to the source versus the magnetic rigidity of the incident ions normalized by the central value. Here, the current setting of SOLENO was adjusted to focus the $^{14}C^{6+}$ ions of 29.7 MeV expected to be emitted by ^{223}Ra. For this current setting, the focussed $^{14}C^{6+}$ fall at the maximum of the transmission curve, while degraded α^+ particles and various fragments which could be emitted by ^{227}Ac and its daughters fall at the positions indicated by the arrows.(From ref. 2, used by permission of the American Physical Society).

Fig. 3 - Bidimensional E x ΔE spectrum displaying the results of the measurement performed at Orsay with an ^{227}Ac source and a telescope E x ΔE placed in the image plane of the spectrometer SOLENO. In this plot, E is the energy measured in the detector labelled E and is not the total energy E + ΔE. Each dot represents an event. In regions where a large number of events is present, contour lines are drawn.

(a) The results of the measurement and of the corresponding calibration. Dashed lines labelled ^{12}C and ^{14}C give the location, according to the calibration, of ^{12}C and ^{14}C expected to be emitted by ^{223}Ra while lines labelled ^{16}O give the location of ^{20}O or ^{18}O expected from ^{227}Ac or ^{227}Th. At low energy E, events are due to α particles. At high energy E, a group of events coincide with the location of ^{14}C nuclei expected from ^{223}Ra and six dispersed events are due to a background.

(b) The results of the background measurement. Dispersed events are observed revealing the existence of a previous contamination of the detectors. (From ref. 2, used by permission of the American Physical Society).

Fig. 4 - Transmission curve of SOLENO established in the experimental conditions of the measurement with ^{230}U source. The current of SOLENO was set to focus $^{14}C^{6+}$ of 30.48 MeV ($\langle B\rho \rangle$ = 0.496 T.m) expected to be emitted by ^{222}Ra. The positions of some α^{++} and α^{+} particles are given. (From ref. 6, used by permission of North-Holland Publishing Co).

Fig. 5 - Results of the measurement performed with ^{230}U source and with the spectrometer SOLENO. At low energy E, there are events originating from α-particles. At high energy E, a group of events coincides with the location of ^{14}C nuclei emitted by ^{222}Ra. The location given in dashed lines is the result of the calibration with ^{14}C beam. (From ref. 6, used by permission of North-Holland Publishing Co).

Fig. 6 - Transmission curve of SOLENO established under the experimental conditions of the measurement with ^{226}Ra source. The current of SOLENO was set to focus $^{14}C^{5+}$ of 22.12 MeV ($\langle B\rho \rangle = 0.507$ T.m). The dashed line is the momentum distribution of $^{14}C^{5+}$ ions expected from the rather thick source of ^{226}Ra. Labels 1-6 are the $^{14}C^{5+}$ positions of the six events observed at high energy in the E x ΔE plot of fig. 7. The position of source α^{++} and α^{+} are given : the α^{+} particles in the range 2.6-3.4 MeV fall inside the transmission curve. (From ref. 6, used by permission of North-Holland Publishing Co).

Fig. 7 - Results of the measurement performed with a source of ^{226}Ra and with the spectrometer SOLENO. In E x ΔE-type event spectrum, dots labelled 1-4 *represent* ^{14}C *nuclei emitted by* ^{226}Ra, *while all other dots represent* random coincidences between ΔE-type and E-type events ; the continous and dashed lines give the location of the ^{14}C nuclei expected from ^{226}Ra according to the calibration with ^{14}C beams. In the single E-type and ΔE-type spectra, events originate from α particles either of low-energy focussed directly on the telescope or of high energy (5-7 MeV) emitted by Rn and its daughters deposited around the detectors. (From ref. 6, used by permission of North-Holland Publishing Co).

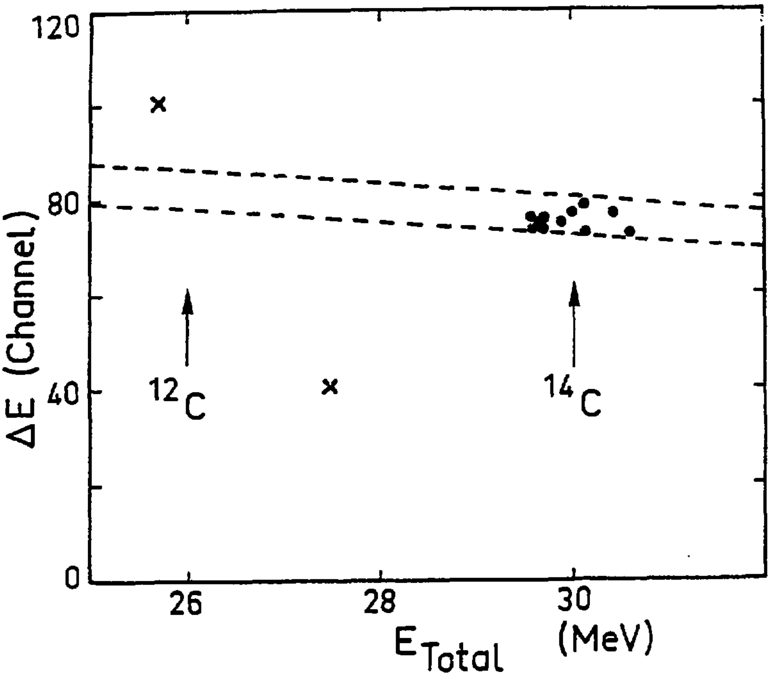

Figure 1
(Hourani)

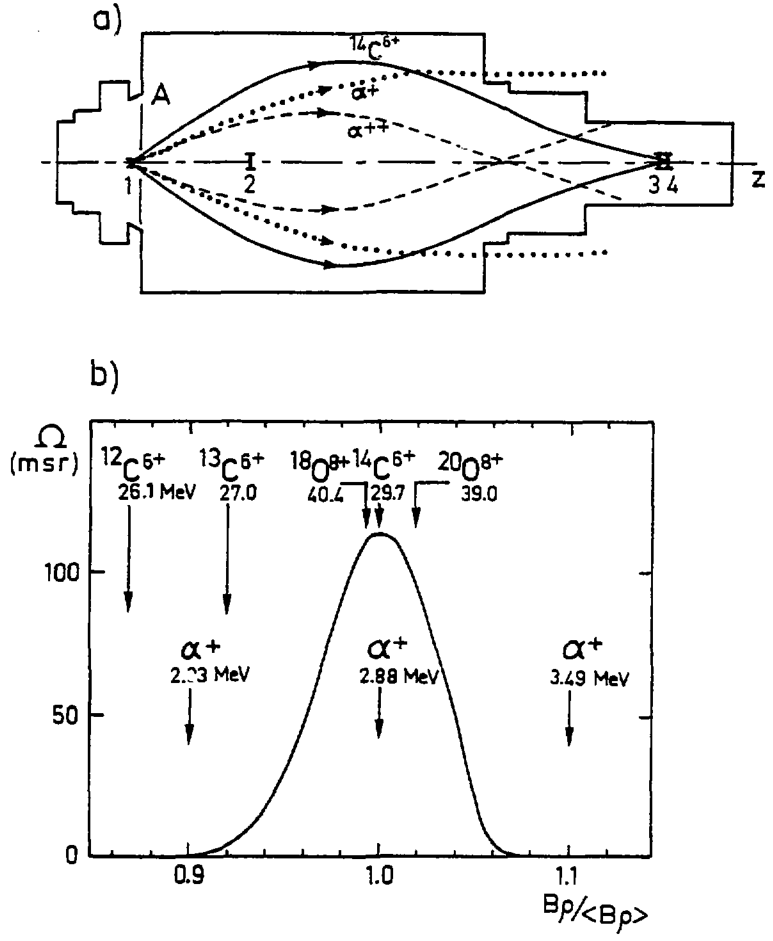

Figure 2
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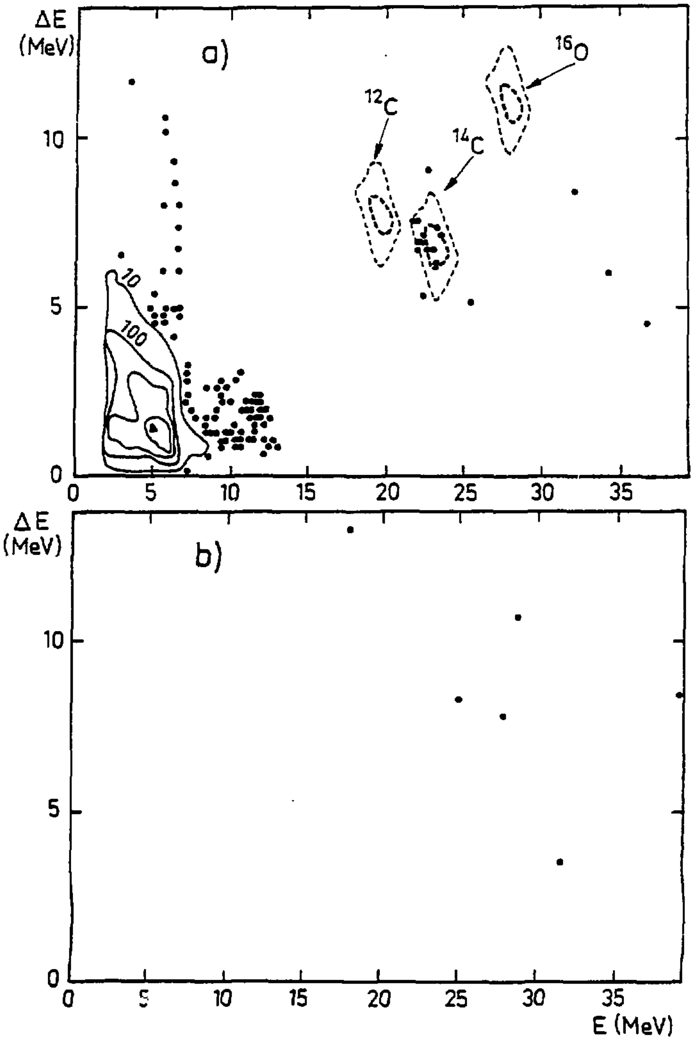

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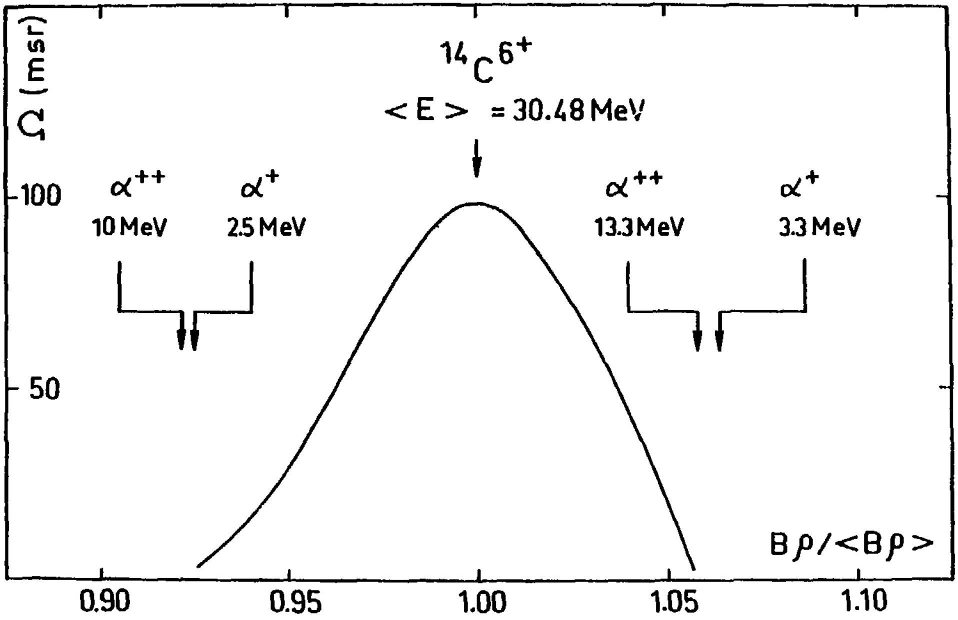

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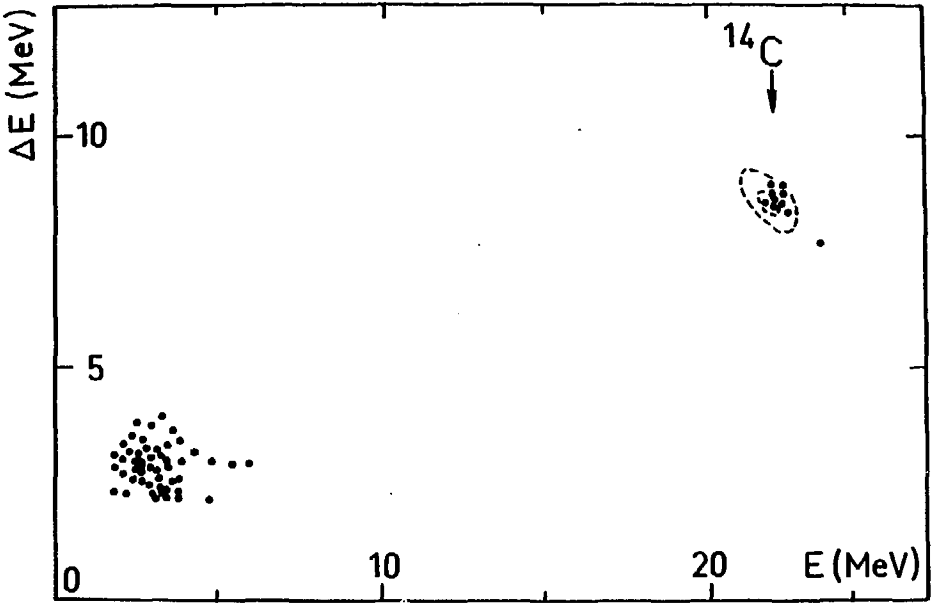

Figure 5
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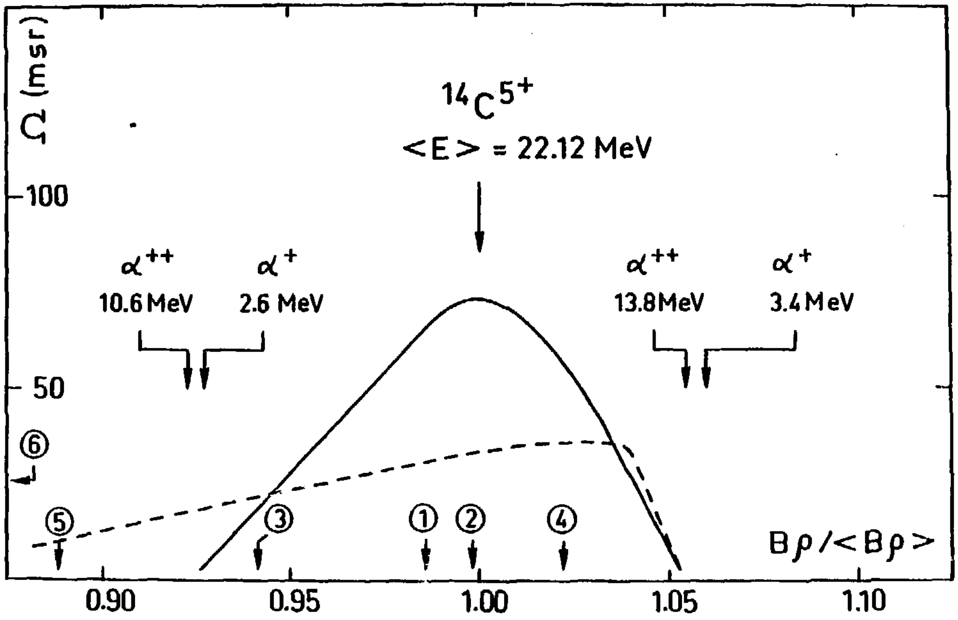

Figure 6
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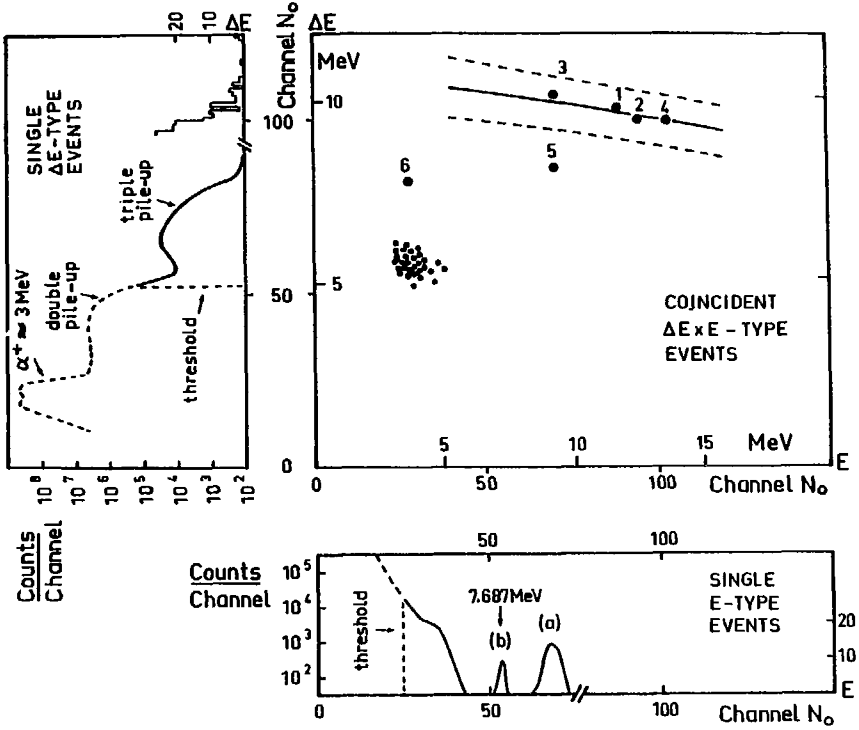

Figure 7
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