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Search for a fine structure in the ^{14}C decay of ^{222}Ra

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The ^{14}C decay of ^{222}Ra has been reinvestigated, using an 85 MBq activity ^{230}U source radiochemically separated, a Si barrier detector, and a superconducting magnetic spectrometer. In a 16 days counting run, 210 ^{14}C events were recorded. The ^{14}C particles energy value 30.930±0.090 MeV was found and an upper limit $b \leq 2\times10^{-12}$ was estimated for the ^{14}C branching ratio to the ^{208}Pb first excited state; the branching-ratio value to the ^{208}Pb ground state, thus equal to the total branching ratio, was found to be $b=\lambda_c/\lambda_\alpha=(2.31\pm0.31)\times10^{-10}$.

I. INTRODUCTION

The discovery of a fine structure in the ^{14}C decay of the ^{223}Ra nucleus[1] has revived the idea that there might exist a similitude between ^{14}C and α emissions. Indeed, from the suite of measurements of ^{14}C emissions from radium isotopes, with mass numbers from $A=222$ to 226 (Refs. 2–5), an odd-even effect staggering the half-life values was deduced,[6] in analogy to the one observed in α decays of odd nuclei; a hindrance factor value of about 100 could be deduced from the only ^{14}C half-life measurements of ^{223}Ra and ^{225}Ra (^{225}Ra half-life upper limit), which exceeds the value by one order of magnitude observed in α decays.

Besides, as with the α-decay features, the recent measurements of the ^{223}Ra ^{14}C energy spectrum by Brillard *et al.*[1] revealed a fine-structure spectrum involving an intense ^{14}C branch to an excited state of the ^{209}Pb daughter nucleus. Then, the low hindrance factor (HF) values deduced for the first and second ^{209}Pb excited states have been interpreted[7] as a consequence of the existence of common shell-model configurations describing the odd neutron in both the parent and daughter nuclei.

Nevertheless, the above HF calculations are based on the knowledge of the even-even radium half-lives for which the measured values were hitherto assumed to be only related to ^{14}C decay to the ground state of the daughter nuclei. So, it seemed useful to measure the ^{14}C spectrum of even radium isotopes, particularly ^{222}Ra, for which the previously measured value of the branching ratio $b=\lambda_{^{14}C}/\lambda_\alpha=(3.6\pm0.5)\times10^{-10}$ (Refs. 4 and 5) is the most favorable. Moreover, it could be interesting to check the possible influence of the static octupole deformation[8–10] ($\beta_3\cong0.1$) of the ^{222}Ra nucleus on the intensity of the ^{14}C transition to the collective octupole $I^\pi=3^-$ first excited state of 2.614 MeV energy of ^{208}Pb.

The measurements of the ^{222}Ra ^{14}C energy spectrum were performed with an intense ^{230}U source, combined with a long counting run with the magnetic spectrometer Soleno facility at Orsay, the measurements lead to an improvement by a factor of 20 in the number of recorded ^{14}C events, with respect to our earlier experiment.[5]

II. EXPERIMENTAL DETAILS

A. Source preparation

The short-lived ($T_{1/2}=37.5$ s) ^{222}Ra nucleus is generated from its long-lived ($T_{1/2}=20.8$ d) ^{230}U parent through the α-decay chain $^{230}U \Longrightarrow {}^{226}Th \Longrightarrow {}^{222}Ra$. Strong ^{230}Pa sources have been prepared by irradiation of thick targets ($e=3$ mm, $\phi=26$ mm) of natural thorium metal with the 34 MeV proton beam delivered by the CERI isochronous cyclotron. With a 20-μA proton beam and irradiation times of 30–40 h, ^{230}Pa activities of some GBq were produced by the $^{232}Th(p,3n)$ reaction. The competitive (p,n) reaction generates ^{232}Pa ($T_{1/2}=1.31$ d), decaying to ^{232}U, which could disturb our measurements. So, in order to obtain ^{230}U sources free from interfering radioactive impurities, the following multistep radiochemical separation has been performed.

After a 12–15 day cooling time, to allow ^{232}Pa to decay, the target was dissolved in 180 cm^3 of 12 M HCl and the solution, diluted to 10 M, was passed through a Dowex 1X-8 anion exchange column ($\Phi=10$ mm, $l=250$ mm). Under these conditions, Pa and U, as well as some fission nuclides, like Ru and Nb, were fixed on the column in the form of anionic complexes, while Th^{4+} and other fission products passed through. After washing the column with 50 cm^3 of 10 M HCl, protactinium was eluted with 125 cm^3 of 4.5 M HCl; finally, the uranium fraction was eluted with 100 cm^3 of 0.1 M HCl. All steps of the elution were controlled by γ-spectroscopic measurements of aliquots. After evaporation to a small volume, the Pa fraction was purified again on a small anionic column.

From 18 up to 44 days after the last purification, the ^{230}U ingrowth activity remained at more than 90% of its maximum activity value. Meanwhile ^{230}U was separated from ^{230}Pa on a Dowex 1X-8 anionic column. Then it was purified on a column filled with di(2-ethylhexyl) phosphoric acid (HDEHP) sorbed on Teflon. After fixation in 0.2 M HCl, the column was washed successively by 2 M and 6 M HCl to eliminate impurities and finally ^{230}U was eluted with 8 M HCl. The uranium was electroplated from a NH_4Cl medium on a platinum disk as a source of 0.5 cm^2 area, with a yield better than 95%.

Two sources were prepared for two separated experiments; the strongest one was obtained by cumulating the activities of two successive irradiations, ^{230}U being separated respectively for 42 and 18 days after the purification of the ^{230}Pa produced in the first and second irradiations.

The α activity of the ^{230}U sources, measured with a Si detector in a well-calibrated geometry, before and after the experiments, was found to be respectively 85±2 MBq and 22.5±0.5 MBq when starting the measurements. The α-energy resolution of these sources was better than 35 keV. Each source was mounted on an aluminum holder, closed by a 20-μg cm^{-2} carbon foil at a distance of 3 mm, to prevent the possible escape of the source material and to assure ^{14}C ion charge state equilibrium (Sec. IV).

B. Magnetic spectrometer Soleno

The magnetic spectrometer Soleno is a solenoidal superconducting coil described elsewhere.[11] The source and the detector were placed in the geometric axis of the spectrometer, with an obturator ($\phi = 20$ mm) between to exclude the direct view of the source by the Si(Au) detector. The transmission curve of the Soleno was determined using a thin thoron deposit source. Figure 1 shows the variation of the Soleno nominal geometrical solid angle Ω, measured as the ratio of the α-counting rate of the 8784.4 keV line of ^{212}Po to the source strength, versus the current I_α.

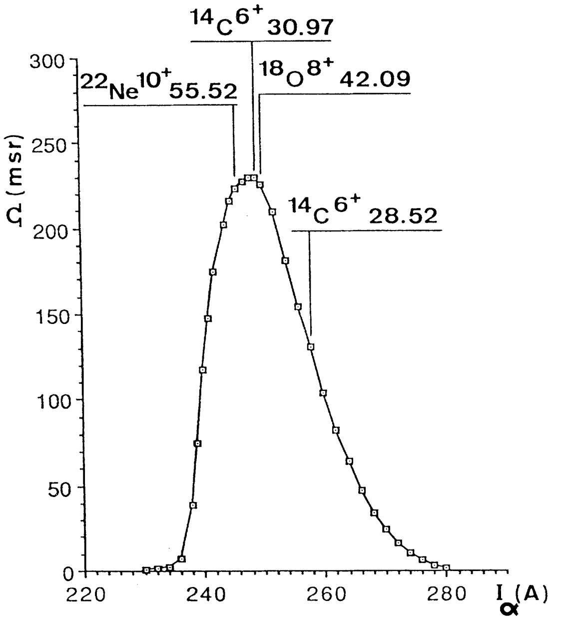

FIG. 1. Transmission curve of the Soleno spectrometer. The solid angle Ω, measured on the 8.784 MeV α line of ^{212}Po, is displayed vs the current I_α. For $I = 290$ A [formula (1)], the $^{14}C^{6+}$ ions to ^{208}Pb (g.s.) ($\langle B\rho \rangle = 0.500$ T m) are transmitted with $\Omega_{max} = 230 \pm 4$ msr; the transmission of those leading to the 2.614 MeV excited state ($\langle B\rho \rangle = 0.480$ T m) is then 130 msr. Transmissions for $^{22}Ne^{10+}$ ($\langle B\rho \rangle = 0.503$ T m) and $^{18}O^{8+}$ ($\langle B\rho \rangle = 0.496$ T m) ions expected to be emitted from respective ^{230}U and ^{226}Th decays are indicated. Energies are in MeV.

The current values I_C and I_α leading to the maximum transmission for $^{14}C^{q+}$ ions and α^{2+} particles of respective energy E_C and E_α being proportional to their magnetic rigidities $(B\rho)_C$ and $(B\rho)_\alpha$ can be written, with a good approximation,

$$I_C = I_\alpha (14E_C/4E_\alpha)^{1/2} 2/q \ . \tag{1}$$

So, with a 290 A current, the transmission of $^{14}C^{6+}$ ions emitted by ^{222}Ra to the ^{208}Pb ground state is maximum while the one for the first excited state feeding is 56% of this maximum. We can notice that, at this current, possible emissions of 42.09 MeV $^{18}O^{8+}$ by ^{226}Th or 55.52 MeV $^{22}Ne^{10+}$ by ^{230}U may be detected with good efficiency.

C. Counting spectrometers

The detector, manufactured in the laboratory, was a large single Si(Au) detector of 450 mm^2 area. Its choice became evident since previous experiments[5] have demonstrated without ambiguity that ^{14}C ions were the clusters emitted in the ^{222}Ra exotic decay. This detector was associated to a preamplifier Canberra and the pulses were amplified with a 672 spectroscopy amplifier (EG&G Ortec). The pulses, in 1 μs shaping time, were simultaneously analyzed with two 4096-channel amplitude analyzers interfaced to a PC-AT microcomputer and a Lecroy multichannel analyzer. The energy resolution (FWHM) of the detector was 19 keV on the 8.784 MeV α line of a thin ^{212}Po source. Besides, a high-precision energy pulser, properly normalized with respect to the energy of the long-range α group of ^{212}Po, was used to generate pulses with energies between 10 and 50 MeV to check the linearity response of the electronic setup. In order to control eventual shift of the electronics, spectra were saved at regular intervals and summed afterwards.

III. MEASUREMENTS AND RESULTS

The first experiment was carried out mainly to determine the kinetic-energy value of the ^{14}C emitted by ^{222}Ra from the energies of the ^{14}C ions from ^{223}Ra decay used as an external standard. Besides, we also tested the influence of the window thickness on the ^{14}C detection efficiency. These comparative measurements were performed with the same amplifier gain, using an energy dispersion of about 10 keV per channel.

With this scope, a 22-MBq ^{227}Ac source was measured, covered with a Mylar foil of about 50 μg/cm^2 thickness. The Soleno current was fixed to $I = 280$ A to focus the ^{14}C cluster group to the ^{209}Pb first excited state at the maximum of the transmission curve; the ^{14}C groups leading to the ^{209}Pb ground and second excited states were then detected with more than 95% of the Ω max. (Fig. 1).

As seen in the spectrum shown in Fig. 2(a), 59 events were recorded in this 4.1 day experiment. Three peaks were observed, like in our previous measurement,[1] which can be attributed unambiguously to ^{14}C transitions to the ^{209}Pb ground state and to its two first excited states.

In a second run, the ^{227}Ac source, covered with a 20-μg/cm^2 carbon foil, was measured with the same amplifier gain. The histogram of the 44 stored events shown in Fig. 2(b) is similar to the one represented in Fig. 2(a), except for a channel shift due to the difference of the energy losses in the two different windows. By summing the event numbers recorded in each peak of these two runs, we deduced the relative ^{14}C transition intensity values of 11%, 85%, and 4% respectively to the ground, first, and second excited states of ^{209}Pb, in good agreement with our previous determination[1] performed with better statistics.

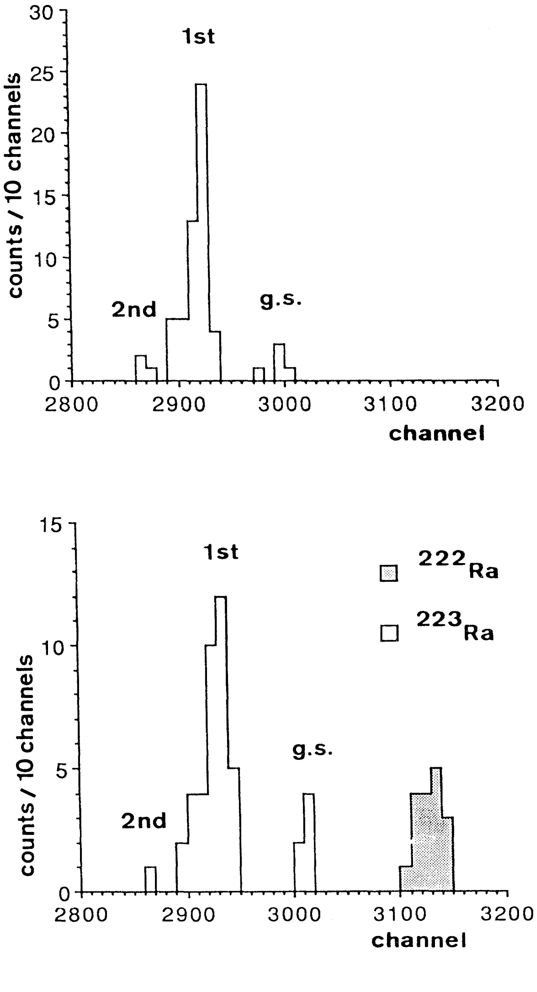

FIG. 2. ^{14}C spectra of radium isotopes. (a) ^{14}C spectrum of the ^{227}Ac source (22 MBq) covered with a 50-μg cm^{-2} Mylar foil. The energy dispersion is about 10.0 keV/channel. The three peaks labeled g.s., 1st, and 2nd correspond to ^{14}C groups emitted from ^{223}Ra to the respective ^{209}Pb g.s., first excited state (0.779 MeV), and second excited state (1.423 MeV). (b) ^{14}C spectrum of the ^{227}Ac source covered with a 20-μg cm^{-2} carbon foil and the same dispersion than in (a). The small energy shift between the two spectra is due to the difference in the energy losses throughout the different windows. The 17 ^{14}C events, recorded with the ^{230}U source (22.5 MBq), are displayed at the same scale (hatched histogram).

A third run has then been performed with the 22.5 MBq activity ^{230}U source, covered with a 20-μg/cm^2 carbon foil, and a Soleno current of $I = 290$ A, which is necessary to focus the $^{14}C^{6+}$ ions of 30.97 MeV energy emitted by ^{222}Ra to the ^{208}Pb ground state. In this 2.8 day experiment, 17 ^{14}C events were recorded [Fig. 2(b)].

From the three ^{14}C cluster energies emitted by ^{223}Ra calculated from the mass tables of Audi and Wapstra[12] and the level energies of the ^{209}Pb nucleus,[13] we deduced, using a least-squares analysis, a 30.930±0.090 MeV energy for the ^{14}C emitted by ^{222}Ra, in good agreement with the one calculated[12,14] for the ^{14}C decay to the ^{208}Pb ground state, i.e., $E = 30.969 \pm 0.008$ MeV.

In the second experiment, the 85±2 MBq activity ^{230}U source was used for searching a possible ^{222}Ra ^{14}C feeding to the ^{208}Pb excited state ($I^{\pi} = 3^-$) of 2.614 MeV energy. The Soleno current was set to 290 A to detect $^{14}C^{6+}$ ions to the ^{208}Pb g.s. with the maximum solid angle ($\Omega = 230$ msr) and consequently a $\Omega = 130$ msr solid angle for $^{14}C^{6+}$ ions to the ^{208}Pb first excited state.

Figure 3 represents a typical single spectrum recorded in a 20-h duration counting run. In this spectrum, the 14 ^{14}C events at 30.93 MeV are clearly separated from the other events recorded by the Si(Au) detector, i.e., the degraded α^+-particle distribution peaked at $\cong 3.0$ MeV, the 6.555, 7.133, and 7.687 MeV α^{++} lines from respective ^{222}Ra, ^{218}Rn, and ^{214}Po nuclei, the 7.13+7.68 MeV summing lines and finally some triple pile-up events with a maximum energy of 23 MeV. So, using the α^{++} lines and the summing ones, we were able to recalculate the energy of each ^{14}C event before summing the individual spectrum.

The 104 events recorded in a first run of 6 days, with an energy dispersion of about 10 keV per channel, are grouped in a single peak, except for three single events recorded in two successive runs; in spite of their low statistical significance, a tentative interpretation was searched for each of these events.

A single event located at $E = 28.83 \pm 0.10$ MeV, could be referred to (i) either a ^{14}C emission to the first excited state of ^{208}Pb, but the observed energy is higher than the expected one, i.e., 28.519±0.008 MeV, even taking account of the total linewidth of about 0.5 MeV, (ii) or a ^{14}C particle from ^{226}Th decay to ^{212}Po g.s., for which the expected energy 28.77±0.01 MeV is closer, (iii) or a degraded energy ^{14}C particle involved in the decay of ^{222}Ra to the ^{208}Pb g.s.

In the second part of this experiment, the amplifier gain was divided by a factor of 2 in order to observe possible higher-energy events, like ^{22}Ne and ^{18}O ions expected from ^{230}U and ^{226}Th decays, respectively. This run was carried out for nine days, and 106 additional ^{14}C events were recorded in the energy range 30.6–31.4 MeV. One event at 34.3±0.1 MeV was interpreted as due to the summing of a ^{14}C event with the degraded α^+-particle distribution of $\langle E \rangle \cong 3$ MeV energy.

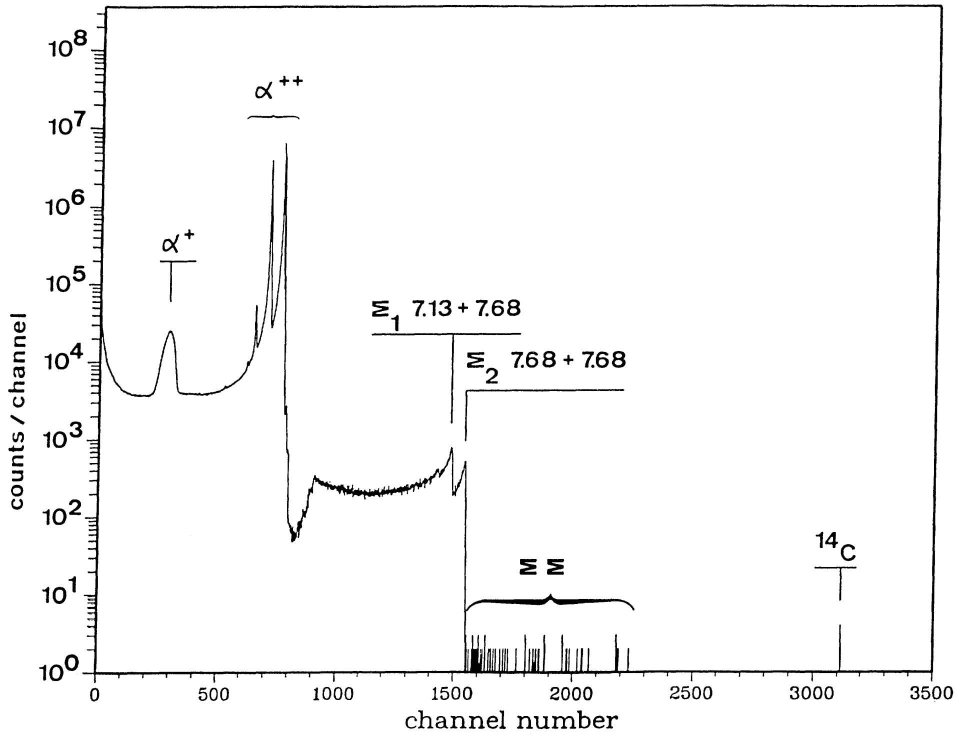

FIG. 3. Typical spectrum of all events measured with the Si detector from the 85 MBq ^{230}U source; counting time = 20 h. The ^{14}C peak contains 14 events clearly separated from all other events. The degraded α^+ particles of $\langle E \rangle \cong 3$ MeV ($\langle B\rho \rangle \cong 0.499$ T m) transmitted by Soleno give a broad distribution; the intensities of the transmitted α^{2+} lines of 6.555, 7.133, and 7.687 MeV from ^{222}Ra, ^{218}Rn, and ^{214}Po, respectively, may be compared to the α-particle number, i.e., $\cong 10^{11}$ emitted in the same solid angle at the same time. Σ means double pile-up peak; $\Sigma\Sigma$ indicates the triple pile-up events.

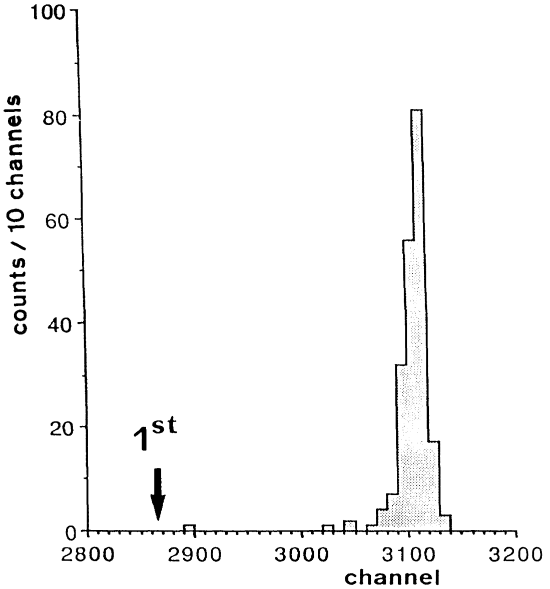

FIG. 4. Total spectrum of the 210 ^{14}C events recorded from the 85 MBq ^{230}U source. Each event detected in the two experiments, with different amplifier gains (see text), has been recalibrated using α^{++} line energies as well as their double summing energies. The position of the ^{14}C group expected to feed the first excited state of ^{208}Pb is indicated by an arrow.

One more event was recorded in both analyzers at $E = 44.05 \pm 0.20$ MeV. Inspection of all possible sources of spurious pulses led to negative results; the energy of this event seemed to be too high to correspond to the ^{18}O decay of ^{226}Th, for which the cluster energy is expected to be 42.09 MeV.

The total spectrum of the 210 events recorded with the intense ^{230}U source, displayed in Fig. 4, exhibits only one peak with a small low-energy tail, like α spectra.

IV. DISCUSSION

The branching ratio $b = \lambda_{^{14}C}/\lambda_\alpha$ for the ^{14}C decay to the ^{208}Pb ground state was calculated from the following expression:

$$b = (N_{^{14}C}/N_\alpha) 4\pi/\Omega\rho \qquad (2)$$

in which the ^{14}C total number of events was $N_{^{14}C} = 210$, Ω is the effective solid angle of Soleno in steradian, N_α is the total number of α particles emitted by ^{222}Ra in equilibrium with ^{230}U during the experiment, and ρ is the fraction of the 6+ charge state in the charge distribution. Allowing for this charge-state equilibrium being reached[17] for 36 MeV ^{12}C ions with carbon foils of 16 $\mu g\, cm^{-2}$, the value $\rho = 0.67$ was deduced for $E/A = 2.21$ MeV/nucleon from a compilation of experimental data.[18,19]

The branching-ratio value determined in this experiment, i.e., $b=(2.31\pm0.31)\times10^{-10}$, is slightly lower than the values given by Price *et al.*,[4] i.e., $b=(3.7\pm0.5)\times10^{-10}$, and Hourani *et al.*,[5] i.e., $b=(3.1\pm1.0)\times10^{-10}$, although it overlaps with the last value in the confidence level 1σ.

From this branching ratio, corresponding to the ^{14}C decay of ^{222}Ra to the ^{208}Pb ground state, and the ^{222}Ra α-decay half-life $T_{1/2}=37.5\pm2.0$ s (Ref. 20), the logarithm of the ^{222}Ra ^{14}C partial half-life was deduced to be $\log T_{1/2}=11.20\pm0.02$.

Accounting for the 28.83 MeV ^{14}C event, the upper limit for the ^{14}C branching ratio to the $I^{\pi}=3^-$ ^{208}Pb excited state at 2.6145 MeV is given to be $b\leq2\times10^{-12}$ and the partial half-life is deduced to be $\log T_{1/2}(\mathrm{s})\geq13.3$.

Besides, within the framework of the cluster preformation theory, the hindrance factor relevant to the ^{14}C exotic decay can be evaluated in a similar way to the one available for the α-decay hindrance factor calculation. Indeed, in α decay, the hindrance factor is defined for an even-even nucleus as $\mathrm{HF}=\gamma_0^2/\gamma_e^2$, where γ_0^2 and γ_e^2 are the reduced widths to the respective ground and excited states of the daughter nucleus; they are given by $\gamma^2=\hbar\ln2/(2T_{1/2}P)$, $T_{1/2}$ being the half-life in seconds and P the penetrability. As noticed by Price,[21,22] a good agreement can be observed in exotic decays between measured and calculated half-lives, allowing the simplest shape for the Gamow barrier, viz., a Coulomb potential truncated by a square well. The barrier penetrability P is expressed as

$$P=\exp(-2G) \tag{3}$$

and the Gamow factor G is given, in the standard WKB approximation, by

$$G=(1/\hbar)(2m)^{1/2}e^2Z_dZ_c(A_dA_c/AQ)^{1/2} \times\{\arccos x^{1/2}-[x(1-x)]^{1/2}\}\ , \tag{4}$$

where $A=A_d+A_c$ is the mass number of the parent nucleus, m is the nucleon mass, and x is the ratio of the touching radius R_t to the distance R_b for which the kinetic energy of separated fragments is equal to the Coulomb energy.

In his model, Price[21,22] is able to reproduce all measured exotic decay half-lives within $\Delta(\log T_{1/2}(\mathrm{s}))=\pm0.2$, assuming a radius value $r_0=0.928$ fm and knocking frequency values $\nu_{\mathrm{even}}=4.3\times10^{26}$ s^{-1} and $\nu_{\mathrm{odd}}=1.1\times10^{25}$ s^{-1}. However, the penetrabilities obtained by Price, with Q values deduced from atomic masses,[12] are slightly modified (by a factor $\cong3$), if we correct atomic masses for the electron binding energies,[14] as was noticed by Blendowske and Walliser,[23] then, a better fit of the experimental half-lives can be obtained with a single frequency $\nu=2.7\times10^{26}$ s^{-1} irrespective of the oddness or evenness of the parent nucleus. Table I gives some half-life values calculated using these parameter values, compared to experimental and calculated ones with the unified cluster model of Blendowske *et al.*,[24] in which the spectroscopic factor (i.e., the preformation probability) is taken as $S_C=(S_\alpha)^{(a-1)/3}$ ($S_\alpha=6.3\times10^{-3}$) and $v/2R=1.9\times10^{21}$ s^{-1}. A good agreement is noticed between measured and calculated half-life values for both even and odd nuclei. In ^{223}Ra decay, the favored ^{14}C transitions are well reproduced and the "unfavored" transition to the ^{209}Pb ground state can be explained by detailed structural considerations.[7]

Although the reduced widths calculated with this model do not have a physical meaning, due to the unrealistic r_0 and ν values used, they allow one to determine a value

TABLE I. Experimental and calculated half-life values for exotic decays. Column 3: lower index 1,2 indicates first or second excited state of the daughter nucleus. Column 4: Q values calculated from bare nuclei masses (Ref. 14). Column 5: (a) this work; (b) Ref. 4; (c) Ref. 6; (d) Refs. 1 and 7; (e) Ref. 26; (f) Ref. 27. Column 6: calculated half-lives using a (square well+Coulomb) potential tuned with a radius $r_0=0.928$ fm and a single frequency $\nu=2.7\times10^{26}$ s (see text). Column 7: half-lives calculated by Blendowske and Walliser (Ref. 23) using their unified cluster model. Column 8: hindrance factors estimated as $\mathrm{HF}=\gamma_0^2/\gamma_e^2$ for even-even nuclei and $\mathrm{HF}=\frac{1}{2}[\gamma_0^2(A+2)+\gamma_0^2(A)]/\gamma_0^2(A+1)$ (Ref. 7) for odd-mass nuclei.

Parent	Cluster	Daughter	Q (MeV)	$\log T_{1/2}$(s) Exp.	Calc.	Calc. (Ref. 23)	HF
^{222}Ra	^{14}C	^{208}Pb	33.164	11.20^{a}	11.2		1
^{222}Ra	^{14}C	^{208}Pb$_1$	30.548	>13.3^{a}	16.2		>0.001
^{224}Ra	^{14}C	^{210}Pb	30.646	15.90^{b}	16.0		
^{226}Ra	^{14}C	^{212}Pb	28.325	21.33^{c}	21.0		
^{223}Ra	^{14}C	^{209}Pb	31.963	16.01^{d}	13.4		442
^{223}Ra	^{14}C	^{209}Pb$_1$	31.184	15.28^{d}	14.9		2.5
^{223}Ra	^{14}C	^{209}Pb$_2$	30.540	16.59^{d}	16.2		2.5
^{226}Th	^{18}O	^{208}Pb	45.884	>15.3^{a}	18.3		
^{226}Th	^{14}C	^{212}Po	30.669	>15.3^{a}	17.9		
^{230}U	^{24}Ne	^{206}Pb	61.550	>18.2^{a}	22.1	22.2	
^{230}U	^{22}Ne	^{208}Pb	61.590	>18.2^{a}	20.7	20.4	
^{236}Pu	^{28}Mg	^{208}Pb	79.900	21.7^{e}	21.1	21.3	
^{238}Pu	^{28}Mg	^{210}Pb	76.160	25.7^{f}	25.8	25.6	
^{238}Pu	^{30}Mg	^{208}Pb	77.260		25.4	25.8	

$HF > 1\times10^{-3}$ to the ^{14}C transition to ^{208}Pb excited state (Table I). The 10^5 hindrance predicted for this state by Landowne and Dasso[25] in their calculations is in fact equal to the probability ratio of $\lambda_{3-}/\lambda_{0+}$ for which we deduced a 1.1×10^5 value (Table I, column 6). As concerns the odd nucleus ^{223}Ra the $F_{^{14}C}$ values calculated in Table I agree well with those deduced[7] using a more realistic radius $r_0 = 1.2$ fm.

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$$\Delta E_{scr} = zd[E_e(Z)]/dZ - fz^2/2d^2[E_e(Z)]/dZ^2$$

is about 99 keV, a value which practically cancels out the kinetic-energy increase of $111\times208/222 \cong 104$ keV for $^{14}C^{6+}$ ions. Therefore, the kinetic energy of clusters emitted in exotic decay can be calculated with Audi and Wapstra's tables (Ref. 12) with a good approximation.

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