Observation of the spontaneous emission of ^{14}C nuclei from ^{223}Ra

D. V. Aleksandrov, A. F. Belyatskiĭ, Yu. A. Glukhov, E. Yu. Nikol'skiĭ, B. G. Novatskiĭ, A. A. Ogloblin, and D. N. Stepanov
I. V. Kurchatov Institute of Atomic Energy, Moscow

(Submitted 29 May 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **40**, No. 4, 152–154 (25 August 1984)

The decay of ^{223}Ra through the emission of ^{14}C nuclei has been observed. The results agree with data reported by Rose and Jones.

Energy considerations allow the spontaneous emission of light nuclei for most nuclides heavier than lead. There have been several studies of this type of decay, which occupies a position intermediate between α activity and fission. For example, it is concluded in the review in Ref. 2 that the emission of ^{24}Ne or ^{28}Mg nuclei is the most probable event for thorium isotopes, while the emission of ^{32}Si or ^{34}S is most probable for uranium isotopes.

Our analysis, in which the emission of light nuclei is treated in a model analogous to the Gamow theory for α decay, shows that the decay which should have the highest probability is that which leads to the production of the doubly magic nucleus ^{208}Pb or neighboring isotopes. An estimate of the penetrabilities of the Coulomb barrier for the nuclei of naturally radioactive families shows that the isotopes $^{223,224,226}Ra$ present the best conditions for the decay of the type under consideration here, with the emission of a ^{14}C nucleus. Table I lists the nuclear isotopes which may be regarded as the most likely candidates for a search for decay accompanied by the emission of heavy particles. The second column shows the decay half-lives of these isotopes, and the third gives the total energy of the decay. The fourth column shows the penetrabilities of the Coulomb barrier for the parameter value $r_0 = 1.2$ fm, which appears in the expression for the channel radius, $R = r_0 (a^{1/3} + A^{1/3})$, where a and A are the mass numbers of the emitted nucleus and the daughter nucleus, respectively. The fifth column shows the

TABLE I.

Decay	$T_{1/2}$	Q, MeV	D	D_α / D_a
$^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$	11.7 d	31.83	$4{,}07\times10^{-30}$	6.21×10^{3}
$^{224}Ra \rightarrow {}^{14}C + {}^{210}Pb$	3.64 d	30.53	$1{,}59\times10^{-32}$	$1{,}92\times10^{5}$
$^{226}Ra \rightarrow {}^{14}C + {}^{212}Pb$	1.62 yr	28.21	$2{,}55\times10^{-37}$	$6{,}35\times10^{4}$
$^{233}U \rightarrow {}^{25}Ne + {}^{208}Pb$	$1.6\cdot10^{5}$yr	60.82	$1{,}32\times10^{-33}$	$1{,}76\times10^{-1}$
$^{233}U \rightarrow {}^{24}Ne + {}^{209}Pb$	—	60.49	$1{,}05\times10^{-33}$	$2{,}21\times10^{-1}$
$^{236}Pu \rightarrow {}^{28}Mg + {}^{208}Pb$	2.7 yr	79.67	1.64×10^{-29}	$8{,}35\times10^{-1}$
$^{240}Cm \rightarrow {}^{32}Si + {}^{208}Pb$	26.8 d	97.57	$1{,}08\times10^{-27}$	$7{,}57\times10^{-1}$
$^{246}Cf \rightarrow {}^{38}S + {}^{208}Pb$	1,5 d	112.72	$9{,}7\times10^{-29}$	$1{,}82\times10^{2}$

ratios of the penetrabilities of the channels for α decay and decay through the emission of a heavy particle.

Since we do not have access to the isotope ^{223}Ra, for which the probability for radiocarbon activity is highest, we carried out our first measurements with ^{224}Ra and ^{226}Ra sources. The results of these measurements were summarized in Ref. 3, where only upper limits were estimated on the emission of ^{14}C with respect α decay.

For the search for the decay $^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$ we selected the source ^{227}Ac ($T_{1/2} = 22$ yr), in which the isotope ^{223}Ra is at a radioactive equilibrium with the actinium decay products.

The charged particles emitted from the source were identified by the standard ΔE-E method. The telescope of semiconductor detectors, which detected the products of the radioactive decay, consisted of a ΔE detector 16 μm thick and an E detector 500 μm thick. The solid angle monitored by the telescope was ≈ 0.1 sr. To prevent radioactive recoil nuclei from striking the surface of the ΔE detectors, we placed a gold foil 200 $\mu g/cm^2$ thick between the source and the detector. The count rate at the ΔE detector was 3×10^4 counts/s, and that at the E detector was 2.5×10^4 counts/s. To reduce the number of multiple coincidences of α pulses in the ΔE and E detection systems, we introduced rejectors to prevent the detection of double coincidences of pulses if the latter were separated by a time interval exceeding 40–50 ns. A pulse-height-discrimination arrangement in the ΔE system prevented the detection of α pulses and of the doubly coincident heights of the pulses of these particles. The effect was to sharply reduce the dead time of the analog-to-digital converter. The experimental procedure is described in more detail in Ref. 4.

By using this procedure we were able to detect very rare events in the face of the vast background of α particles and to reliably distinguish radiocarbon ions from multiple coincidences of pulses. Figure 1a shows a two-dimensional spectrum of ΔE-E events found from a 30-day exposure with an ^{227}Ac source. The solid lines in the lower part of this figure show a region of quadruple coincidences of α pulses (there were a

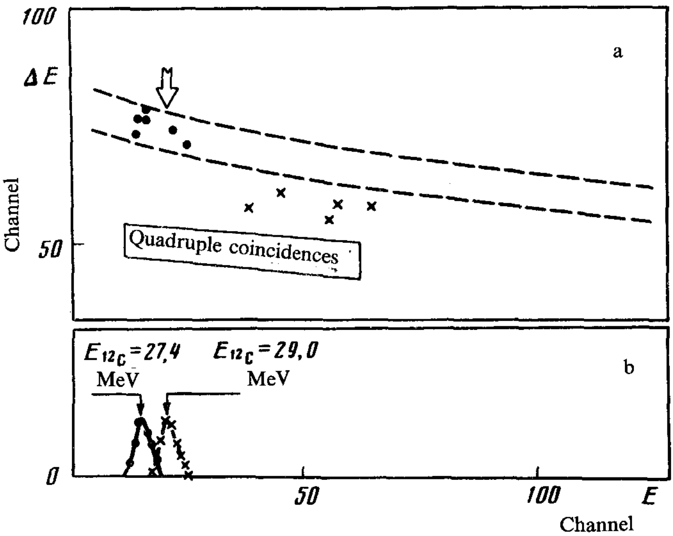

FIG. 1. a—Two-dimensional ΔE-E spectrum from measurements with the ^{223}Ra source. The filled circles are events corresponding to the emission of ^{14}C nuclei. The dashed lines bound the zone of possible ^{14}C events. The crosses show quintuple coincidences of α pulses. The solid lines bound the region of quadruple coincidences. The arrow shows the calculated position $E_{^{14}C} = 29.8$ MeV according to the calibration shown in part b; b—energy spectrum of the elastic scattering of ^{12}C ions by ^{12}C target (the peak at the left) and an ^{197}Au target.

total of 1800 events in this region). The crosses above this region show events corresponding to quintuple superpositions of α particles; five such events were detected.

Above the region of quintuple coincidences we detected, over the course of all the measurements, the seven counts shown by the filled circles. The positions of these events agree approximately with that calculated for the carbon-14 hyperbola according to the specific ionization loss. Calculations to accurately reflect the many corrections for the inhomogeneous thickness of the source and the energy loss of the ions as they cross the protective foil, the layers of gold on the surfaces of the detectors, and a possible dead layer on the E detector are extremely problematical. For a reliable determination of the region of the carbon hyperbola and an energy calibration of the ΔE-E spectrum, we carried out measurements in beams of ^{14}N ions with $E_N = 70$ MeV and of ^{12}C ions with $E = 29.2$ MeV, produced in the cyclotron of the Kurchatov Institute of Atomic Energy. In the former case we accurately determined the region of the carbon-14 hyperbola, which is the region bounded by the dashed lines in Fig. 1. In the second experiment, we used elastic scattering by ^{197}Au and ^{12}C targets for an energy calibration of the carbon ion spectrum in the energy interval 27–30 MeV (Fig. 1b).

From the control measurements we concluded that these seven events should be identified as the detection of ^{14}C nuclei with an energy of 29–30 MeV from the decay $^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$ (the decay energy is 31.84 MeV). We found the ratio of the yield of ^{14}C nuclei to that of α particles in ^{223}Ra to be $(7.6 \pm 3.0) \times 10^{-10}$.

While the search for the emission of ^{14}C from ^{223}Ra was being carried out at the Kurchatov Institute, Rose and Jones[1] reported observing this decay. Our own results agree completely with theirs.[1] Our study is thus independent confirmation of the discovery of a new type of spontaneous emission of radiocarbon in ^{223}Ra.

We have now undertaken an experimental search for the decay of ^{233}U accompanied by the emission of neon nuclei.

We thank V. I. Dukhanov for assistance in adjusting the electronics.

[1]H. J. Rose and G. A. Jones, Nature **307**, 245 (1984).
[2]A. Sandulescu, D. N. Poenaru, and W. Greiner, Fiz. Elem. Chastits At. Yadra **11**, 1334 (1980) [Sov. J. Part. Nucl. **11**, 528 (1980)].
[3]D. V. Aleksandrov, A. F. Belyatskiĭ, Yu. A. Glukhov, B. G. Novatskiĭ, and D. N. Stepanov, Tezisy XXXIV Soveshch. po yadernoĭ spektroskopii i strukture atomnogo yadra (Thirty-Fourth Conference on Nuclear Spectroscopy and Nuclear Structure), Nauka, Leningrad, 1984.
[4]V. I. Dukhanov and I. B. Mazurov, Prib. Tekh. Eksp. No. 6, 114 (1981).

Translated by Dave Parsons
Edited by S. J. Amoretty