Cluster decay of ^{236}Pu and correlations of the probabilities of α decay, cluster decay, and spontaneous fission of heavy nuclei

M. Hussonnois, J. F. Le Du, and D. Trubert
Institut de Physique Nucleaire, 91406 Orsay, France

R. Bonetti and A. Guglielmetti
Instituto di Fisica Generale Applicata dell'Universita degli Studi di Milano, I. N. F. N., Sezione di Milano, Milano, Italy

T. Guzel'
Faculty of Science, University of Istanbul, Turkey

S. P. Tret'yakova, V. L. Mikheev, A. N. Golovchenko, and V. A. Ponomarenko
Joint Institute of Nuclear Research, 141980 Dubna, Moscow Region, Russia

(Submitted 22 September 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **62**, No. 9, 685–689 (10 November 1995)

Fifteen cases of the decay of ^{236}Pu accompanied by the emission of magnesium nuclei were recorded. The probability of this decay mode relative to α decay is $(2.7\pm0.7)\times10^{-14}$. A partial half-life of $(1.25\pm0.12)\times10^9$ yr was obtained for spontaneous fission of ^{236}Pu. It was shown that the total probability of cluster decay and spontaneous fission for even-even nuclei with $88\leqslant Z\leqslant100$ has a lower limit of $\approx10^{-9}$ relative to α decay. © *1995 American Institute of Physics.*

Since its discovery, cluster radioactivity has been regarded as a process which possesses features in common with α decay and spontaneous fission. A rather complete exposition of experiments and theory can be found, for example, in the reviews in Refs. 1–3 . All cluster emitters in the region of nuclei heavier than lead emit α particles with a probability that is 10^8-10^{16} times higher than that of cluster decay. For nuclei with atomic numbers $Z\leqslant92$ the probability of cluster decay is comparable to or higher than that of spontaneous fission. For nuclei with $Z>92$ the existing experimental data show that the probability of cluster decay drops sharply with respect to that of spontaneous fission. For ^{242}Cm, for example, we have[4] $\lambda_{cl}/\lambda_{s.f.}<10^{-9}$. To clarify the possible relation between the cluster decay and spontaneous fission probabilities, it is useful to accumulate and refine the data on cluster decay of nuclei with a relatively high probability of spontaneous fission. One such nucleus is ^{236}Pu. The partial half-life of the α decay of this nucleus is 2.85 yr and the partial half-life for spontaneous fission is $\approx10^9$ yr. The first data on the decay of this nucleus with the emission of ^{28}Mg were obtained in Ref. 5 . The experiments were performed with a source containing at the start of the measurements 1.3×10^{15} plutonium nuclei, of which 66% consisted of the isotope ^{236}Pu and 34% consisted of ^{238}Pu. After 690 days of measurements performed with the use of solid-state track detectors made of polyethylene terephthalate (lavsan, a polyester), we recorded two tracks produced by 70±3-MeV magnesium nuclei. From them we determined the probability of cluster decay relative to α decay of $\approx 10^{-14}$ and relative to spontaneous fission $\approx 1.5\times 10^{-5}$. Our objective in this study is to refine the data on cluster decay of ^{236}Pu and to search for possible correlations between different modes of hadronic decay of nuclei. The isotope ^{236}Pu was obtained on the cyclotron at the Research Center in Orleans (France) via the reaction $^{238}U(p, 3n) \rightarrow {}^{236}Np \xrightarrow{(\beta-)} {}^{236}Pu$ by bombarding uranium with 34-MeV protons. The procedure used at the Radiochemical Laboratory of the Institute of Nuclear Physics in Orsay (France) is described in detail in Ref. 6 . After plutonium was chemically separated, it was deposited by electrolysis on a platinum substrate as a source 11 mm in diameter. The intensity of the source at the start of the measurements was equal to 2.0±0.2 mCi with respect to ^{236}Pu (9.6×10^{15} nuclei). The admixture of ^{238}Pu atoms amounted to 46 wt. % (1.5% according to the α activity). Solid-state track detectors made of phosphate glass, prepared at the State Optics Institute in St. Petersburg (Russia), were used to detect the clusters. The detectors were calibrated on the U-400 cyclotron at the Joint Institute for Nuclear Research (Dubna), using ^{22}Ne, ^{24}Mg, ^{27}Al, and ^{28}Si ions with energies in the range 1–5 MeV/nucleon. Calibrations were performed on the Legnaro tandem Van de Graaf (Italy), using 60-MeV ^{24}Mg ions and 70-MeV ^{28}Si ions. To obtain track images which are visible under an optical microscope, the detectors were etched in 48% HF at a temperature of 35°C. The tracks were identified by measuring the etch selectivity $S=V_T/V_M$, which is equal to the ratio of the etch rate V_T along a track to the etch rate V_M of an undamaged surface of the detector. The etching was therefore conducted in several stages until a track etched along its entire length was obtained. The track parameters were measured at each stage with the help of silicone rubber replicas.[7,8] Each stage was characterized quantitatively by the residual range R_r, which is equal to the difference of the total range and the track length revealed at the given stage of etching. The method is described in detail in Refs. 1 , 8 , and 9 . To perform cluster decay measurements we placed the ^{236}Pu source at the center of a hemisphere 16 cm in diameter. The inner surface of the hemisphere was formed by the track detectors. The total area of the detectors, equal to 402 cm^2, gave a detection efficiency of 86% of 2π. A vacuum was maintained in the measuring chamber throughout the entire exposure time in order to prevent energy losses of the recorded particles. After 216 days of exposure, 15 cases of cluster decay were registered. The results of the measurements of the parameters of the defected tracks are shown in Fig. 1. The tracks can be linked quite reliably to the particles with $Z=12$. Because of the short length of the cluster tracks obtained by using a high-density glass (2.9 g/cm^3), the mass number is determined with an accuracy no better than ±2. The measurement results presented in Fig. 1 are consistent with the results predicted for ^{28}Mg. The fact that the most likely cluster in the case of the decay of ^{236}Pu is ^{28}Mg, whose emission leads to ^{208}Pb, follows from the energy balance Q (Ref. 12) and is justified in detail in Ref. 5 . It follows from measurements of the total range that the energy of the ^{28}Mg particles which we recorded is 65±7 MeV, in reasonably good agreement with the value 70.2 MeV obtained from Q for the decay[12] $^{236}Pu \rightarrow {}^{28}Mg + {}^{208}Pb$. The probability of observing one track of Mg or Si from cluster decay of the ^{238}Pu admixture in our source is equal to $\approx 10^{-3}$ in accordance with the data of Ref. 7 . The probability of observing one magnesium track as a result of the formation of magnesium in triple spontaneous fission does not exceed[13] $\approx 10^{-2}$. In our experiments

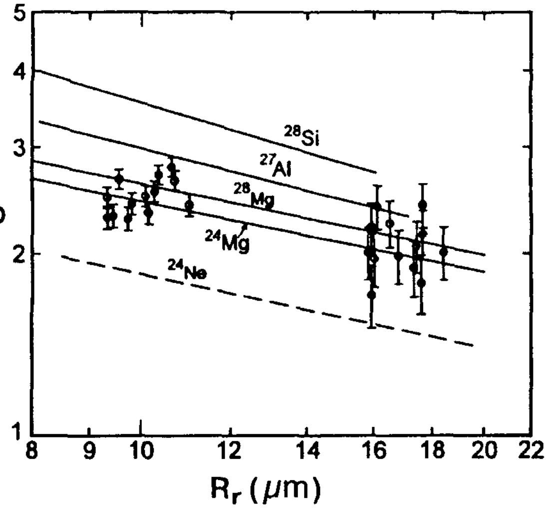

FIG. 1. Etch selectivity $S=V_T/V_M$ versus the residual range R_r. Solid lines — results of calibration with accelerated heavy ions. The dashed line for ^{24}Ne is presented in order to demonstrate the magnitude of the etch selectivity of the tracks of particles with $Z<12$. Since this glass does not detect neon, the etch selectivity was determined from the relation $S=0.0851(dE/dx)^{1.365}$ which is used to approximate the experimental data for ^{24}Mg, ^{27}Al, and ^{28}Si. The specific energy losses dE/dx in glass were calculated in accordance with Refs. 10 and 11 . Dots — measurements of the parameters of the tracks of particles from cluster decay of ^{236}Pu at two stages of etching of the detectors.

we could have observed fifteen ^{24}Ne tracks from cluster decay of ^{232}U which accumulated from the α decay of ^{236}Pu (Ref. 14). However, the threshold of detection with respect to Z for the phosphate glass and the etch conditions prevented Ne particles, whose specific kinetic energy losses are lower than for Mg particles, from being recorded. The total α-particle flux detected by the detectors in our experiments is equal to 1.4×10^{12} cm^{-2}. The sensitivity of the phosphate glass in this case remains constant.[7,15] It follows from our data that the probability for the cluster decay of ^{236}Pu accompanied by emission of ^{28}Mg relative to the α decay is equal to $(2.7\pm0.7)\times10^{-14}$, in good agreement with the data of Ref. 5 . We recorded 6.85×10^3 cm^{-2} fission fragments. If the entire effect is attributable to the spontaneous fission of ^{236}Pu, then the corresponding partial half-life is equal to $(1.13\pm0.13)\times10^9$ yr. This is ≈1.8 times lower than the value reported in Ref. 16, which was used in a review article in Ref. 17 . The contribution of spontaneous fission of ^{238}Pu to the total number of tracks of fission fragments is equal to $\approx10^{-3}$. The contribution from spontaneous fission of daughter products of α decay of plutonium isotopes and induced fission by background neutrons is negligible. The results of this study agree, within the limits of error, with the data obtained in Ref. 5 on the fission fragments, which give a partial half-life of ^{236}Pu $T_{1/2}=(1.36\pm0.20)\times10^9$ yr. The preparation of the plutonium, the fabrication of the sources, and the exposure of the detectors were conducted by different methods in different countries. The geometries of the measurements employing detectors (plastics, glass) were also different. The reasons for the disagreement with the results of Ref. 16 ($T_{1/2}=(2.09\pm0.06)\times10^9$ yr) are not clear to us.

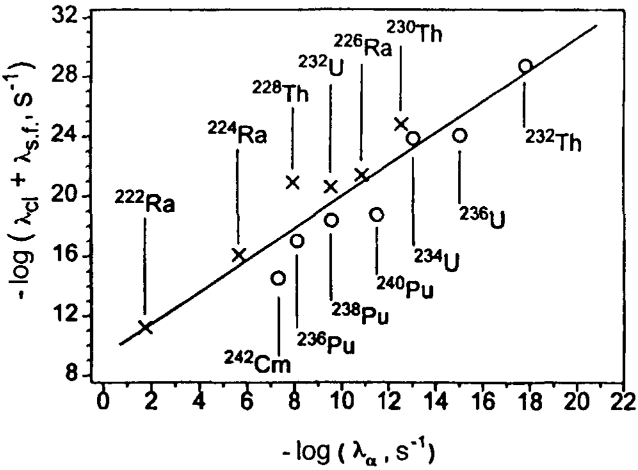

FIG. 2. Total probability for the cluster decay and spontaneous fission of the nuclei indicated in the figure as a function of the probability of their α decay: $\times$ — $\lambda_{cl} > \lambda_{s.f.}$; O — $\lambda_{s.f.} > \lambda_{cl}$. The data for spontaneous fission of ^{232}Th were taken from Ref. 18 .

In Ref. 5, it was hypothesized that there was uncontrollable contamination of the source or of the detectors by ^{252}Cf in the amount of $\approx 10^7$ atoms. However, it is strange that the amount of ^{252}Cf was found to be identical in completely different experiments (this study and Ref. 5). We note that in the experiments performed by our group[18] on the spontaneous fission of ^{232}Th it was shown that ^{252}Cf contamination in a track detector with an area of 1000 cm^2 did not exceed $\approx 10^2$ atoms. Averaging the results obtained by us and the results of Ref. 5 shows that the spontaneous fission half-life of ^{236}Pu is $(1.25 \pm 0.12) \times 10^9$ yr. The first measurements of the spontaneous fission of ^{236}Pu gave[19] $T_{1/2} = (3.5 \pm 1) \times 10^9$ yr.

Figure 2 shows the total probability for the spontaneous fission and cluster decay of heavy nuclei versus the probability of α decay. Experimental data for even-even nuclei were used in order to eliminate exclusion effects for the decay of odd nuclei. The data presented here include the isotopes of elements from Ra to Cm, for which the experimental results on cluster decay are available.[1-3] Least-squares fit makes it possible to relate all points by a linear relation of the form

$$\log(\lambda_{cl} + \lambda_{s.f.}, s^{-1}) = -(9.3 \pm 1.3) + (1.06 \pm 0.12) \times \log(\lambda_\alpha, s^{-1}). \quad (1)$$

When data for odd nuclei are included, a slightly larger scatter of the points is obtained, but the correlation which is established is not destroyed. Data for the probabilities of spontaneous fission of nuclei with $Z \leqslant 100$ (Ref. 17), which are not listed in Fig. 2, would give points that fall below the straight line (1). This makes it possible to interpret the obtained correlation as a limitation on the smallness of the probability for the decay of nuclei accompanied by the formation of particles heavier than α particles relative to the probability for the decay with the formation of α particles. Relation (1) can be represented in the form

$$\log[(\lambda_{cl}+\lambda_{s.f.})/\lambda_\alpha]\geq-(\approx 9). \quad (2)$$

The physical basis for the correlation which we obtained could be that both the α decay and the spontaneous fission and cluster decay are connected with the tunneling through the corresponding potential barriers formed in a given nucleus as it undergoes spontaneous oscillations.

We thank N. F. Orlov and N. Z. Andreeva for developing the formulas and for fabricating the phosphate glass which was used as the material for the track detectors.

This work was supported by the Russian Fund for Fundamental Research (Grant 93-02-37-19).

[1] Yu. S. Zamyatnin, V. L. Mikheev, S. P. Tret'yakova *et al.*, Fiz. Élem. Castits At. Yadra **21**, 537 (1990) [Sov. J. Part. Nucl. **21**, 231 (1990)].
[2] P. B. Price, Nucl. Phys. A **502**, 41C (1989).
[3] R. Bonetti and A. Guglielmetti, *Nuclear Decay Modes*, edited by D. N. Poenaru, IOP, Bristol, in press.
[4] V. L. Mikheev, S. P. Tretyakova, A. A. Ogloblin *et al.*, *FLNR Scientific Report 1991–1992*, E7-93-57, JINR, Dubna, 1993, p. 48.
[5] A. A. Ogloblin, N. I. Venikov, S. K. Lisin *et al.*, Phys. Lett. B **235**, 35 (1990).
[6] G. Ardisson, M. Hussonnois, J. F. Le Du *et al.*, Phys. Rev. C **49**, 2963 (1994).
[7] S. Wang, D. Snowden-Ifft, P. B. Price *et al.*, Phys. Rev. C **39**, 1647 (1989).
[8] D. Weselka, P. Hille, and A. Chalupka, Phys. Rev. C **41**, 778 (1990).
[9] R. L. Fleischer, P. B. Price, and R. M. Walker, *Nuclear Tracks in Solids*, University of California Press, Berkeley, 1975.
[10] L. C. Northcliffe and R. F. Shilling, Nucl. Data Tables **7**, 233 (1970).
[11] J. F. Ziegler, J. P. Birsack, and U. Littmark, *The Stopping and Range of Ions in Solids*, Pergamon Press, New York, 1985.
[12] D. N. Poenaru, D. Schnabel, W. Greiner *et al.*, Atomic Data and Nuclear Data Tables **48**, 231 (1991).
[13] F. Gonnenwein, B. Borsig, U. Nast-Linke *et al.*, in *Proceedings of the 6th International Conference on Nuclei Far from Stability and 9th International Conference on Atomic Masses and Fundamental Constants*, Bernkastel-Kues, Germany, 1992, edited by R. Neugart and A. Wohr, IOP Publishing, 1993, p. 453.
[14] R. Bonetti, C. Chiesa, A. Guglielmetti *et al.*, Phys. Rev. B **44**, 888 (1991).
[15] K. J. Moody, E. K. Hulet, S. Wang *et al.*, Phys. Rev. C **36**, 2710 (1987).
[16] Yu. A. Selitskiĭ, V. V. Funshteĭn, and V. A. Yakovlev in *Abstracts of Reports at 38th Conference on Nuclear Spectroscopy and Nuclear Structure*, Baku, 1988 [in Russian], Nauka, Leningrad, 1988, p. 131.
[17] N. E. Holden in *Proceedings of the International Conference "50 Years with Nuclear Fission,"* Gaithersburg/Washington, April 25–28, 1989, edited by J. W. Behrns and A. D. Carlson, American Nuclear Society, p. 465.
[18] R. Bonetti, C. Chiesa, A. Guglielmetti *et al.*, Phys. Rev. C **51**, 2530 (1995).
[19] A. Ghiorso, G. H. Higgins, A. E. Larsch *et al.*, Phys. Rev. **87**, 163 (1952).

Translated by M. E. Alferieff