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Systematics of spontaneous emission of intermediate mass fragments from heavy nuclei

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We have used polycarbonate track-recording films to confirm the rare decay mode of ^{226}Ra by ^{14}C emission and to set stringent upper limits on ^{14}C-emission rates of ^{221}Fr, ^{221}Ra, and ^{225}Ac. The ^{14}C-emission rate exhibits a pronounced odd-even effect. For Ra isotopes the hindrance factor for odd-even parents relative to even-even parents is at least 10 times higher for ^{14}C emission than for α emission.

Impressive progress has been made in the two years since Rose and Jones[1] first reported the novel spontaneous decay mode $^{223}\mathrm{Ra} \rightarrow {}^{14}\mathrm{C} + {}^{209}\mathrm{Pb}$. Since then, the isotopes ^{222}Ra and ^{224}Ra were observed by Price *et al.*[2] to emit ^{14}C, and Hourani *et al.*[3] have recently reported the observation of four events consistent with ^{14}C emission from ^{226}Ra. This new phenomenon of spontaneous emission of nuclei intermediate in mass between alpha particles and fission fragments was shown to be quite general when Barwick *et al.*[4] discovered ^{24}Ne emission from ^{232}U. A Dubna group has presented evidence that three additional isotopes emit ^{24}Ne ions—^{231}Pa (Ref. 5), ^{233}U (Ref. 6), and ^{230}Th (Ref. 7).

Two theoretical models have been proposed which share the assumption that intermediate-mass spontaneous emission can be described by a barrier penetration process similar to spontaneous fission. The model of Sandulescu *et al.*[8] was developed prior to the experimental discovery of Rose and Jones and was later modified[9,10] to bring predicted intermediate-mass decay rates into closer agreement with experimental data. Sandulescu, Poenaru, Ivascu, and Greiner were able to show that their model, termed the analytic superasymmetric fission model, describes alpha decay systematics and intermediate-mass emission within a unified framework. Shi and Swiatecki[11,12] have independently developed a barrier penetration model based on the proximity plus Coulomb potential and having the virtue of no adjustable parameters.

In this paper we report stringent new upper limits on the branching ratio for the emission of ^{14}C by ^{221}Ra, ^{221}Fr and ^{225}Ac relative to α decay, along with a confirmation of ^{14}C emission from ^{226}Ra with improved statistics. These new data on the ^{14}C emission mode permit systematic analysis of isotopes predicted to have the most favorable branching ratios. Trends established by this analysis can be used to gain additional insight into the physical mechanisms involved in intermediate-mass emission. When the measured branching ratios for ^{14}C emission and for ^{24}Ne emission are examined, it is seen that the analytic superasymmetric fission model in its present form underestimates intermediate-mass emission rates from parent nuclei with even numbers of protons and neutrons (even-even) and overestimates predicted rates from even-odd nuclei.

Three experiments were performed. In the first, the ISOLDE on-line isotope separator at CERN was used to produce combined beams of 60 keV ^{221}Fr and ^{221}Ra ions. In the second, combined beams of 60 keV ^{225}Fr and ^{225}Ra with half-lives of 3.9 min and 14.6 days, respectively, beta decayed to produce the ^{225}Ac source. Radioisotope production techniques and the methods used to determine the number of atoms collected at ISOLDE were disucssed in Ref. 2. Our detector geometry differs from that described in Ref. 2. In the present work, nearly 4π steradian collection was obtained by surrounding a 0.9 μm thick aluminum collector foil with a 15 cm diameter hollow sphere lined on the inside with Rodyne-*P* polycarbonate track-recording plastic film. The beam of radioactive ions entered a hole in the front of the hollow sphere and stopped in the thin collector at the center of the sphere. Alpha particles and ^{14}C ions subsequently emitted could escape from the collector in all directions with negligible energy loss except nearly in the plane of the collector foil.

In the third experiment, two Rodyne-*P* polycarbonate foils of ~100 cm^2 area were placed 10 cm from a 2.1 mCi source of ^{226}Ra and exposed for 1 day in moderate vacuum (~1 mm of Hg) at the Institute of Nuclear Physics, Orsay.

All Rodyne detectors were etched for 8 h at 70 °C in a 6.25 normal solution of NaOH. Measurements of the dimensions of conical etchpits produced along the trajectories of the ^{14}C ions were used to determine range and to make two independent measurements[2] of charge. Charge identification was based on calibrations of Tuffak polycarbonate detectors irradiated with ^{11}B, ^{12}C, and ^{20}Ne ions produced at the Lawrence Berkeley Laboratory Superhilac.

From the ^{226}Ra source, we identified tracks of 23 carbon nuclei for which the range, track-etch rate, and trajectory were consistent with the values expected for the emission of 26.5 MeV ^{14}C ions. (A kinetic energy of 26.5 MeV corresponds to a Q value of 28.21 MeV and a transition to the ground states of both ^{212}Pb and ^{14}C.) Figure 1 compares the histogram of measured ranges with the spread of values calculated from a range-energy table, taking into account the thickness of the ^{226}Ra source (1.77 mg/cm^2 thick, or ~20% of the expected ^{14}C range) and the allowed angles of emission from the source. Carbon-14 ions originating near the surface of the source emerge with nearly full kinetic energy, whereas the ions originating near the bottom emerge with reduced energy. A consequence of the spread in measured ranges is that the mass number of the carbon ion cannot be as well determined as it was in the earlier experiments, which utilized much thinner sources.[2,4]

The observed 23 carbon events from ^{226}Ra lead to a branching ratio with respect to alpha decay $B(\lambda_c/\lambda_\alpha) = (2.9 \pm 1.0)\times 10^{-11}$, in excellent agreement with the result of Hourani *et al.*[3] based on four events. The error takes into account uncertainty of source strength as well as statistical error. The absence of observed carbon emission from ^{221}Ra, ^{221}Fr, and ^{225}Ac leads to upper limits (90% C.L.) on branching ratios of 1.2×10^{-13}, 5×10^{-14}, and 4×10^{-13}, respectively. Table I gives further properties of the discussed decay modes.

Alpha decay rates for even-odd parent nuclei are known to be hindered by up to an order of magnitude with respect to the rate expected from neighboring even-even nuclei. Figure 2 shows the dependence of lifetime on kinetic energy for both alpha emission and ^{14}C emission from Ra isotopes. The ^{14}C decay lifetimes exhibit an even-odd hindrance much greater than is typical of alpha decay. For a given decay energy, the lifetimes for ^{14}C emission from even-even nuclei are at least a factor 100 less than those observed from even-odd nuclei, based on one positive measurement and one lower limit, whereas for α decay of Ra isotopes the average hindrance is only a factor ~10.

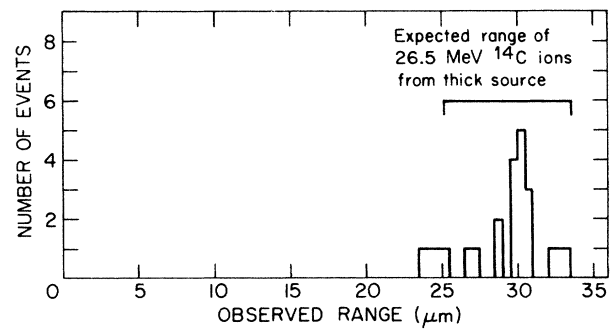

FIG. 1. Distribution of ranges of carbon fragments after emerging from the 1.77 mg/cm^2 $RaSO_4$ source. All 23 events are clustered within the range interval expected for ^{14}C ions with initial kinetic energy given by the Q value for $^{226}Ra \rightarrow {}^{14}C + {}^{212}Pb$ decay.

An even more striking systematic behavior is seen when measured branching ratios from intermediate-mass particle emission are compared with theoretical predictions of the analytic superasymmetric fission model. The bold line in Fig. 3 traces the sawtooth behavior of the ratio of theoretical to experimental branching ratios. The sawtooth trend also occurs in the case of ^{24}Ne emitters but is less dramatic than for ^{14}C emitters. This plot suggests that it might be beneficial to use different expressions for zero-point vibration energy E_v which distinguish between nuclei that have paired nucleons and nuclei that do not. Using the same functional form for E_v chosen in Ref. 9, we determined better values for the parameters in E_v by minimizing the root mean square of the difference between the known intermediate-mass branching ratios (or the measured upper limit) and the theoretical predictions.

The results of these calculations give E_v as

$$E_v = Q\left[0.060 + 0.035\exp\left(\frac{4-A_e}{2.5}\right)\right];\ A_e \geq 4,\ \text{e-e}\ , \quad (1)$$

$$E_v = Q\left[0.048 + 0.047\exp\left(\frac{4-A_e}{2.5}\right)\right];\ A_e \geq 4,\ \text{e-o}\ , \quad (2)$$

where A_e is the mass number of the light emitted fragment. The equations leave E_v unaltered for alpha decay.

The results obtained using Eqs. (1) and (2) with the analytic superasymmetric fission model have also been plotted in Fig. 3. The amplitude of the fluctuations has been diminished. The root-mean-square deviation σ can be de-

TABLE I. Properties of decay modes.

Z	A	Q	Atoms collected or source strength	$B = [\lambda(^{14}C)/\lambda(\alpha)]$	$\tau_{1/2}$ (sec)	Number of events
87	221	31.26	4.6×10^{13}	$< 5.0\times 10^{-14}$	$> 6.3\times 10^{15}$	0
88	221	32.39	1.8×10^{13}	$< 1.2\times 10^{-13}$	$> 2.4\times 10^{14}$	0
88	226	28.21	2.1 mCi	$(2.9\pm 1.0)\times 10^{-11}$	$(1.7\pm 0.7)\times 10^{21}$	23
89	225	30.47	1.2×10^{13}	$< 4.0\times 10^{-13}$	$> 2.5\times 10^{18}$	0

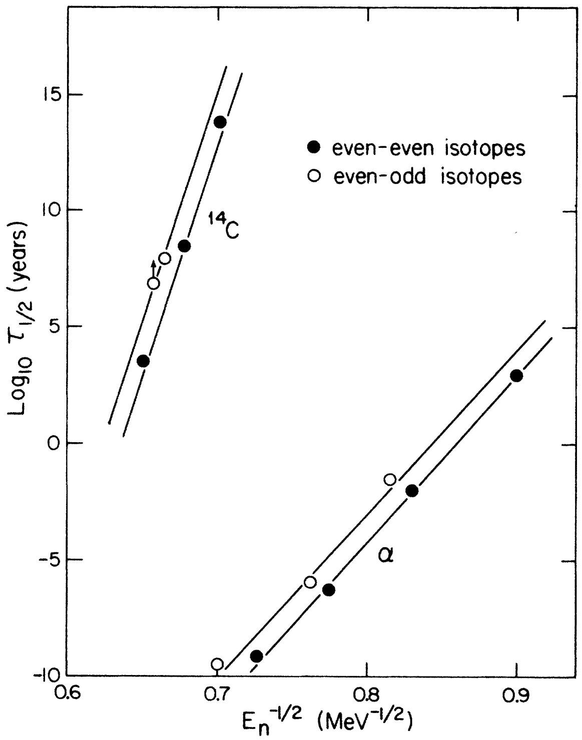

FIG. 2. Dependence of lifetime on $E_n^{-1/2}$, where E_n is the Q value per nucleon, for α emission and ^{14}C emission from radium isotopes. Hindrance factor for even-odd parents relative to even-even parents is ~100 for ^{14}C emission compared with ~10 for α emission.

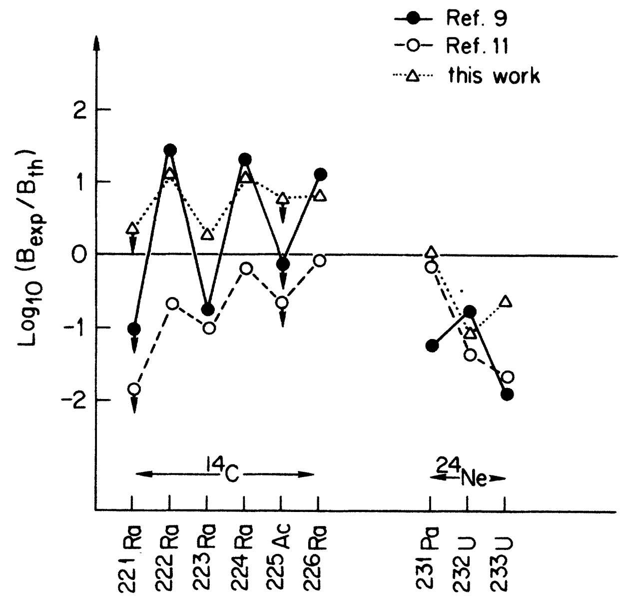

FIG. 3. Comparison of observed branching ratios for ^{14}C emission relative to α emission with values calculated by Shi and Swiatecki (Ref. 11), by Poenaru *et al.* (Ref. 10), and by use of Eqs. (1) and (2) for E_v.

fined by

$$\log_{10}\sigma(B) \equiv \left\{ \frac{1}{N} \sum_{i=1}^{N} [\log_{10}(B^i) - \log_{10}(B^i_{\exp})]^2 \right\}^{1/2}, \quad (3)$$

where N is the number of nuclei for which intermediate mass decay modes have been measured and B is the theoretically predicted branching ratio. We utilized branching ratio limits for those decays which have not been detected excluding those limits which are greater than the predicted values. The rms deviation is reduced from $\sigma(B) = 13.9$ to $\sigma(B_{e\text{-}o}) = 7.0$ if one uses Eqs. (1) and (2) for E_v instead of the expression found in Ref. 10.

The new expressions for E_v significantly decrease the predicted branching ratios for parent nuclei with an unpaired nucleon and the effect becomes more pronounced as the mass of the intermediate-mass decay fragment increases. Table II lists a comparison between theoretical branching ratios (including our modification of the analytic superasymmetric fission model) and experimental

TABLE II. Comparison of theoretical branching ratios and experimental values.

Decay	Q	Theoretical predictions $-\log_{10}(B)$ (Ref. 9)	$-\log_{10}(B)$ (Ref. 11)	$-\log_{10}(B_{e\text{-}o})$ (this work)	Measured $-\log_{10}B$
$^{221}Fr \to {}^{14}C + {}^{207}Tl$	31.26	12.5	11.1	13.2	> 13.1[a]
$^{221}Ra \to {}^{14}C + {}^{207}Pb$	32.39	11.9	11.1	13.3	> 12.9[a]
$^{222}Ra \to {}^{14}C + {}^{208}Pb$	33.05	11.0	8.8	10.7	9.43[b]
$^{223}Ra \to {}^{14}C + {}^{209}Pb$	31.85	8.5	8.2	9.4	9.21[b]
$^{224}Ra \to {}^{14}C + {}^{210}Pb$	30.54	11.8	10.2	11.5	10.37[b]
$^{225}Ac \to {}^{14}C + {}^{211}Bi$	30.47	12.2	11.8	13.2	> 12.4[a]
$^{226}Ra \to {}^{14}C + {}^{212}Pb$	28.21	11.7	10.5	11.4	10.5[a]
$^{231}Pa \to {}^{24}Ne + {}^{207}Tl$	60.42	10.0	11.0	11.1	11.22[c]
$^{232}U \to {}^{24}Ne + {}^{208}Pb$	62.31	10.9	10.3	10.5	11.7[d]
$^{233}U \to {}^{24}Ne + {}^{209}Pb$	60.50	10.3	10.4	11.5	12.12[e]
$^{240}Pu \to {}^{34}Si + {}^{206}Hg$	90.95	13.3	15.6	12.7	. . .
$^{241}Am \to {}^{34}Si + {}^{207}Tl$	93.84	12.4	14.3	14.4	. . .

[a]This work.
[b]Reference 2.
[c]Reference 5.
[d]Reference 4.
[e]Reference 6.

values. The list includes two favorable candidates for ^{34}Si emission. The branching ratio for ^{34}Si emission from ^{241}Am as predicted by the superasymmetric fission model drops from 4×10^{-13} to 4×10^{-15} if Eqs. (1) and (2) are employed, rendering the experimental observation far more difficult. Because the second ^{34}Si candidate, ^{240}Pu, is an even-even parent the branching ratio increases with the use of (1) for E_v. Searches for ^{34}Si emission from these two nuclides are in progress.

As Fig. 3 shows, the predictions made by Shi and Seiatecki are much less sensitive to nucleon pairing effects than are those of the superasymmetric fission model. However, the overall agreement with experimental branching ratios is not as good as in the superasymmetric fission model with our modified expressions for E_v. The Shi-Swiatecki model gives $\sigma(B_{ss})=15.1$.

To summarize, we confirm that ^{14}C is emitted from ^{226}Ra at a branching ratio which agrees with the previously published observation.[3] This result, along with our upper limits on the branching ratios for ^{14}C emission from ^{221}Ra, ^{221}Fr, and ^{225}Ac, indicates that there is a large systematic even-odd effect relative to the branching ratios predicted by the analytic superasymmetric model. A refinement of the expression for the zero-point vibration energy depresses the expected branching ratios for even-odd parent nuclei, especially those nuclei which are presumed to emit fragments heavier than neon, and leads to results in better agreement with experiment.

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