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Isomeric States in ^{250}Fm and ^{254}No†

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A preliminary report on the discovery of isomeric states in ^{250}Fm and ^{254}No was included in a recent article on α-emitting isotopes of element 104. The existence and assignments of the 1.8 ± 0.1-sec isomer to ^{250}Fm and the 0.28 ± 0.04-sec isomer to ^{254}No have now been confirmed by cross-bombardment techniques. Isomeric ratios based on measurements of collection efficiency of recoil atoms from the decay of isomeric states are given. An interpretation of the even-even isomers as high-spin two-quasiparticle states is discussed.

I. INTRODUCTION

This study was initiated by the appearance of the well-known α-particle groups of 30-min ^{250}Fm and 55-sec ^{254}No with much shorter apparent half-lives in the α-particle spectra measured in the course of studies of ^{257}Rf.[1] In bombardments of ^{249}Cf with ^{12}C ions the 7.44-MeV α group of ^{250}Fm and 8.1-MeV α group of ^{254}No appeared to have half-lives of about two seconds and a fraction of a second, respectively. Further studies of these short-lived components have indicated that they most likely arise from the decay of isomeric states in ^{250}Fm and ^{254}No. All the experimental evidence reported here is based on α-particle spectroscopy; no radiation from the isomer has been detected. Some of the results presented here have been included in our article in *Nature*.[2]

II. EXPERIMENTAL

The experimental apparatus and techniques have been described in some detail in earlier papers.[3,4] The recoil atoms knocked out of a target were thermalized and transferred onto the rim of a wheel by helium gas flowing rapidly through a small chamber adjacent to the target. The wheel was periodically rotated to place the recoil products deposited on the wheel next to a series of Si(Au) detectors for α-particle measurements.

Most of the experiments were performed with a five-detector-station system, the stations being located at 39° intervals. The wheel was digitally rotated in steps of 1.5°. During a measurement it was advanced 26 steps or 39° with an extra step added to every 119th movement. Such a scheme allowed for an effective discrimination against long-lived activities accumulating on the wheel. Some of the later experiments were performed with a seven-detector-station system with 45° separation between two adjacent stations. The wheel was then advanced 30 steps at a time and an extra step was given at chosen intervals.

In both experimental arrangements the detector stations were identical, each one having four detectors. Two movable detectors alternately faced the wheel or were shuttled to face the two stationary detectors in an off-wheel position. In the off-wheel position one would observe the decay of atoms recoiled from the wheel onto the movable detectors as a result of the impulse given by α, β, or γ emission. A schematic diagram of the seven-detector-system is shown in Fig. 1.

A variety of target and heavy-ion projectile combinations were used in the experiments. However, the primary emphasis was on bombarding a 290-μg/cm^2 ^{249}Cf target, electrodeposited on a 2.2-mg/cm^2 Be foil, with ^{12}C ions. The 10.4-MeV/nucleon heavy-ion beams were supplied by the Berkeley heavy-ion linear accelerator (HILAC). Beryllium metal foils were used to degrade the beam, whose energy was measured by a Si(Au) detector looking at the particles scattered from the target at an angle of 30°.

Signals from the detectors were amplified by modular units developed at our laboratory, processed by a PDP-9 computer, and stored on an

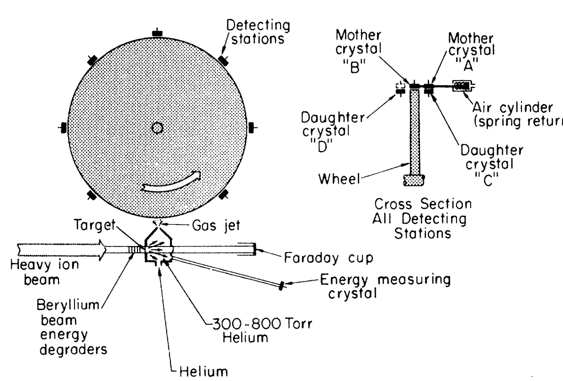

FIG. 1. A schematic diagram of the seven-detector-station system. The cross section at right shows the arrangement of the two movable mother detectors and the two stationary daughter detectors.

IBM tape. Each wheel-cycle and shuttle period was divided into four time subgroups of equal length to facilitate half-life determinations.

III. RESULTS

A. ^{250m}Fm

In our preliminary report[2] the evidence for the existence of a 2-sec isomeric state in ^{250}Fm was derived from α-particle spectra resulting from bombardments of ^{249}Cf with ^{12}C ions. The 2-sec half-life was shown to be associated with a parent activity of the 30-min, 7.44-MeV ^{250}Fm. In later experiments Fm isotopes were produced by ^{242}Pu

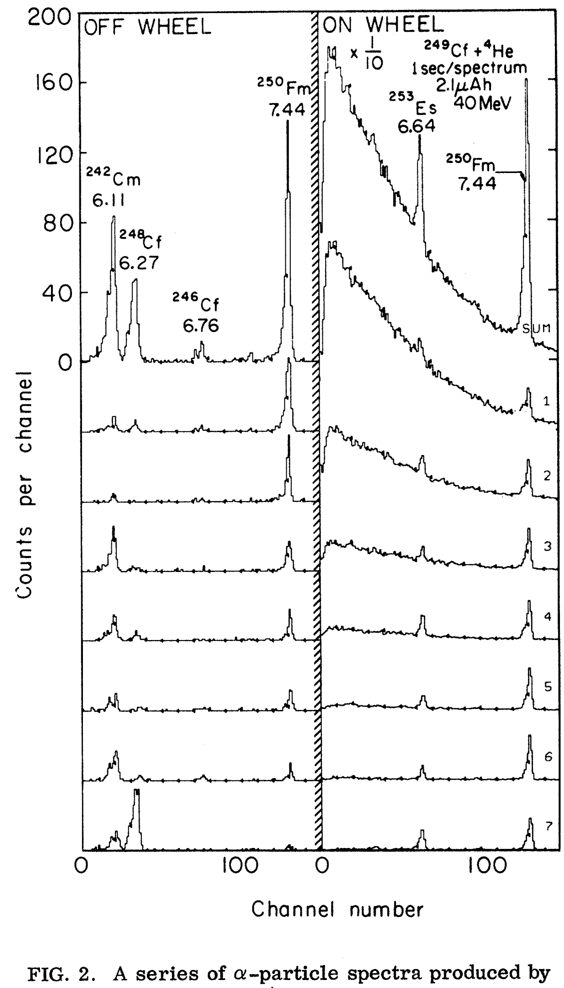

FIG. 2. A series of α-particle spectra produced by bombardments of Cf with ^{4}He ions. The spectra on the right-hand side were recorded by the detectors in the on-wheel position and those on the left-hand side by the detectors in the off-wheel position. Individual spectra show the total of counts recorded at each of the seven stations and the sum of the seven spectra is plotted topmost. The wheel-stepping interval, the integrated beam reading, and the bombardment energy are indicated in the figure.

$+{}^{12}C$, ${}^{243}Am + {}^{11}B$, and ${}^{249}Cf + {}^{4}He$ reactions. In all of them the 2-sec predecessor of ${}^{250}Fm$ appeared and thus it was proved that the atomic number of the parent is ≤ 100.

The spectra displayed in Fig. 2 resulted from a bombardment of a ${}^{249}Cf$ target by 40-MeV ${}^{4}He$ ions. The spectra recorded by the detectors in the on-wheel position are plotted on the right-hand side and the corresponding spectra recorded by the detectors in the off-wheel position are plotted on the left half of the figure. The 7.44-MeV peak is assigned to 30-min ${}^{250}Fm$ produced by the $({}^{4}He, 3n)$ reaction. Both ${}^{251}Fm$ and ${}^{249}Fm$ decay predominately by electron capture and consequently do not appear in the α-particle spectra. The large background in the lower channels is due to the high-energy tail of the β spectrum of ${}^{8}B$ produced in the Be backing foil of the target. Both ${}^{242}Cm$ and ${}^{248}Cf$, as well as part of the ${}^{246}Cf$ present, are residues of earlier experiments; the 6.64-MeV ${}^{253}Es$ was used as a calibration source.

The apparent 2-sec half-life of the 7.44-MeV peak in the spectra on the left-hand side belongs to the parent that provided the necessary recoil energy. A least-squares analysis of the half-life data from several experiments gives a half-life value of 1.8 ± 0.1 sec for the parent. By following the decay of the 7.44-MeV activity at an individual detector one finds that it has a half-life of 30 min, consistent with its assignment to ${}^{250}Fm$. A further indication of the activity being due to ${}^{250}Fm$ is the appearance of the 6.76-MeV, 36-h ${}^{246}Cf$ daughter activity on the stationary detectors in numbers proportional to those of the 7.44-MeV α activity.

After having excluded both ${}^{254}No$ and ${}^{250}Md$ as parental candidates the somewhat remote possibility of ${}^{250}Es$ having a high-energy, β-emitting isomer was all but eliminated by the fact that bombardment of ${}^{243}Am$ with neither ${}^{12}C$ nor ${}^{13}C$ produced the 1.8-sec activity. Especially in the ${}^{13}C$ bombardment one would expect a high cross section for ${}^{243}Am({}^{13}C, \alpha 2n){}^{250m}Es$ reaction. Since the Q_{β}- value for ${}^{250}Es$ is about -0.8 MeV,[5] a 1.8-sec isomeric state with a typical favored $\log ft$ value of 5 would have to be situated unreasonably high, some 8–9 MeV, above the ${}^{250}Es$ ground state. It thus seems that the 1.8-sec activity is ${}^{250}Fm$ itself, i.e., that it has a 1.8-sec isomeric state which decays to the 30-min ground state by a series of γ transitions.

The transfer of the ${}^{250}Fm$ atoms from the wheel onto the movable detectors must then be caused by the feeble recoil resulting from the isomeric transition or other accompanying γ rays and conversion electrons in the cascade that leads to the ground state. For a 500-keV γ ray the recoil energy of a ${}^{250}Fm$ atom is about 0.5 eV. The recoil atoms may be highly charged as a consequence of the vacancy cascade that follows the creation of a single inner-shell vacancy by internal conversion. Pleasonton and Snell[6] have measured the charge distribution for ${}^{131m}Xe$-${}^{131}Xe$ and shown that in this case a loss of eight electrons is most probable. The small recoil energy of the atoms and the possibility of their being highly charged suggested that the number of recoils reaching the detectors could be controlled by introducing gas or by setting up an electric field in the space between the detectors and the wheel. Results of such experiments are shown in Fig. 3. The ${}^{213}Fr$ and ${}^{250}Fm$ atoms were transferred from the wheel onto the detectors by the recoil resulting from electron capture (EC) of ${}^{213}Ra$ and decay of the isomeric state of ${}^{250}Fm$, respectively. One can see that biasing the wheel negatively by 10 V reduces the transfer of both activities by approximately an order of magnitude, both in the presence of a 2-Torr argon atmosphere and in vacuum. The α-particle and α-recoil yields are unaffected in all cases. According to Valli, Treytl, and Hyde,[7] the EC branching of ${}^{213}Ra$ to ${}^{213}Fr$ is $(20 \pm 5)\%$. Using this value, a relative intensity of $(49 \pm 2)\%$ for the 6.623-MeV peak of ${}^{213}Ra$, and a calculated ratio to take into account the wheel-stepping and shuttle periods we estimate a transfer efficiency ϵ_r of 0.5 ± 0.2 for the EC-recoil atoms of ${}^{213}Fr$ in the case of no Ar and no bias voltage. The ϵ_r for the recoils resulting from the decay of the ${}^{250}Fm$ isomer could not be determined in a similar fashion, because the relative cross sections for the production of the ground state and the isomer are not known. However, the similar behavior of the yields in the two cases of Fig. 3,

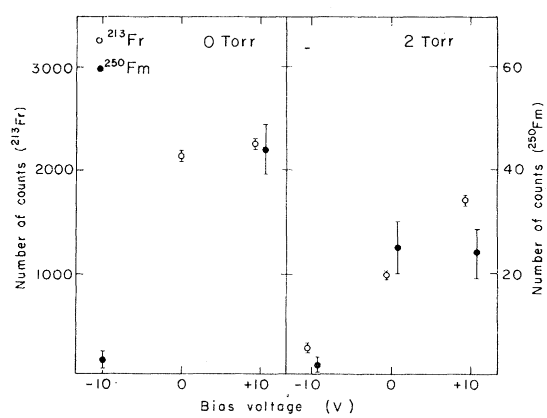

FIG. 3. The dependence of the recoil yield on the bias voltage applied between the wheel and the detectors and on the pressure of Ar gas introduced into the space between the detector faces and the rim of the wheel. Open circles refer to ${}^{213}Fr$ from EC of ${}^{213}Ra$, and the black circles to ${}^{250}Fm$ from ${}^{250m}Fm$.

as well as the lower limit $\epsilon_r \geq 0.3$ for an isomer[8] in ^{222}Ac, suggest that, at least for these cases, ϵ_r is about the same for both EC and isomeric transition. Assuming a value of $\epsilon_r = 0.5$ and taking into account the known geometry and time-dependent factors we obtain approximate value for the isomeric production ratio σ_m/σ_g of 0.3 for bombardments of ^{249}Cf with 80-MeV ^{12}C, and 1.2 using 40-MeV ^{4}He ions. We did not see any evidence for the decay of ^{250m}Fm by α emission or spontaneous fission; the upper limits for such decay modes are of the order of 20%.

B. ^{254m}No

While studying the properties of ^{257}Rf we observed the transfer of the 55-sec ^{254}No with a very short apparent half-life onto the movable detectors.[1] Results from a subsequent more detailed study of the short-lived activity are displayed in Figs. 4 and 5. The series of α-particle spectra shown in Fig. 4 were produced by bombarding a ^{249}Cf target with ^{12}C ions; the spectra were recorded by the detectors in the off-wheel position. The labeled peaks present well-known activities and have been discussed more thoroughly in other reports.[1,2] In Fig. 5 the 8.10-MeV peak decays with an apparent half-life of 0.28 ± 0.04 sec. The distribution of the counts in the four time subgroups of the 6-sec shuttle period was 68, 80, 63, and 53, consistent with the 55-sec half-life of ^{254}No. In another experiment with a 100-sec shuttle period the corresponding distribution yielded a half-life of 55 ± 20 sec for the 8.1-MeV peak. A further proof of the activity being ^{254}No was the detection of 7.44-MeV α particles from the ^{250}Fm daughter by the stationary off-wheel detectors when the movable detectors were not facing them. The total of such counts in the experiment depicted in Figs. 4 and 5 was 23 with a stationwise distribution of 16, 5, 1, 1, and 0. The observed total is quantitatively in agreement with the 264 counts of the 8.1-MeV α particles detected by both the stationary and movable detectors when facing each other. The loss factor of 11 is consistent with geometry and time-dependent factors. We concluded that the 7.44-MeV α events are from the decay of ^{250}Fm atoms recoiled from the faces of the movable detectors as a result of α decay of the parent ^{254}No.

The 8.1-MeV α activity was also observed on

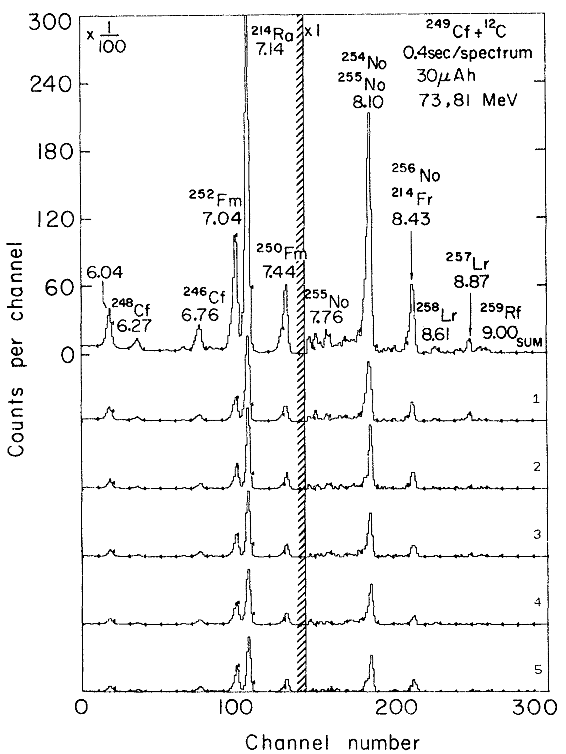

FIG. 4. A series of α-particle spectra resulting from bombarding ^{249}Cf with ^{12}C ions. The spectra were recorded by the detectors in the on-wheel position. The arrangement of the spectra and the data pertinent to the bombardment correspond to those in Fig. 1.

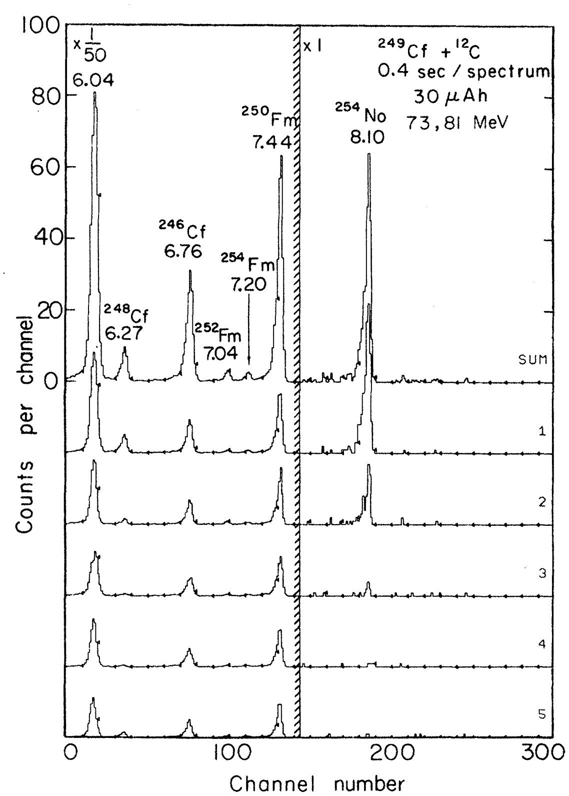

FIG. 5. The α-particle spectra resulting from the same bombardments as those in Fig. 3 but recorded by the detectors in the off-wheel position.

the detectors in the off-wheel position with an apparent half-life of 0.3 sec in bombardments of ^{246}Cm with 73-MeV ^{12}C ions. The isomeric ratio σ_m/σ_g, derived from experimental values with the same assumptions as in the case of ^{250}Fm, was about 0.4. The ratio was 0.2 when the isomeric pair was produced by bombarding ^{249}Cf with 73- and 81-MeV ^{12}C ions. As was the case for the ^{250m}Fm isomer, no α emission or spontaneous fission attributable to ^{254m}No was observed; the upper limit for such branching is 20%.

IV. DISCUSSION

Although we were unable to study the details of the decay of the two even-even isomers ^{250m}Fm and ^{254m}No we can suggest a plausible explanation for their existence in terms of two-quasiparticle states. A number of such low-lying high-spin isomeric states have been found[9] in the mass range $A = 170-190$, including the well-known series of isomers in even $N = 106$ isotones and even Hf isotopes.[10] Many of these states have been interpreted as being $K^\pi = 8^-$ two-quasiparticle states with Nilsson configurations $\frac{7}{2}^-[514]_n$, $\frac{9}{2}^+[624]_n$ or $\frac{7}{2}^+[404]_p$, $\frac{9}{2}^-[514]_p$.

There are single-particle levels near the Fermi surface for both neutrons near $N = 150$ and protons near $Z = 100$ such that coupling of two neutron or proton orbitals can give rise to high-spin two-quasiparticle states. According to the single-particle level scheme of Nilsson *et al.*[11] the 149th, 151st, and 153rd neutrons occupy $\frac{7}{2}^+[624]$, $\frac{9}{2}^-[734]$, and $\frac{7}{2}^+[613]$ levels, respectively. This order of single-particle states is generally supported by the ground-state assignments of nuclides with mentioned neutron numbers, and in particular, by the study of Braid *et al.*[12] of single-particle states in odd-mass Cm isotopes. Possible two-neutron configurations for isomeric states in ^{250}Fm and ^{254}No would thus be $K^\pi = 8^-$ two-quasiparticle states with Nilsson configurations $\frac{7}{2}^+[624]_n$, $\frac{9}{2}^-[734]_n$ and $\frac{9}{2}^-[734]_n$, $\frac{7}{2}^+[613]_n$. Fields *et al.*[13] have observed the decay of the 39-min isomer of ^{246}Am to proceed via such a $K^\pi = 8^-$ state of ^{246}Cm, which is an isotone of ^{250}Fm.

In the case of protons the relevant 99th, 101st, and 103rd protons are in the orbitals $\frac{7}{2}^+[633]$, $\frac{7}{2}^-[514]$, and $\frac{9}{2}^+[624]$.[11] Probable two-proton two-quasiparticle isomeric states would then be $K^\pi = 7^-$ or 8^- states arising from Nilsson configurations $\frac{7}{2}^+[633]_p$, $\frac{7}{2}^-[514]_p$ in ^{250}Fm and from $\frac{7}{2}^-[514]_p$, $\frac{9}{2}^+[624]_p$ in ^{254}No.

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