

## Alpha-Decay Properties of Some Francium Isotopes Near the 126-Neutron Closed Shell\*

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Several new alpha-emitting isotopes of francium have been produced by the reactions  $\text{Au}^{197}(\text{O}^{16}, xn)\text{Fr}^{213-x}$ ,  $\text{Tl}^{203}(\text{C}^{12}, xn)\text{Fr}^{215-x}$ ,  $\text{Tl}^{205}(\text{C}^{12}, xn)\text{Fr}^{217-x}$ , and  $\text{Pb}^{208}(\text{B}^{11}, xn)\text{Fr}^{219-x}$ . The nuclides studies lie on or below the 126-neutron closed shell. Their alpha-decay properties are as follows:

Nuclide	$Q_\alpha$ (MeV)	Half-life (sec)
$\text{Fr}^{213}$	$6.90 \pm 0.01$	$33.7 \pm 1.5$
$\text{Fr}^{211}$	$6.68 \pm 0.02$	$186 \pm 4.0$
$\text{Fr}^{210}$	$6.68 \pm 0.02$	$159 \pm 5.0$
$\text{Fr}^{209}$	$6.78 \pm 0.02$	$54.7 \pm 1.0$
$\text{Fr}^{208}$	$6.78 \pm 0.02$	$37.5 \pm 2.0$
$\text{Fr}^{207}$	$6.90 \pm 0.02$	$18.7 \pm 0.8$
$\text{Fr}^{206}$	$6.92 \pm 0.02$	$15.8 \pm 0.4$
$\text{Fr}^{205}$	$7.05 \pm 0.02$	$3.7 \pm 0.4$
$\text{Fr}^{204}$	$7.16 \pm 0.03$	$2.0 \pm 0.5$

Alpha-decay reduced widths  $\delta^2$  were calculated by using beta-decay systematics from this region to estimate the beta branching ratios. These reduced widths are compared to those of the isotonic polonium isotopes.

### I. INTRODUCTION

ALPHA radioactivity of spherical nuclei has been observed in two different mass regions. These are the rare-earth region near the 82-neutron shell and the  $\text{Pb}^{208}$  region near the 82-proton and 126-neutron shells. However, the data in these regions are in many cases incomplete and rather limited.

Theoretical calculations of alpha decay rates are much simpler to carry out for spherical nuclei than for spheroidally deformed nuclei. Various formulations based on shell-model wave functions of the nucleons outside the closed shells have been made,<sup>1-5</sup> and Mang has very successfully compared some numerical calculations with available experimental data.<sup>6</sup> It is hoped that a careful study of new spherical alpha emitters will give useful data for further testing and refining the theory.

Results have recently been reported by us on new holmium and erbium isotopes in the rare-earth region.<sup>7,8</sup> Our purpose in the present paper is to report results obtained for the neutron-deficient francium isotopes,  $\text{Fr}^{204}$  to  $\text{Fr}^{211}$  and  $\text{Fr}^{213}$ .

### II. EXPERIMENTAL PROCEDURES

Several new isotopes of francium have been produced by heavy-ion compound-nucleus reactions involving  $\text{Au}^{197}$  with  $\text{O}^{16}$  ions,  $\text{Tl}^{205}$  and  $\text{Tl}^{203}$  with  $\text{C}^{12}$  ions, and  $\text{Pb}^{208}$  with  $\text{B}^{11}$  ions. The bombardments were done at the Berkeley heavy-ion linear acceleration (HILAC), which accelerates heavy ions to 10.38 MeV per atomic mass unit. Aluminum absorbers were used to degrade the energy of the heavy ions, and the energy of the beam was determined from the absorber thicknesses and the range-energy data of Northcliffe.<sup>9</sup> The gold target con-

TABLE I. Isotopic analysis of targets.

$\text{Tl}^{205}$	$\text{Tl}^{203}$	$\text{Pb}^{208}$
98.7% $\text{Tl}^{205}$	92.6% $\text{Tl}^{203}$	90.7% $\text{Pb}^{208}$
1.3% $\text{Tl}^{203}$	7.4% $\text{Tl}^{205}$	4.4% $\text{Pb}^{207}$
		4.9% $\text{Pb}^{206}$
		0.1% $\text{Pb}^{204}$

sisted of a 0.0025-mm-thick gold foil. The thallium and lead targets consisted of salts deposited on a 0.006-mm aluminum backing. The isotopic analysis of these targets is given in Table I. The energy loss of the heavy ions in the gold target was obtained from the range-energy curves of Hubbard, and these curves were also used to estimate that the energy loss in the lead and thallium targets was equivalent to that in 0.006-mm-thick Al.<sup>10</sup>

Three different techniques were used in preparing samples for alpha-particle analysis. For the longer-lived activities, reaction recoils ejected from the target

<sup>9</sup> L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).

<sup>10</sup> E. L. Hubbard, Lawrence Radiation Laboratory Report UCRL-9053, 1960 (unpublished).

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<sup>1</sup> H. J. Mang, Z. Physik **148**, 582 (1957).

<sup>2</sup> D. J. Brussard and H. A. Tolhoek, Physica **24**, 263 (1958).

<sup>3</sup> H. J. Mang, Theory of  $\alpha$ -Decay, Lawrence Radiation Laboratory Report UCRL-8931, 1959 (unpublished).

<sup>4</sup> K. Harada, Progr. Theoret. Phys. (Kyoto) **26**, 667 (1961).

<sup>5</sup> J. O. Rasmussen, Nucl. Phys. **44**, 93 (1963).

<sup>6</sup> H. J. Mang, Phys. Rev. **119**, 1069 (1960).

<sup>7</sup> R. D. Macfarlane and R. D. Griffioen, Phys. Rev. **130**, 1491 (1963).

<sup>8</sup> R. D. Macfarlane and R. D. Griffioen, Phys. Rev. **131**, 2176 (1963).

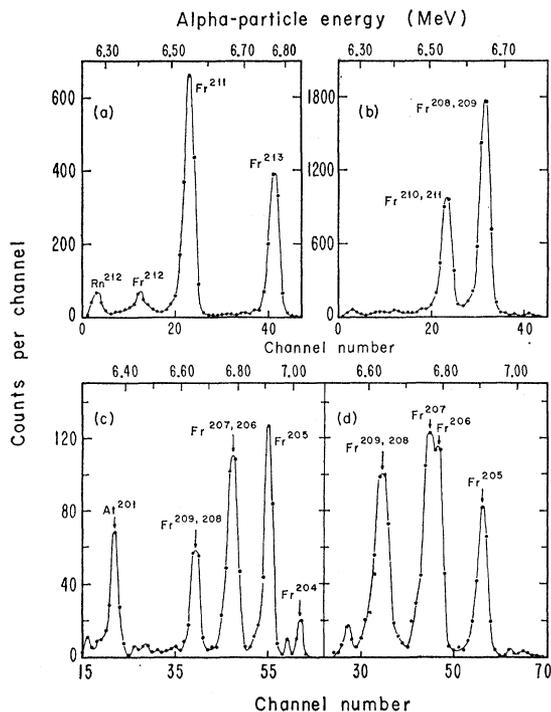


FIG. 1. Alpha-particle spectra recorded under conditions of continuous bombardment, collection and counting, showing the presence of the francium isotopes from mass number 204 through 213: (a)  $Tl^{205}$  plus 86 MeV  $C^{12}$  ions; (b)  $Tl^{203}$  plus 96 MeV  $C^{12}$  ions; (c)  $Au^{197}$  plus 153 MeV  $O^{16}$  ions; (d)  $Au^{197}$  plus 153 MeV  $O^{16}$  ions (high resolution).

were thermalized by helium and collected on a charged plate. The plate was then placed in a Frisch-grid ionization chamber for alpha-particle analysis. The details of this procedure have been described.<sup>11</sup> The "recoil-milking" apparatus, which was used to establish parent-daughter relationships, is also described in this earlier paper.<sup>11</sup> Primary samples were collected electrostatically in a helium atmosphere. Alpha-decay recoils from this sample were then electrostatically collected on first one and then another secondary plate during a measured time interval. Identification of these recoil daughters was accomplished by energy and half-life determinations. The relative amounts of daughter activity on the two secondary plates, and the duration of the collecting time on these plates, gave the half-life of the parent of a particular group and thus related the alpha-decay daughter to its parent.

A second procedure was used only incidentally in the study of these nuclides. Primarily this technique was used for studying very short-lived activities ( $t_{1/2} \leq 20$  msec).<sup>12</sup> However, in our study of these short-lived activities some of the nuclides reported in the present paper were also produced and so these data are also included. Instead of being thermalized in a helium

atmosphere, the reaction recoils were stopped in a thin aluminum leaf in vacuum and this sample was observed between the beam pulses of the MILAC by means of a gold-surface barrier detector. The alpha-particle spectra obtained in this way have much poorer resolution because of the thickness of the aluminum leaf. Thus, these results were used mainly as a corroboration of those obtained by the other techniques.

We have studied all of the activities reported in this paper by the third and principal procedure. Only the general features of this method are given here since the details are reported elsewhere.<sup>13</sup> Reaction recoils ejected from the target were slowed down in helium at 1 atm pressure in the target assembly. They were then swept through a small orifice into a chamber under vacuum, adjacent to the target assembly, where they were deposited on the surface of a collector. By using a gold-surface barrier detector, we recorded alpha-particle spectra of the recoil samples at the same time that we collected activity. For the shorter-lived activities ( $t_{1/2} \leq 1$  min), the level of activity was allowed to build up to an equilibrium value before we recorded alpha-particle spectra. This was convenient for cross section measurements since no corrections for decay were required. Relative excitation functions were obtained from the intensities of the various alpha-particle groups after the counting data were normalized to a constant integrated beam current. Excitation energies of the compound systems were calculated using the semi-empirical mass data of Seeger.<sup>14</sup>

Various gold-surface barrier alpha-particle detectors

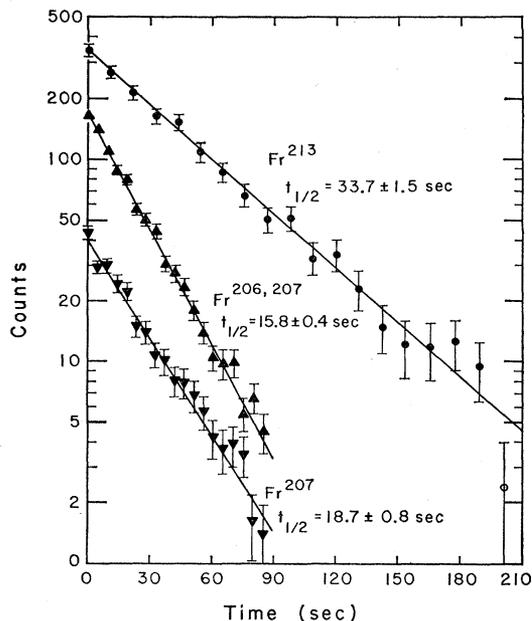


FIG. 2. Alpha-decay curves for  $Fr^{213}$  and  $Fr^{207}$  and  $Fr^{206}$ .

<sup>11</sup> R. D. Macfarlane, Phys. Rev. **126**, 274 (1962).

<sup>12</sup> R. D. Griffioen and R. D. Macfarlane, Bull. Am. Phys. Soc. **7**, 541 (1962).

<sup>13</sup> R. D. Macfarlane and R. D. Griffioen, Nucl. Instr. Methods (to be published).

<sup>14</sup> P. A. Seeger, Nucl. Phys. **25**, 1 (1961).

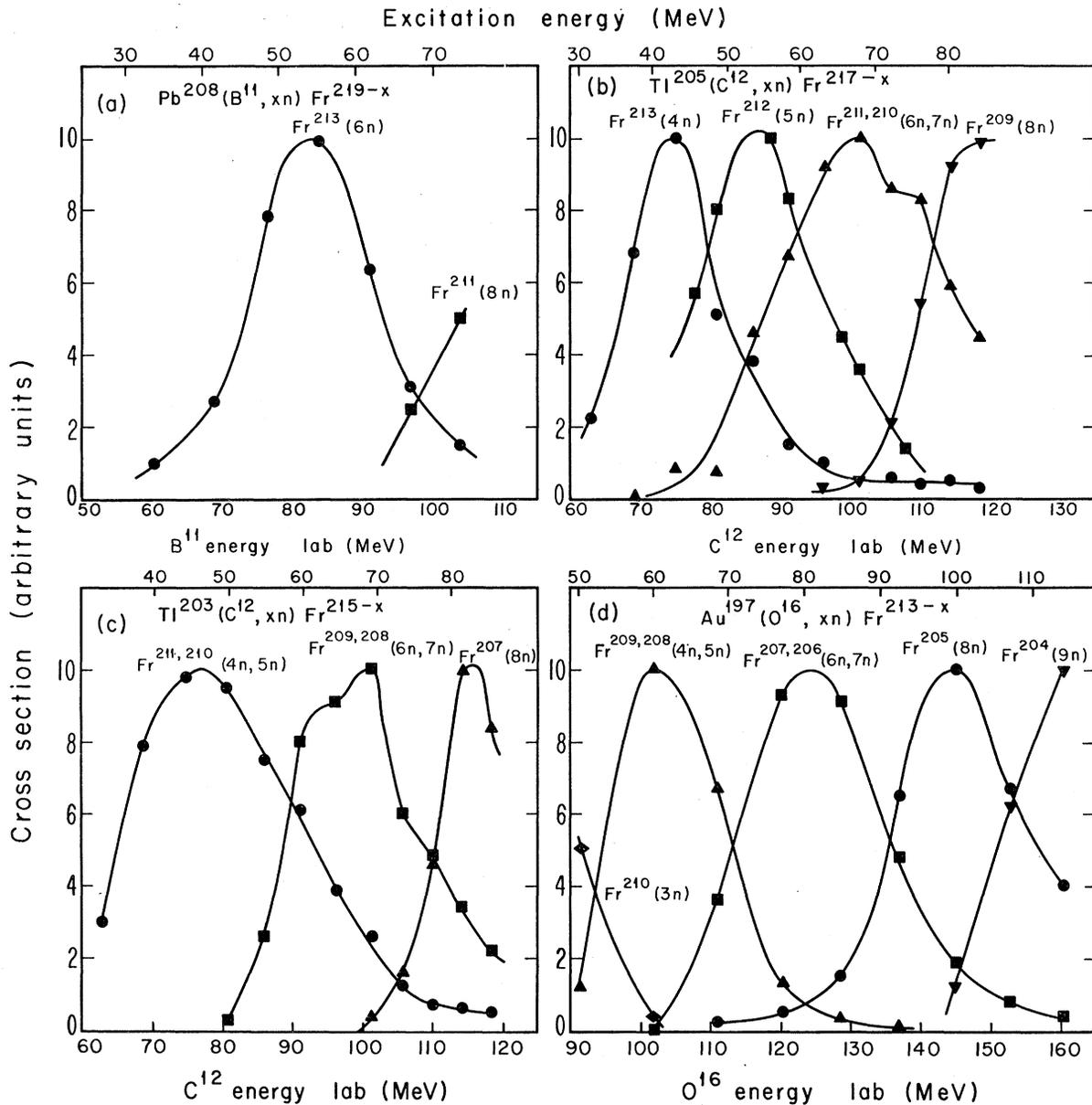


FIG. 3. Excitation functions for the production of francium isotopes by neutron evaporation reactions from various compound nuclei: (A)  $Pb^{208} + B^{11} \rightarrow Fr^{219-x}$ ; (B)  $Tl^{205} + C^{12} \rightarrow Fr^{217-x}$ ; (C)  $Tl^{203} + C^{12} \rightarrow Fr^{215-x}$ ; (D)  $Au^{197} + O^{16} \rightarrow Fr^{213-x}$ .

were used. These were made from *n*-type silicon with nominal resistivity in the range 1800–3200  $\Omega$ -cm. The reverse operating bias was usually about 25 V and the active surface area ranged from 10 to 50 mm<sup>2</sup>. Energy calibration was obtained from the alpha particles of Po<sup>210</sup>, Th<sup>228</sup> and its daughters, and Ra<sup>226</sup> and its daughters.

The determination of half-lives was greatly facilitated by the use of a timing circuit which controlled both the Hilac beam and a time-to-height converter and allowed the data from repeated cycles of bombardment and counting to accumulate.<sup>13</sup>

### III. RESULTS

Various compound nuclei of francium were formed by bombarding Au<sup>197</sup> with O<sup>16</sup> ions, Tl<sup>203</sup> and Tl<sup>205</sup> with C<sup>12</sup> ions, and Pb<sup>208</sup> with B<sup>11</sup> ions. The subsequent reactions gave rise to several new alpha groups in addition to some known alpha emitters of astatine and radon and the known alpha groups of Fr<sup>212</sup>. Each new group was characterized by its energy and half-life. None of the groups had been observed by others<sup>15</sup> when astatine compound nuclei were produced from C<sup>12</sup> bombardments of Au<sup>197</sup>

<sup>15</sup> T. D. Thomas, G. E. Gordon, R. M. Latimer, and G. T. Seaborg, Phys. Rev. 126, 1805 (1962).

and  $N^{14}$  bombardments of  $Pt^{195}$ ,  $Pt^{196}$ , and  $Pt^{198}$ , although their procedure limited this data to half-lives greater than about one minute. We also bombarded  $Tl^{205}$  with  $B^{11}$  ions to form radon compound nuclei and observed none of these new groups. This seemed to indicate that these groups result from new isotopes of francium that are formed by neutron evaporation from the various compound nuclei. Excitation functions for these activities were obtained for each of the compound systems studied, which provided information on the mass assignments of the different groups. In the case of some of the longer-lived nuclides, a simple chemical separation confirmed that these were isotopes of francium; milking the astatine daughters substantiated the mass assignments.

### A. $Fr^{213}$

Figure 1(a) shows an alpha-particle spectrum of the activity collected while bombarding  $Tl^{205}$  with 86-MeV  $C^{12}$  ions. A strong group is seen at  $6.77 \pm 0.01$ -MeV alpha-particle energy. This activity decays with a half-life of  $33.7 \pm 1.5$  sec (Fig. 2). Excitation functions for producing this activity from  $Tl^{205}$  with  $C^{12}$  ions and from  $Pb^{208}$  with  $B^{11}$  ions are shown in Figs. 3(a) and 3(b). In the  $Tl^{205}$  and  $C^{12}$  bombardments the peak of the excitation function falls at 43 MeV. The excitation function for the known  $Fr^{212}$ , which is formed by a  $(C^{12}, 5n)$  reaction, peaks at 54 MeV. This indicates very strongly that the 6.77-MeV group is produced by a  $(C^{12}, 4n)$  reaction and is therefore obtained from  $Fr^{213}$ . The peak for the  $Pb^{208}$  and  $B^{11}$  bombardments falls at an excitation energy of 54 MeV which is reasonable for a  $(B^{11}, 6n)$  reaction, and which is again consistent with this activity being  $Fr^{213}$ .

To identify this activity as an isotope of francium, we attempted a simple chemical separation from radon and astatine. A mixture of known isotopes of radon and astatine was produced by bombarding first gold and then mercury targets with  $C^{12}$  ions.  $Fr^{213}$  and  $Fr^{212}$  were then produced from the  $Tl^{205}$  target and deposited on the collector containing the radon and astatine isotopes. This mixture was put through a simple ion-exchange separation (see  $Fr^{211, 210}$  below) but due to the short half-life of the francium the results were inconclusive. However, it was found that a "chemical separation" could be accomplished by flaming fairly strongly with a Bunsen burner a platinum plate on which the activity was deposited. In this way, essentially all of the radon and over 90% of the astatine were removed while more than 90% of the  $Fr^{212}$  and also the  $Fr^{213}$  (corrected for decay) were left on the plate. Since the chemical identity of  $Fr^{212}$  is known with certainty, the chemical identity of  $Fr^{213}$  is substantiated.<sup>16</sup>

Confirmation of the mass assignment was obtained from "milking experiments." After collecting a sample

of the 6.77-MeV activity, alpha-decay recoils were electrostatically collected on two plates for 30 sec each.  $At^{209}$  was present on both plates and was identified by a half-life of  $5 \pm 1$  hours and an alpha-particle energy of 5.62 MeV. Due to the short half-life of the parent, the target chamber was not pumped out between the bombardments and the milking (see Ref. 11). Thus, there was some contamination in the daughter samples from primary products, resulting in a rather imprecise determination of the parent half-life. The ratio of  $At^{209}$  activity on the two secondary plates indicated a half-life of  $47 \pm 12$  sec for its parent, which is consistent only with the 6.77-MeV activity assigned to  $Fr^{213}$ .

$Fr^{213}$  should also be unstable with respect to  $\beta^+$  and  $EC$  decay, and should decay to the 19-msec  $Rn^{213}$ , which has an alpha-particle energy of 8.13 MeV.<sup>12</sup> A measure of the relative amounts of 6.77- and 8.13-MeV alpha groups would give the ratio of beta decay to alpha decay of  $Fr^{213}$ . One problem in this measurement was the possibility that part of the radon might have left the sample. However, in the case of  $Fr^{212}$  and its daughter  $Rn^{212}$ , it was found that very little, if any, of the radon escaped. Assuming that none of the  $Rn^{213}$  had escaped, a beta branching ratio for  $Fr^{213}$  of  $0.52 \pm 0.03\%$  was determined. The error quoted is only the statistical error from the counting rates.

### B. $Fr^{211}$ and $Fr^{210}$

Figure 1(a) shows another new alpha group at  $6.55 \pm 0.02$  MeV. Figure 1(b) shows the spectrum of the activity from the bombardment of  $Tl^{205}$  with 96-MeV  $C^{12}$  ions. This new group is again present here. When this activity was produced from  $Tl^{205}$  with 91-MeV  $C^{12}$  ions, it was found to decay with a half-life of  $186 \pm 4$  sec. Figure 3 shows the excitation functions for the formation of this alpha group from all four of the compound systems studied. Looking first at Fig. 3(b) we see that this excitation function is much different from those for  $Fr^{213}$  and  $Fr^{212}$ . Starting from the lower energy  $C^{12}$  ions, we see that it appears to be an activity produced by a  $(C^{12}, 6n)$  reaction. However, another new alpha group did not appear until much higher  $C^{12}$  energies; also, the excitation function for the 6.55-MeV group is seen to be exceptionally broad with almost a second peak appearing at higher energies. These facts seem to indicate that this group is due to two different isotopes,  $Fr^{211}$  and  $Fr^{210}$ , which were formed by  $(C^{12}, 6n)$  and  $(C^{12}, 7n)$  reactions, respectively. The bombardment of  $Tl^{205}$  with  $C^{12}$  ions again produced a very broad excitation function, corresponding quite well to what would be expected for the  $(C^{12}, 4n)$  and  $(C^{12}, 5n)$  reactions (Fig. 3C). Figure 3(D) shows that at the lowest possible  $O^{16}$  energies (near the Coulomb barrier) a small amount of this activity is formed from  $Au^{197}$ . This would almost certainly be due to  $Fr^{210}$ , formed by a  $(C^{12}, 3n)$  reaction, rather than to  $Fr^{211}$ , which would have to be formed by an  $(O^{16}, 2n)$  reaction.

<sup>16</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).

We further checked  $\text{Fr}^{211}$  and  $\text{Fr}^{210}$  by determining the half-life of the 6.55 MeV group at different points on the excitation function. When  $\text{Tl}^{203}$  was bombarded with 75-MeV  $\text{C}^{12}$  ions, the group again had a 186-sec half-life (Fig. 4). However, when  $\text{Tl}^{203}$  was bombarded with 96-MeV  $\text{C}^{12}$  ions, the activity produced had a half-life of  $159 \pm 5$  sec (Fig. 4). This value is reported as the half-life for  $\text{Fr}^{210}$ , but involves the assumption that no  $\text{Fr}^{211}$  was present in the sample. Since the excitation functions tail more on the high-energy side and since the cross section decreases as the number of neutrons boiled off increases (see Sec. IV) there may possibly be some contamination from  $\text{Fr}^{211}$ . This would have the effect of making the reported half-life of  $\text{Fr}^{210}$  too long. However, the half-life was also determined for  $\text{Fr}^{210}$  (presumably free from  $\text{Fr}^{211}$ ) formed from the  $\text{Au}^{197}$  plus low-energy  $\text{O}^{16}$  bombardments. The half-life measured was  $162 \pm 20$  sec, which is consistent with the reported half-life and which shows that the half-life cannot be very much shorter than the reported value. Attempts to resolve the alpha group into two components with a high resolution detector indicated that the energies must differ by less than 10 keV.

In order to confirm the chemical identity of this activity ( $\text{Fr}^{211}$ ), a sample containing a mixture of astatine, radon, and francium isotopes was produced from gold and thallium targets and collected on palladium leaf. The leaf was dissolved and the solution was passed through an ion-exchange column. The activity assigned to  $\text{Fr}^{211}$  followed  $\text{Fr}^{212}$ , although the separation from astatine was not complete. A much better separation was accomplished by collecting the sample on a platinum plate and flaming it (see  $\text{Fr}^{213}$  above). This showed rather conclusively that this activity too is an isotope of francium.

The mass assignment for  $\text{Fr}^{211}$  was confirmed by electrostatically collecting the alpha-decay daughter as recoils from a thin sample of the parent. Alpha-decay recoils were collected on one plate for 3 min and then on a second for 3 min. The most prominent activity found on these plates was identified as  $\text{At}^{207}$  by its half-life of  $1.6 \pm 0.2$  h and its alpha-particle energy of 5.75 MeV. All other activities on these plates were lower than this one by at least a factor of 10. The relative amounts of  $\text{At}^{207}$  on the two plates indicated that the parent half-life was  $195 \pm 15$  sec, which is consistent with the value obtained for the 6.55-MeV group assigned to  $\text{Fr}^{211}$ .

No attempt was made to milk the activity assigned to  $\text{Fr}^{210}$  since its daughter,  $\text{At}^{206}$ , has an alpha branch of less than 1%.<sup>17</sup> Also, one would have to go to fairly low cross sections to try to produce  $\text{Fr}^{210}$  relatively free from  $\text{Fr}^{211}$ . Thus the expected counting rates would be too low to detect. The assignment of  $\text{Fr}^{210}$  is made primarily on the basis of the excitation functions.

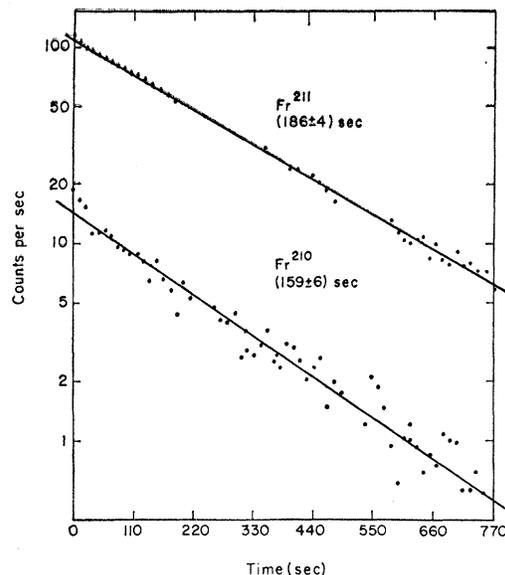


FIG. 4. Alpha-decay curves for  $\text{Fr}^{211}$  and  $\text{Fr}^{210}$ .

### C. $\text{Fr}^{209}$ and $\text{Fr}^{208}$

During the bombardment of  $\text{Tl}^{205}$ , as the energy of the  $\text{C}^{12}$  ions was increased above 95 MeV, an alpha group having an energy of  $6.65 \pm 0.02$  MeV appeared. This activity was also produced in the bombardments of  $\text{Tl}^{208}$  and  $\text{Au}^{197}$  as is shown in Figs. 1(B), 1(C), and 1(D). The excitation function from the  $\text{Tl}^{205} + \text{C}^{12}$  system appears to be due to a  $(\text{C}^{12}, 8n)$  reaction, which would assign this activity to  $\text{Fr}^{209}$  [Fig. 3(B)]. Once again there were indications that this alpha group is a result of two different isotopes. The  $\text{Tl}^{203} + \text{C}^{12}$  excitation function is somewhat broadened and distorted and no other alpha groups with an excitation function corresponding to a  $(\text{C}^{12}, 7n)$  reaction were found. The excitation function from the  $\text{Au}^{197} + \text{O}^{16}$  system is also consistent with the assignment to  $\text{Fr}^{209}$  and  $\text{Fr}^{208}$ . The fact that the excitation function does not appear to be broadened—although the spacing between the excitation functions is very large—is probably due to the fact that the peak cross section for the various neutron-evaporation reactions from the  $\text{O}^{16}$  bombardments decreases by a factor of about 4 for each additional neutron boiled off (see Sec. IV).

The half-life of this group was determined to be  $54.7 \pm 1.0$  sec when formed by 102-MeV  $\text{O}^{16}$  ions on  $\text{Au}^{197}$  and  $37.5 \pm 2.0$  sec when formed by 120-MeV  $\text{O}^{16}$  ions on  $\text{Au}^{197}$  (Fig. 5). These two values are reported here as the half-lives of  $\text{Fr}^{209}$  and  $\text{Fr}^{208}$ , respectively. There is the possibility that the  $\text{Fr}^{208}$  sample contained some  $\text{Fr}^{209}$  even though the sample was produced on the high-energy tail of the excitation function [Fig. 3(D)]. The decay, however, looks good and the half-lives are sufficiently different that a large amount of  $\text{Fr}^{209}$  would have been detected.

Recoils from samples collected from the bombard-

<sup>17</sup> R. M. Latimer, G. E. Gordon, and T. D. Thomas, J. Inorg. Nucl. Chem. 17, 1 (1961).

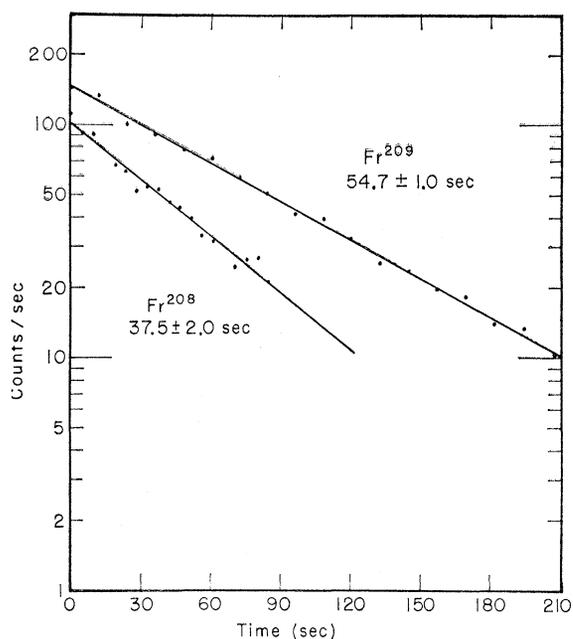


FIG. 5. Alpha-decay curves for  $\text{Fr}^{209}$  and  $\text{Fr}^{208}$ .

ment of  $\text{Tl}^{203}$  with 93-MeV  $\text{C}^{12}$  ions were electrostatically milked. There were two daughter groups present in the secondary samples. These were 26-min  $\text{At}^{205}$  with a 5.92-MeV alpha-particle energy, and a small amount of 1.6-h  $\text{At}^{207}$  with a 5.76-MeV alpha-particle energy. Again, due to the short half-life, we had interference from the primary recoils that collected on the secondary plates. From some of the "cleanest" samples, the half-life of the parent of  $\text{At}^{205}$  was found to be about 1 min, which is consistent with the value obtained for the alpha group we have assigned to  $\text{Fr}^{209}$ , and which indicates therefore that this group is the parent of  $\text{At}^{205}$ . The  $\text{At}^{207}$  present was a result of the small amount of  $\text{Fr}^{211}$  formed at this energy. Once again the decreased yield of  $\text{Fr}^{208}$  and the small alpha branch of its daughter,  $\text{At}^{204}$ , made it impractical to attempt to establish this parent-daughter relationship.

#### D. $\text{Fr}^{207}$ and $\text{Fr}^{206}$

The next alpha group that was studied has an energy of  $6.77 \pm 0.02$  MeV, which is the same as that of  $\text{Fr}^{213}$ . Figures 1(C) and 1(D) show spectra containing this group. The excitation function for this activity from the  $\text{Tl}^{203} + \text{C}^{12}$  system is very similar to that for the  $\text{Tl}^{205}(\text{C}^{12}, 8n)\text{Fr}^{209}$  reaction. If this is also a  $(\text{C}^{12}, 8n)$  reaction, then the new activity would belong to  $\text{Fr}^{207}$ . The excitation function for the  $\text{Au}^{197} + \text{O}^{16}$  system is also consistent with this being  $\text{Fr}^{207}$ , formed now by an  $(\text{O}^{16}, 6n)$  reaction. After the experience which we had with the previous two groups, it was quite natural to ask if this group, too, might belong to a pair of isotopes,  $\text{Fr}^{207}$  and  $\text{Fr}^{206}$ . Since, as has been mentioned before and will be discussed later, the cross section for the

$\text{Au}^{197}(\text{O}^{16}, 7n)\text{Fr}^{206}$  reaction is probably about one fourth the value of the  $\text{Au}^{197}(\text{O}^{16}, 6n)\text{Fr}^{207}$  reaction cross section, we cannot see any great effect on the excitation function for this group. Several things have led us to the conclusion that this is both  $\text{Fr}^{207}$  and  $\text{Fr}^{206}$ .

Figure 2 shows the half-lives obtained on the low-energy side of the excitation function for  $\text{Fr}^{207}$  and on the high-energy side for  $\text{Fr}^{206}$ . Because of the low yield, this latter half-life was taken on the side of the excitation function rather than way down on the tail. These half-lives of 18.7 sec and 15.8 sec are quite close and do not therefore clearly show a second isotope. A less accurate measurement on the high-energy tail at full  $\text{O}^{16}$  energy gave a half-life of  $14.4 \pm 2.0$  sec for  $\text{Fr}^{206}$ , which is consistent with the previous value but which indicates that the reported value may be a little high.

The use of a high-resolution detector enabled us to partially resolve these two alpha groups, as is shown in Fig. 1 (d). At the low bombarding energies, the group was narrow [like that of  $\text{Fr}^{205}$  in Fig. 1(d)] and had an energy of 6.77 MeV. At higher  $\text{O}^{16}$  energies, the additional peak at 6.79 MeV became evident. Even at full energy  $\text{O}^{16}$  the peak is broadened, indicating either that some  $\text{Fr}^{207}$  is still produced at this energy or that  $\text{Fr}^{206}$  has two alpha groups itself.

No milking experiments were attempted on these short-lived activities. However, the mass assignment of  $\text{Fr}^{207}$  is strengthened by the fact that  $\text{At}^{203}$  was found in the sample at the peak of the  $\text{Fr}^{207}$  excitation function. This activity could hardly be produced directly at this energy and is most probably produced from the decay of  $\text{Fr}^{207}$ . Additional evidence for  $\text{Fr}^{206}$  comes from the identification of the next alpha group as  $\text{Fr}^{205}$  (see Sec. III E).

#### E. $\text{Fr}^{205}$ and $\text{Fr}^{204}$

The bombardment of  $\text{Au}^{197} + \text{O}^{16}$  at high energies produced two more alpha groups which are shown in

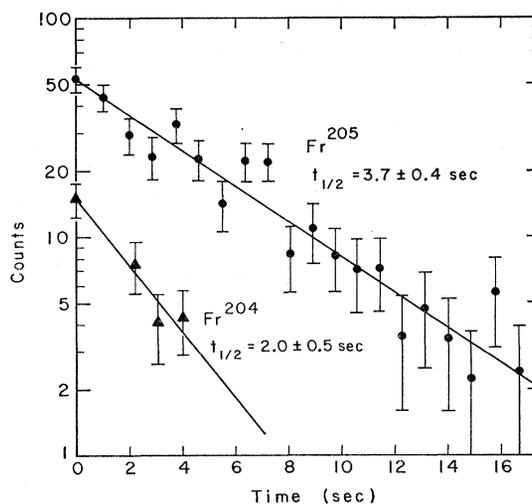


FIG. 6. Alpha-decay curves for  $\text{Fr}^{205}$  and  $\text{Fr}^{204}$ .

Figs. 1(c) and 1(d). The energies of these groups are  $6.91 \pm 0.02$  MeV and  $7.02 \pm 0.03$  MeV. The excitation functions were determined and are shown in Fig. 3(d) as the  $(O^{16}, 8n)$  and  $(O^{16}, 9n)$  reactions. The half-lives of these groups are  $3.7 \pm 0.4$  sec and  $2.0 \pm 0.5$  sec, respectively (Fig. 6).

The identification of the 6.91-MeV group as  $Fr^{205}$  was based on its excitation function and also on the excitation function of  $At^{201}$ . One can see  $At^{201}$  in the spectrum of Fig. 1(c). This activity followed an excitation function similar to the 6.91-MeV group. Again one would not expect any direct formation of  $At^{201}$  with an excitation function peaking at these energies and it is felt that it is produced from the decay of  $Fr^{205}$ . Since it follows the excitation function for the 6.91-MeV group, this would identify this group as the parent of  $At^{201}$ , namely,  $Fr^{205}$ . The assignment of the 7.02-MeV group to  $Fr^{204}$  is based on the excitation-function data and on alpha-decay systematics.

#### IV. DISCUSSION

##### A. Alpha-Decay Energies

The variation of alpha-decay energy of the isotopes of francium with mass number—or more specifically with neutron number—is seen to be quite similar to that of the three elements just below it, Po, At, and Rn. The isotope with 125 neutrons is seen to have the lowest alpha-decay energy. The energy increases rather sharply in going to larger neutron numbers and increases more slowly in going to the lighter isotopes. The phenomenon of pairs of isotopes having similar alpha energies is seen to occur in all four of these elements.<sup>16–18</sup> Polonium has two such pairs, astatine has three, and radon has two or possibly three. Now we find that francium also has three such pairs of isotopes and that these occur for the same neutron numbers as the others: 119 and 120, 121 and 122, and 123 and 124.

##### B. Cross Sections

Each of the excitation functions shown in Fig. 3 has its own arbitrary units and no attempt has been made to indicate the relative magnitude of the units used in different cases.

These relative cross sections for the various activities produced from a given compound system have been determined, however. In the  $C^{12}$  bombardments the peak cross section decreases rather slowly as the number of neutrons boiled off increases. In comparing the  $(C^{12}, 4n)$  with the  $(C^{12}, 8n)$  reaction, the magnitude of the peak cross section of the latter is about four times smaller. In the  $O^{16}$  bombardments the effect is much greater. To compare the cross sections, we assume that the peak cross section for a given group is due solely to the group formed by the lower number of neutrons emitted; e.g., the peak of the 6.77-MeV excitation func-

<sup>18</sup> W. Forsling, T. Alväger, L. W. Holm, O. Melin, J. Uhler, and B. Aström, *Arkiv Fysik* **19**, 83 (1961).

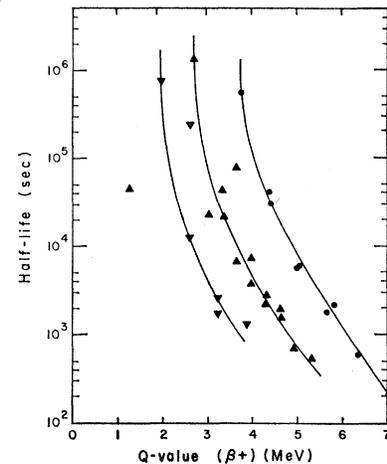


FIG. 7. Beta-decay half-lives of  $\bullet$  even,  $\blacktriangle$  odd  $A$ , and  $\blacktriangledown$  odd nuclides of Bi, Po, At, and Rn as functions of  $Q$  value for beta-plus decay.

tion is assumed to be due only to the  $(O^{16}, 6n)$  reaction and to have a negligible contribution from the  $(O^{16}, 7n)$  reaction. The cross section for the  $4n$  reaction is then 6 times greater than the  $6n$  reaction and would appear to be greater yet if it were not for the Coulomb barrier. The cross section for the  $8n$  reaction is  $\frac{1}{17}$  the  $6n$  cross section and that for the  $9n$  reaction is reduced to  $\frac{1}{16}$  the value of the  $8n$  reaction. Here the  $9n$  reaction may not have peaked yet at the highest available energies from the Hilac and this factor may then be lower. In this case then, there is a factor of 600 in going from the  $4n$  to the  $9n$  reaction. If this factor is roughly the same for each step—which looks reasonable if the complications at the highest and lowest energies are taken into account—then there is a decrease in cross section by a factor of about 3.6 for each additional neutron emitted. This large factor is presumably due to the fission competition which is going on. Since charged-particle emission is very small in this region and other conditions affecting the cross section would be small, we assume this factor is due solely to the fission competition. If fission occurs with comparable probability in each of these nuclides in the evaporation chain, then  $\Gamma_f/\Gamma$  (ratio of level width for fission to the total level width) is about 0.7. These data would indicate that the fission process is more important for the  $O^{16}$  than for the  $C^{12}$  bombardments; we explain this by the suggestion that fission-ability increases with increasing angular momentum.<sup>19,20</sup> Using the same assumptions as above, the ratio  $\Gamma_f/\Gamma$  for the  $C^{12}$  case would be about 0.3. This value is almost the same as that reported for the bombardment of  $Au^{197}$  with  $C^{12}$  ions.<sup>15</sup>

##### C. Beta Branches

To calculate alpha-reduced widths for these nuclides, one must know the alpha decay half-life and not just the total half-life. The beta branch of  $Fr^{212}$  is known to

<sup>19</sup> G. A. Pik-Pichak, *Zh. Eksperim. i Teor. Fiz.* **34**, 341 (1958) [English transl.: *Soviet Phys.—JETP* **7**, 238 (1958)].

<sup>20</sup> J. R. Huizenga and R. Vandenbosch, in *Nuclear Reactions* (North-Holland Publishing Company, Amsterdam, 1962), Vol. 2.

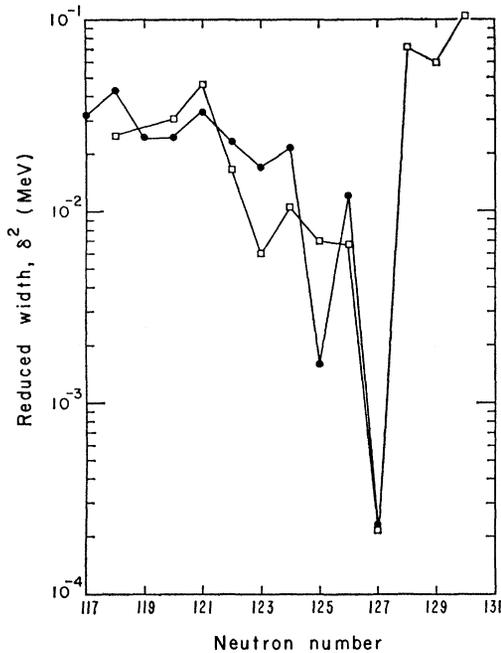


FIG. 8. Alpha reduced level widths for isotopes of ● Fr and □ Po.

be 56% of the total decay,<sup>21</sup> and we have measured the beta branch of Fr<sup>213</sup> to be 0.5% (see Fr<sup>213</sup> above). Thus, the alpha half-lives of these two isotopes can be calculated.

By looking at the systematics of beta decay in this region of the chart of the nuclides, one would qualitatively predict that the beta branches would be small and the alpha half-lives would be close to the total half-lives. However, in order to get a quantitative estimate of the beta branches, the beta decay half-lives of all the known bismuth, polonium, astatine and radon beta-plus emitters in this region were plotted versus the  $Q$  values for beta-plus decay. This resulted in the graph shown in Fig. 7 and the results fall on a family of three curves, depending on whether the nuclide is an even, an odd, or an odd- $A$  nuclide. The  $Q$  values are taken from the semi-empirical mass data of Seeger.<sup>14</sup> We determined the beta-decay half-lives for the francium isotopes from these curves, again using the predicted  $Q$  values of Seeger. Beta branches were then calculated from these half-lives and from the measured total half-lives, and are listed in Table II. The alpha-

TABLE II. Calculated beta-decay branches.

Nuclide	Beta branch	Nuclide	Beta branch
Fr <sup>211</sup>	0.10	Fr <sup>207</sup>	0.12
Fr <sup>210</sup>	0.19	Fr <sup>206</sup>	0.14
Fr <sup>209</sup>	0.10	Fr <sup>205</sup>	0.05
Fr <sup>208</sup>	0.15	Fr <sup>204</sup>	0.07

<sup>21</sup> E. K. Hyde, A. Ghiorso, and G. T. Seaborg, Phys. Rev. **77**, 765 (1950).

TABLE III. Summary of results.

Nuclide	$Q_\alpha$ (MeV)	Half-life (sec)	$\delta^2$ (MeV)
Fr <sup>213</sup>	6.90±0.01	33.7±1.5	0.0122
Fr <sup>211</sup>	6.68±0.02	186 ±4	0.0215
Fr <sup>210</sup>	6.68±0.02	159 ±5	0.0172
Fr <sup>209</sup>	6.78±0.02	54.7±1.0	0.0230
Fr <sup>208</sup>	6.78±0.02	37.5±2.0	0.0328
Fr <sup>207</sup>	6.90±0.02	18.7±0.8	0.0244
Fr <sup>206</sup>	6.92±0.02	15.8±0.4	0.0243
Fr <sup>205</sup>	7.05±0.02	3.7±0.4	0.0427
Fr <sup>204</sup>	7.16±0.03	2.0±0.5	0.0318

decay half-lives used below were also calculated from these beta-decay estimates.

#### D. Alpha Reduced Widths

Alpha-decay reduced level widths ( $\delta^2$ ) were calculated for each of the francium isotopes using Ramussen's method.<sup>22</sup> The alpha reduced width is defined by the expression

$$\lambda = \delta^2 P / \hbar,$$

where  $\lambda$  is the alpha-decay constant,  $P$  is the barrier penetrability, and  $\hbar$  is Planck's constant. The potential used in the calculation of  $P$  is the sum of the Coulomb potential and the real part of the alpha-nuclear potential derived from optical-model analysis of alpha-particle elastic-scattering data and given by

$$V(r) = -1100 \exp\{-[r - (1.17A^{1/3}/0.574)]\} \text{ MeV},$$

where  $r$  is the distance in F and  $A$  is the mass number.<sup>23</sup> Because of our lack of knowledge of the spin assignments of parent and daughter, the centrifugal potential was taken to be zero; in other words, values of the reduced width were calculated for  $l=0$  waves only. The calculated values are given in Table III and Fig. 8 compares these values with those of the analogous polonium isotopes.<sup>22,24</sup> The variation with neutron number is seen to be similar for the two cases. The 127-neutron isotope, which decays across the closed shell, has a very small reduced width.<sup>12</sup> As the neutron number decreases, moving away from the closed shell, the reduced width follows a generally increasing trend. Table III summarizes the results obtained for the francium alpha emitters.

#### ACKNOWLEDGMENTS

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<sup>22</sup> J. O. Rasmussen, Phys. Rev. **113**, 1593 (1959).

<sup>23</sup> G. Igo, Phys. Rev. Letters **1**, 72 (1958).

<sup>24</sup> J. O. Rasmussen, Phys. Rev. **115**, 1675 (1959).