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NEW ISOTOPES OF THORIUM STUDIED WITH AN IMPROVED HELIUM-JET RECOIL TRANSPORT APPARATUS

Kalevi Valli and Earl K. Hyde

May 1968

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ABSTRACT

Five neutron-deficient isotopes of thorium lighter than mass 218 were studied at the Berkeley Heavy Ion Linear Accelerator by bombardment of enriched ^{206}Pb with ^{16}O. Silicon (Au) surface-barrier detectors were used in on-line measurements performed to determine α-decay characteristics. Mass number assignments of ^{213}Th through ^{217}Th were made on the basis of excitation functions and systematic trends in α-decay properties. Half lives and α-particle energies were determined. New information was also obtained about the α-decay characteristics of ^{216}Ac, ^{215}Ra, ^{214}Fr, and ^{212}At. Systematic occurrence of complex structure in the α-decay of even-Z 125-neutron nuclei is discussed and spins and parities are suggested for three lowest energy levels of ^{211}Ra, ^{209}Rn, and ^{207}Po. Systematic occurrence of isomerism in odd-Z 127-neutron nuclei is also discussed and spins and parities are suggested for several levels. On the basis of the extreme regularity of the experimental α-energies, predictions are made for the energies of several unknown neutron-deficient isotopes of thorium, protactinium, uranium, and neptunium. In the experimental work the helium jet transport method was used to remove the alpha emitting products from the target region. Technical details concerning the application of the method to millisecond activities are discussed.

I. INTRODUCTION

This report deals chiefly with the discovery of 5 thorium isotopes with mass numbers ranging from 213 to 217 produced at the Berkeley Heavy Ion Linear Accelerator (HILAC) by bombardment of ^{206}Pb targets with beams of ^{16}O and studied by on-line techniques of α-spectroscopy. For nuclei with Z = 90 these are extremely deficient in neutrons but the extra stability associated with the 126-neutron shell decreases the α-decay energies enough so that half-lives are lengthened to the range of milliseconds to seconds. This study represents an extension of our previous studies of the alpha decay properties of isotopes lying near or below the 126 neutron shell of the following elements: polonium and astatine,[1] radon,[2] francium,[3] radium,[4] and actinium.[5] This series of studies together with previously available information[6] makes it possible with considerable accuracy to interrelate many nuclei in the group of nuclides with more than 82 protons and 127 or fewer neutrons and to derive important properties such as atomic masses, neutron and proton binding energies, etc.

In the present work it was necessary to make some modification in the experimental techniques to permit faster collection of the recoil nuclei. As a result of this greater collection speed it was possible to obtain new information on several isotopes containing 127 neutrons, which were unobserved or incompletely studied in the papers published previously. In a separate section below there is presented a discussion of results on ^{214}Fr, ^{215}Ra, ^{216}Ac, and ^{217}Th.

Compound nucleus reactions involving as their first step the complete fusion of the ^{16}O projectile and ^{206}Pb target were required for the preparation of the thorium isotopes and the characterisitics of these reactions (principally the shape of the excitation function) were used in the identification of the

atmospheres. The helium flow rate was 60 cm^3/sec. From this and the cross section of the capillary (0.13 mm^2) the velocity of the helium at exit was 460 m/sec and the kinetic energy of an atom of mass 216 swept along with it was 0.24 eV.

In the system illustrated in Fig. 1 the time between production and detection is determined by only two factors: one, how long it takes to slow down and sweep the recoil nucleus out of the chamber in the gas jet and two, how soon after the beam burst the detector can be gated on. When the target chamber was operated at the minimum size, the volume was about 1.0 cm^3. To remove this amount of gas through the capillary required 17 msec. However, some fraction of the activity was collected in a much shorter time. This 17 msec turnover time meant that nuclides with half lives much shorter than 17 msec underwent considerable decay before collection and the observed intensities of these peaks do not reflect the true production rates for these nuclides. It also meant that in any half-life determination performed during the interval (~20 msec) between two subsequent beam bursts, both collection and decay of the activity had to be considered.

The long capillary made it possible to shield the detector from the highly radioactive target area. The counter could be gated on immediately after the beam burst and in fact, it could be used even during the beam bursts although the alpha spectra taken during the beam-on period had much greater background caused by neutrons and gamma rays.

Some aspects in the design of the apparatus are discussed in an appendix at the end of the report.

B. Bombardment Techniques

The reactions used to produce the thorium activities were:

$$^{206}Pb(^{16}O,xn)^{222-x}Th$$

where x refers to the number of evaporated neutrons. The target consisted of 2.8 mg/cm^2 of separated ^{206}Pb electroplated on 1.7 mg/cm^2 copper foil. The ^{206}Pb was obtained from Oak Ridge National Laboratory and had the following isotopic composition: 97.22% ^{206}Pb, 1.34% ^{207}Pb, and 1.39% ^{208}Pb.

The 166 MeV ^{16}O beam of the HILAC was bent 15° in a steering magnet and directed through 20 feet of beam pipe before entering the target chamber (Fig. 1) through the entry window consisting of the ^{206}Pb target, its copper foil backing and a 1.7 mg/cm^2 nickel foil, the lead deposit being on the inside surface of the window. The beam energy was varied by inserting stacks of 1.72 mg/cm^2 aluminum foils in front of the target. The range-energy relationships of Northcliffe[10,11] were used to calculate the beam energy degradation in the absorber and in the chamber window.

In a typical set of experiments a steady beam current of about 350 nA ^{16}O was passed through the chamber and measured in a Faraday cup. The run was terminated when the integrated beam reached some selected value. This usually took about 30 min. The products of the reaction were continuously collected and the amplitudes of the α pulses from the detector-amplifier system were measured with a 1024-channel ADC unit interfaced to a PDP-7 computer. The output of the amplifier was gated off during the 5 msec beam bursts. At the end of the run the number of absorbers was changed and a new run began, starting about 10 min after the end of the preceding run. Ten such runs were made until

the beam energy was reduced to the Coulomb barrier value which is about 75 MeV. The spectra from all runs were plotted and the areas under individual alpha peaks computed and replotted as excitation functions (yield-versus-beam-energy curves).

The energy scales for the alpha spectra were determined in two steps. Primary alpha standards from the actinium and thorium series, in particular ^{211}Bi 6.6222 MeV, ^{219}Rn 6.8176 MeV, ^{215}Po 7.3841 MeV, and ^{212}Po 8.7854 MeV,[12] were used to determine the energies of prominent peaks assigned in this study to ^{216}Th, ^{215}Ra, and ^{214}Fr. The reaction products and the primary standards were measured simultaneously to eliminate any possibility of shifts due to unknown factors. Then the peaks for those three reaction products plus ^{215}Ac, 7.605 MeV[5] and ^{214}Ra, 7.136 MeV[4] were used as internal secondary standards to calibrate the scale for the other peaks in the spectra.

C. Half-life Determinations

The measurement of decay periods was done with a special electronic programmer which synchronized the operation of the beam with the recording of spectra during selected time periods and controlled the storage of these spectra in separate parts of the computer memory. The programmer could operate in several modes. In the case of relatively long half lives such as the 1.2 sec of ^{215}Th or the 125 msec of ^{214}Th, analysis was first blocked for a bombardment period of several half lives to allow the activity to reach a saturation value. Then the beam was turned off by the central programmer and data were collected during 16 equal time periods and stored in 16 sections of the computer memory. Then the beam was turned on again by the programmer and the process was repeated

until sufficient statistical accuracy was obtained. A normal overall running time for the half-life determination was one or two hours.

In a faster mode of operation analysis was blocked during the beam bursts and then, during the intervals between subsequent bursts, alpha pulses were collected in 16 equal time periods and stored in 16 sections of the computer memory. This process was continued for as many beam bursts as necessary to build up sufficient statistical accuracy in the sixteen 256-channel spectra.

In still another mode of operation analysis was not blocked during the beam. Instead a timing signal from the accelerator turned on the amplifier output just as the beam pulse started. Then spectra were recorded for 8 or 16 successive time periods spread over the beam pulse and the time between beam pulses. The successive spectra were stored in separate sections of the computer. In this case the spectra from the counting periods covering the beam pulse contained a higher general background. However, many peaks could be resolved and in particular important information on certain very short lives peaks, notably ^{217}Th, could be obtained from an examination of the spectra taken just before and just after the end of the beam pulses.

Owing to the 17 msec turnover time in our chamber the observed count-rate-versus-time curves in the case of the last two modes of operation have a complex shape similar to the growth and decay of a daughter product of radioactive decay. Half lives determined from the descending parts of these curves are slightly longer than the true half lives.

III. RESULTS

A. General Comments

Figure 2 shows sample spectra taken at representative beam energies. The alpha emitting products are quite numerous because the nuclear reactions are complex and because many radioactive daughter products are formed in the rapid decay of the primary products.

The compound nucleus is ^{222}Th and the excitation energies at the Coulomb barrier and at the maximum beam energy are sufficient for the evaporation of 3 and 10 neutrons, respectively. Hence we can expect to form thorium isotopes of mass 219 down to 212 each with a characteristic excitation function. Each of these should reach a maximum at a beam energy most favorable for the evaporation of a certain number of neutrons and decrease at higher beam energies as the probability for the evaporation of one more neutron increases. A series of excitation functions having these characteristics does appear in Figs. 3 and 4. From the extreme regularities discussed in our previous papers for the variation of alpha energy versus neutron number near the magic number 126 we can predict with confidence the alpha energies and half lives of the thorium isotopes. These predictions help us to identify individual isotopes. The predicted and observed half lives of ^{216}Th and the lighter isotopes were in a convenient range for our helium transport system. It was borderline whether our collection time would be fast enough for us to observe ^{217}Th but we did succeed in identifying it. We expected the half lives of ^{218}Th and ^{219}Th to be too short for detection with our techniques and in fact we did not observe them. We discuss the individual thorium isotopes in Sec. IIIB below.

A second main group of products is the isotopes of actinium, radium, francium, radon, astatine, and polonium—particularly those with 126 or fewer neutrons—made by a variety of reactions. Actinium and radium isotopes can be formed by the rapid electron capture or α-decay of thorium parent isotopes but, owing to the large loss of the parent nuclei by nuclear fission, a more likely mode of formation is the direct emission of a proton or an alpha particle together with several neutrons during the de-excitation of the compound nucleus. The high intensities of the actinium and radium peaks compared to the thorium peaks, as well as the broad excitation functions, are in agreement with this conclusion. One consequence of this is that the presence of actinium and radium isotopes of known mass number is not helpful in the assignments of the thorium mass numbers. Some of the francium, astatine, and polonium activities come from radioactive decay of precursors but a considerable fraction may come from reactions not involving a complete fusion of the target and projectile.

A third category of products is a group of very short-lived nuclides containing 127 neutrons. We obtained some new information on ^{217}Th, ^{216}Ac, ^{215}Ra, and ^{214}Fr which we discuss in Sec. IIIC.

The fact that nuclear fission accounts[13,14] for about three quarters of the total reaction cross section deserves mention for two reasons. First of all fission occurs primarily in the excited thorium nuclei formed in the de-excitation of the compound nucleus so that the yields of thorium isotopes are greatly reduced. The nuclei of lower atomic number do not suffer so much from this fission competition and hence contribute relatively much more to the alpha spectra. A second remark is that the range of the fission products is much larger than that of the α-emitting reaction products so that most of the fission products are not stopped in the helium before they strike the walls of the target chamber and hence are not transported out by the helium stream.

B. Thorium Isotopes with 126 or Fewer Neutrons

1. Thorium 216

From the regularities in alpha decay properties displayed in Fig. 9 the alpha energy of ^{216}Th was expected to be between 7.90 and 7.95 MeV and on the basis of the known half lives of ^{215}Ac, ^{214}Ra, ^{213}Fr, ^{212}Rn, and ^{211}At—all nuclei with 126 neutrons—the half-life of ^{216}Th could be predicted to lie between 10 and 50 msec. In Fig. 2 an alpha group is presented at 7.921 ± 0.008 MeV. The measured decay period for this group is 28 ± 2 msec. Supporting evidence for the assignment of this activity to ^{216}Th comes from the excitation function shown in Figs. 3 and 4 which has the shape characteristic of a compound nucleus reaction product. The yield maximum at 114 MeV beam energy corresponds to an excitation energy of 69 MeV for the ^{222}Th compound nucleus (Table I), which is reasonable for the evaporation of 6 neutrons. Additional support for the assignment comes from the fact that the yield maximum occurs at a beam energy 14 MeV lower than that for the yield maximum of ^{215}Th which isotope can be identified with certainty on the basis of its distinctive decay properties. It is probable that ^{216}Th undergoes decay by electron capture to ^{216}Ac to a slight extent. Our results give an upper limit of 0.6 percent for such branch decay. The true value could be substantially lower.

2. Thorium-215

From the regularities in properties of other nuclides with 125 neutrons, namely, ^{214}Ac, ^{213}Ra, ^{212}Fr, and ^{211}Rn, we can expect the alpha spectrum of ^{215}Th to have several alpha groups and thus to be distinguished from neighboring thorium isotopes which are expected to have a single prominent alpha transition. Furthermore, we can expect the lowest energy group to have an energy between 7.50 and 7.55 MeV. In addition, we can expect ^{215}Th to have the longest half life of any thorium isotope prepared in this study.

We assign three groups in our spectra (Fig. 2) to ^{215}Th: 7.522 ± 0.008 MeV, 40 ± 3%; 7.393 ± 0.008 MeV, 52 ± 3%; and 7.331 ± 0.010 MeV, 8 ± 3%. A half life of 1.2 ± 0.2 sec was measured for all three groups. The excitation functions of the two most prominent groups are shown in Figs. 3 and 4. The shapes of the curves are typical for a compound nucleus reaction. The maximum yield occurs at a beam energy of 128 MeV, which corresponds to an excitation energy of 83 MeV for the ^{222}Th compound nucleus. This amount to a reasonable average of 11.9 MeV (Table I) removed in the evaporation of each of the seven neutrons.

The third ^{215}Th group at 7.331 MeV is most clearly visible at about 125 MeV beam energy because at higher beam energies it is obscured by the intense 7.37 MeV peak that belongs to ^{212}Ac and ^{213}Ac.

We looked for a possible electron capture branching of ^{215}Th by measuring the half life for ^{215}Ac observed in bombardments carried out with 125 MeV ^{16}O ions (i.e. at the peak of the ^{215}Th excitation function). The ^{215}Ac alpha group decayed with its previously reported[5] half life of 170 msec and there was no 1.2 sec component. From the data we can set an upper limit of 1.5 percent to the EC branching of ^{215}Th.

3. Thorium-214 and Thorium-213

Regularities in alpha decay properties suggest that ^{214}Th and ^{213}Th should have closely similar energies between 7.65 and 7.73 MeV (see Fig. 9) and similar half lives in the range 50 to 200 msec. The small peak at 7.68 MeV shown in Fig. 2 is assigned to these isotopes. The maximum of the excitation function shown in Figs. 3 and 4 falls in the proper energy range for a thorium isotope of lower mass than 215. The width of the excitation maximum is some-

what greater than expected for one isotope but on the other hand is not so broad as to indicate clearly that the 7.68 MeV alpha activity is a mixture of two isotopes. However, when the alpha particle energy and the half life were carefully measured at two widely spaced points of the excitation function different values were obtained. At a beam energy of 142 MeV an alpha energy of 7.680 ± 0.010 MeV and a half life of 125 ± 25 msec were found and assigned to ^{214}Th. At a beam energy of 157 MeV an alpha energy of 7.690 ± 0.010 and a half life of 150 ± 25 msec were obtained and assigned to ^{213}Th. These determinations are less certain than those for the higher mass thorium isotopes.

C. Short-Lived Isotopes with 127 Neutrons

In Fig. 9 a pronounced break occurs in the variation of alpha energy with neutron number at the 126 neutron shell. The nuclides with 127 neutrons have much higher decay energies and hence shorter half lives than do the nuclides with 126 or fewer neutrons. The half lives of most of the 127 neutron nuclides are so short that in our previous studies with a longer helium transport time[1,5] we were able to observe them only if they were produced by the decay of a long-lived parent. In the present study, however, the transport time was sufficiently short that we were able to collect and identify alpha groups from several of these nuclides including ^{217}Th, ^{216}Ac, ^{215}Ra, and ^{214}Fr. The nuclide ^{213}Rn was not observed because it is gaseous and does not stick to the collector foil. A summary of our results on the 127-neutron nuclides is given in Table III. We include revised alpha energies of ^{212}At and ^{212m}At in the list although that isotope was not a subject of further study. The new information that we collected on these nuclides came as a result of our study of

the thorium isotopes and is incomplete. A more definitive study would require additional work with a faster system and the investigation of other projectile target combinations.

1. Thorium-217

^{217}Th contains one neutron more than the filled 126-neutron shell. From the known half lives of ^{216}Ac, ^{215}Ra, ^{214}Fr, and ^{213}Rn, which also have 127 neutrons each, the half life of ^{217}Th can be estimated to be between 50 and 200 μsec. From the systematic regularities in alpha-decay properties displayed in Fig. 9 the alpha energy of ^{217}Th is predicted to lie between 9.20 and 9.35 MeV.

The speed characteristics of our chamber were not suitable for the study of such a short-lived activity and under the operating conditions used to measure most of our spectra we did not observe this activity. However, in half-life measurements that were run continuously during and between beam bursts (see the third mode of operation in Sec. IIC), a very short-lived group was observed at 9.250 ± 0.010 MeV. It was visible only in spectra taken during a few milliseconds before and after the end of each beam burst. The spectrum shown in Fig. 5 was obtained by combining two such spectra. We obtained an upper limit of 0.3 msec for the half life.

Similar measurements during and just after the beam burst were made at other beam energies and the results were used to determine a rough excitation function for the 9.250 MeV group. This curve is drawn with a broken line in Fig. 3 to indicate that it comes from a separate set of measurements. The curve has a shape typical for a compound nucleus reaction. The yield maximum at 102 MeV beam energy corresponds to an excitation energy of 57 MeV for the

compound nucleus ^{222}Th (Table I). This is a proper amount for the evaporation of five neutrons in the de-excitation process, indicating strongly that the 9.250 MeV activity belongs to ^{217}Th.

2. Actinium-216

Rotter et al.[17] reported a half life of 0.39 ± 0.03 msec and an alpha energy of 9.14 ± 0.03 MeV for ^{216}Ac prepared by the reaction of ^{12}C ions with ^{209}Bi in the Dubna cyclotron. In our α spectra from products of the ^{206}Pb + ^{16}O reaction we observed two weak peaks of approximately equal intensity at 9.020 and 9.105 MeV. We verified that this was the same activity observed by Rotter et al. by identifying the same two groups in samples prepared by their method; when a ^{209}Bi target was bombarded with 88 MeV ^{12}C ions the 9.020 and 9.105 MeV peaks were observed in good yield. On such samples the half life was measured by the technique discussed in Sec. IIC with spectra recorded for 1 msec intervals during and after the beam burst. The result, shown in Fig. 6, indicates an apparent half-life of 0.56 msec. The ratio of the 9.020 and 9.105 MeV peaks remained 1:1 throughout the measurement. This half life is an upper limit because of the influence of the transport time and the true value is about 0.5 msec. We do not completely understand why our method gives a value so much longer than the 0.39 msec figure reported by Rotter et al.,[17] but as their value was determined in a more direct way it should be the more correct.

In the course of these half life measurements we observed that two small peaks at 8.198 and 8.283 MeV also had the same growth and decay characteristics. See Fig. 6.

For confirmation of the results on ^{216}Ac we prepared additional samples by the bombardment of ^{205}Tl with ^{16}O. Based on our previous excitation functions[5] for ^{214}Ac and ^{215}Ac produced by ^{205}Tl(^{16}O,xn) reactions we chose a bombardment

energy of 100 MeV for maximum production of ^{216}Ac. Under these conditions the 9.020 and 9.105 MeV peaks were observed in good yield and in equal abundance (±2 percent) as shown in Fig. 7. In addition the small peaks at 8.198 and 8.283 MeV were reproduced (see also Fig. 7). A half life measurement gave results for the combined 9.020 and 9.105 MeV peaks and the combined 8.198 and 8.283 MeV peaks identical to those shown in Fig. 6.

The energy separation between the 8.198 and 8.283 MeV peaks is 85 keV which is the same as the energy separation of the prominent 9.020 and 9.105 MeV peaks. Isomerism is to be expected in ^{216}Ac for the reasons discussed in Sec. IVC below and it is our conclusion that the 4 peaks under discussion here can be attributed to the decay of two isomeric forms of ^{216}Ac separated in energy by 87 keV and having nearly identical half lives. The radiations assigned to the two forms are as follows: ^{216m}Ac (8.283 MeV 3±0.5%, 9.105 MeV 97±0.5%) and ^{216}Ac (8.198 MeV 2±0.5%, 9.020 MeV 98±0.5%). All of our observations of the behavior of these 4 peaks in the ^{209}Bi + ^{12}C and ^{205}Tl + ^{16}O reactions are consistent with these assignments.

In the case of the ^{206}Pb + ^{16}O reaction we encountered some interference in our study of the 9.020 and 9.105 MeV peaks owing to the presence of an additional peak at 9.01±0.02 MeV with a half life of 25±2 msec. For example at an ^{16}O ion energy of 96 MeV the 9.01 MeV peak with a 25 msec half life was 4-fold greater than the 9.105 MeV peak, the latter having a half life of 0.5 msec. Preliminary work, not discussed in this report, indicates that this 9.01 MeV α activity is ^{216}Fr kept alive by a 25 msec ^{220}Ac parent. The possibility that this activity could be an isomer of ^{216}Ac formed by the electron-capture decay of 28 msec ^{216}Th was eliminated by noting that the yield variations of the 9.01-MeV activity and of ^{216}Th with ^{16}O ion energy were markedly different.

3. Radium-215

The earliest information on ^{215}Ra came from an unpublished study of Griffioen and Macfarlane[18] who reported a half life of 1.6 msec and an alpha energy of 8.7 MeV. Rotter et al.,[17] reported an energy value of 8.73 MeV. In our previous study of actinium isotopes[5] we observed a peak at 8.70 ± 0.02 MeV which we attributed to ^{215}Ra formed by electron capture of ^{215}Ac.

In the present study ^{215}Ra was produced in abundance at all beam energies (see Fig. 2). Our revised value of the alpha energy is 8.698 ± 0.005 MeV. The excitation function of this group is shown in Fig. 3.

We have evidence for two other groups in lesser intensity. At a beam energy of 95 MeV where ^{215}Ra is produced in greatest intensity two weak groups where observed at 8.168 ± 0.008 MeV and 7.880 ± 0.008 MeV (Fig. 2). Insofar as the excitation functions for these peaks could be followed (Fig. 3) they were parallel to that of the intense 8.698 MeV group. Interference from the 7.921 MeV group of ^{216}Th made it impossible to observe the 7.880 MeV group at beam energies above 100 MeV. Measurement of the half lives of the three ^{215}Ra groups is illustrated in Fig. 8. Within the statistical scatter of the data points the decay curves of the 8.168 MeV and 7.880 MeV groups are similar to that of the 8.698 MeV group. Additional evidence for the assignments came from bombardments of separated ^{206}Pb and ^{208}Pb with ^{20}Ne. The 8.168 MeV and 7.880 MeV peaks were observed in those spectra only at beam energies where the yield of the 8.698 MeV group was at maximum.

The relative intensities of the three ^{215}Ra groups are: 8.698 MeV 95.7 ± 1.0%, 8.168 MeV 1.3 ± 0.5%, and 7.880 MeV 3.0 ± 0.5%. The half life is 1.7 ± 0.2 sec.

4. Francium-214

The first information about ^{214}Fr came from an unpublished study by Griffioen and Macfarlane[18] according to which the half life is 3.9 msec and the alpha energy 8.55 MeV. Rotter et al.[17] reported an alpha energy of 8.53 MeV. In our previous study of radium isotopes[4] we found a weak alpha peak at 8.430 ± 0.008 MeV with an excitation function and apparent half life similar to 2.6 sec ^{214}Ra and assigned it to ^{214}Fr formed by electron capture decay of ^{214}Ra. Somewhat later Torgerson, Gough, and Macfarlane[19] reported to us their observation of two alpha groups from ^{214}Fr, one at 8.546 MeV and another at 8.478 both with a half life of about 3.4 msec and called attention to the possibility of the occurrence of ^{214}Fr isomers similar to the ^{212}At isomers reported by Jones.[20]

In the present study we observed four alpha peaks which we assign to ^{214}Fr with energy values 8.549, 8.477, 8.426, and 8.353 MeV, all with error limits of 0.008 MeV. The groups are shown in Figs. 2 and 5. A half life of 3.6 ± 0.5 msec was determined for the 8.549 and 8.477 MeV groups and 5.5 ± 0.5 msec for the other two groups. The excitation functions of the 8.549 and 8.477 MeV groups (Fig. 4) differ considerably from those of the 8.426 and 8.353 MeV groups. This difference is analogous to the 15 MeV displacement of the excitation function for ^{149m}Tb relative to that for ^{149}Tb in reactions induced in ^{139}La by ^{16}O ions.[21] Shifts of this type can be caused by selective formation of the high spin isomer from compound nuclei of high angular momentum.[21,22,23] Because of these differences in the half lives and excitation functions, we assign the 8.426 and 8.353 MeV groups to the ground state of ^{214}Fr and the other two groups to an isomer ^{214m}Fr. Relative intensities of the α groups are given in Table III. The decay scheme of ^{214}Fr is discussed further in Sec. IVC.

IV. DISCUSSION

A. α-Energy Systematics

It is of some interest to see how the new data on 23 alpha groups listed in Tables II and III fit in with the α data on neighboring nuclides. In Fig. 9 we present a revised plot of α energies versus neutron number. The systematic trends in this figure have been discussed in our previous reports.[4] By extrapolation of these curves it is possible to estimate unknown alpha energies for several nuclides with greater accuracy than is possible with existing mass formulas. In Table IV we list estimated α energies for a few unknown isotopes of thorium, protactinium, uranium, and neptunium. These estimated points are also shown in Fig. 9.

B. Complex Alpha Decay of 125-Neutron Nuclei

Even-Z nuclei with 125 neutrons have pronounced complex structure in their alpha spectra, which is of interest because it provides information on levels in the daughter nuclei. Figure 10 summarizes the data we wish to discuss. The data on ^{215}Th come from the present study, those for ^{213}Ra come from one of our previous papers,[4] and those for ^{211}Rn and ^{209}Po come from the references listed in The Table of Isotopes by Lederer, Hollander, and Perlman.[24] In the case of the daughter nuclei ^{205}Pb and ^{207}Po many other excited levels are known but we show only the lowest-lying ones, which are the ones populated by alpha decay. The properties of the levels of ^{205}Pb are well established and there seems little question that the 1/2-, 5/2-, and 3/2- levels can be assigned to the $p_{1/2}$, $f_{5/2}$, and $p_{3/2}$ wave functions of the independent particle

shell model. The 5/2 and 1/2 levels are also well established in ^{207}Po and, in particular, the ground state spin has been established to be 5/2 by the work of Axensten and Olsmats.[25]

Figure 10 reveals a regular trend in the relationship of the three lowest levels and while the alpha spectra do not establish the spins of the levels in ^{209}Rn and ^{211}Ra it is tempting to make the assignments shown in parentheses and to regard the shifts in the level positions as a reflection of the operation of residual internucleon forces as the proton pairs are increased beyond the 82-proton shell. No one has treated this particular problem with any of the current theoretical models for the residual internucleon forces although the shift in these levels in going from ^{207}Pb to ^{205}Pb has been treated by several authors (see for example Ref. 26 and 27).

Assuming that this identification of the levels shown in Fig. 10 is correct we can make the predictions of the levels of ^{213}Th and the α-decay pattern of ^{217}U shown in Fig. 10 and in Table IV.

C. Isomerism in Odd-Odd Nuclei with 127 Neutrons

As a result of the work of Jones[20] on ^{212}At, of Torgerson, Gough, and Macfarlane[19] on ^{214}Fr of ourselves in the present study on ^{214}Fr, and of the authors cited in The Table of Isotopes[24] on ^{210}Bi we now have proof of the systematic occurrence of isomerism in nuclides of odd elements above lead containing 127 neutrons. The cases are summarized in Fig. 11.

The isomerism can be explained by the high spins of the odd neutron and proton. The shell model predicts $g_{9/2}$ and $h_{9/2}$ respectively so that close lying levels of 0- and 9- are possible. The case of ^{210}Bi is particularly

interesting from the standpoint of theory[28] because it provides an opportunity to test the np interaction for a single neutron and single proton of high spin beyond a double-closed-shell core. The residual interaction between these two particles gives rise to a multiplet of 10 negative-parity levels with spins from zero to nine. The correct ordering of these levels seems to occur only if a short-range tensor force is added to a central force of the type successfully used to explain the nn interaction in such nuclei as ^{206}Pb. A particularly interesting feature in ^{210}Bi is that the ground state is 1- rather than 0-. The 0- level occurs at 47 keV and then a wide spin gap occurs because the next state is 9- at 250 keV. Theoretical calculations on ^{212}At, ^{214}Fr, and ^{216}Ac would be more complex because of the extra pairs of protons beyond the closed shell but the experimental results prove that a wide gap in spin is preserved in the ground state multiplet, at least for the first two. The data also suggest strongly that 1- rather than 0- is preserved as the ground state in ^{212}At and ^{214}Fr. This conclusion comes from the fact that the pair of alpha groups from the isomer have the same energy separation as the pair from the ground state indicating that the ground and first excited states are being populated in both instances. But this could not be the case if the ground state of ^{212}At or ^{214}Fr were 0- because the selection rules of alpha decay would forbid a 0- to 4+ transition and it is highly likely that the ground state and first excited state of the daughter nuclei are 5+ and 4+, respectively, resulting from the coupling of $p_{1/2}$ neutron and $h_{9/2}$ proton.

In the case of ^{216}Ac the systematic trends would suggest the occurrence of isomerism. As mentioned in Sec. IIIC-2, we have observed 4 α-groups which we tentatively attribute to isomeric decay in ^{216}Ac. The proposed scheme is shown in Fig. 11.

ACKNOWLEDGMENTS

We thank Dr. Albert Ghiorso for his interest in and support of this work. We are indebted to Mr. Richard Leres for the design of the electronic programmer used for the selection of mode of data collection and routing of data to the PDP-7 computer memory. We used the ADC unit and other interface hardware to the computer designed by Dr. Lloyd Robinson and employed his programs during data taking. Mr. Charles Corum contributed his usual excellent ideas to the mechanical design. Finally we acknowledge the contributions of Professor R. D. Macfarlane to our program during a period of collaboration with one of us (KV) in the summer of 1967.

FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

Fig. 1. Schematic diagram of the present apparatus.

Fig. 2. Alpha spectra showing activities from the $^{206}Pb + ^{16}O$ reactions at three beam energies. The spectra were recorded continuously between beam bursts. The beam current, 0.3 μA, was integrated over the measuring time, 40 min, to the same total in each measurement. These spectra and others were used to construct the excitation functions shown in Figs. 3 and 4.

Fig. 3. Excitation functions for the thorium and radium isotopes produced in the $^{206}Pb + ^{16}O$ reactions. The experiments were run from high to low beam energies with about 10 min intervals between measurements. See the caption of Fig. 2 for other details. The ^{217}Th - curve was obtained from a separate set of experiments as discussed in Sec. IIC1.

Fig. 4. Excitation functions for the thorium actinium and some francium activities produced in the $^{206}Pb + ^{16}O$ reactions. See the captions of Figs. 2 and 3 for details.

Fig. 5. Alpha spectrum showing the 9.250 MeV group of ^{217}Th and a few other groups. The spectrum was obtained by adding together two runs made at 96 MeV and 106 MeV beam energies, respectively. The counting was performed during a period of 1.8 msec after the end of each beam burst.

Fig. 6. Measurement of half life of ^{216}Ac prepared in the reaction of ^{209}Bi with 88 MeV ^{12}C ions. Experimental points show activity registered in counter during 1 msec periods starting at the beginning of the 4 msec beam burst and continuing 6 msec past the end of the beam burst. Upper curve shows sum of 9.020 and 9.105 MeV peaks. Lower curve shows sum of 8.198 and 8.283 MeV peaks.

Fig. 7. Alpha spectrum showing the four alpha groups of ^{216}Ac prepared in the reaction of ^{205}Tl with 100 MeV oxygen ions.

Fig. 8. Measurement of half life of ^{215}Ra prepared by the reaction of ^{206}Pb with 96 MeV ^{16}O ions. Experimental points show activity registered in detector during 1.8 msec periods starting at the beginning of a 4 msec beam burst and continuing in the period between beam bursts. The apparent half life is 2.1 msec but correction for the effect of transport time reduces this to 1.7 msec.

Fig. 9. Alpha energy versus neutron number for isotopes of elements above lead in the vicinity of the 126 neutron shell.

Fig. 10. Alpha decay schemes of the 125-neutron isotopes of the even elements polonium, radon, radium, and thorium. The framed spin values have been measured directly, those shown without parentheses have been derived indirectly from extensive data, and those given in parentheses are our suggestions based principally on the apparent similarities between the schemes. The predicted decay scheme of ^{217}U is indicated by broken lines.

Fig. 11. Alpha decay schemes of the 127-neutron isotopes of the odd elements bismuth, astatine, francium, and actinium. The framed spin value has been measured directly, those shown without parentheses have been derived indirectly, and those given in parentheses are our suggestions based on the shell model predictions and the apparent similarities between the schemes.

Table I. Excitation energies of the compound nucleus ^{222}Th at the Coulomb barrier and at the yield maxima of the thorium isotopes. The beam energies corresponding to the maxima were taken from Fig. 3; masses of ^{16}O and ^{206}Pb were obtained from Mattauch et al.[15] and the estimated mass of ^{222}Th from Viola et al.[16]. The last column gives the average energy release per evaporated neutron in the de-excitation process.

	E_{beam} (MeV)	$E_{excit.}$ (MeV)	n	$E_{excit.}/n$ (MeV)
Coulomb barrier	74.7	29.8	-	-
^{217}Th	102	57	5	11.4
^{216}Th	114	69	6	11.5
^{215}Th	128	83	7	11.9
^{214}Th	142	97	8	12.1
^{213}Th	157	112	9	12.4

Table II. Summary of thorium results.

Isotope	Alpha energy (MeV)	Half life (sec)	%
^{217}Th	9.250 ± 0.010	< 0.0003	
^{216}Th	7.921 ± 0.008	0.028 ± 0.002	
^{215}Th	7.522 ± 0.008	1.2 ± 0.2	40 ± 3
	7.393 ± 0.008	1.2 ± 0.2	52 ± 3
	7.331 ± 0.010	1.2 ± 0.5	8 ± 3
^{214}Th	7.680 ± 0.010	0.125 ± 0.025	
^{213}Th	7.690 ± 0.010	0.150 ± 0.025	

Table III. Summary of results on nuclides with 127 neutrons.

Isotope	This work			Previous report		
	Alpha energy (MeV)	Half life (msec)	%	Alpha energy (MeV)	Half life (msec)	Ref.
^{217}Th	9.250 ± 0.010	< 0.3	-	-	-	-
^{216m}Ac	9.105 ± 0.010	~ 0.5	97 ± 0.5	9.14	0.390	17
	8.283 ± 0.010	~ 0.5	3.0 ± 0.5	-	-	-
^{216}Ac	9.020 ± 0.010	~ 0.5	98 ± 0.5	9.14	0.390	17
	8.198 ± 0.010	~ 0.5	2.0 ± 0.5	-	-	-
^{215}Ra	8.698 ± 0.005	1.7 ± 0.2	95.7 ± 1.0	8.70	1.6	5,18
	8.168 ± 0.008	~ 1.7	1.3 ± 0.5	-	-	-
	7.880 ± 0.008	~ 1.7	3.0 ± 0.5	-	-	-
^{214m}Fr	8.549 ± 0.008	3.6 ± 0.5	51 ± 2	8.546	3.42	19
	8.477 ± 0.008	3.6 ± 0.5	49 ± 2	8.478	3.4	19
^{214}Fr	8.426 ± 0.008	5.5 ± 0.5	94.5 ± 2.0	8.430	5.3	4
	8.353 ± 0.008	5.5 ± 0.5	5.5 ± 0.5	-	5.3	-
^{213}Rn	-	-	-	8.090	19	3,18
^{212m}At	7.899 ± 0.008	-	34 ± 3	7.88	120	20
	7.837 ± 0.008	-	66 ± 3	7.82	120	20
^{212}At	7.678 ± 0.008	-	80 ± 3	7.66	305	20
	7.616 ± 0.008	-	20 ± 3	7.60	305	20

Table IV. Estimated alpha energies of a few unknown neutron-deficient isotopes of thorium, protactinium, uranium, and neptunium. The predictions are based on the systematic regularities observed in the alpha-decay of the light isotopes of the elements polonium through thorium.

Isotope	Alpha energy (MeV)	%
^{211}Th	7.82 ± 0.03	
^{212}Th	7.83 ± 0.03	
^{214}Pa	8.11 ± 0.03	
^{215}Pa	8.10 ± 0.03	
^{216}Pa	7.91 ± 0.03	~ 35
	7.79 ± 0.03	~ 35
	other groups	
^{217}Pa	8.36 ± 0.03	
^{218}Pa	9.50 ± 0.10	
^{215}U	8.39 ± 0.05	
^{216}U	8.38 ± 0.05	
^{217}U	8.21 ± 0.05	~ 40
	8.07 ± 0.05	~ 50
	8.03 ± 0.05	~ 10
^{218}U	8.63 ± 0.05	
^{218}Np	8.51 ± 0.07	
	8.41 ± 0.07	
	other groups	

NOTE: We have observed a weak short-lived alpha group at 8.340 ± 0.010 MeV in spectra obtained by bombardment of ^{203}Tl and ^{206}Pb with ^{20}Ne-ions, which on the basis of the evidence can be assigned to ^{217}Pa.

APPENDIX

A. Range Considerations

The recoil energies of the reaction products and their ranges play a role in the choice of chamber dimensions, helium pressure, and target thickness. Since we can be sure that the thorium isotopes are prepared by compound nucleus reactions involving full momentum transfer we can compute their kinetic energies and use the data obtained by others on recoil characteristics of reaction products from reactions induced by complex projectiles. If we assume isotropic emission of particles in the center of mass system of the compound nucleus we can write

$$E_R = E_B \, A_R A_B / (A_B + A_T)^2 \qquad (1)$$

where kinetic energy and mass are represented by E and A and the subscripts R, B, and T refer to the recoil nucleus, bombarding projectile, and target nucleus, respectively. According to this equation the recoil energies for the thorium isotopes of mass 213-217 produced in the $^{206}Pb + ^{16}O$ reactions are about 7 percent of the energy of the bombarding ^{16}O ions. Hence from an ^{16}O ion energy of 75 MeV corresponding to the barrier energy up to the full energy of 160 MeV the recoil energies of the thorium isotopes can vary from 5 to 11 MeV.

We can use the range data of Gilat and Alexander[22] on the range in helium of dysprosium recoil nuclei to estimate the range in helium of our products. For example dysprosium ions of 5.8 MeV have a helium range of 453 $\mu g/cm^2$ equivalent to 2.6 cm at one atmosphere pressure and room temperature, and 8 MeV dysprosium ions have a range of 560 $\mu g/cm^2$ equal to 3.3 cm. The higher atomic number throium ions should have somewhat shorter ranges. A comparison of the ranges in aluminium absorbers of 5-11 MeV recoil ions of Dy, Sm, Tb, At, and Po indi-

cates[30,31,32,33] that the range difference in light element absorbers (helium in our case) for Dy and Th ions should be about 20-30%.

In most experiments our helium stopping path was 2.0 cm and the helium pressure was 1.9 atmospheres so that we had sufficient helium to stop the thorium recoils before they reached the back window, even without considering the slowing down in the target itself.

The stopping power of lead for heavy element recoils is less than that of helium by a factor of 1.5 or 2. We have no data on recoil atoms as heavy as thorium but we can get some guidance from the measurements of Alexander and Winsberg[34] on the range of astatine recoil nuclei in gold. For example 8.1 MeV ions have a range of 1.14 mg/cm^2. Our target thickness of 2.8 mg/cm^2 is greater than this so that only those nuclei produced in the rear layer of the target emerged into the helium atmosphere.

Some of the other products such as the radium isotopes probably also have ranges corresponding to full momentum transfer. This is true whether these isotopes are made by the alpha decay of thorium parents or by α-emission in the de-excitation of the compound nucleus. The work of Kaplan[35] is pertinent to this conclusion. For nuclei lighter than radium we may expect a variety of reactions not involving full momentum transfer to account for a considerable fraction of the yield. For example the recoil ranges of several polonium, astatine, and radon isotopes produced in the bombardment ^{197}Au, ^{208}Pb or ^{209}Bi with ^{16}O have been measured by Alexander and Winsberg[34] and by Croft and Alexander[36]. In most cases the recoil ranges were far lower than expected on the basis of full momentum transfer, typically something of the order of a factor of 3 lower. We can expect in our case that the overall yield for such reaction products is reduced because only those originating in a thin back layer of the target escape into the helium.

There is one small class of products for which an opposite trend occurs. In reactions of ^{209}Bi with ^{16}O near the Coulomb barrier some of the astatine and polonium products are known[36] to be ejected with energies substantially greater than those corresponding to full momentum transfer. Such a result can occur for reactions of low impact parameter if the incoming projectile transfers a few nucleons to the target and the remainder of the projectile is then directed in the backward direction by Coulombic repulsion. It is possible that the ^{211}Po observed in our spectra at the lower bombarding energies is formed by such a mechanism from ^{206}Pb or from the 1.4 percent ^{208}Pb contamination in the target.

Still another class of products is the fission products which, judging from previous measurements,[13,14] account for three quarters of the total reaction. The fission products have energies comparable to those for the products of ^{235}U fission induced by neutrons and hence their ranges are far greater than the ranges of the other reaction products. Most of them should penetrate the target and the helium gas to imbed themselves in the walls of the chamber.

B. Chamber Design Notes

The range considerations covered in the preceeding section play a role in the design of a chamber for work with short-lived nuclides. In principle the collection process can be accelerated by reducing the chamber volume or by increasing the helium flow rate. No particular problem arises for products of heavy ion reactions with half lives longer than a few tenths of a second because a chamber sufficiently small to permit an effective collection time of substantially less than one half life is still large enough to accomodate a helium stopping path greater than the longest of the recoil ranges. However, when the half lives are of the order of milliseconds or shorter, it is difficult to avoid a compromise between speed and range requirements in a choice of chamber dimensions. The key problem is the fact that the probability that the activity will stick to the collector foil drops quickly when the helium flow rate is increased beyond a certain point. Hence the use of a larger diameter capillary to increase speed or of a pressure higher than about 2 atm to decrease the range (and hence the chamber dimensions) is not permissable.

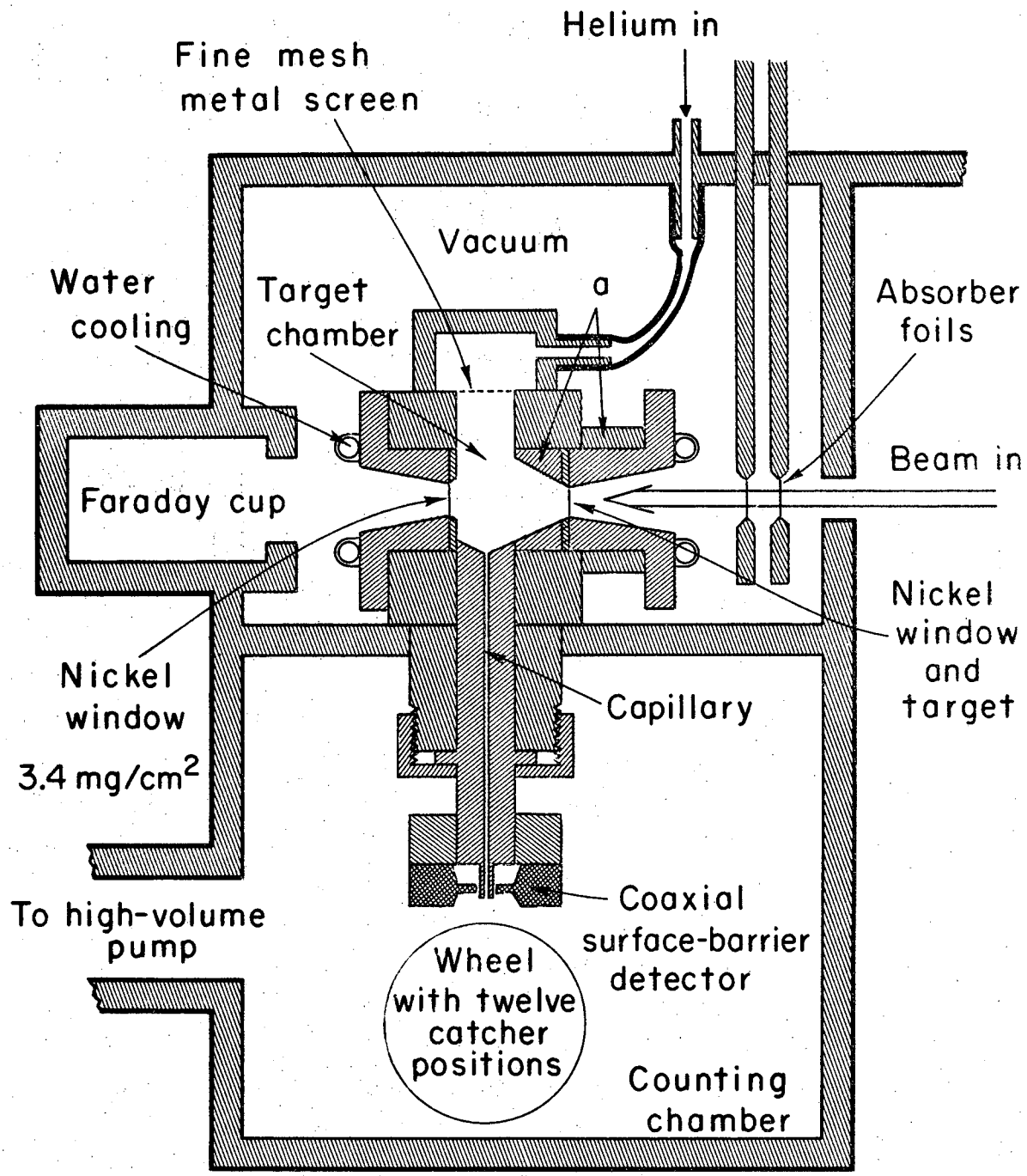
Nonetheless the range considerations suggest some ideas to reduce the problem. For example it may be possible to choose reaction conditions to make the desired product with a lower recoil energy. If it is made by a compound nucleus reaction, then according to Eq. (1) the range can be reduced by using a low bombarding energy or a light projectile. For example ^{214}Fr can be made by the reactions $^{206}Pb + ^{16}O$ and $^{206}Pb + ^{11}B$. In the first case the yield maximum occurs at a bombarding energy of 103 MeV (Fig. 4) and the recoil energy from Eq. (1) is 7.3 MeV. In the second case the peak of the excitation function would occur at about 55 MeV beam energy and the recoil energy would be about 2.7 MeV. If a spherical target chamber with a radius equal to half of the re-

coil range were used in each case and other conditions were kept unchanged, then the chamber volume in the $^{206}Pb + ^{11}B$ case would be about 1/8 of that in the $^{206}Pb + ^{16}O$ case, and accordingly, the smaller chamber would be 8 times faster than the bigger one. In reactions induced by such particles as protons and deuterons the recoil ranges are so short that a reaction chamber with a volume of the order of 0.1 cc could be used to study nuclides with half lives in the microsecond range.

In some cases the same product can be made in one reaction by a mechanism involving full momentum transfer or by another reaction in which only partial momentum transfer occurs and hence be produced with much less kinetic energy.

Another option is to use a thick target so that the recoiling nuclei are slowed down in the target itself and spend only a fraction of their range in the gas. The chamber volume can be decreased to take advantage of this but this, of course, involves some loss of yield because those products made in the back target layer are lost to the back wall of the chamber.

The present design of the target chamber has some advantages that may be valuable in other applications. The capillary can be made at least 20 cm long (probably much longer) without essential loss of speed or collection efficiency. This provides enough room for heavy shielding around the detector. Because of the small size of the target chamber most fission products are buried in the walls of the chamber which reduces the build-up of beta and gamma activity in the counting chamber. These properties were at considerable value in the present experiments, and they would be much more important in the application of the helium jet transport system to the collection of recoil nuclei for on-line study of gamma activity.



XBL683-2006

Fig. 1.

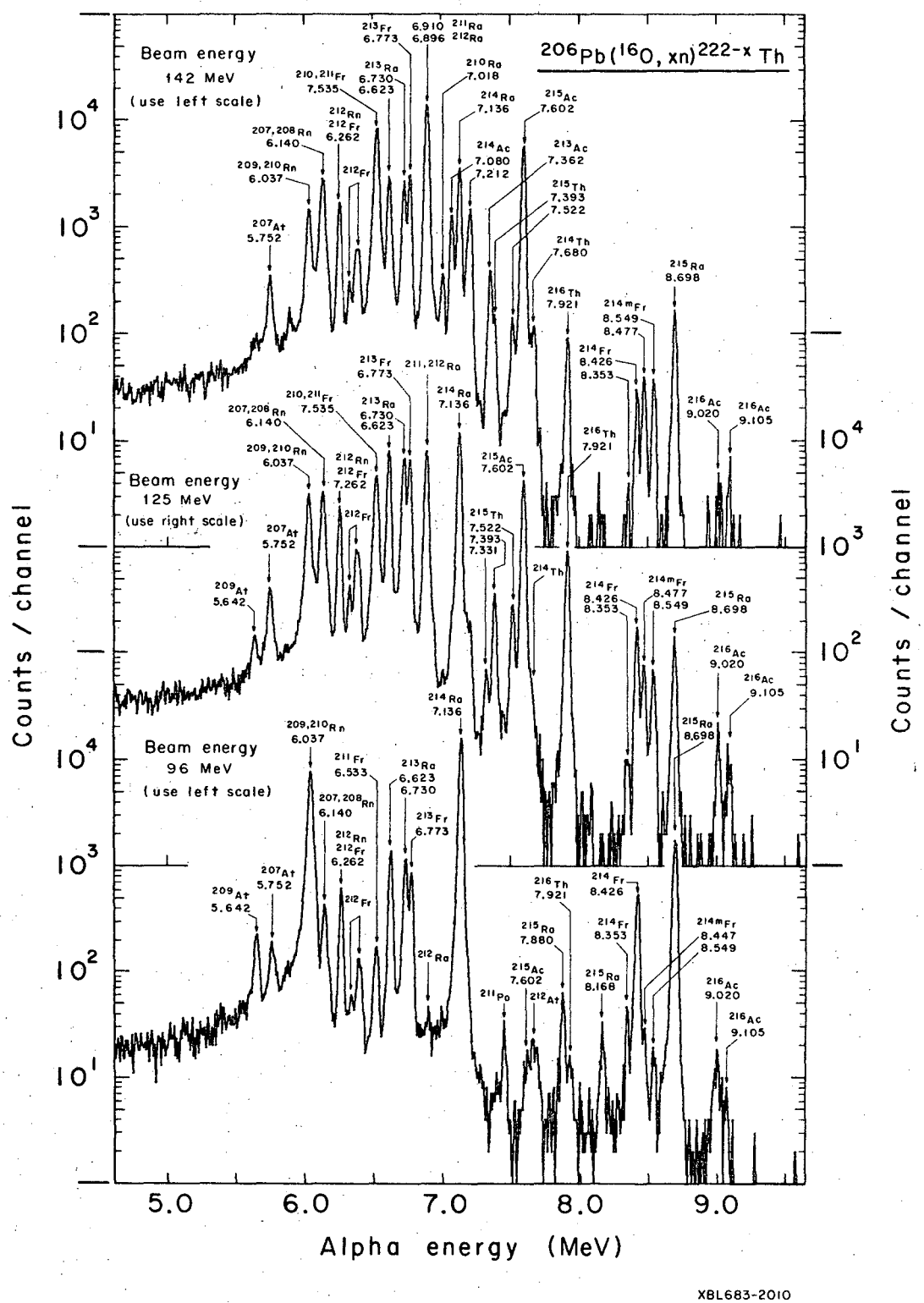
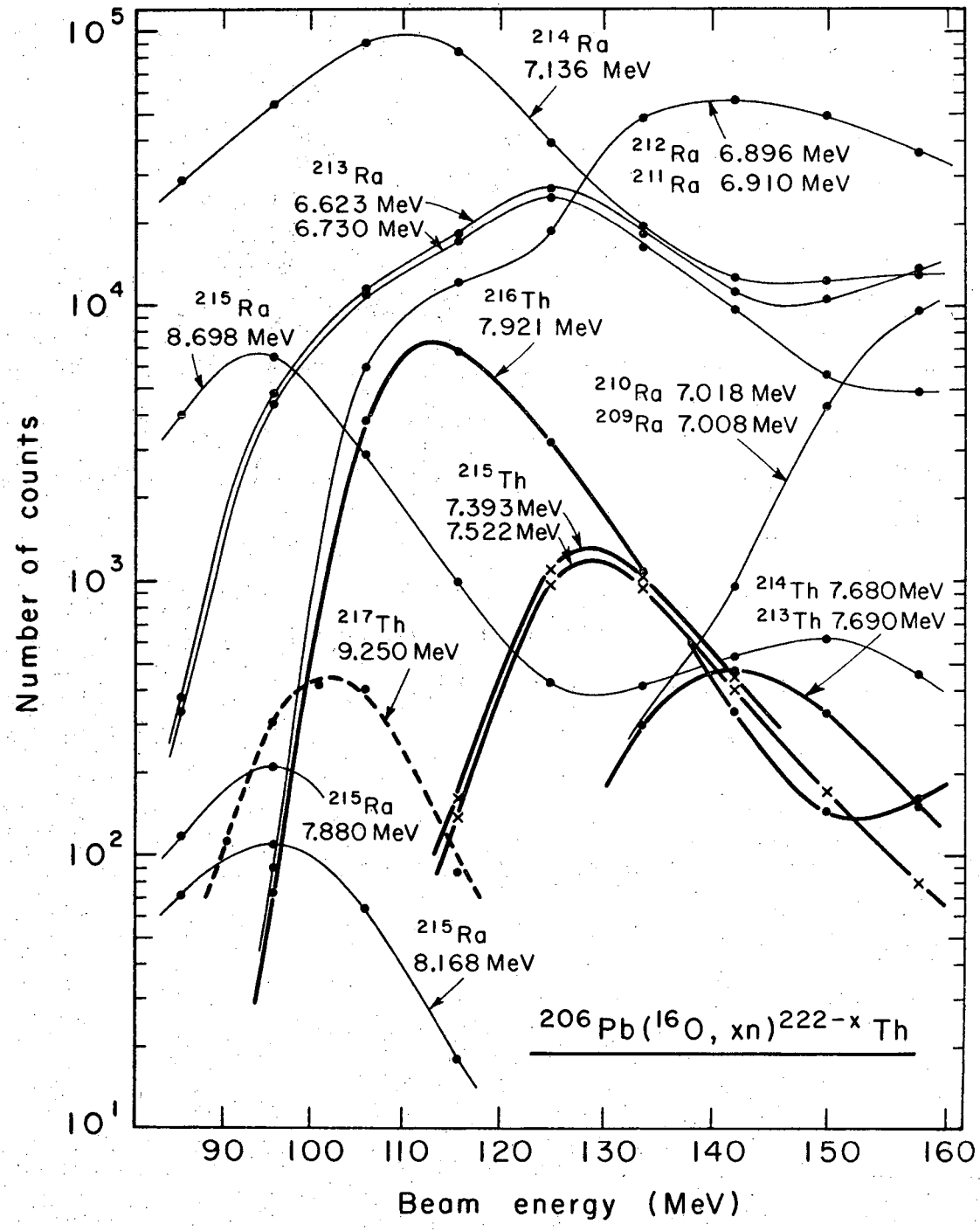
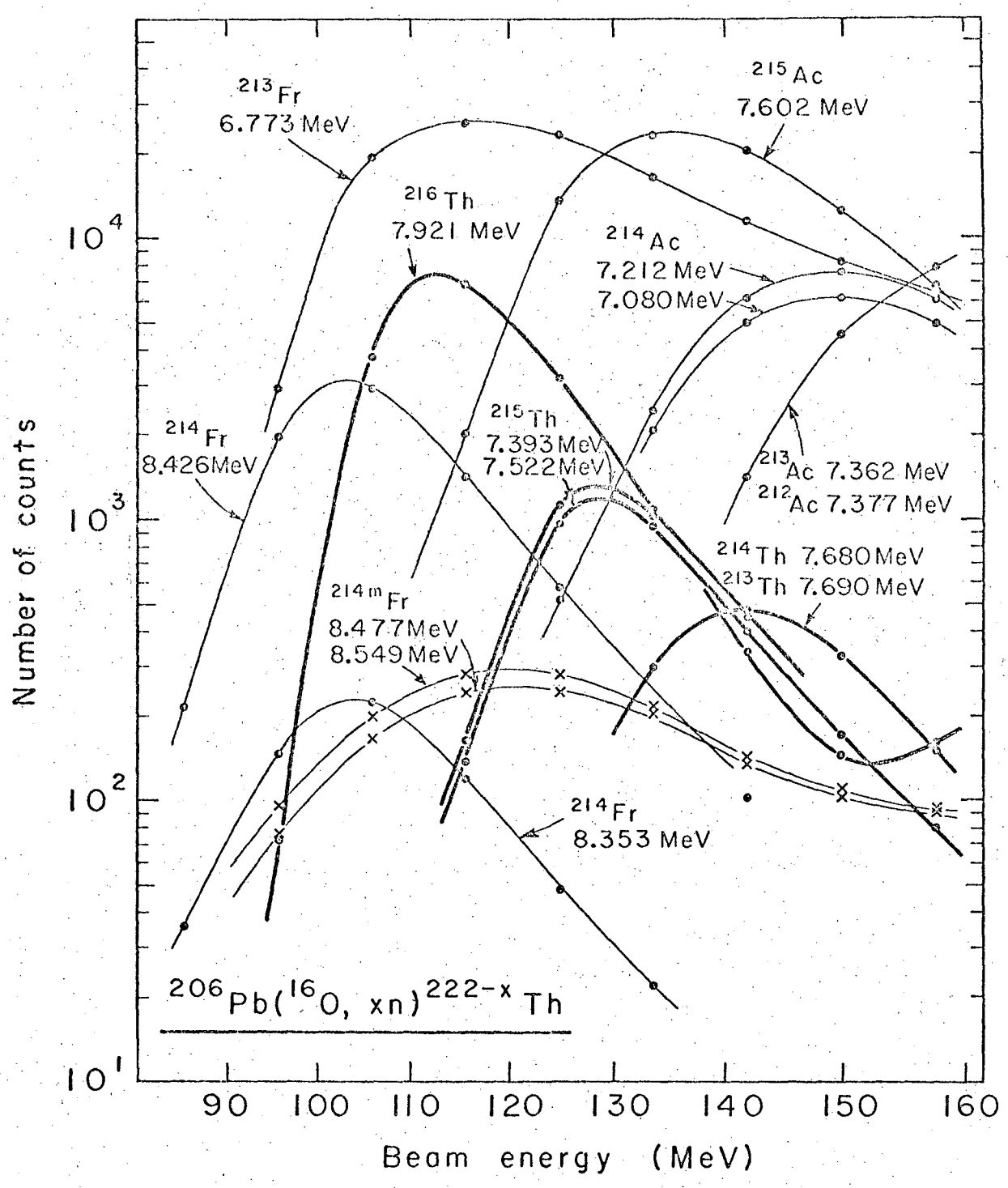

206Pb(16O, xn)222-x Th
Beam energy
142 MeV
(use left scale)
Beam energy
125 MeV
(use right scale)
Beam energy
96 MeV
(use left scale)
Counts / channel
Counts / channel
Alpha energy (MeV)
5.0
6.0
7.0
8.0
9.0
XBL683-2010

Fig. 2.



XBL683-2009

Fig. 3.



XBL683-2008

Fig. 4.

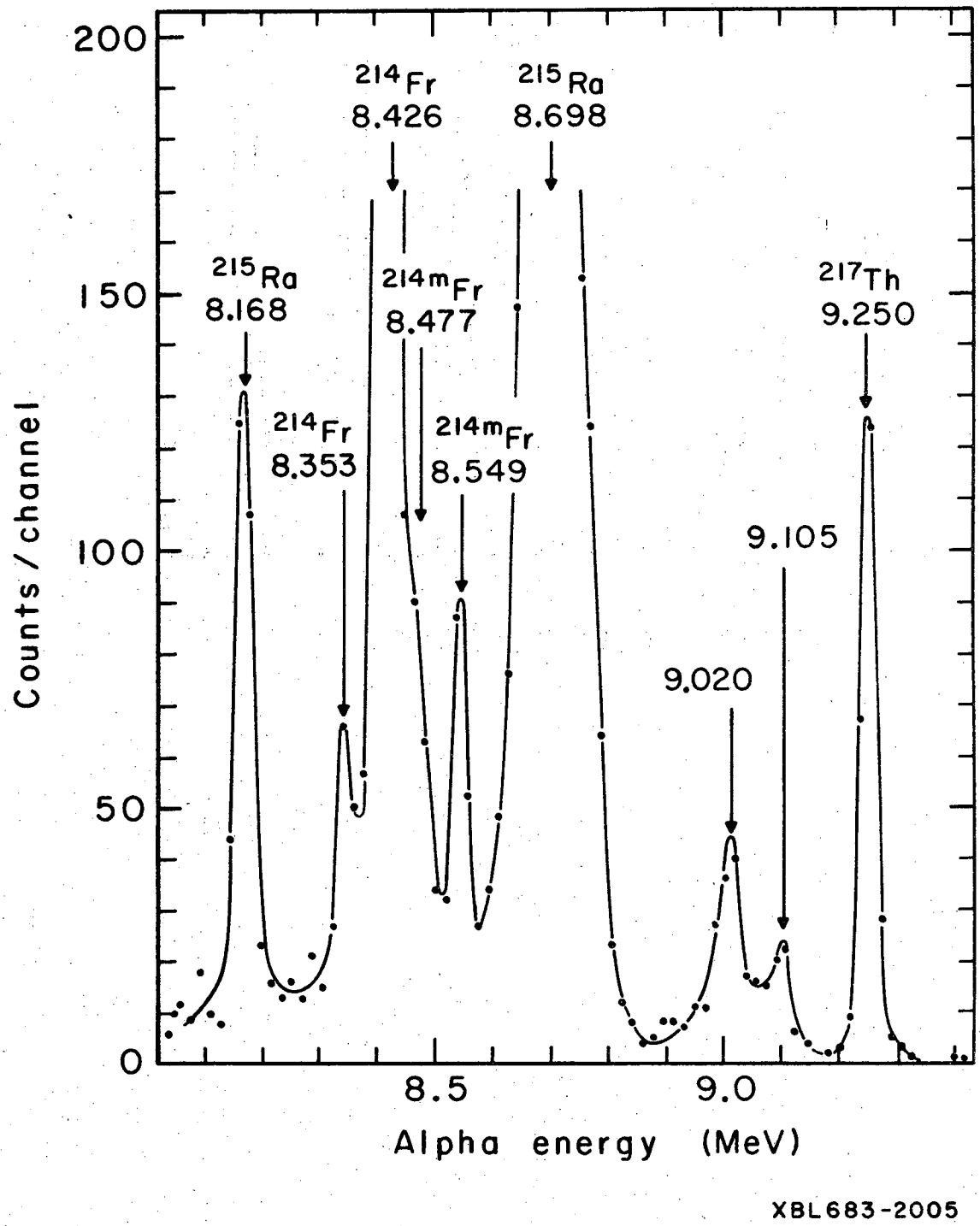

200
150
100
50
0
Counts / channel
8.5
9.0
Alpha energy (MeV)
215Ra
8.168
214Fr
8.353
214Fr
8.426
214mFr
8.477
214mFr
8.549
215Ra
8.698
9.020
9.105
217Th
9.250
XBL683-2005

Fig. 5.

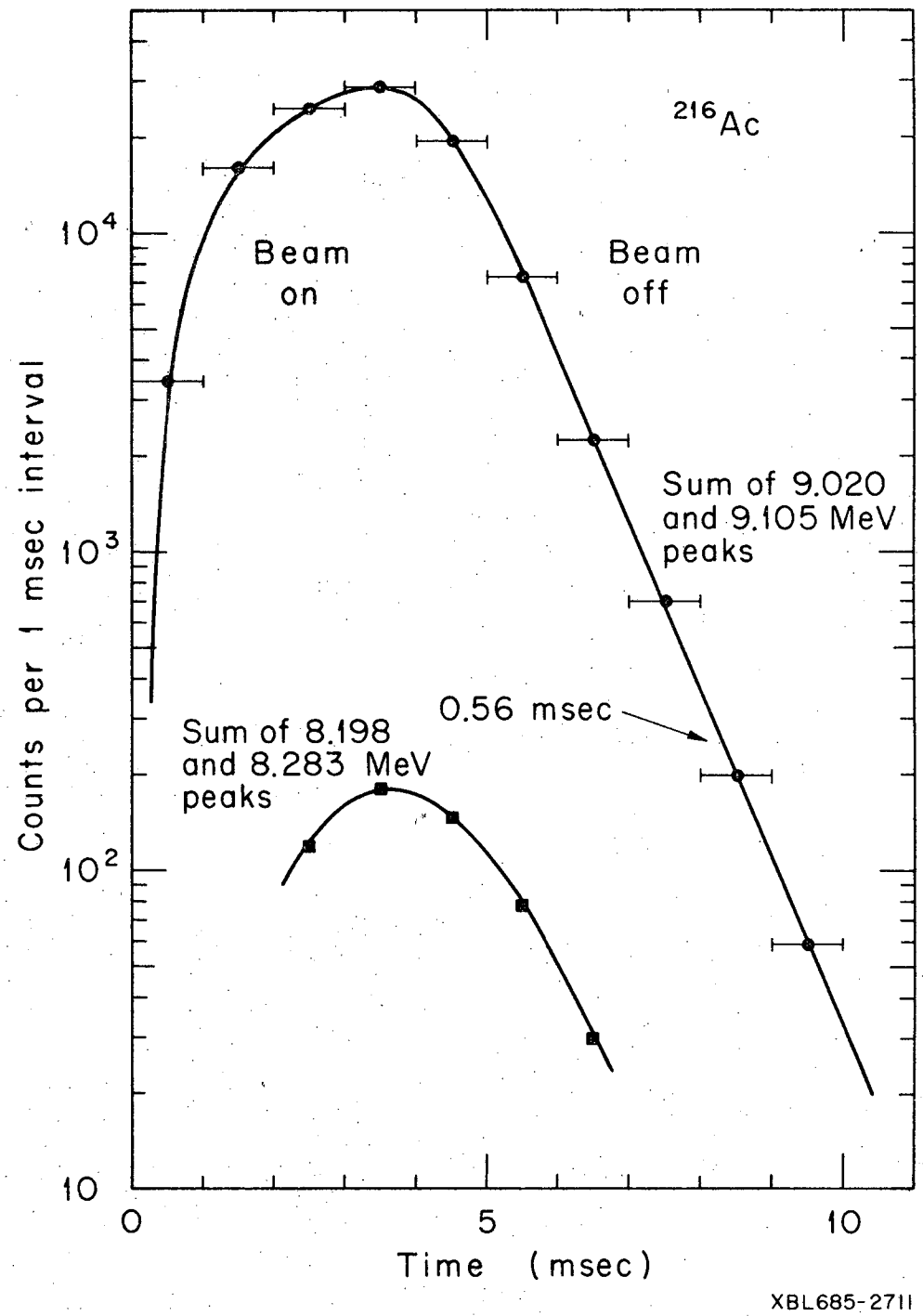

^{216}Ac
Beam on
Beam off
Sum of 9.020 and 9.105 MeV peaks
0.56 msec
Sum of 8.198 and 8.283 MeV peaks
Counts per 1 msec interval
10^4
10^3
10^2
10
0
5
10
Time (msec)
XBL685-2711

Fig. 6.

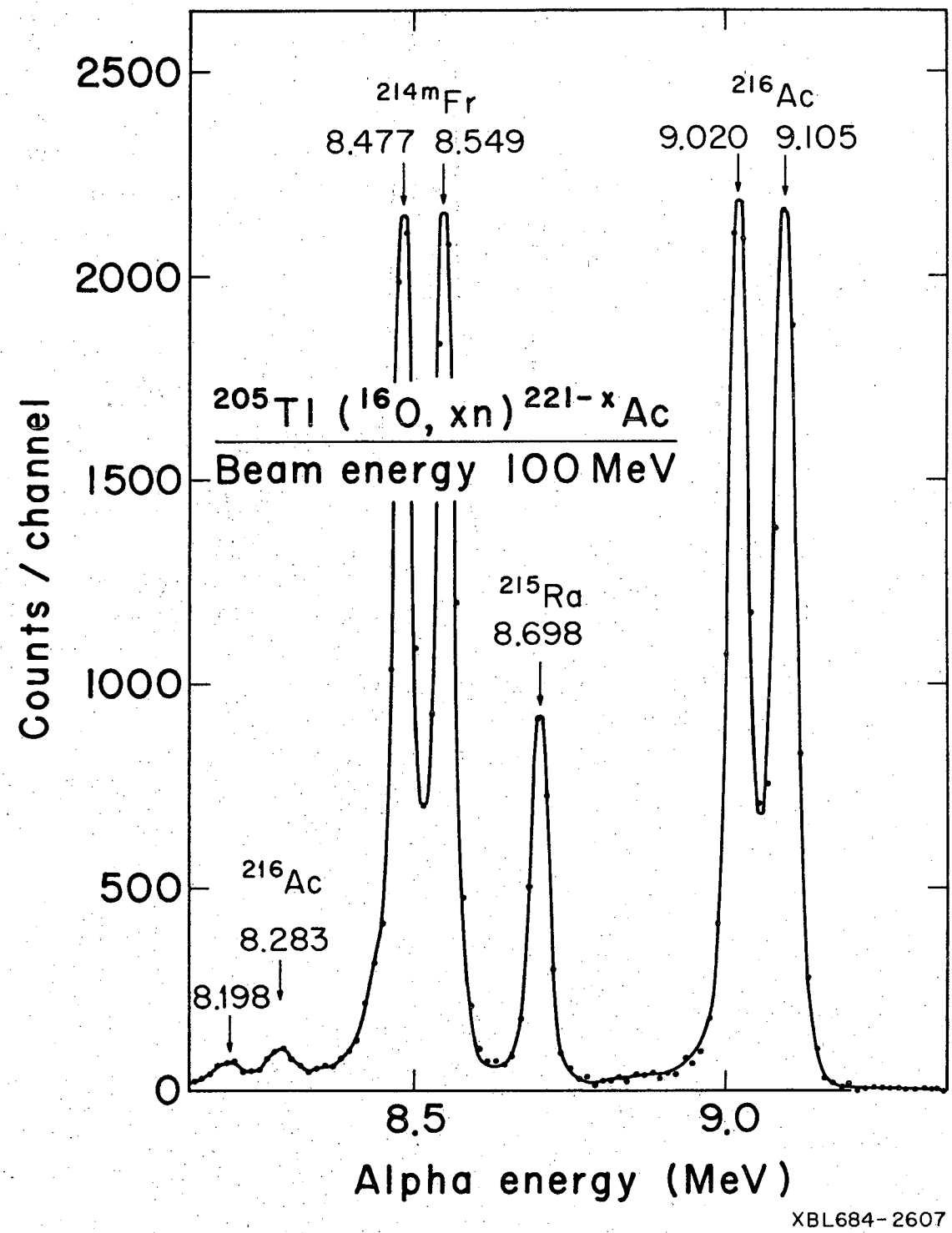

2500
2000
1500
1000
500
0
Counts / channel
214mFr
8.477 8.549
216Ac
9.020 9.105
205Tl (16O, xn) 221-xAc
Beam energy 100 MeV
215Ra
8.698
216Ac
8.283
8.198
8.5
9.0
Alpha energy (MeV)
XBL684-2607

Fig. 7.

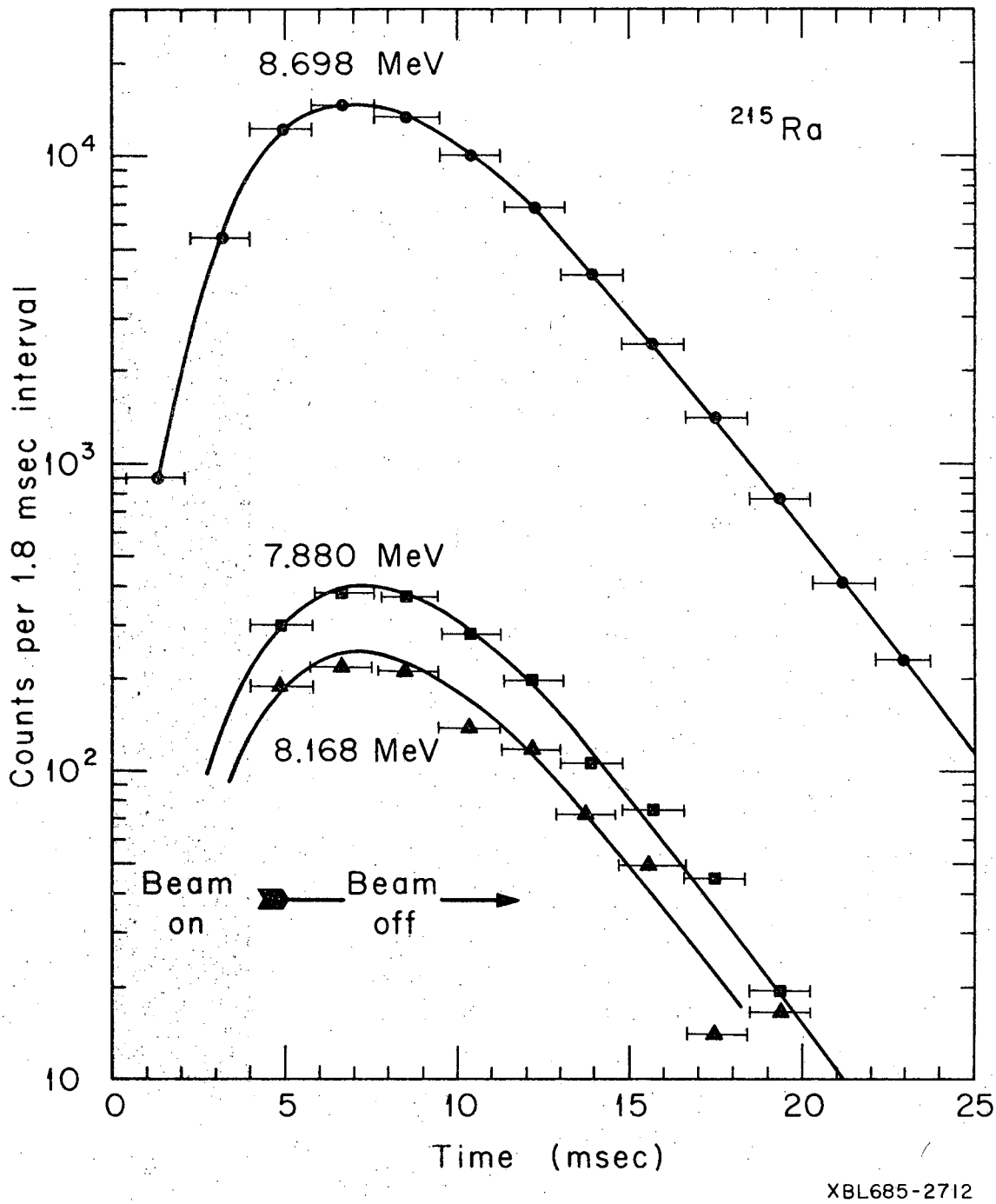

8.698 MeV
215 Ra
7.880 MeV
8.168 MeV
Beam on
Beam off
Counts per 1.8 msec interval
Time (msec)
10
10²
10³
10⁴
0
5
10
15
20
25
XBL685-2712

Fig. 8.

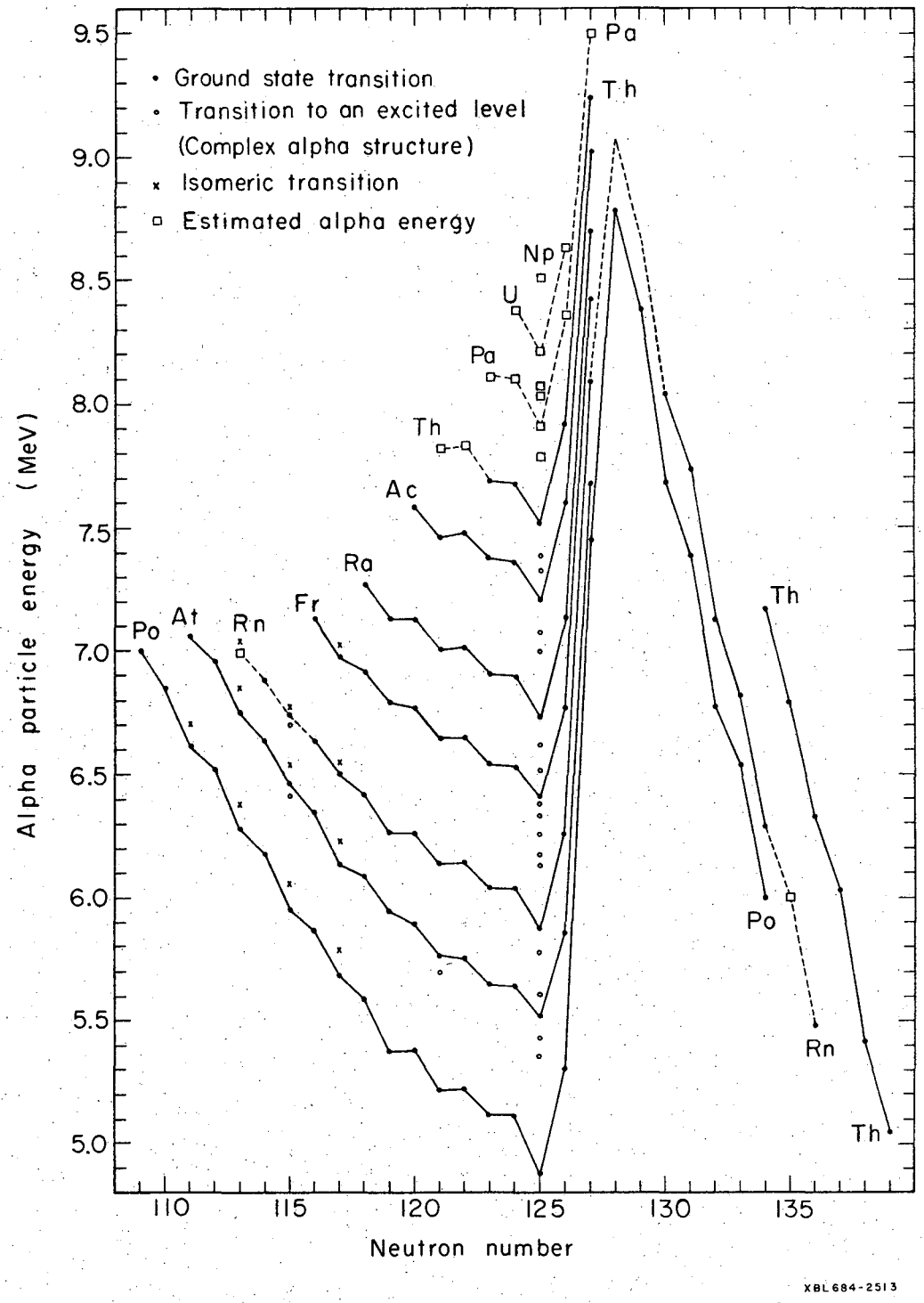

• Ground state transition
• Transition to an excited level
(Complex alpha structure)
× Isomeric transition
□ Estimated alpha energy
Alpha particle energy (MeV)
Neutron number
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
110
115
120
125
130
135
Po
At
Rn
Fr
Ra
Ac
Th
Pa
U
Np
Pa
Th
Th
Po
Rn
Th
XBL684-2513

Fig. 9.

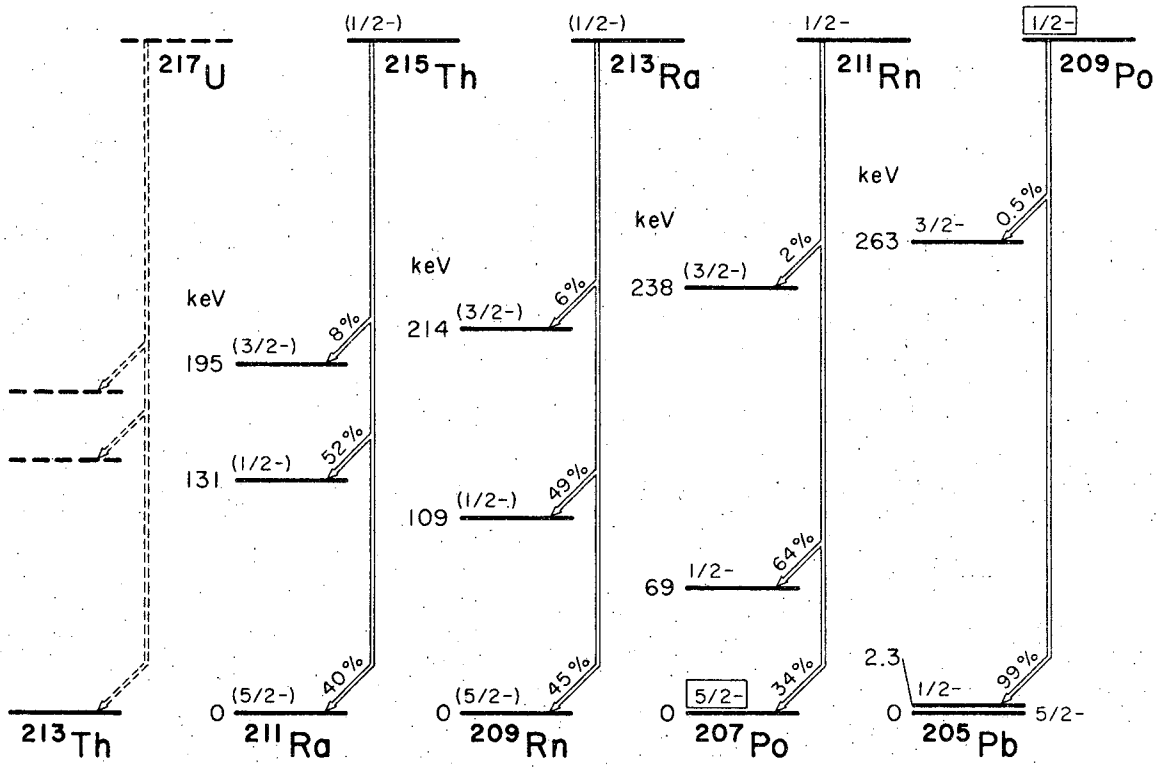

217U
215Th
213Ra
211Rn
209Po
(1/2-)
(1/2-)
1/2-
1/2-
keV
keV
keV
keV
195 (3/2-) 8%
131 (1/2-) 52%
0 (5/2-) 40%
214 (3/2-) 6%
109 (1/2-) 49%
0 (5/2-) 45%
238 (3/2-) 2%
69 1/2- 64%
0 5/2- 34%
263 3/2- 0.5%
2.3 1/2- 99%
0 5/2-
213Th
211Ra
209Rn
207Po
205Pb
XBL684-2608

Fig. 10.

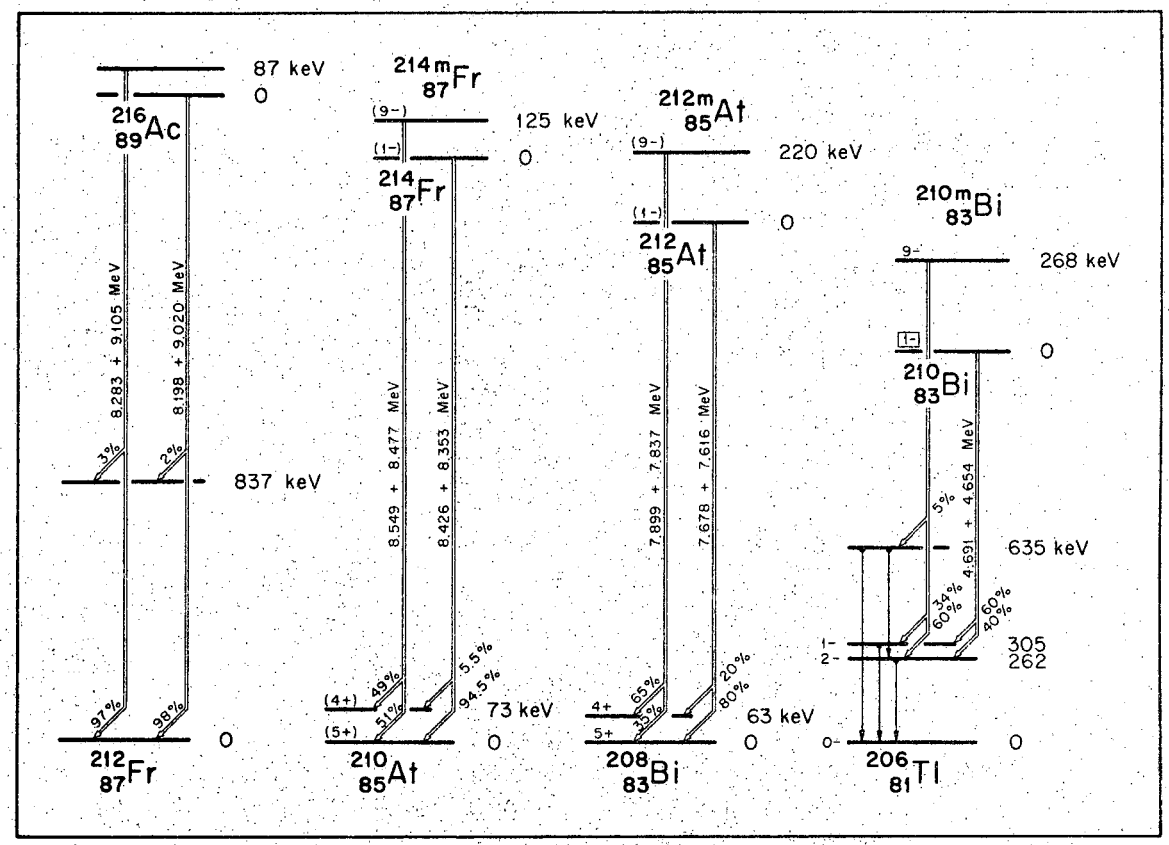

87 keV
0
216 89 Ac
8.283 + 9.105 MeV
8.198 + 9.020 MeV
3%
2%
837 keV
97%
98%
0
212 87 Fr
214m 87 Fr
(9-)
125 keV
(1-)
0
214 87 Fr
8.549 + 8.477 MeV
8.426 + 8.353 MeV
(4+)
49%
51%
5.5%
94.5%
73 keV
(5+)
0
210 85 At
212m 85 At
(9-)
220 keV
(1-)
0
212 85 At
7.899 + 7.837 MeV
7.678 + 7.616 MeV
4+
65%
35%
20%
80%
63 keV
5+
0
208 83 Bi
210m 83 Bi
9-
268 keV
1-
0
210 83 Bi
4.691 + 4.654 MeV
5%
635 keV
34%
60%
60%
40%
1-
2-
305
262
0-
0
206 81 Tl
XBL685-2713

Fig. 11.

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