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ABSTRACT

An isomeric state of ^{222}Ac with a half-life of 66±3 sec has been discovered in bombardments of lead with ^{18}O ions. The isomer decays predominantly by alpha-particle emission and has a complex spectrum with major alpha-particle groups at 6.46 (2%), 6.71 (8%), 6.75 (15%), 6.81 (27%), 6.84 (10%), 6.89 (15%), 6.97 (8%), and 7.00 MeV (15%). Evidence for the decay of the isomeric state both by isomeric transition (< 10%) and by electron capture (~1%) is presented.

I. INTRODUCTION

The similarity of both alpha-particle energies and apparent half-lives of nuclides in the lead region with those in the heavy actinide or trans-actinide region is well known. Because reaction cross sections are typically higher by several orders of magnitude for reactions induced in lead by heavy ions in comparison with those induced in heavy actinide targets, the presence of even minute quantities of lead gives rise to activities that interfere with the interpretation of alpha-particle spectra from heavier elements. Trace quantities of lead are almost inevitably present in heavy-element targets. Thus a detailed knowledge of the decay properties of alpha-active isotopes produced in heavy-ion bombardments of lead is essential for the study of the heaviest elements. Results of an extensive survey of alpha-active isotopes between lead and uranium have been reported by Valli and Hyde et al. in a series of articles.[1-8] The

66-sec alpha-emitting isomeric state, ^{222}Ac, reported here was discovered as a byproduct of a study of ^{261}Rf.[9]

Earlier studies of ^{222}Ac[10-12] have been carried out by producing the 1.8-min ^{226}Pa in bombardments of thorium with high-energy deuterons and following its alpha-decay through the chain ^{226}Pa - ^{222}Ac - ^{218}Fr - ^{214}At - ^{210}Bi. By use of recoil collection method Meinke et al.[12] were able to separate ^{222}Ac from its parent, ^{226}Pa, and obtained a value of 5.5 ± 0.5 sec for its half life.

With similar techniques but using Au-Si surface-barrier detectors, McCoy[13] associated two alpha-particle groups, one at 6.998 MeV (94%) and the other at 6.952 MeV (6%), with the decay of the 5.5-sec ^{222}Ac. We have assigned the 66-sec alpha-activity with a complex alpha-particle spectrum to ^{222m}Ac by showing that the alpha particles are promptly followed by 7.87-MeV and 8.81-MeV alpha particles of short-lived ^{218}Fr and ^{214}At.

II. EXPERIMENTAL

An approximately 100 $\mu g/cm^2$ lead target of natural composition and plated on a Be foil was used in the experiments. The target was bombarded by ^{16}O and ^{18}O ions accelerated by the Berkeley heavy-ion linear accelerator. The energy of the 10.4 MeV/nucleon particles was adjusted by a stack of Be metal-foil degraders and the energy of the particles scattered from the target at an angle of 30° was measured by a solid-state detector.

The experimental techniques and apparatus used have been described in some detail in References 14 and 15. A gas jet was used to collect and transfer the reaction recoil atoms from the vicinity of the target onto a vertically mounted wheel. The wheel was periodically rotated 45° to place the accumulated recoil products next to a series of peripherally mounted Si-Au surface-barrier detectors to measure their alpha-particle spectra. There were seven detector stations along the periphery of the wheel with 45° separation between adjacent stations. At each detector station there were four detectors: two movable ones which alternately faced the wheel or were shuttled to face one of the two stationary detectors in off-wheel position. This arrangement made it possible to observe the decay of daughter alpha activities without interference from the mother activity in a series of genetically-related alpha activities.

Alpha-decay events recorded by the detectors were amplified by modular units developed in our laboratory, processed by a PDP-9 computer and stored on IBM tape. Each wheel-cycle and shuttle period was divided into four time subgroups of equal length. Besides the pulse height, a detector identification signal, wheel cycle and shuttle time subgroups, the wheel position, and the time of occurrence of each event with an accuracy of 50 milliseconds were recorded.

III. RESULTS

The 66-sec alpha activity has been observed in bombardments of Pb targets with both ^{16}O and ^{18}O ions and a Bi target with ^{15}N and ^{18}O ions. Recently the activity has also been observed by Eppley et al.[16] when bombarding Th and U targets by 5-GeV protons from the Bevatron. The series of alpha-particle spectra displayed in Fig. 1 resulted from bombardment of the Pb target with 97-MeV ^{18}O ions. Most of the activities belong to members of alpha-decay chains originating from Ac and Ra isotopes. However, it is readily seen that the 7.87-MeV and 8.81-MeV peaks assigned to ^{218}Fr and ^{214}At, respectively, decay with a half-life of about a minute instead of the 5.5-sec half-life expected on the basis of the known half-life of ^{222}Ac. This suggests that there is an isomeric state in either ^{218}Fr or its precursor, ^{222}Ac, that feeds the ground state of ^{218}Fr. The decay of the 7.87-MeV alpha-particle group is shown in Fig. 2. A half-life of 66 ± 3 sec is derived for the activity by a least squares analysis.

A search for a possible precursor to the 7.87-MeV alpha-particle activity naturally leads to a closer study of the complex cluster of peaks in the 6.5-7.0 MeV energy region, where one would expect to find an Ac alpha emitter with a half-life of a minute. As shown in Fig. 1 the 2.2 min ^{211}Bi, 2.7-min ^{213}Ra and its EC daughter 35-sec ^{213}Fr, 30-sec ^{221}Ra, 2.2-min ^{223}Ac and 20-h ^{255}Fm also contribute to the alpha-particle spectrum in this energy region. The ^{255}Fm was produced in unrelated bombardments preceding the one resulting in the spectrum shown in Fig. 1. To single out the contribution of the possible ^{222}Ac alpha activity from a host of interfering activities, we looked for time-correlated alpha-decay events where an alpha-particle was followed within 50 milliseconds by an alpha-particle in either the 7.87-MeV or 8.81-MeV peak. The alpha-particle spectra resulting from the search are plotted in Fig. 3. Accidental correlations due to a high counting rate have been accounted for by subtracting from the above correlations the ones where a 7.87-MeV or 8.81-MeV alpha-particle preceded an alpha particle in the 6.4-7.1 MeV range. On the left hand side in Fig. 3 the correlations detected at the first station, or within the first 60 seconds after the irradiation, are plotted in four 15-sec time subgroups. To the right these same counts are summed up in the first spectrum and the three spectra underneath arise from correlated events found at the three succeeding stations. The fast decaying component in the prominent 7.01-MeV peak is associated with the previously known 5.5-sec alpha activity of ^{222}Ac. The rest of the complex alpha-particle spectrum is assigned to the new 66-sec isomer of ^{222}Ac. The energies, intensities and hindrance factors for alpha-particle groups contributing to the composite spectrum are presented in Table I. The shape of the 7.87-MeV ^{218}Fr peak was used as a standard when resolving the spectrum into its components. The hindrance factors were calculated using the spin-independent ($\ell = 0$) equations of Preston.[17] A value of 9.29 fm was chosen for the radius parameter R.

The alpha-particle spectra recorded by the movable detectors when in the off-wheel position combined with the spectra recorded by the stationary detectors facing them provided information as to which of the two isomeric levels of ^{222}Ac lies higher. The number of counts observed in the 7.01-MeV peak at the seven detector stations were 60, 30, 17, 8, 6, 2 and 2. The distribution of these same counts by the quadrants of the 60-sec shuttle period was 111, 11, 2 and 1. On the basis of the two distributions we obtain a half-life of 5 ± 1 seconds for the 7.01-MeV activity and a half-life of 65 ± 10 seconds for its precursor. The interpretation of these results is that the 66-sec metastable state also decays by gamma-ray emission and feeds the 5.5-sec state, which probably is the ground state. As a result of the recoil energy imparted to the residual atoms by the gamma rays or associated conversion electrons, part of the atoms are transferred onto the detectors facing the wheel and their subsequent decay by alpha-particle emission is observed in the off-wheel position.

The ratio of counts attributed to the 66-sec ^{222m}Ac in the spectra recorded in the on-wheel position to the counts in the 7.01-MeV peak recorded in the off-wheel position was approximately 400:1. On the other hand a calculation taking into account the effect of detector geometry and timing conditions gives a value of 12 to the ratio of isomeric transitions to observed alpha decays of the ground state. On the basis of the calculated and observed ratios, we obtain a lower limit of 3 ± 1% for the decay of the 66-sec metastable state by isomeric transition. However, because the transfer efficiency of the recoil atoms onto the detectors facing the wheel after the isomeric transition is not known, but certainly is less than the 100% assumed in the calculation, the actual branching for the decay by isomeric transition is certainly higher than this 3% lower limit. An upper limit of 10% can be set by comparing the number of counts in the 7.01-MeV peaks in Figs. 1 and 3.

In addition to its decay by alpha-particle and gamma-ray emission, the 66-sec isomer also seems to decay by EC to the levels of 38-sec ^{222}Ra. The evidence for such branching by EC is again obtained from the spectra recorded in the off-wheel position. The number of counts observed in either the 7.13-MeV peak of ^{218}Ra or the 7.68-MeV peak of ^{214}Po at the seven stations were 52, 21, 16, 10, 4, 3, and 1. By quadrants of the 60-sec shuttle period, these counts were distributed as 29, 38, 24, and 20. By the former distribution, the half-life of the precursor is 64 ± 10 sec, and by the latter, the half-life of the daughter is 50 ± 15 sec. The precursor half-life agrees well with that of the 66-sec ^{222m}Ac and the daughter half-life is in accordance with the 38-sec ^{222}Ra. The 6.55-MeV alpha-particle group of ^{222}Ra was masked in the daughter spectra by the prominent 6.62-MeV group of ^{211}Bi and only the descendants of ^{222}Ra were easily distinguished.

Approximate values of 0.7% and 2% of the lower and upper limits for branching by EC were obtained by comparing the observed number of counts in the 7.13-MeV peak both in on and off-wheel positions with the observed number of counts attributable to ^{222m}Ac.

Because the event times were only measured with an accuracy of 50 milliseconds, an accurate half-life measurement for the daughter, ^{218}Fr, was not possible. However, a value of 0.7 ± 0.6 msec was deduced on the basis of the observed ratio, 0.019 ± 0.017, between those pairs of events where ^{222}Ac and

^{218}Fr alpha decays followed each other within the same 50 msec interval and those where the two events were recorded in two consecutive 50 msec intervals.

There seems to be a discrepancy of some 10-40 keV between the alpha-particle-energy-values reported in this work and those given by McCoy.[13] Thus we have found the energies 7.01, 7.87, and 8.81 MeV for the 5.5-sec ^{222}Ac, 0.7-msec ^{218}Fr and 2-μsec ^{214}At, respectively, while the corresponding energies of McCoy are 6.998, 7.845, and 8.777 MeV. Our values are based on the analysis of the spectra by SAMPO computer program[18] using the 7.136-MeV ^{214}Ra and 8.675-MeV ^{215}Rn as internal energy standards.[4,7]

The relative cross sections for some of the more prominent reaction products in bombardments of the lead target by ^{18}O ions are shown in Fig. 4. Because of the natural isotopic composition of the lead target, several reactions may substantially contribute to the production of an isotope. The predominant component in each case seems to come from the most abundant isotope, ^{208}Pb, in the target, i.e., from the reactions ^{208}Pb(^{18}O,3n)^{223}Th, ^{208}Pb(^{18}O,p2n)^{223}Ac, ^{208}Pb(^{18}O,p3n)^{222m}Ac, ^{208}Pb(^{18}O,αn)^{221}Ra, and ^{208}Pb(^{18}O, ^{14}C)^{212m}Po.

The cross section ratio of the 66-sec state to the 5.5-sec state of ^{222}Ac was measured to be 3.9 ± 0.5 for the 97-MeV ^{18}O ions. The isomeric ratio was found to be relatively constant near the peak of the cross section curve. It was not studied more closely at lower bombarding energies because of the low yields.

IV. DISCUSSION

Of the earlier studied isotopes of actinium only ^{216}Ac has been found to have an alpha-emitting metastable state.[6,19] The isomerism is caused by the close proximity of 9^- and 1^- states, which are members of a multiplet arising from the $(2g\ 9/2)_n\ (1h\ 9/2)_p$ single-particle configuration. No evidence of the existence of a similar high-spin isomer in ^{218}Ac was seen by Torgerson and Macfarlane[19] in their recent study of the odd proton, N = 127 isotones. Borggreen et al. also did not find any direct indication of the existence of isomers in ^{218}Ac or ^{220}Ac in connection with their studies of protactinium isotopes with mass numbers 222-225.[8] It thus seems that this particular type of isomerism is confined to N = 127 isotones.

A possible explanation for the presence of a long-lived alpha-emitting isomer in ^{222}Ac is derived from the Nilsson model level scheme. Borggreen and his coworkers[8] observed that the alpha-particle energies of N = 133 isotones were systematically lower than expected on the basis of the regular behavior in neighboring isotones. They suggested that this anomalous trend might be caused by the occurrence of some individual particle level of high spin. Such a level could well be the $13/2^+[606\uparrow]$ level predicted to be favored by the 131st and 132nd neutron in the recent level scheme calculation by S. G. Nilsson et al.[20] However, for the scheme to be applicable one has to postulate the onset of stable ground-state deformation for the nuclei in the region of A near 225 with deformation parameters $\epsilon \sim 0.2$ and $\epsilon_4 = -0.04$. The coupling of $13/2^+[606\uparrow]$-neutron with the odd proton, which according to the calculations should occupy $3/2^+[651\uparrow]$ level, would provide a high-spin multiplet that could be the source of the 66-sec metastable state. An application of the Gallagher-Moszkowski coupling rule gives a spin of 5^+ for the lowest state of the spin multiplet.

On the basis of the measured isomeric ratio it seems likely that the 66-sec level has a higher spin than the 5.5-sec level. Although the cross sections for the production of the two isomeric states are relatively small, a more detailed study of their properties by use of alpha-gamma and alpha-conversion electron coincidence measurements is feasible and could provide valuable information on the applicability of the Nilsson model level scheme in this transition region. A careful search might also result in discovery of similar isomerism in nearby odd-odd isotopes.

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FIGURE CAPTIONS

1. A series of alpha-particle spectra produced by bombardments of Pb with ^{18}O ions. The individual spectra show the total of counts recorded at each of the seven stations by the two movable detectors when facing the wheel. The sum of the seven spectra is plotted topmost. The wheel-stepping interval, the integrated beam reading and the bombardment energy are indicated in the figure.

2. Decay of the 7.87-MeV alpha-particle group associated with 0.7-msec ^{218}Fr. The ^{218}Fr is replenished by the alpha-decay of ^{222}Ac, the half-life of which is thus given by the decay curve. The first point is above the curve because of a contribution from the 5.5-sec ^{222}Ac.

3. The spectrum of alpha-decay events which were followed within 50 milliseconds by another event recorded at either the 7.87-MeV or 8.81-MeV peak. The events observed at the first detector station are displayed in four consecutive 15-sec subgroups in the left hand side of the figure. To the right the correlated events from the first four stations are plotted.

4. Excitation curves for some of the activities produced in the bombardments of Pb by ^{18}O ions.

TABLE I. Alpha-particle groups associated with the decay of the 66-sec ^{222m}Ac.

Alpha-Particle energy [MeV]	Intensity %	Hindrance factor
6.46 ± 0.02	2 ± 1	13
6.71 ± 0.02	8 ± 4	33
6.75 ± 0.02	15 ± 5	25
6.81 ± 0.01	27 ± 10	24
6.84 ± 0.02	10 ± 5	85
6.89 ± 0.02	15 ± 5	85
6.97 ± 0.02	8 ± 3	310
7.00 ± 0.02	15 ± 5	220

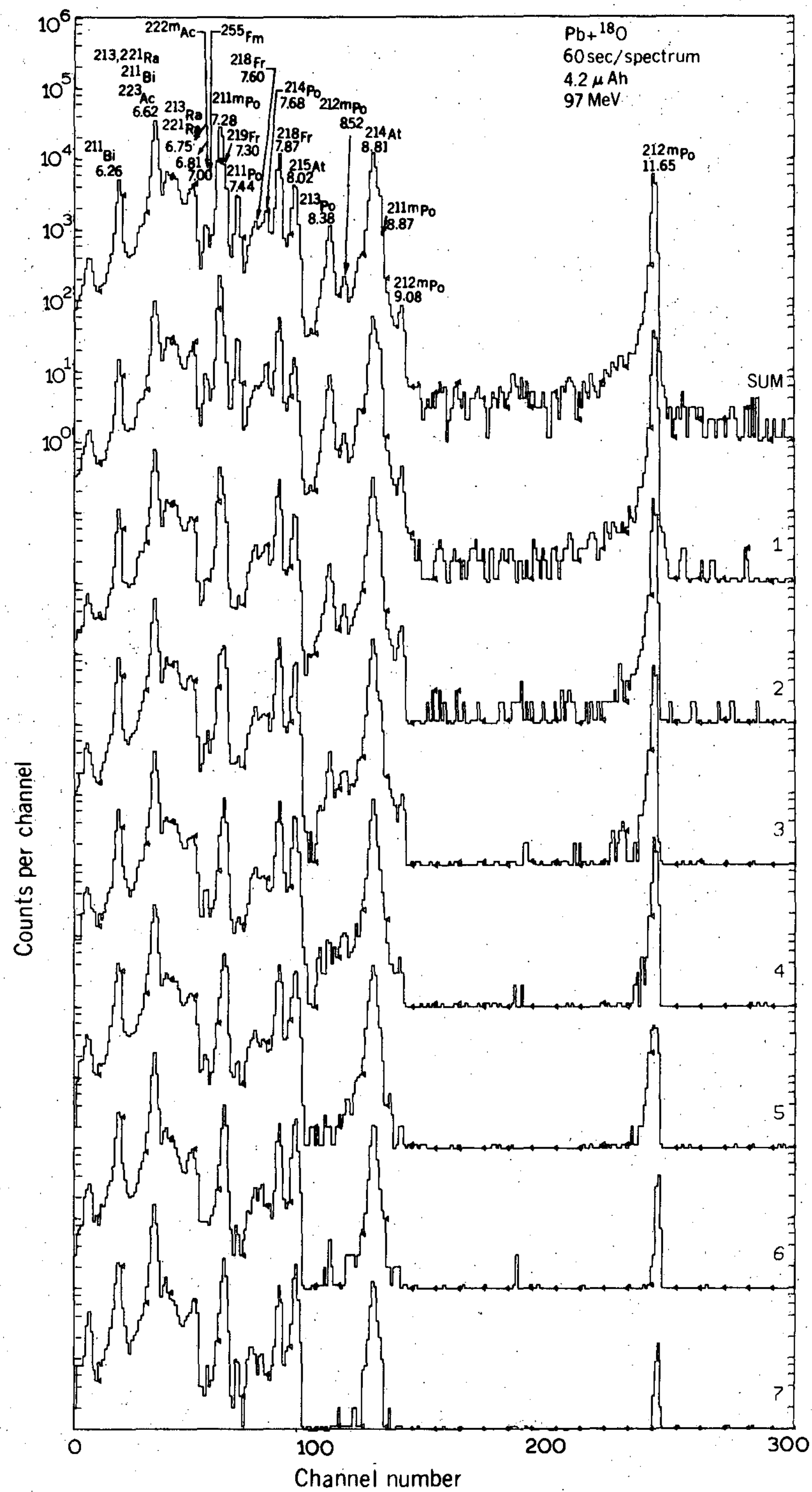

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Fig. 1

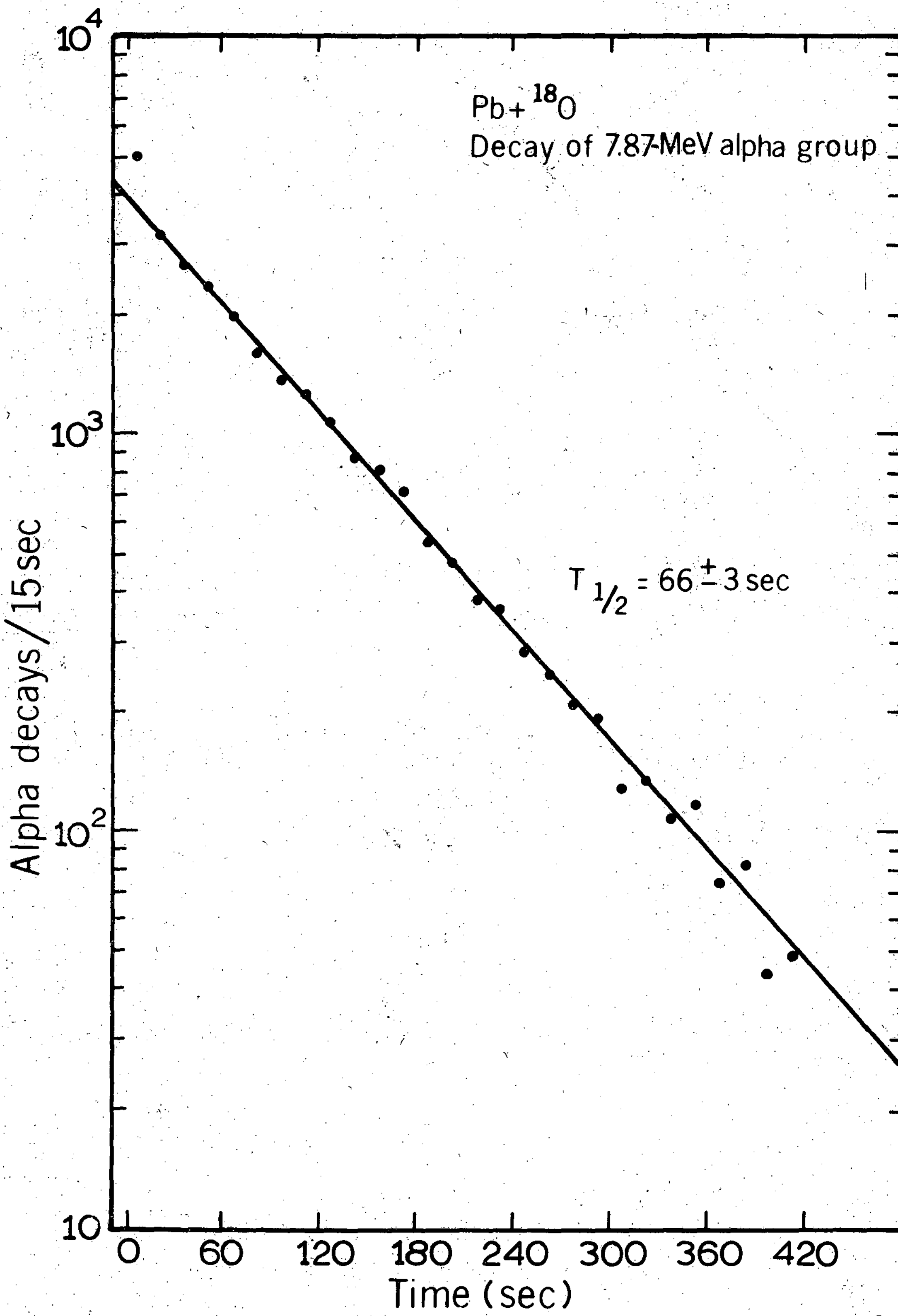
Pb+ 18O
Decay of 7.87-MeV alpha group
T 1/2 = 66 ± 3 sec
Alpha decays / 15 sec
10^4
10^3
10^2
10
0
60
120
180
240
300
360
420
Time (sec)
XBL 718 6368

Fig. 2

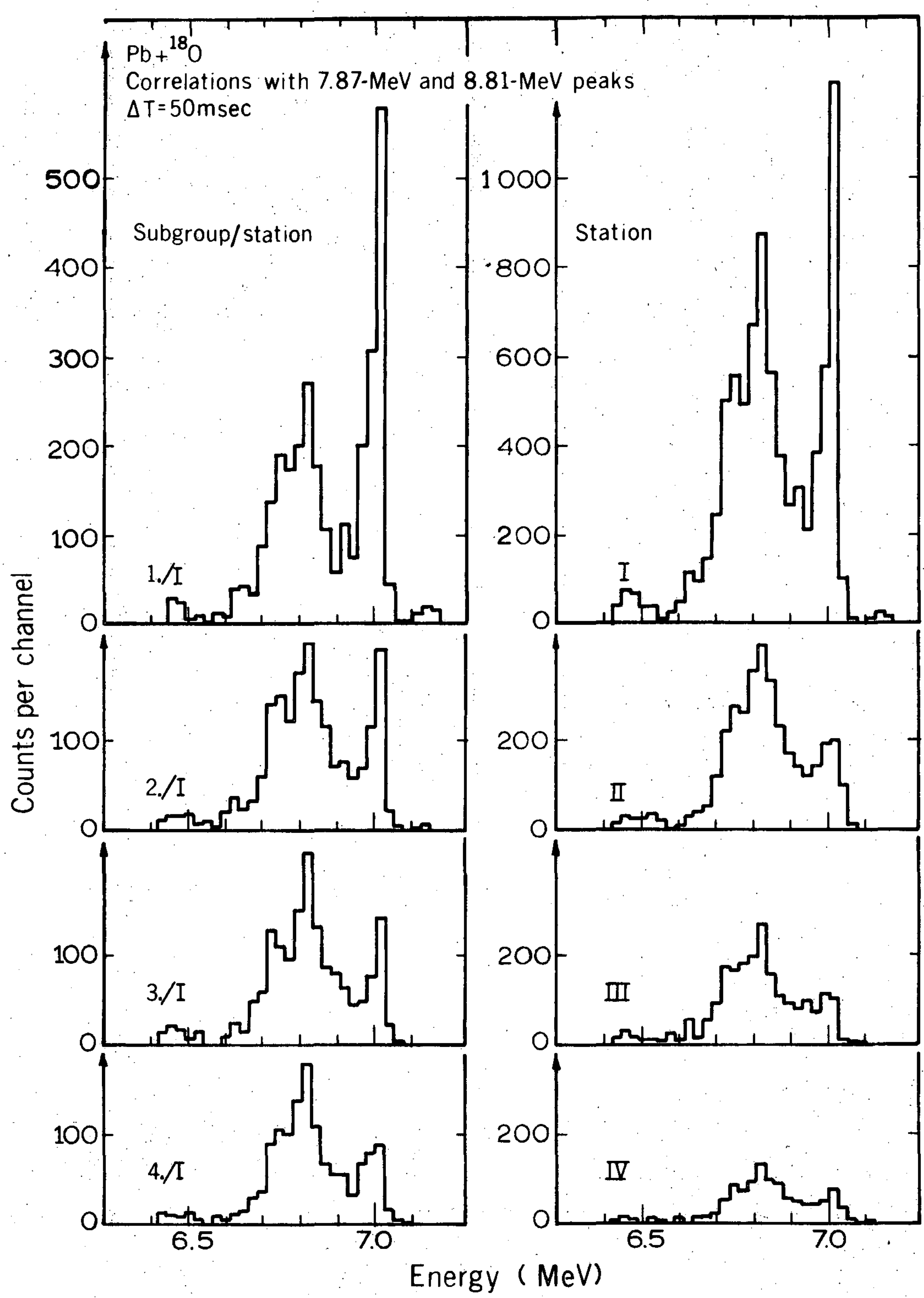

Fig. 3

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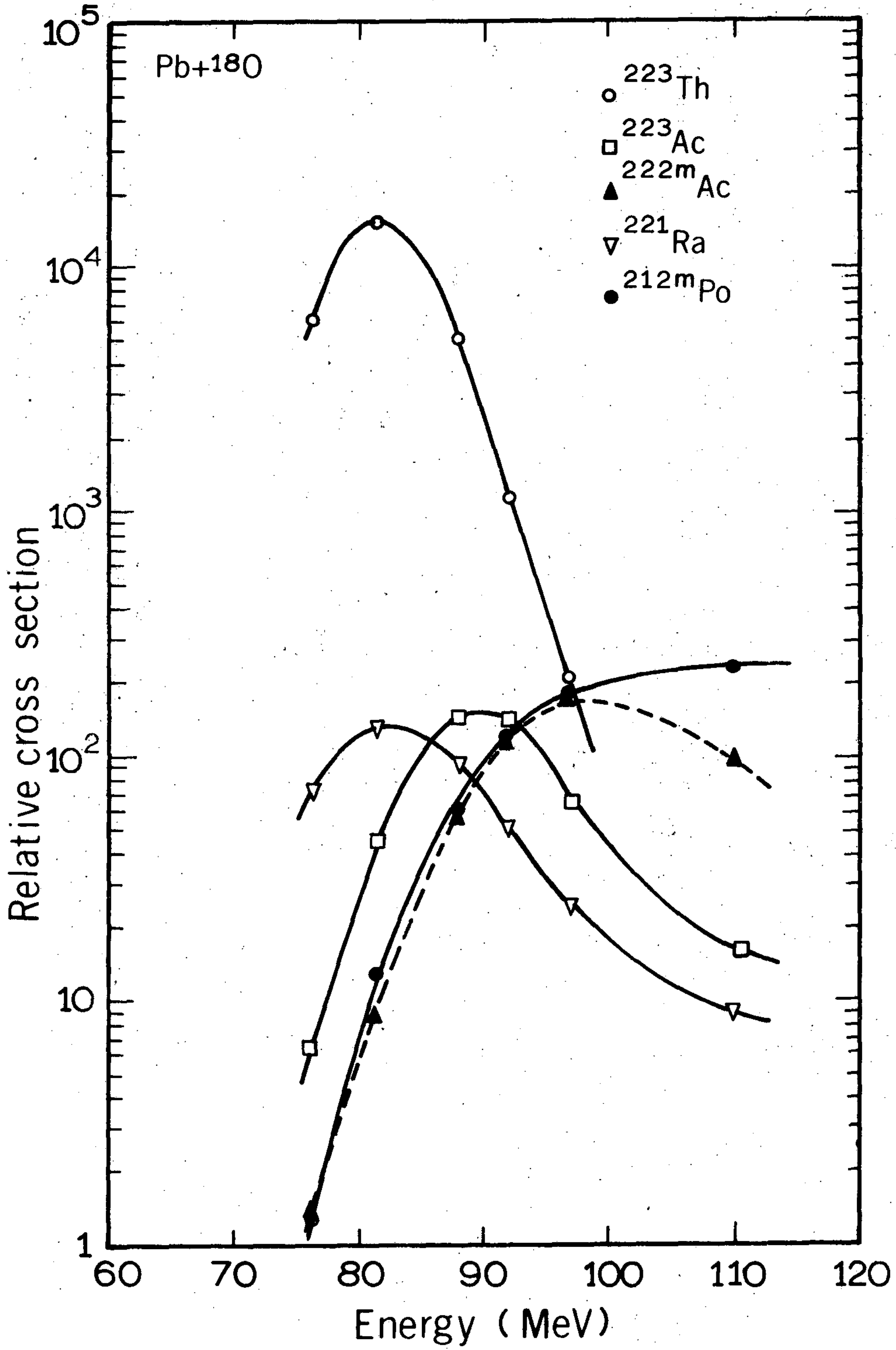

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Fig. 4

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