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AN ISOMERIC STATE OF Po^{212}

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ABSTRACT

An isomer of Po^{212} with a half life of (45.1±0.6)sec and an excitation energy of 2.93 Mev has been positively identified by chemical and nuclear-spectroscopic techniques. The isomer decays to known states of Pb^{208} by the emission of alpha particles with energies and intensities of (11.65±0.02)Mev-97%, (9.08±0.015)Mev-(1.00±0.04)%, and (8.52±0.015)Mev-(2.05±0.09)%. In coincidence with Po^{212m} alpha particles we found gamma rays with energies and intensities of (2.61±0.02)Mev-(2.6±0.3)% and (0.57±0.015)Mev - ~2%. An immense alpha decay hindrance factor for the 11.65 Mev group of 4×10^{13} is explained by spin and parity assignments of 18+ for Po^{212m} in conjunction with a theoretical shell model analysis made by N. K. Glendenning.

The alpha particle energies and intensities of previously known alpha groups of Po^{211m} [(25.5±0.3)sec] were measured yielding values of (8.87±0.01)Mev-(7.04±0.14)%, (7.99±0.015)Mev-(1.66±0.03)% and (7.27±0.015)Mev-91%. A new alpha group was found with an energy and intensity of (8.30±0.015)Mev-(0.25±0.02)%. In coincidence with Po^{211m} alpha particles was a gamma ray of (0.90±0.015)Mev-(1.65±0.11)%. These Po^{211m} radiations are all consistent with the known energy level scheme of Pb^{207}.

AN ISOMERIC STATE OF Po^{212}*

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I. INTRODUCTION

Investigations associated with the use of accelerated heavy ions for the preparation of new elements have turned up an alpha-emitter of very high energy shown to arise from lead and bismuth in the targets. In this laboratory, Ghiorso and co-workers[1] have irradiated bismuth and lead with carbon and oxygen ions in the Heavy Ion Linear Accelerator (HILAC) and used ionization chambers and silicon counters as detectors. Karnaukhov, Khalizev and Flerov[2] bombarded lead with carbon and oxygen ions in the Moscow Heavy Ion Cyclotron and observed the high energy alpha tracks in photographic emulsions.

The Moscow group reported the properties of the high-energy emitter to be (11.8±0.4)Mev with a half life of about 1 minute. Among the other alpha groups seen was one of (9.0±0.3)Mev having a half life of (35±10)sec. From excitation functions they deduced that both of these groups belonged to emitters in the polonium-radium region and suggested that they were "long-range alpha groups" for which the half lives measured are those of the parents which decay by K-capture.

In the present work, the alpha activities produced by heavy ion and alpha particle bombardments of bismuth and lead were examined by radiochemical methods, and their decay spectra were measured. These studies definitely show that the alpha particles at 11.6 Mev, as well as several groups of lower energy, arise directly from the alpha decay of an isomer of Po^{212} with a half life of 45 sec. A half life of 45 sec. for an 11.6 Mev alpha group in this region corresponds

to a retardation or hindrance in the alpha emission process by a factor of about 10^{13}. This obviously implies an unusual structure for Po^{212m} and a severe restriction on the possible excited states of Po^{212} which lie between Po^{212m} and the ground state.

II. BOMBARDMENT CONDITIONS AND CHEMICAL SEPARATIONS

The HILAC delivers ions with energies of 10.4 Mev per nucleon which can be degraded in energy by the use of absorbers placed in front of the targets. In the present experiments ions of C^{12}, B^{10} and He^{4} were used. Although all three of these projectiles were employed in identifying the isotope responsible for the high energy alpha groups, the He^{4} irradiations proved most illuminating when it was found that the α groups in question appeared with bismuth (Bi^{209}) as the target but not with lead (Pb^{208}). With lead as the target, the alpha groups of Po^{211m} were readily observed.

The lead and bismuth targets were prepared by melting the oxides onto thin nickel (0.003 inch) or stainless steel (0.001 inch) foils. The oxide films varied from 50 to 500 mg/cm^2 thickness. In the initial experiments the alpha groups and their half-lives were measured on preparations obtained merely by catching the recoils from the heavy-ion reactions. The energy measuring device consisted of a gold-silicon surface barrier detector, amplifiers and a 400-channel pulse height analyzer.

When it was found that the high-energy alpha groups could be prepared with helium ions on bismuth, some simple and rapid chemical processing was employed to limit further the element which was responsible. The oxide targets were heated with a gas torch in such a way that volatile elements such as polonium and astatine would be volatilized onto a platinum plate without

vaporizing the bismuth or lead appreciably. It was thus possible to show that the activity was not isotopic with the target elements. Furthermore, it was possible to volatilize the astatine fraction from the platinum plate selectively leaving the polonium fraction behind, and when this was done the 45 sec. activity remained with the polonium. The internal monitors for these separations were At^{211} and Po^{211m} which could be followed through their characteristic half-lives and alpha-group energies.

A more definitive chemical identification resulted from the following experiment. A lead oxide target was irradiated with a 0.5 μamp beam of 116 Mev B^{11} ions for 2 minutes and processed with the "blowtorch chemistry" just described. The residue on the platinum plate was dissolved in hydrochloric acid and transferred to a small Dowex A1 anion exchange column. The column was washed with 12M HCl which should strip off such elements as lead, bismuth and francium.[3] (The astatine removal had already been accomplished by selective heating as described.) The polonium faction was then stripped with concentrated perchloric acid and the alpha particle spectrum determined. After correcting intensities for decay, the ratio of the 45 sec activity (11.6 Mev group) to the 25 sec period of Po^{211m} was 7 ± 1. This is in excellent agreement with the ratio 7.3 ± 0.1 found for these same activities following an identical irradiation but for which no further chemical separation was made.

Further discussion of chemical separations will appear where the isotopic identification is explained. The experiments just cited prove that the 45-sec period belongs to a polonium isotope but do not in themselves rule out the possibility that the associated alpha groups belong to a shorter-lived decay product.

III. EXPERIMENTAL RESULTS

A. Confirmation of 11.6 Mev Alpha Particles

Alpha energies were again determined with a gold-silicon surface barrier detector, amplifiers, and a 400-channel pulse height analyzer. The 400-channel analyzer could be divided into four 100-channel sections, and the spectrum from a single preparation could be displayed on these successively in order to determine the decay periods. The energy scale was calibrated using the 8.780 Mev ground state transition of Po^{212} and the rare long-range alpha groups of 9.492 and 10.543 Mev. The response was assumed to be linear out to ~12 Mev. The best energy for the high energy group with 45-sec half-life was (11.65 ± 0.02) Mev.

An alpha group of ~12 Mev energy with measurable half-life is sufficiently unusual that one may well be cautious to the extent of eliminating the possibility of some coincidence or electronic stack-up effect. A simple energy-degradation experiment was employed to show that an 11.6 Mev alpha group was indeed present. A thin source of the 45-sec activity was measured interposing an aluminum foil of 9.5 mg/cm^2 thickness between it and the gold-silicon detector. The peak was much broadened on the low energy side but gave the correct energy of 8.0 Mev expected for an 11.6 Mev alpha particle. For comparison, a 10 Mev alpha particle would be degraded to 6.0 Mev and a 6 Mev alpha particle would be completely absorbed.

B. Isotopic Assignment

The initial observations that the 11.6 Mev alpha emitter could be produced with heavy ions on lead and bismuth left much uncertainty as to its identity since no element from polonium to radium could be ruled out. The production of this activity with helium ions on bismuth not only limited the

atomic number but the mass number must also be 212 or less. The most prominent products to be expected from targets of Bi^{209} and Pb^{208} are listed in Table I. The crude excitation functions shown in Fig. 1 in conjunction with the chemical evidence indicated that Po^{212} was a likely assignment. The 45-sec activity appeared as it should according to an (α,p) reaction on Bi^{209} and is shown in conjunction with the 25-sec Po^{211m} produced by the (α,pn) reaction. With Pb^{208} as the target, Po^{211m} was produced by an (α,n) reaction but the 45-sec period was found only in extremely low intensity.

Table I. Prominent α induced reactions of Bi^{209} and Pb^{208}.

$Bi^{209}(\alpha,n)$	At^{212}	0.2 sec
$(\alpha 2n)$	At^{211}	7.0 hours
(αp)	Po^{212}	3.0×10^{-7} sec
(αpn)	Po^{211}	0.52 sec
	Po^{211m}	25 sec
$Pb^{208}(\alpha,n)$	Po^{211}	0.52 sec
	Po^{211m}	25 sec
$(\alpha,2n)$	Po^{210}	138 days
(α,p)	Bi^{211}	2.16 min
(α,pn)	Bi^{210}	5.0 days
	Bi^{210m}	2.6×10^{6} years

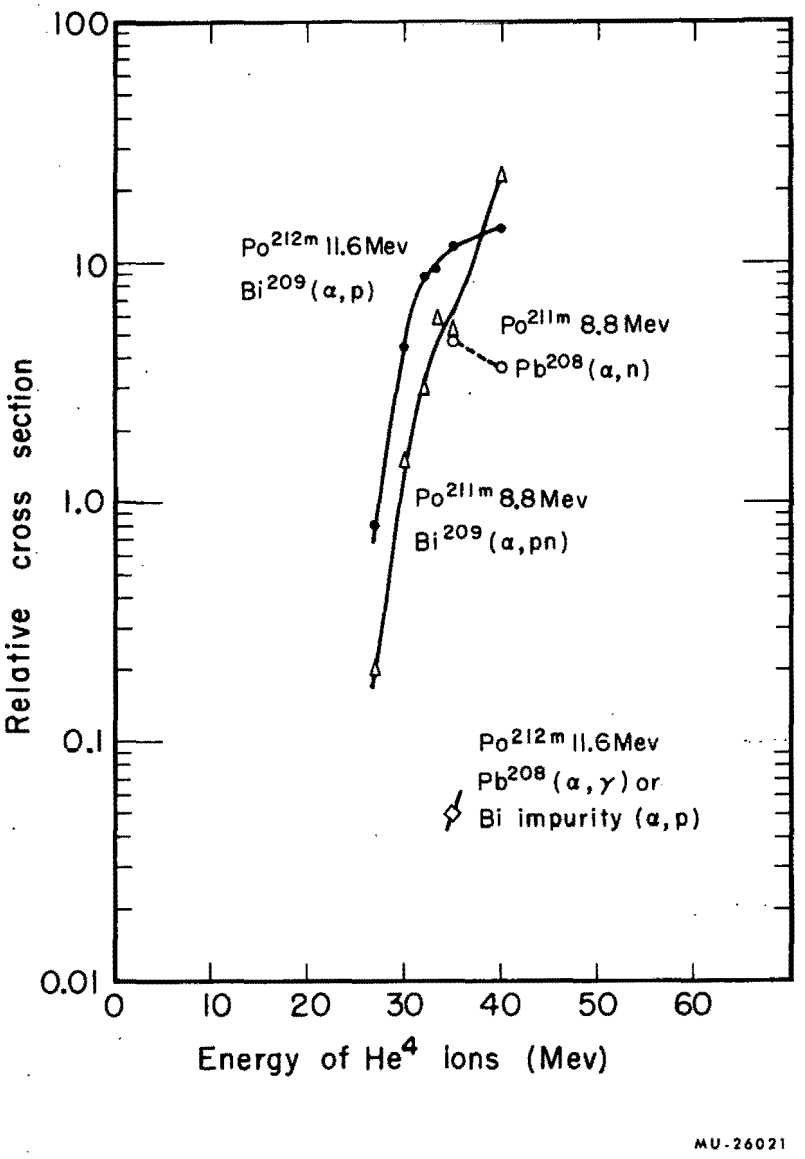

Fig. 1. Relative cross sections for the production of Po^{212m} and Po^{211m} from α bombardments of Bi and Pb.

Normally the above information would be sufficient to assign the activity to Po^{212m}. As this nucleus must have an extremely high spin to explain the highly hindered alpha decay, however, it is possible that a high-spin state might be reached with Bi^{209} (spin 9/2) as the target but not with Pb^{208} (spin 0). Furthermore, the highly unlikely possibility that the actual α particles arise from the daughter of a 45-sec β-emitter is not ruled out from this evidence. The concluding experimental proof that the 45-sec period is the alpha emitter Po^{212m} came from a study of the complex alpha spectrum and the attendant gamma rays.

C. Complex Alpha Spectrum of Po^{212m}

Figure 2 shows a logarithmic plot of alpha peak intensities from the polonium fraction of a bismuth target irradiated with 40 Mev helium ions. On the assumption that the 11.65 Mev alpha group leads to the ground state of Pb^{208}, particular attention was given to a search for alpha-groups leading to known excited states of Pb^{208}. Two such groups were found: one at 9.08 Mev corresponding to a transition to the known 3- state of Pb^{208} at 2.62 Mev and another at 8.52 Mev assigned as the transition to the 5- state at 3.20 Mev. The other alpha-groups agree well with the known α-spectrum of 25-sec Po^{211m}. The peak at 8.30 Mev had not been observed by Jentsche et al.[4] in Po^{211m} decay, presumably because of its low intensity. It corresponds, however, with a known energy level in Pb^{207}. Very small peaks for At^{211} and Po^{211} represent a small residue of astatine left in the polonium fraction.

The assignment of the various groups to either the 45-sec activity or to 25-sec Po^{211m} was completed by checking their half-lives. The decay curves of Fig. 3 are the summation of two runs for which the chemistry was sufficiently rapid and clean to obtain good intensity data on the various peaks.

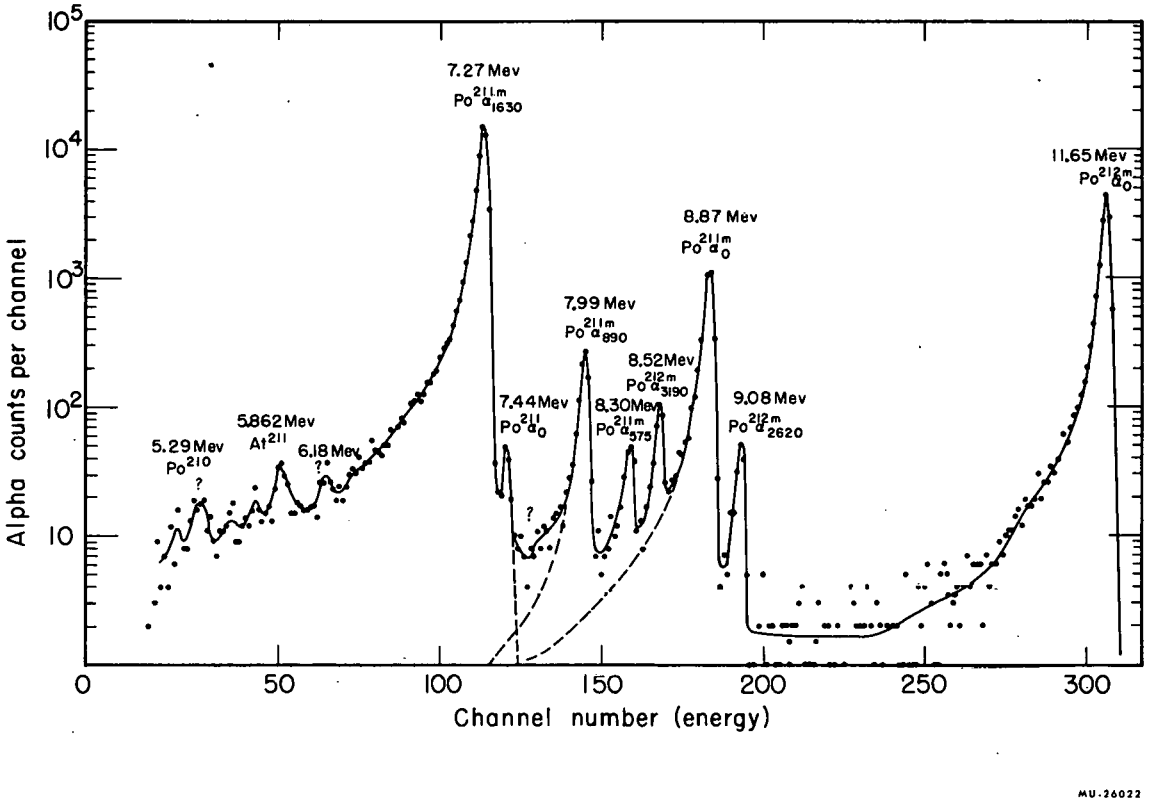

Fig. 2. Alpha spectrum of Po^{212m} and Po^{211m}.

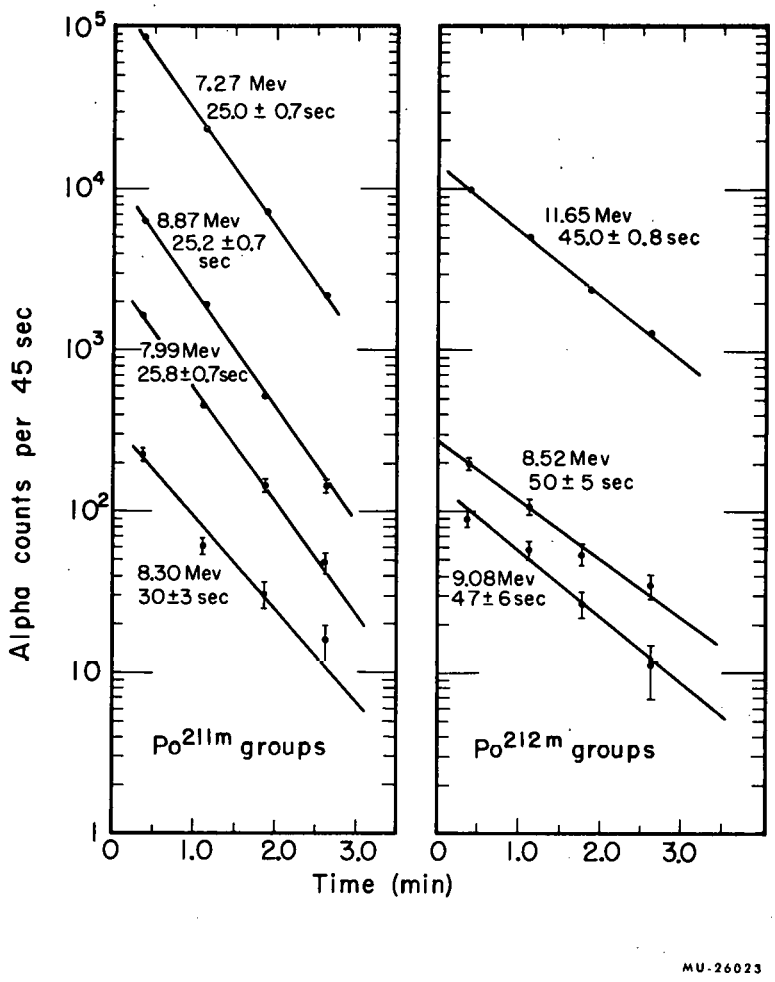

Fig. 3. Decay curves for Po^{211m} and Po^{212m} α groups.

The energy and intensity values for the α-groups of both Po^{211m} and Po^{212m} are summarized in Table II. The data on Po^{211m} are in reasonably good agreement with values reported previously. The indicated errors are standard deviations.

Table II. Po^{212m} and Po^{211m} alpha spectra data.

Alpha group	Alpha particle energy (Mev)	Excited state energy (Mev)	Intensity (%)	Jentschke et al.[4] Energy (Mev)	Jentschke et al.[4] Intensity (%)
Po^{212m}	11.65±0.02	0	97		
	9.08±0.015	2.62	1.00±0.04		
	8.78		< 1.5		
	8.52±0.015	3.19	2.05±0.09		
Po^{211m}	8.87±0.01	0	7.04±0.14	8.70±0.05	7.0
	8.30±0.015	0.575	0.25±0.02	---	---
	7.99±0.015	0.89	1.66±0.03	7.85±0.05	2.5
	7.44		< 0.2	7.43	< 1
	7.27±0.015	1.63	91	7.14±0.05	90.5

Energy standards: At^{211} 5.862 Mev
Po^{212} 8.780 Mev

The γ-ray measurements carried out in this study can best be visualized in terms of the suggested decay scheme based upon the α-particle and γ-ray data. This is shown in Fig. 4 along with the energy level diagram of Pb^{208} obtained largely from the study of Tl^{208} β-decay.[5] The objective was to show that alpha particles with 45-sec half-life are in coincidence with γ-rays of the required

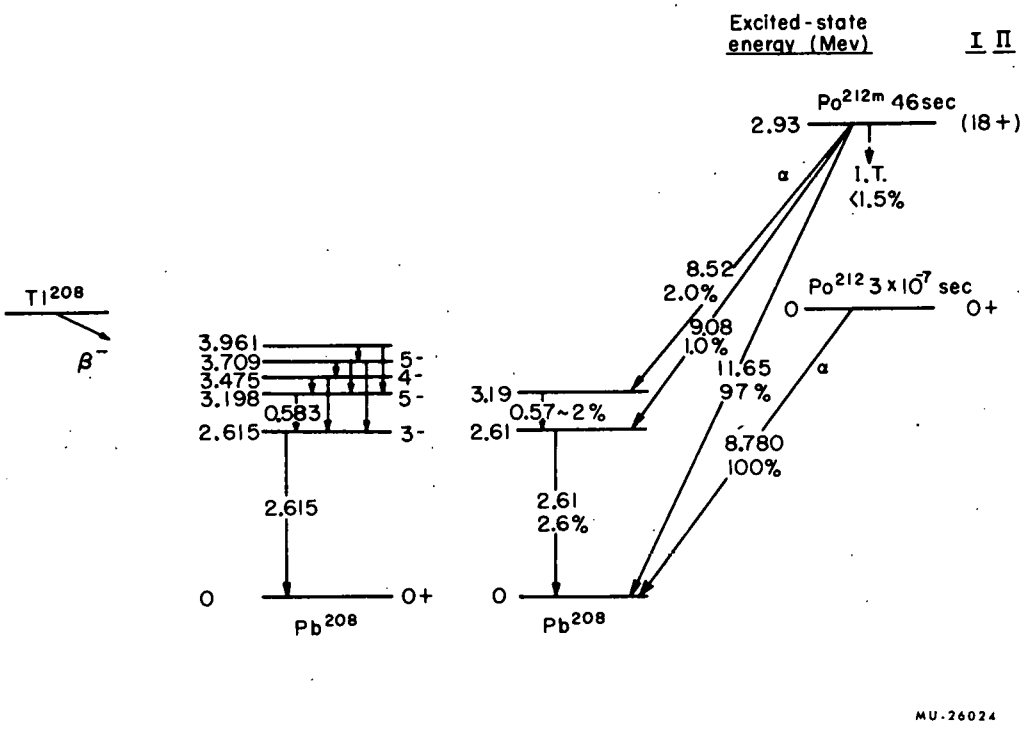

Fig. 4. Decay schemes of the Po^{212} isomers and Tl^{208}.

energies and intensities. Since Po^{211m} was present in all measurements its decay scheme is also reproduced here (Fig. 5).

A number of experiments were performed in which the polonium fraction was isolated after a short irradiation of bismuth with 40 Mev helium ions and the γ-ray spectrum measured with a scintillation counter. The data on each experiment were printed out for successive 0.7 min measuring intervals so as to obtain decay rates for the several peaks. In addition the alpha decay rate was measured simultaneously.

The γ-ray peaks which appeared had energies of 0.57, 1.06, 1.62 and 2.61 Mev. According to the decay scheme of Jentschke et al. for Po^{211m} (Fig. 5), there should be γ-rays of 0.56 and 1.06 Mev in high intensity (almost one gamma-ray per alpha-particle). These were found in about the requisite intensities when compared with the 25-sec component of the alpha particles and also had the appropriate half life. The peak at 1.62 Mev has the required energy and intensity to be attributed to coincidence stack-up of the 0.57 and 1.06 Mev peaks. The peak at 2.61 Mev was present in very low intensity and appeared not to decay with a 25-sec half-life.

The same type of experiment was performed, but this time the γ-ray detector signal was placed in coincidence with the signal from an α-particle detector. It will be noted that the strong γ-rays of 1.06 and 0.57 Mev arise from the 1.63 Mev metastable state in Pb^{207} (0.8 sec half-life). Consequently, they should disappear in the coincidence counting. The peak at 1.06 Mev did disappear except for a small component which had the expected intensity for accidental coincidences. With this removal of the intense 1.06 Mev peak a true coincidence peak at 0.90 Mev showed up. This is a low intensity (1.7%) γ-ray in Pb^{207} which should be in prompt coincidence with Po^{211m} alpha particles (see Fig. 5), and as expected it decayed with a half-life close to 25-sec [(26.5 ± 1.1)sec].

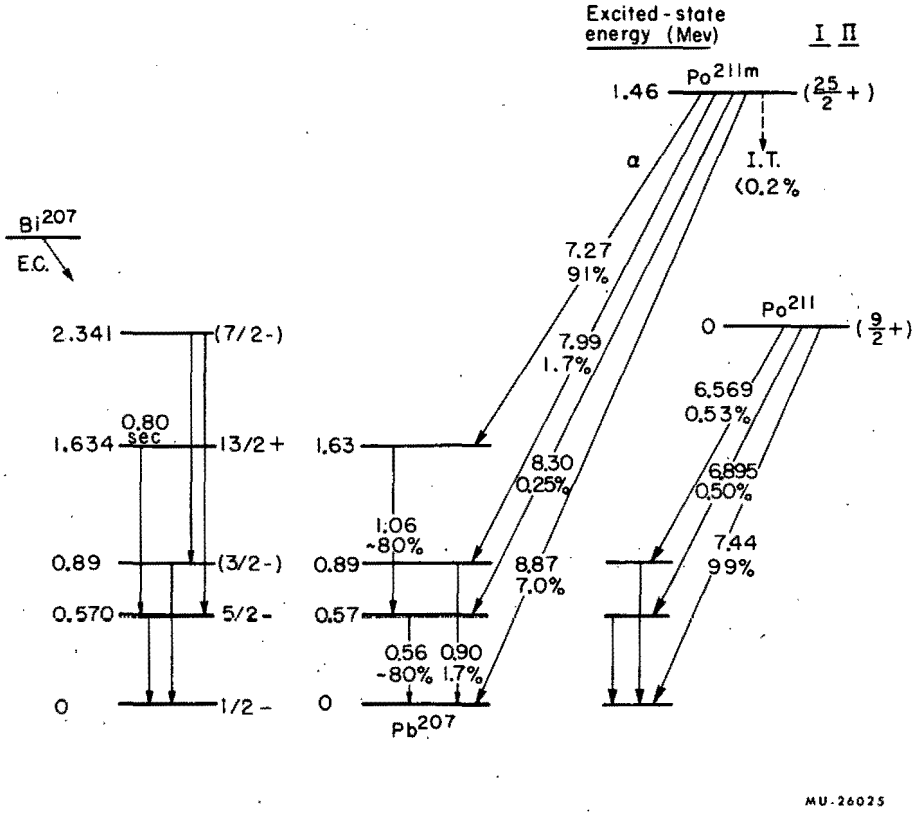

Fig. 5. Decay schemes of the Po^{211} isomers and Bi^{207}.

The peak at 0.57 Mev was strongly diminished in the α-γ coincidence counting but a considerable intensity still remained. According to the decay schemes of Figs. 4 and 5 there are three sources of α-γ coincidences involving a photon of this energy. One of these comes from chance coincidences from the delayed cascade through the 1.63 Mev state in Pb^{207}. Another comes from prompt coincidences involving the newly found α-transition of Po^{211m} which goes directly to the 0.57 Mev state in Pb^{207}. The third is the transition between the 3.19 and 2.61 Mev states in Pb^{208} arising from Po^{212m} decay. The chance coincidence rate for the 0.57 Mev gamma ray could be determined from the remaining peak height of the 1.06 Mev transition in Pb^{207} which can only arise through chance coincidences. When this was subtracted, there resulted an α-0.57 Mev γ coincidence component which decayed with a 45 ± 5 sec half-life. This would appear to be the expected coincidences from the Po^{212m} decay. However, there should still be present the 25-sec component from the true coincidence involving the 0.57 Mev state in Pb^{207}. With this uncertainty we can only give an approximate value for the intensity of the Po^{212m} coincidences.

The 2.61 Mev coincidence peak decayed with the proper half-life to arise from Po^{212m} decay and the intensity conformed rather well with the alpha-spectrum. The summary of the coincidence data is given in Table III and the intensities may be compared with the demands of the decay schemes in Figs. 4 and 5.

Table III. Po^{212m} and Po^{211m} alpha particle-gamma ray coincidences.

Isotope	γ energy (Mev)	Half-life (seconds)	Intensity (%)
Po^{211m}	0.90±0.015	26.5±1.1	1.65±0.11
Po^{212m}	0.57±0.015	45 ±5	~2%
	2.61±0.02	49 ±6	2.6 ±0.3%

IV. INTERPRETATION

In the present picture of the alpha decay process, it has proved useful to consider separately: (a) the barrier penetration of a particle in a one-body model, and (b) the intrinsic nuclear factors having to do with the initial and final nuclear states and the creation of the alpha particle on the nuclear surface. The ground state transitions of even-even nuclei serve as an important guide line since they obey the one-body barrier penetration features of this problem without consideration of intrinsic nuclear factors. The common feature of all such transitions is that the alpha-particles comprise an $\ell=0$ wave and the initial and final nuclear states are simply ground-state even-even nuclei. Such transitions are spoken of as "unhindered".

The emission of an alpha-particle which carries off angular momentum will not proceed so rapidly because the barrier penetrability is reduced, and in general intrinsic nuclear factors come into play. It is possible to remove from these considerations the effect of angular momentum per se on the barrier penetrability by introducing a "centrifugal barrier" term. The retardation which remains is sometimes termed the "reduced hindrance factor".

In considering the decay of the 11.65 Mev transition of Po^{212m} to the ground state of Pb^{208} comparison is made with the (unhindered) decay of ground-state Po^{212} for which the hindrance factor is taken to be unity. The hindrance factor for the 11.65 Mev transition and for the other groups of Po^{212m} (as well as those for the Po^{211} isomers) are listed in Table IV. We shall consider primarily the 11.65 Mev transition.

It was likely at the outset that Po^{212m} has a huge spin because no case has yet been found in which intrinsic nuclear factors produce hindrance factors of more than several thousand. Indeed, before the isotopic assignment had been made experimentally, it was deduced to be Po^{212m} because of the large spin which

Table IV

Isotope	Half-life (seconds)	Alpha-particle energy (Mev)	Intensity (%)	Hindrance factor	L	L Hindrance factor	Reduced hindrance factor
Po^{212}	3.0×10^{-7}	8.78	100	1.000	0	1	1
Po^{212m}	45.1±0.6	11.65	97	4.04×10^{13}	18	6×10^{9}	6×10^{3}
		9.08	1.0	7.3×10^{10}	15	5×10^{7}	1.5×10^{3}
		8.52	2.05	1.7×10^{9}	13	1×10^{6}	1.5×10^{3}
Po^{211}	0.52	7.44	99.0	1.1×10^{2}	5	12	9
		6.88	0.53	2.7×10^{2}	3	2.8	1.0×10^{2}
		6.56	0.50	18	3	2.8	6
Po^{211m}	25.5±0.3	8.87	7.0	7.1×10^{8}	13	9.6×10^{5}	7×10^{2}
		8.30	0.25	6×10^{8}	11	3×10^{4}	2×10^{4}
		7.99	1.66	1.23×10^{7}	11	3.5×10^{4}	4×10^{2}
		7.27	91	1.60×10^{3}	6	3	5×10

could result in a nucleus which has two neutrons and two protons beyond the doubly closed shell of Pb^{208}. In addition, Po^{212m} with a large spin might have to decay principally to the ground state of Pb^{208} because of the absence of levels of very high spin within several Mev of the ground state. The effect of the centrifugal barrier on the hindrance factor has been calculated by Glendenning[6] using the formulation given by Bethe.[7] The relation between the angular momentum carried off in the alpha decay and the angular momentum hindrance factor for the 11.65 Mev group is shown in Fig. 6. It is seen that the observed hindrance factor could be explained by I ~ 21 for the emitting state. As we shall see, the maximum spin due to particle configuration is likely to

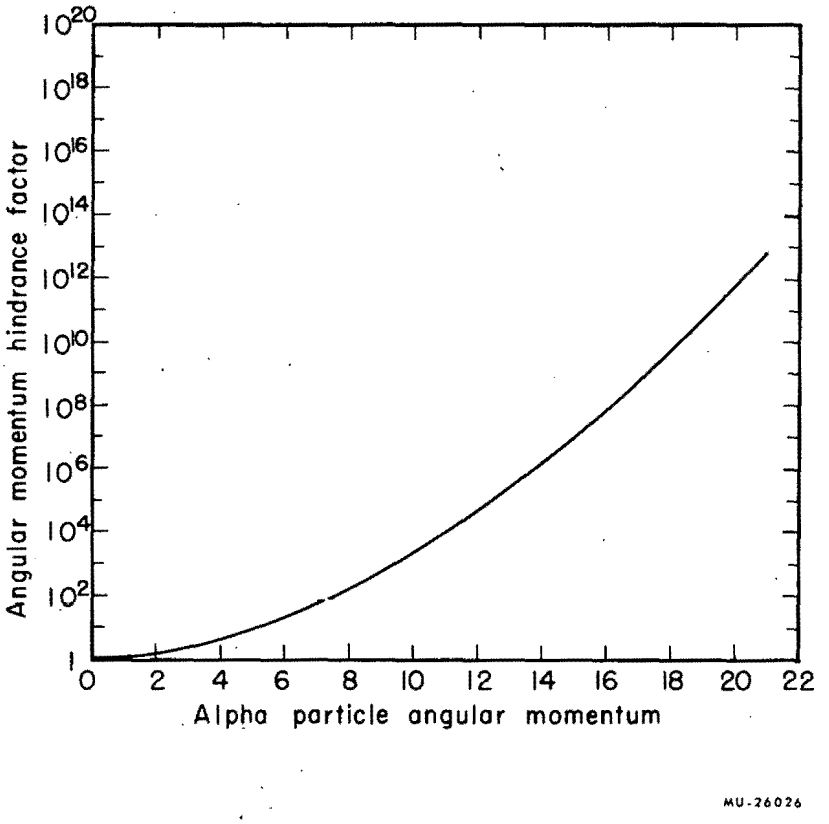

Fig. 6. Angular momentum hindrance factors for an 11.6 Mev α group of Po^{212}.

be 18. This would imply that the decay of Po^{212m} involves some intrinsic hindrance beyond the centrifugal barrier retardation, a conclusion not at variance with what one might expect.

The shell model states for the two protons beyond 82 are h9/2 and the neutrons, i 11/2 or g 9/2. The maximum angular momentum for a configuration $(h\ 9/2)^2\ (i\ 11/2)^2$ is 18. The same particle states can, of course, couple to lower total angular momentum and in addition there are other configurations which must be considered. If Po^{212m} does indeed have spin 18 (or some other high spin) there is another condition which must be met in explaining its decay properties. We can set a minimum half-life of 3×10^3 sec for the isomeric transition of Po^{212m}, which, in essence, means that there cannot be another state between 2.9 Mev and the ground state which has a spin of 4 units or fewer removed from that of Po^{212m}. When one considers the multitude of four-particle configurations possible, this criterion presents an unusual challenge for shell model calculations.

The accompanying paper by Glendenning[8] is concerned with shell model calculations for Po^{212}. He concludes that the $(h\ 9/2)_p^2\ (i\ 11/2)_n^2$ configuration is the one responsible for Po^{212m} because the order of spin states expected is 0,2,4,6,8,10,18,16, etc., and no other configuration gives a state of $I > 10$ below the spin 18 state. If the Po^{212} isomer does have a spin of 18, the "spin gap" between 10 and 18 is just what is required to explain the long alpha-decay half-life and the absence of the isomeric transition.

Glendenning has calculated the angular momentum hindrance factors for each of the alpha transitions shown in Figs. 4 and 5 (see Table IV). If spin 18 is the correct assignment for Po^{212m}, one might consider if the reduced hindrance factor of 6×10^3 is reasonable for the 11.65 Mev alpha group. Shell model calculations of the type made by Mang[9] would be desirable for a detailed

analysis of the alpha transition probabilities. A simple comparison, however, with the Po^{211} isomers can be fruitful. Po^{211} has been assigned a configuration of $\left[(h^2_{9/2})_0 g_{9/2}\right]_{I=9/2}$ and Po^{211m} has been assigned[9] a configuration of $\left[(h^2_{9/2})_8 g_{9/2}\right]_{I=25/2}$. As seen from Table IV, the Po^{211m} reduced hindrance factors are about a factor of 100 larger than those for Po^{211} decay to the same states. This factor of 100 should then be related to the partial decoupling of the proton configuration from $(h^2_{9/2})_0$ to $(h^2_{9/2})_8$. The same difference in proton configuration occurs in Po^{212} and Po^{212m}, and in addition, a pair of $(i^2_{11/2})_0$ neutrons are partially decoupled to $(i^2_{11/2})_{10}$. If the hindrance associated with each of these decouplings is the same as for the Po^{211} isomers, the resulting reduced hindrance factor is $\sim 10^4$ — in reasonable agreement with the value of 6×10^3 derived from the Po^{212m} data.

FOOTNOTES AND REFERENCES

* This work was done under the auspices of the U. S. Atomic Energy Commission.

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