UCRL-2305
UNCLASSIFIED

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

TWO-WEEK LOAN COPY

This is a Library Circulating Copy which may be borrowed for two weeks. For a personal retention copy, call Tech. Info. Division, Ext. 5545

BERKELEY, CALIFORNIA

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

UCRL-2305
Unclassified-Chemistry Distribution

UNCLASSIFIED

UNIVERSITY OF CALIFORNIA
Radiation Laboratory
Contract No. W-7405-eng-48

I. ISOMERIC STATES OF BISMUTH 210
II. RELATIVE YIELDS IN THE FORMATION OF NUCLEAR ISOMERS

Harris Benjamin Levy

(Thesis)

August 1953

Berkeley, California

TABLE OF CONTENTS

		Page
	ABSTRACT	3
I.	ISOMERIC STATES OF BISMUTH 210	4
	A. Introduction	4
	B. Confirmation of the Mass Assignment of the Long-lived Isomer	6
	C. Alpha Decay Energy of Long-lived Bi^{210}	8
	D. Decay of the Long-lived State of Bi^{210} to Po^{210}	12
	E. Search for Long-lived Bi^{210} in the Decay of RaD (Pb^{210})	17
	F. Summary and General Discussion	20
II.	RELATIVE YIELDS IN THE FORMATION OF NUCLEAR ISOMERS	25
	A. Introduction	25
	B. Experimental Procedures	29
	C. Treatment of Data	32
	D. Results	33
	E. Discussion	35
III.	ACKNOWLEDGMENTS	41
IV.	REFERENCES	42

I. ISOMERIC STATES OF BISMUTH 210

II. RELATIVE YIELDS IN THE FORMATION OF NUCLEAR ISOMERS

Harris Benjamin Levy
Department of Chemistry and Radiation Laboratory
University of California, Berkeley, California

August 1953

ABSTRACT

In Part I, the previous work on a long-lived alpha-emitting isomeric state of Bi^{210} has been revised and extended. The mass assignment has been confirmed by separation of mass fractions, and the half-life of this isomer is now estimated to be $\sim 10^6$ years. The alpha decay energy was redetermined to be 5.031 ± 0.020 Mev, indicating that the long-lived state of Bi^{210} lies 25 ± 40 kev below RaE. The branch decay of the long-lived state to Po^{210} was determined and found to be about one part in 270. A search resulted in an upper limit of one part in 10^6 being set for the amount of the long-lived state present in uranium ores relative to the amount of RaE present. A discussion of the relationship of the long-lived state of Bi^{210} to Pb^{210}, RaE, and Po^{210} is presented, showing that a likely designation for the long-lived state is 4-.

In Part II, the formation of nuclear isomers in reactions is examined on the basis of results obtained for the isomers of Co^{58} from an (α, n) reaction on Mn^{55}. The results on the Co^{58} isomers prove that the ratio $(2I_m + 1)/(2I_g + 1)$ cannot act as a limiting value for the ratio σ_m/σ_g. A qualitative analysis of the mechanism for the formation of isomers in nuclear reactions is presented.

I. ISOMERIC STATES OF BISMUTH 210

A. Introduction

In 1950 Neumann, Howland, and Perlman[1] reported the existence of a long-lived isomeric state of Bi^{210} formed by prolonged neutron bombardment of bismuth. After all the RaE had decayed out and the bismuth fraction had been repurified, a long-lived alpha activity remained. This alpha activity was assigned to Bi^{210} on the basis of experiments in which the daughter, Tl^{206}, was milked from the bismuth fraction.

The above mentioned authors looked for the growth of Po^{210} in the sample and found none, indicating that alpha emission was the principal mode of decay for the long-lived isomer with little branching decay. This is in marked contrast with RaE, the well-known naturally occurring state of Bi^{210}, which decays with a 5-day half-life by β^- emission and has only a very small amount of alpha decay branching.

The above authors also measured the alpha particle energy of this long-lived Bi^{210} by means of an ionization chamber connected to a differential pulse height analyzer. From data then available it was also possible to calculate the alpha energy for RaE. Comparison of the two alpha energies indicated that the long-lived isomer was the metastable excited state, while RaE was the ground state of Bi^{210}.

Palevsky, Hughes, and Eggler[2] reported the total absorption cross section for pile neutrons on bismuth to be 30 millibarns. The activation cross section for the formation of 5-day RaE is only 15 millibarns. It is probable that the difference between these two

values represents the cross section for the formation of the long-lived isomeric state of Bi^{210}. If the cross sections for formation of the two states and one of the half-lives is known, it is possible to estimate the half-life of the other state by comparison of the amounts of activities of the two states formed in a bombardment. If the value of 15 millibarns is used for the cross section for formation of the long-lived Bi^{210}, one obtains a half-life of $\sim 10^6$ years for this state.

Further considerations and developments made it desirable to go over (and revise, if necessary) and to extend the work done on the long-lived Bi^{210}. The energy separation of the two isomeric states made the apparent absence of isomeric transition seem rather puzzling. Then better samples became available making it possible to determine with greater accuracy some of the characteristics of the long-lived bismuth alpha emitter. The mass assignment was confirmed by means of separated mass fractions; the alpha energy was redetermined with greater accuracy; and a successful search was made for the branch decay to Po^{210}. The results of this work led to a revision of some of the previous conclusions concerning the relationship between the long-lived state of Bi^{210} and the 5-day RaE, and also led to a search for the long-lived isomer in uranium ores as the product of a possible branch decay of RaD.

In conclusion, the possible relationship of the long-lived isomer to RaE and to RaD is discussed in the light of the results obtained in this work.

B. Confirmation of the Mass Assignment of the Long-lived Isomer

All the available samples of the long-lived Bi^{210}, since it is formed by an (n, γ) reaction on natural bismuth, contain a considerable bulk of stable Bi^{209}. If this long-lived alpha emitter is actually an isomeric state of Bi^{210}, then any separation of the bismuth fraction into various mass fractions should result in an increase in the specific activity of the 210 mass fraction and a corresponding decrease in the specific activities of the other mass fractions.

A sample of the long-lived bismuth alpha emitter, having a specific alpha activity of ~2 counts per minute per mg of bismuth, was electromagnetically separated into different mass fractions on the Calutron at Oak Ridge National Laboratory. The mass fractions 208, 209, and 210 were collected and examined in the manner described in the following section.

1. Experimental procedure. -- The various bismuth mass fractions were received in nitric acid solutions. Samples of each mass fraction were taken and prepared for counting by electroplating bismuth metal onto tared platinum disks.

The electroplating of the bismuth was carried out as follows. A portion of the solution containing a given bismuth mass fraction was taken and first evaporated to dryness several times with hydrochloric acid. This was done in order to destroy the nitric acid which appeared to interfere with formation of an adherent coating of bismuth metal. The residue was then taken up in 12 to 15 ml of sulfuric acid solution containing 6 ml of H_2SO_4 to 100 ml

of solution.

The sulfuric acid solution containing the bismuth was then transferred to a small cylindrical electrolysis cell. The anode was a rotating platinum rod which also served to keep the solution stirred during electrolysis. The cathode was a tared platinum disk. The various solutions were electrolyzed at an average current density of 1 to 3 milliamps per cm^2. The voltage across the cell ranged from 2.0 to 2.5 volts. The deposit of bismuth metal covered a circular area about one inch in diameter. After electrolysis was completed, the platinum disk was washed with dilute sulfuric acid, water, and finally alcohol, and then dried under a heat lamp.

The platinum disks were then weighed to determine the amounts of bismuth deposited. The deposits ranged in weight from 1.6 mg to 2.5 mg.

2. Results. -- The amount of alpha activity on each of the samples was determined by counting them in an argon alpha counter at 52% geometry. Two samples were taken from each mass fraction. The specific alpha activities of the various samples are given below in Table 1.

Table 1

The specific alpha activity of the various bismuth mass fractions

Mass Fraction	Counts per min. per mg. of bismuth Sample 1	Sample 2
208	0.6 ± 0.3	0.9 ± 0.3
209	0.3 ± 0.3	<0.1
210	64 ± 5	71 ± 5

The unseparated bismuth had a specific alpha activity of ~2 counts per min. per mg. of bismuth.

3. Conclusions. -- The increase of the specific activity in the 210 mass fraction over that of the unseparated bismuth, along with the corresponding decrease of the specific activity in the 208 and 209 mass fractions gives direct proof that the assignment of the long-lived alpha activity to Bi^{210} was correct.

C. Alpha Decay Energy of Long-lived Bi^{210}

The alpha decay energy of the long-lived Bi^{210} was reported by Neumann, et al.[1] to be 5.12 ± 0.05 Mev. From data available at that time the alpha decay energy of RaE was calculated to be 4.94 ± 0.10 Mev. Since the publication of that paper, data has been published making possible a better calculation of the alpha decay energy of RaE. Availability of enriched Bi^{210} samples from the Calutron made possible the preparation of thin samples of Bi^{210} of higher specific activities than heretofore. This, along with the use of better alpha energy standards, made possible a more accurate determination of the alpha decay energy of the long-lived isomer.

1. Experimental procedure. -- Thin samples of enriched Bi^{210} were prepared in the following fashion. The samples from the separated 210 mass fraction described in the previous section were dissolved from the platinum disks with concentrated HNO_3. Portions of this solution were treated in the same manner as described previously to get thin samples of bismuth electrodeposited onto platinum disks.

The kinetic energy of the alpha particles emitted by the long-lived Bi^{210} was then measured by counting the samples in an ionization chamber connected to a 48-channel differential pulse height analyzer. The distribution of pulse heights from the alpha particles of the Bi^{210} compared to those from alpha particles of known energy standards enables one to determine the alpha particle energy of the Bi^{210}.

2. Results. -- The results of the five runs made on the alpha pulse analyzer are listed in Table 2.

Table 2

Determination of Alpha Particle Energy of Long-lived Bi^{210}

Run No.	Alpha Energy (Mev)
1	4.92
2	4.94
3	4.94
4	4.93
5	4.945

On runs 1 to 3, inclusive, the pulse analyzer was calibrated by making separate runs with samples of Ra^{226} plus its daughters. On runs 4 and 5, Po^{208} and Po^{210} were run simultaneously with the Bi^{210}, providing an internal standard for the energy calibration. Figure 1 shows the pulse analyzer curve for run 5.

A weighted average of these results yields a value of 4.935 ± 0.020 Mev for the alpha particle energy of the long-lived Bi^{210}. Correcting for the recoil of the residual nucleus results in

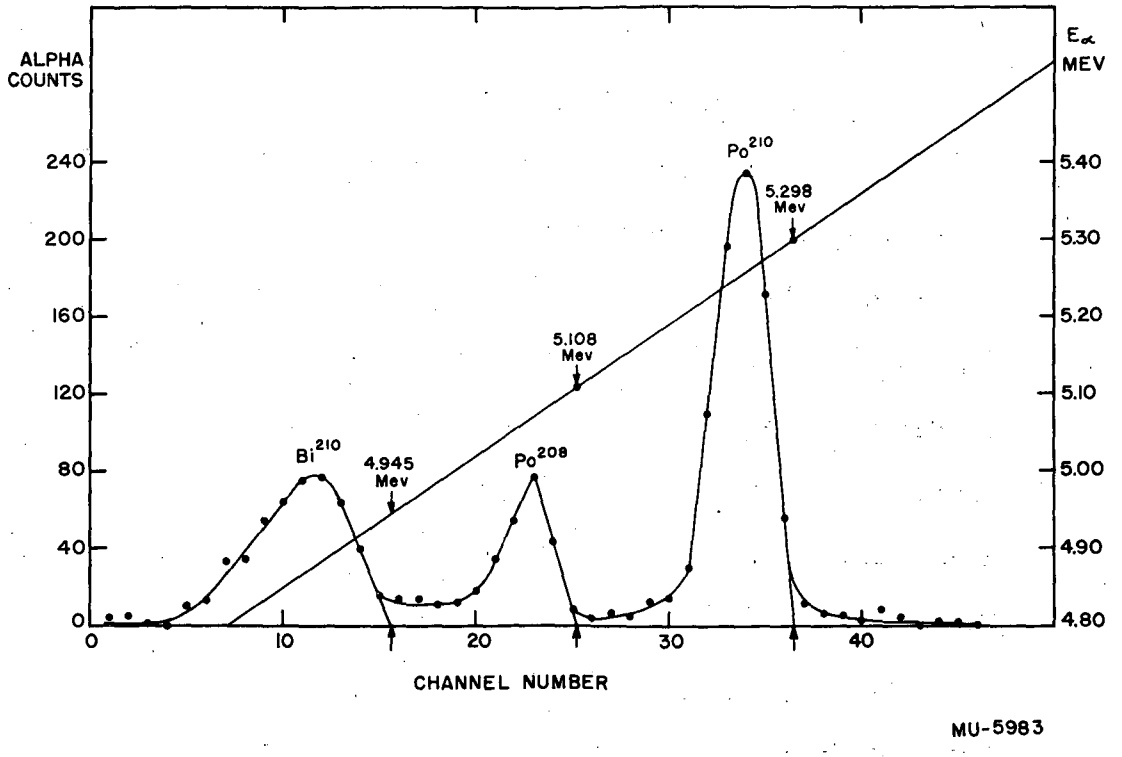


Fig. 1. Alpha pulse analysis of long-lived Bi^{210}.

an alpha decay energy of 5.031 ± 0.020 Mev.

If the observed alpha decay of the long-lived Bi^{210} did not go to the ground state of Tl^{206}, then it should be followed by an isomeric transition. Judging from other alpha emitters, it seems likely that if the decay does go to an excited state of Tl^{206}, it would go to a low-lying excited state, and the transition to the ground state would be internally converted sufficiently to be easily observed. The following experiment was conducted by D. C. Dunlavey of this laboratory. An Ilford G5 photographic plate was impregnated with a solution containing some of the enriched Bi^{210}. After a suitable time, the plate was developed, and the number of alpha tracks which had in coincidence with them electron tracks of <150 kev energy was determined. Dunlavey reported that the number of these low energy electrons in coincidence with alpha particles was very small and could be attributed entirely to the beta spectrum of the Tl^{206} daughter. Thus, in all probability the value 5.031 ± 0.020 Mev does represent the total alpha decay energy of the long-lived Bi^{210}.

3. Discussion. -- By using a closed energy cycle it is possible to calculate from known decay data the alpha decay energy of RaE. The following decay energies were used in the calculation.[3-5]

$$Tl^{206}, \quad E_{\beta^-} = 1.51 \pm 0.01 \text{ Mev}$$
$$RaE, \quad E_{\beta^-} = 1.165 \pm 0.005$$
$$Po^{210}, \quad E_{\alpha} = 5.401 \pm 0.005.$$

These are all total disintegration energies. These values yield a total alpha decay energy for RaE of 5.056 ± 0.020 Mev.

Comparison of the alpha decay energies for both isomeric states of Bi^{210} yields the result that the long-lived Bi^{210} lies 25 ± 40 kev below the 5-day RaE state. The two states lie so close together in energy that although the long-lived isomer appears to be the ground state, our present experimental errors make it impossible for one to state with certainty which is the ground state and which the metastable excited state.

It is possible, however, to draw conclusions as to the energetic relationship between RaD (Pb^{210}) and the long-lived Bi^{210}. RaD decays to RaE with a total decay energy of 65 kev.[5] The long-lived Bi^{210} lies 25 ± 40 kev below RaE. Therefore, RaD should be unstable with respect to the long-lived Bi^{210} by 90 ± 40 kev. Regardless of which of the two states of Bi^{210} in question is the lower state, the decay of RaD to the long-lived state of Bi^{210} should be energetically possible.

These considerations led to a search for the long-lived Bi^{210} in uranium ore where it would be in equilibrium with RaD. The results of this search will be described in a later section.

D. Decay of the Long-lived State of Bi^{210} to Po^{210}

The 5-day state of Bi^{210}, RaE, decays principally by beta emission to Po^{210}. The beta decay energy of RaE is 1.165 ± 0.005 Mev.[3] Since the long-lived state of Bi^{210} was determined to lie 25 ± 40 kev below RaE, the long-lived Bi^{210} should be unstable with respect to beta decay to Po^{210} by 1.14 ± 0.05 Mev. Even with a large spin change involved in the beta decay, such a large disintegration energy indicated a good possibility of observing the beta decay.

1. Experimental procedure. -- A sample of the long-lived isomer of Bi^{210} was produced by a prolonged neutron bombardment on a slug of bismuth metal. The bismuth was dissolved and purified, then allowed to stand until all of the RaE had decayed. The Po^{210} was then removed by repeated precipitations of tellurium carrier with stannous chloride.

The purified bismuth fraction as obtained by this author consisted of ~50 grams of BiOCl containing ~10^5 alpha counts per minute of the long-lived bismuth activity. As an added precaution the BiOCl was dissolved in HCl, and further precipitations of tellurium carrier with stannous chloride were made to ensure the removal of all of the Po^{210}.

The experiments to determine the amount of branch decay of the long-lived Bi^{210} to Po^{210} were carried out in the following fashion. The purified bismuth (in HCl solution) was allowed to stand for a period of time sufficient to allow an easily detectable amount of Po^{210} to grow in. The solution, after standing, was made 3-4 N in HCl. A known amount of Po^{208} was added as a tracer so that the chemical yield of polonium could be determined by the amount of Po^{208} that went along with the Po^{210}. Ten to twenty milligrams of tellurium carrier was then added as tellurious acid. The tellurium was precipitated as the metal by the addition of a few milliliters of a $SnCl_2$ solution, the polonium being carried along. The precipitate was allowed to settle and then removed by centrifuging. The precipitate of tellurium (after washing with 3 N HCl) was dissolved in a few drops of concentrated HNO_3. About 5 ml of

6 N HCl were added, and the tellurium was again precipitated by saturating the solution with sulfur dioxide, the polonium now remaining in solution. The solution was then extracted twice with equal volumes of 20% tributyl phosphate in dibutyl ether. The polonium was back extracted from the organic phase with concentrated HNO_3. The HNO_3 fraction was evaporated to dryness, and the residue was taken up in ~10 ml of dilute H_2SO_4 solution (6 ml conc H_2SO_4/100 ml solution). This solution was electrolyzed in exactly the same manner as that described for the deposition of the bismuth samples. The polonium was deposited as a weightless sample onto a platinum disk.

The polonium sample was then placed in an alpha counter and counted at 52% geometry to determine the gross alpha activity of the polonium. The sample was next counted in an alpha differential pulse height analyzer to determine from the ratio of Po^{210} to Po^{208} in the sample how much Po^{210} was in the entire bismuth solution at the time of milking:

$$\text{Amt. of } Po^{210} \text{ in bismuth solution} = \frac{\text{Amt. of } Po^{210} \text{ in sample}}{\text{Amt. of } Po^{208} \text{ in sample}} \times \text{Amt. of } Po^{208} \text{ added.}$$

Furthermore, from the amount of Po^{210} that was present in the bismuth solution and the time allowed for growth, one can calculate how much Po^{210} activity would be present at equilibrium from the following relationship:

$$A = A^*(1 - e^{-\lambda t}),$$

where

$A = Po^{210}$ activity in solution at the time of milking,

$A^* = Po^{210}$ activity in solution at equilibrium,

λ = the decay constant of Po^{210},

t = time allowed for the Po^{210} to grow in.

2. Results. -- In all, three polonium milks were carried out on the solution of the long-lived Bi^{210}. The results are shown in Table 3.

Table 3

Milking of Po^{210} from Long-lived Bi^{210}

No. of milk	A(c/min)	t(days)	A^*(c/min)
1	172	199	272
2	121	83	356
3	116	82	344

It was decided to discard the results of the first milk for the following reason. Insufficient Po^{208} tracer was used, resulting in poor counting statistics in the pulse analysis and leading to unreliable values for A and A^*. If proper corrections for background and overlapping of the peaks could have been made, the value for A^* would have approached more closely those obtained in the second and third milkings. Statistics in the second and third milks were much better. Taking an average of milks two and three, one obtains

$$A^*_{av.} = 350 \text{ c/m}$$

for the amount of Po^{210} that would be present in the bismuth solution at equilibrium.

The total alpha activity of the bismuth solution was determined both before and after the milking experiments were carried out. The results are shown in Table 4.

Table 4

Total Alpha Activity of the Bismuth Solution

		Alpha counts/min.
Before milkings	=	9.71×10^4
After milkings	=	9.45×10^4
Average	=	9.58×10^4

In the determination of the alpha activities of the various samples from these experiments, all of the samples were counted at the same geometry (52%). Therefore, in order to find the ratio of the rate of decay of the long-lived Bi^{210} to Po^{210} to its rate of decay by alpha emission to Tl^{206}, one need only to divide the number A^* by the total alpha activity of the bismuth:

$$\frac{\text{Rate of decay to } Po^{210}}{\text{Rate of decay to } Tl^{206}} = \frac{350}{9.58 \times 10^4} = \frac{1}{273}.$$

In other words, one nucleus undergoes decay to Po^{210} for about every 270 alpha decays that the long-lived Bi^{210} state undergoes.

3. Discussion. -- Since we cannot say with certainty whether the long-lived Bi^{210} or RaE is the ground state, there remain two possible paths by which the long-lived isomeric state might decay to Po^{210}.

If the long-lived Bi^{210} is actually the ground state, then the only possible mode of decay to Po^{210} would be by beta emission. Since the half-life of the long-lived Bi^{210} for alpha emission is $\sim 10^6$ years, the half-life for beta decay would be $\sim 2.7 \times 10^8$ years.

If, on the other hand, RaE is the ground state, the long-lived state may decay to Po^{210} in two ways. It may still decay by direct beta decay, or it may first undergo isomeric transition to RaE with subsequent beta decay of RaE to Po^{210}. Still another possibility is a combination of both these two modes.

E. Search for Long-lived Bi^{210} in the Decay of RaD (Pb^{210})

As was pointed out in a previous section, comparison of the alpha decay energies for the long-lived Bi^{210} and for RaE indicates that the long-lived state lies 25 ± 40 kev below RaE. Since RaD decays to RaE with a total decay energy of 65 kev, RaD should be unstable with respect to the long-lived state of Bi^{210} by 90 ± 40 kev. Therefore, it is energetically possible for the long-lived Bi^{210} to be formed in the beta decay of RaD. Because of the very long life of this state of Bi^{210}, the only place it might be possible to see it as a decay product of RaD would be in uranium ores, where both the RaD and the long-lived Bi^{210} would be present in equilibrium with the U^{238} parent. Accordingly, the bismuth fraction was removed from a sample of uranium ore and examined for the presence of the long-lived Bi^{210}.

1. Experimental procedure. -- A 50 gram sample of uranium ore (~49% U_3O_8) was taken and ground into a fine powder. The ore was then digested in hot concentrated HNO_3 for several hours. The mixture was filtered, and the residue was taken and digested once again in concentrated HNO_3 and finally in aqua regia. The three filtrates were then combined.

To remove most of the nitrate ion from the solution excess NH_4OH was added, the mixture filtered, the filtrate discarded and the precipitate dissolved in HCl. This step was repeated. The acidity of the resultant HCl solution was properly adjusted, and it was then saturated with H_2S and allowed to stand overnight.

The sulfide precipitate was removed by centrifuging. The precipitate was then dissolved in HNO_3. The nitrate ion was removed by twice adding excess NH_4OH, filtering and dissolving the precipitate in HCl. The resultant HCl solution was again saturated with H_2S and allowed to stand for several hours.

The sulfide precipitate was removed by centrifuging, and then dissolved in HNO_3. The solution was diluted, and H_2SO_4 was added to precipitate $PbSO_4$, thus carrying down almost all the RaD. The $PbSO_4$ was centrifuged off and discarded.

To the remaining solution was added 5 mg of inert lead carrier. The solution was then made ammoniacal and excess ammonium carbonate was added (to keep the uranium still remaining in solution). The solution was saturated with H_2S and allowed to stand.

The sulfide precipitate was centrifuged off and washed thoroughly with a saturated $(NH_4)_2CO_3$ solution. The precipitate was dissolved in HNO_3. This solution contained the bismuth fraction of the ore.

The last traces of RaD were removed in the following manner. The dilute HNO_3 solution containing the bismuth fraction was run through a column of Dowex-50 cation resin, both the lead and the bismuth sticking to the column. After being washed with water, the column was eluted with 1 N HCl, most of the bismuth coming off in

the first fraction, while the lead lags far behind.

The bismuth fraction was taken and evaporated to dryness. The beta activity of this sample was followed and showed only the 5-day RaE half-life, indicating that a good separation had been effected. The amount of RaE present at the time at which the bulk of the RaD was removed was 5.5×10^6 disintegrations per minute.

The bismuth sample from the ore was allowed to stand until all of the RaE had decayed. It was then necessary to remove all the Po^{210} from the sample in order to be able to detect a small amount of the long-lived bismuth alpha activity. This was done in the following manner. The sample was taken up in HCl. An aliquot was taken, evaporated to dryness and taken up in 1 ml of concentrated HCl. The concentrated HCl solution was run through a column of Dowex-2 anion resin. The column was then washed with 10 ml of concentrated HCl. The two put-through solutions were combined. In this operation about 70-80% of the bismuth comes through the column while almost all of the polonium sticks to the resin. (This was determined by previous tracer experiments.) This process was repeated once again. The final solution was evaporated to dryness on a platinum disk and checked for gross alpha activity. There were 3 counts per minute at 52% geometry on the platinum plate.

The sample was then counted in an alpha pulse analyzer to determine what fraction, if any, of the alpha activity was due to the long-lived Bi^{210}. All of the alpha particles from the sample had an energy corresponding to those of Po^{210}. The limits of detection were such that the total bismuth fraction taken from the ore must have contained less than 5-6 disintegrations per minute of the

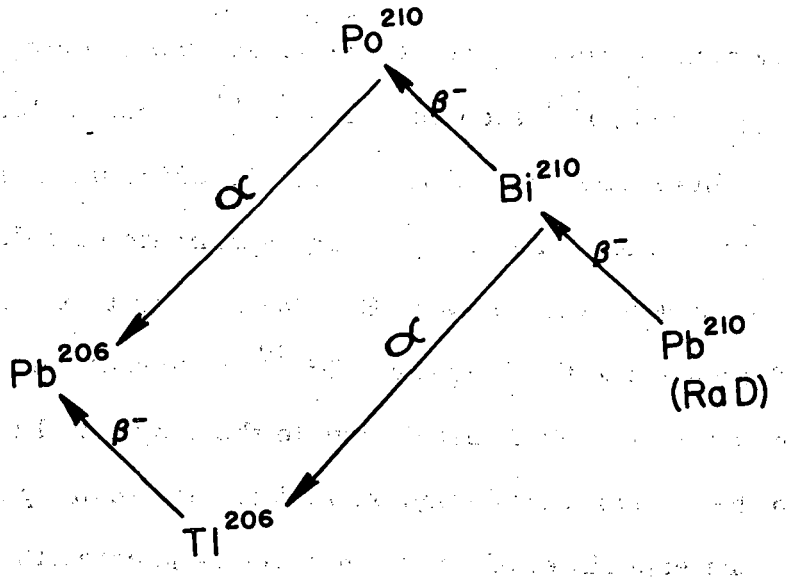
long-lived Bi^{210}. Therefore, we can deduce that the ratio of the amount of long-lived Bi^{210} to that of RaE in the uranium ore was less than 10^{-6}. Correspondingly, the fraction of the decay of RaD which results in the long-lived Bi^{210} is less than 1 part in 10^6.

F. Summary and General Discussion

It may be well to summarize briefly here what is now known about the long-lived state of Bi^{210}. The mass assignment is confirmed to be 210. The half-life of this state is $\sim 10^6$ years. The principal mode of decay is by alpha emission, the alpha decay energy being 5.031 ± 0.020 Mev. The long-lived Bi^{210} also decays to Po^{210}, one nucleus undergoing decay in this fashion for about every 270 which undergo alpha decay. Comparison of alpha decay energies indicate that the long-lived state lies 25 ± 40 kev below RaE. This means that RaD is unstable with respect to the long-lived Bi^{210} by 90 ± 40 kev. However, the amount of RaD that decays to the long-lived Bi^{210} compared to the amount that decays to RaE is less than 1 part in 10^6.

Since the experimental error involved is greater than the actual difference in energy between RaE and the long-lived state, we cannot say with certainty which is the ground state of Bi^{210}. However, discussion of the various possibilities may throw some more light on the long-lived state of Bi^{210}.

For the purposes of the discussion it would be well to remember that RaD decays to RaE with a 22-year half-life, and that RaE decays to Po^{210} with a 5-day half-life. Also, RaE is considered to have zero spin and odd parity.[3, 5] Figure 2, showing the decay paths of Bi^{210} and genetically related nuclides, may help the reader in the following discussion.



MU-5984

Fig. 2. Decay paths of Bi^{210} and genetically related nuclides.

Let us first assume that the long-lived state does lie 25 kev below RaE. This would mean that the decay of the long-lived Bi^{210} to Po^{210} proceeded by direct beta decay. With a beta energy of 1.14 Mev and a half-life of ~2.7×10^8 years, this decay would have a log ft value of ~18.3. There is only one other known nuclide that undergoes beta decay with both an energy and a half-life of this same order of magnitude. Potassium 40 has a maximum beta energy of 1.33 Mev and a partial beta decay half-life of 1.3×10^9 years,[5] giving it a lot ft value of 18. It is known that the beta decay of K^{40} involves a spin change of 4. Thus, it would seem by analogy that the beta decay of the long-lived Bi^{210} would involve a spin change of 4 or possibly greater. Since Po^{210}, being an even-even nucleus, has a spin of zero, the long-lived Bi^{210} would have a spin of 4 or greater.

Furthermore, one would conclude from the data at hand that the beta decay of RaD to the long-lived Bi^{210} must have a half-life greater than 22×10^6 years. Along with a maximum beta energy of 90 kev, this would entail a log ft value greater than 13.6. A log ft value this large would indicate, at the very least, a spin of 3 or greater for the long-lived Bi^{210}.

Still further, the absence of the long-lived isomer in uranium ores means that the half-life for the isomeric transition from RaE to the long-lived state must be greater than 1.4×10^4 years. Using the Weisskopf formula,[6] one can estimate the lifetimes for various types of 25 kev gamma emissions in Bi^{210} (neglecting internal conversion). For 25 kev M3, E4, and M4 transitions, respectively, one obtains $\tau_\gamma(M3) \sim 0.3$ yr., $\tau_\gamma(E4) \sim 2 \times 10^5$ yrs., and $\tau_\gamma(M4) \sim 4 \times 10^7$ yrs.

Taking into account internal conversion, which would speed up the transition, any transition involving a spin change of less than 3 seems completely out of the question. It would be necessary for the transition to have an energy of less than 5 kev before even an M3 transition would have a chance of giving anywhere near a long enough lifetime. With a lifetime greater than 1.4×10^4 yrs. (and again neglecting internal conversion) an E4 transition would have to have an energy of <33 kev, and an M4 transition an energy of <60 kev. The long-lived state, lying 25 kev below RaE, easily fits into this range. Since RaE is a 0- state, this is further indication that the long-lived Bi^{210} has a spin of 4 or possibly greater.

On the other hand, suppose the long-lived state lies above RaE. It is not likely to be more than 15 kev above RaE. If the decay to Po^{210} went by direct beta emission, there would again be a log ft value of ~18.3, and the discussion previously based on this condition would still apply. If, however, the decay of the long-lived state actually went by way of initially undergoing isomeric transition to RaE, then the direct beta decay to Po^{210} would involve a log ft value of greater than 18.3. This in turn would indicate a spin change ≥ 4 in going from the long-lived Bi^{210} to Po^{210}.

Consider, too, the isomeric transition itself. It would have to have a mean lifetime equal to or greater than $\sim 4 \times 10^8$ yrs. For simple gamma ray emission the following lifetimes would apply for a 1 kev gamma ray: $\tau_\gamma(E3) \sim 6 \times 10^6$ yrs., $\tau_\gamma(M3) \sim 10^9$ yrs., and $\tau_\gamma(M4) \sim 3 \times 10^{17}$ yrs. Taking into account internal conversion, here again it seems highly unlikely that a spin change of less than 4 occurs between the long-lived Bi^{210} and the RaE state.

This discussion of the various possibilities still open shows that whether the long-lived Bi^{210} lies above or below RaE, present information strongly indicates that the long-lived state of Bi^{210} has a spin equal to or greater than 4.

Bismuth 210 has 83 protons and 127 neutrons, both being just one beyond closed shells. RaE is designated as a 0- state. Applying Nordheim's rules,[7] and making use of the fact that the odd proton in Bi^{209} is in the $h_{9/2}$ state, one would guess that the most likely configuration for the RaE state would be $h_{9/2} - g_{9/2}$. With the two states of Bi^{210} lying so close together, it is probable that their configurations differ merely by transferring one of the odd nucleons (either proton or neutron) to a different but close lying level. Possibilities for the configuration of the long-lived state would be $h_{9/2} - g_{7/2}$ or $f_{7/2} - g_{9/2}$, both of which would give $1 < I \leq 8$ for the spin by Nordheim's rules. Also possible is $h_{9/2} - i_{11/2}$, which would give $1 < I \leq 10$. All three of these possibilities would give the long-lived state of Bi^{210} odd parity.

RaE with an alpha decay energy of 5.056 Mev has a partial half-life for alpha decay of 2.7×10^4 years.[5] The long-lived state with a decay energy of 5.031 Mev has an alpha half-life of $\sim 10^6$ years. Both of these half-lives are much longer than would be predicted by simple alpha decay theory. However, in the emission of an alpha particle from Bi^{210} both the closed shell of 82 protons and the closed shell of 126 neutrons are broken open. It would not be surprising to find that breaking the closed shells open causes quite a hindrance in the decay. There is no information available whereby one can predict how far the half-life for alpha decay should depart from simple

theory in such a case.

In conclusion, it appears that the most likely designation for the long-lived state of Bi^{210} is 4-. However, a spin of greater than 4 still remains a distinct possibility. A spin of less than 4 or even parity for the long-lived state both seem highly unlikely on the basis of present information.

II. RELATIVE YIELDS IN THE FORMATION OF NUCLEAR ISOMERS

A. Introduction

In studying the fission of bismuth by high energy protons W. F. Biller[8] had occasion to measure the yields of several nuclides that possessed isomeric states. In all cases where it was possible to determine or to estimate primary yields, formation of the isomer with the greater spin, whether it was the ground state or an excited state, was favored over that of the state with the lower spin, sometimes to the apparent exclusion of the latter.

Not very much has appeared in the literature concerning the yields of isomers in nuclear reactions, especially high energy reactions. Most of what does appear is based on the formation of nuclear isomers by means of the (n, γ) reaction at low neutron energies.[9,10] The articles discussing formation of isomers in other types of reactions[11-15] give no clear-cut picture of what might happen as one increases the energy involved in the reaction beyond the range of 5-20 Mev.

In their 1949 review article on nuclear isomerism E. Segrè and A. C. Helmholz[10] presented a possible clue to what one might expect in the formation of isomers at high energies. In discussing

the different yields of some isomers formed by the (n, γ) reaction at different neutron energies, they made the following suggestion: "If the energy of the neutrons captured is increased so that capture occurs over many levels of all possible angular momenta, one might expect that the influence of the level in which the capture occurs will be washed out, and in the limiting case only the statistical weights (2I + 1) of the isomeric states themselves should determine the formation cross section."

In the cases discussed, the neutron energies never were high enough so that the capture could take place over a very wide spread of angular momentum states. Therefore, their idea was not subject to test.

One, however, can extend this reasoning to nuclear reactions in general. The key point is the formation of the final nucleus in highly excited states covering a wide range of angular momentum or spin values. If this condition is achieved, then the relative number of nuclei ending up in each of the isomeric states should be determined simply by their respective statistical weights, (2I + 1), regardless of how the final excited nucleus was formed. In other words we might then write:

$$\frac{\sigma_m}{\sigma_g} = \frac{2I_m + 1}{2I_g + 1},$$

where σ_m and I_m are the cross section and the spin for the metastable excited state, and σ_g and I_g are the corresponding terms for the ground state of the nuclide in question.

If this line of reasoning is valid then $(2I_m + 1)/(2I_g + 1)$ would be a limiting value for the ratio σ_m/σ_g. If one were to plot σ_m/σ_g

versus energy (of the reaction producing the isomers), the curve would approach $(2I_m + 1)/(2I_g + 1)$ asymptotically. The ratio σ_m/σ_g might approach the limiting value from either above or below depending upon the spins of the target nucleus, the bombarding projectile, and the isomers. But it would never cross the line representing the value $(2I_m + 1)/(2I_g + 1)$ and would only reach this value when the reaction energy was high enough to form the excited final nucleus over a wide range of spin states.

In high energy fission or spallation many particles are "boiled out" of the compound nucleus either before or during the actual formation of the excited final nucleus. Even if these particles coming out of the compound nucleus carry off relatively little angular momentum, the fact that there are several of these particles gives rise to a wide range of possible spin states in which the final excited nucleus may be formed. Thus, we might expect that the relative yields of isomers of a particular nuclide formed by high energy fission or spallation would be in proportion to their respective statistical weights.

The results found by Biller for the high energy fission of bismuth do not bear out this hypothesis. However, high energy fission and spallation reactions are rather complex, and one cannot always be certain that he is determining primary yields. The reasoning employed in the above paragraphs would not apply to the yields of isomers formed in decay processes.

A better test of the hypothesis could be provided by a study of the formation of a pair of isomers by the simplest type of nuclear reaction X(a, b)Y. This may be written in more detailed fashion as:

1. $a + X \rightarrow C^*$

2. $C^* \rightarrow Y^* + b$

3. $Y^* \rightarrow Y^m + \gamma$

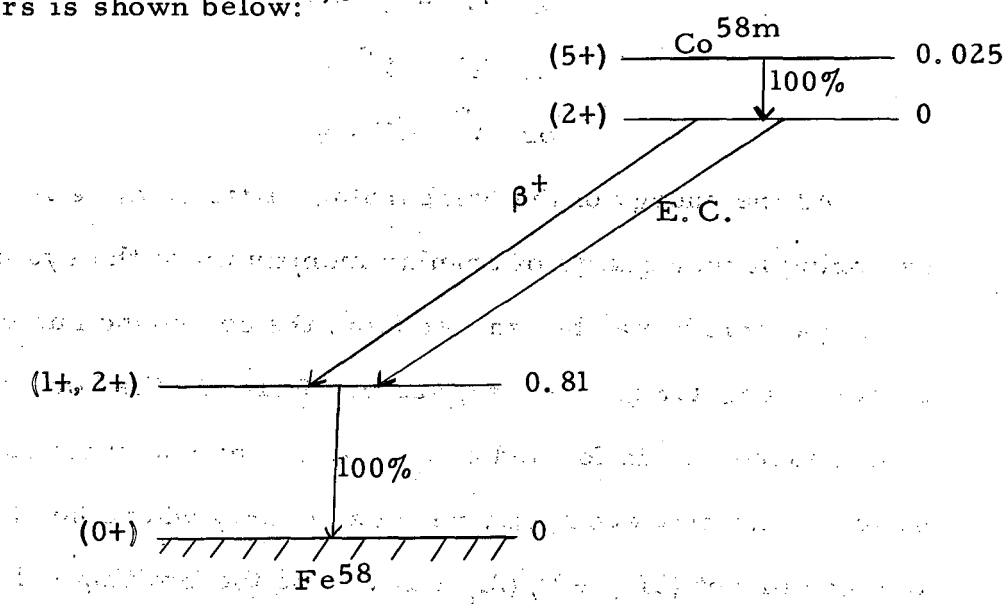
or $Y^* \rightarrow Y + \gamma$.

As the energy of the bombarding particle, a, is increased, a can bring larger quanta of angular momentum to the system. Likewise, particle b may be emitted from the compound nucleus with a larger ℓ (angular momentum) value. Thus, as the bombarding energy is increased, Y^* is formed over a wider and wider range of spin states. One may eventually reach an energy where he can tell whether or not $(2I_m + 1)/(2I_g + 1)$ will be the limiting value for the ratio σ_m/σ_g.

For the purpose of this thesis the isomers of Co^{58} were chosen for study. This isomer pair has several advantages. It can be produced by a simple (α, n) reaction on manganese, which has only one stable isotope. The decay scheme is simple and well known, and the half-lives involved make it easy to work with. A major advantage, as will be pointed out later, is the fact that one can avoid the necessity of estimating counting efficiencies.

For the purpose of accurate analysis of the data, it was necessary to know accurately the half-lives of the two Co^{58} isomers, especially that of the shorter-lived Co^{58m}. Accordingly, these were checked by this author. Two careful independent determinations of the half-life of Co^{58m} each yielded a value of 8.96 hours for the half-life. This checks very well with the latest quoted figure in the literature, 9.0 ± 0.2 hrs.[16] A check on the half-life of the long-lived Co^{58} gave a value of 72 days, which is just what was reported by

Livingood and Seaborg.[5,17] The decay scheme for the two Co^{58} isomers is shown below:[3,5]



Goldhaber and Hill[3] assign a spin of 2+ to the ground state of Co^{58} based upon log ft values of the decays to the excited state of Fe^{58} and the subsequent 810 kev transition to the ground state of Fe^{58}. They further assign a spin of 5+ to Co^{58m} on the basis of the lifetime and conversion coefficients of the 25 kev transition. Comparison with Co^{60} also seems to bear out these assignments. Therefore, for Co^{58}, $(2I_m + 1)/(2I_g + 1) = 2.2$.

B. Experimental Procedures

1. Bombardment procedures. -- The yields of the Co^{58} isomers were studied by bombarding manganese with alpha particles in the Berkeley 60-inch cyclotron. In all the bombardments the target material used was $MnSO_4 \cdot H_2O$ powder.

In most of the bombardments a "thick target" was used. In these cases the $MnSO_4 \cdot H_2O$ powder was pressed wafer-like onto a grooved aluminum backing plate. The powder was then covered with

thin platinum and tantalum foils to keep it in place. In the last few bombardments a "thin" target was desired. A paste was made of the target material. This was then spread on an aluminum foil and dried. The foil with the thin layer of target material was then placed on the aluminum backing plate and covered with thin foils to keep it in place.

The energy of the bombarding alpha particles was varied by placing different thicknesses of aluminum foil in front of the target.

2. Chemical separation. -- In all of the bombardments the cobalt fraction was separated chemically before counting. The chemical separations were carried out in the following manner. Because some MnO_2 was always formed in the bombardment, the target material was dissolved in warm dilute HCl to which some Na_2SO_3 was added. The solution was then neutralized with KOH, and then made 2-3 N in acetic acid. A few milligrams of cobalt carrier was added, and $K_3Co(NO_2)_6$ was then precipitated by the addition of a few milliliters of a saturated KNO_2 solution. The precipitate was centrifuged and washed two or three times. Then the precipitate was dissolved in warm concentrated HCl and manganese carrier was added. The solution was neutralized with KOH, and the cobalt was reprecipitated as $K_3Co(NO_2)_6$ in the manner described above. The precipitate was again centrifuged and washed. It was then slurried on to a depressed aluminum disk and dried. The disk was then mounted on an aluminum plate for counting.

3. Counting methods. -- Co^{58m} decays 100% by a 25 kev isomeric transition to Co^{58}. Co^{58} decays by electron capture and by positron emission to an excited state of Fe^{58}, which then decays

instantaneously to the ground state of Fe^{58} by emission of an 810 kev gamma ray.

It is possible to count the radiations from Co^{58} to the exclusion of those from Co^{58m}. The decay curve for the Co^{58} alone will show a 9-hour growth tailing into 72-day half-life. It is possible to resolve out the two components of the curve and to obtain the amounts of both Co^{58m} and Co^{58} present at the end of the bombardment. The method used to effect this resolution will be explained in the next section.

In the two bombardments at low energies (9.5 Mev and 11.3 Mev) the cobalt fraction was counted in a chlorine tube Geiger-Müller counter. The radiations from the Co^{58m} were stopped by placing an aluminum absorber 222 mg/cm^2 thick between the sample and the counter window. Thus, only the Co^{58} activity was observed.

In the bombardments at higher energies Co^{57} was formed along with Co^{58m} and Co^{58}. Therefore, a different counting method had to be used. Accordingly the gamma rays from the cobalt fractions were counted with a NaI crystal that was connected to a differential pulse height analyzer. This provided a spectrum of the gamma rays of the cobalt fraction, from which the 810-kev gamma rays of Co^{58} could easily be resolved. The intensity of the Co^{58} activity was obtained by merely integrating under the 810 kev gamma ray peak.

These methods avoid the tricky problem of estimating the counting efficiencies of both Co^{58m} and Co^{58}. Here both activities are effectively normalized to the same counting efficiency.

C. Treatment of Data

The first problem in handling the data obtained is to determine the amounts of both Co^{58m} and Co^{58} initially present at the end of the bombardment. The excited state decays 100% by isomeric transition to Co^{58} which then undergoes electron capture and β^+-decay. If we use the subscripts 1 and 2 to denote the states Co^{58} and Co^{58m}, respectively, and A^o is the activity at the end of the bombardment and λ the decay constant, then the equations giving A_1 and A_2, the activities at time t after bombardment are:[18]

$$A_2 = A_2^o \, e^{-\lambda_2 t},$$

and

$$A_1 = A_1^o e^{-\lambda_1 t} + \frac{\lambda_1}{\lambda_1 - \lambda_2} \, A_2^o \, (e^{-\lambda_2 t} - e^{-\lambda_1 t}).$$

In these experiments A_1 alone was determined directly at various times after bombardment. From these data A_1^o and A_2^o were obtained in the following fashion.

Since $\lambda_2 > \lambda_1$, let $-\beta = \lambda_1/(\lambda_1 - \lambda_2)$ and the second equation above may be rewritten as follows:

$$A_1 = (A_1^o + \beta A_2^o) \, e^{-\lambda_1 t} - \beta A_2^o \, e^{-\lambda_2 t}.$$

Multiplying this equation through by $e^{\lambda_1 t}$ gives

$$A_1 e^{\lambda_1 t} = (A_1^o + \beta A_2^o) - \beta A_2^o \, e^{(\lambda_1 - \lambda_2)t}.$$

A_1 is a measured quantity and λ_1, λ_2, and β are known. If $A_1 e^{\lambda_1 t}$ is plotted against $e^{(\lambda_1 - \lambda_2)t}$, a straight line of the form $y = a + bx$ is obtained, where $a = (A_1^o + \beta A_2^o)$ and $b = -\beta A_2^o$. The quantities a and b can be determined accurately by the least squares method, and from these A_1^o and A_2^o may easily be obtained.

To obtain the relative cross sections σ_2 and σ_1 one need only

apply the following equations:[10]

$$A_2^o = \phi\sigma_2(1 - e^{-\lambda_2 t}),$$

$$A_1^o = \phi(\sigma_1 + \sigma_2)(1 - e^{-\lambda_1 t}) + \frac{\lambda_1}{\lambda_1 - \lambda_2}\phi\sigma_2(e^{-\lambda_1 t} - e^{-\lambda_2 t}),$$

where t is the duration time of the bombardment and ϕ represents the flux density of the bombarding particles (and cancels out in obtaining σ_2/σ_1).

D. Results

The majority of the bombardments to produce the Co^{58} isomers by the (α, n) reaction on Mn^{55} were carried out on thick targets. The alpha particles were degraded in energy in passing through the target material. Therefore, instead of obtaining the ratio σ_m/σ_g at the particular bombarding energy, what was actually found was the ratio

$$\frac{\bar{\sigma}_m}{\bar{\sigma}_g} = \frac{\int_{E_i}^{E_f} w\sigma_m dE}{\int_{E_i}^{E_f} w\sigma_g dE}.$$

Here σ represents the cross section at a given alpha particle energy, E; the limits of integration are from the energy of the incident alpha particles, E_i, to the energy the alpha particles have after passing completely through the target, E_f; and w is a weighting factor corresponding to the different amounts of target material that the alpha particles of different energies effectively see. Obviously, if the ratio $\bar{\sigma}_m/\bar{\sigma}_g$ crosses the "limiting" value of $(2I_m + 1)/(2I_g + 1)$, then somewhere in the range between E_f and E_i the ratio σ_m/σ_g crosses this "limiting" value.

Below is a table of values of $\bar{\sigma}_m/\bar{\sigma}_g$ for the formation of Co^{58} isomers by the (α, n) reaction on Mn^{55} at various bombarding energies E_i. In all the cases in Table 5, $E_f = 0$. Also included is a point determined at 17 Mev by K. Strauch.[19]

Table 5

Values of $\bar{\sigma}_m/\bar{\sigma}_g$ for Co^{58} at Various E_i

E_i (Mev)	$\bar{\sigma}_m/\bar{\sigma}_g$
9.5	0.98
$\leq$11.3	1.24
13.6	1.44
17	1.7
21.0	1.96
23.6	2.08

As is seen in Table 5 the ratio $\bar{\sigma}_m/\bar{\sigma}_g$ has not yet gone beyond 2.2 at E_i = 23.6 Mev. However, since the cross section for the (α, n) reaction reaches its peak in this case at ~15 Mev and then falls off, the ratio $\bar{\sigma}_m/\bar{\sigma}_g$ does not give a complete picture of what may be happening at higher alpha energies because the major contribution to $\bar{\sigma}_m/\bar{\sigma}_g$ comes from the region around the cross section peak. Accordingly, an attempt was made to reduce the thickness of the targets. The results of the bombardments with thinner targets are listed below in Table 6.

Table 6

$\bar{E}_i$ (Mev)	E_f (Mev)	$\bar{\sigma}_m/\bar{\sigma}_g$
23.7	~11	2.3 ± 0.1
23.9	~19	2.9 ± 0.2
23.7	~23	3.7 ± 0.3

The last three bombardments show beyond any doubt that the ratio σ_m/σ_g for Co^{58} isomers does go beyond 2.2 for sufficiently high alpha energies.

E. Discussion

It is plainly seen from the last few bombardments on Mn^{55} to form the Co^{58} isomers that the ratio $(2I_m + 1)/2I_g + 1)$ does not act as a limiting value to the ratio σ_m/σ_g. Is there any way, then, that we can predict what might happen to the relative yields of isomers as we increase the bombarding energies? A detailed quantitative analysis of the problem would be unfeasible here, because of the difficulty in calculating such factors as barrier penetrabilities and because of the lack of sufficient knowledge of level densities and spin states in excited nuclei. However, a qualitative treatment may be applied with some success.

Let us consider the formation of nuclear isomers by the simple reaction X(a, b)Y. It is convenient to break the reaction into three principle steps:

1. Formation of the compound nucleus, C^*,

$$X + a \rightarrow C^*.$$

2. Break-up of the compound nucleus to give the excited residual nucleus, Y^*,

$$C^* \rightarrow b + Y^*,$$

3. De-excitation of the excited residual nucleus by successive gamma ray emission ending in either of the isomeric states,

$$Y^* \rightarrow Y^m + y,$$

or $$Y^* \rightarrow Y + \gamma.$$

Let us consider step 1 first. When a bombarding particle, a, is absorbed into a target nucleus, X, it may bring into the system a certain amount of angular momentum, ℓ. We may write the cross section for the capture of a particle with angular momentum, ℓ, as follows:

$$\sigma_c^{\ell} = (2\ell + 1)\, \pi \lambda\!\!\!^{-2} T_{\ell}(E),$$

where $T_{\ell}(E)$ is the factor representing the penetration of the potential barrier (coulombic plus centrifugal). The higher the ℓ value of the incoming particle, the harder it is for the particle to penetrate the potential barrier. However, as one increases the energy of the incoming particle above that of the barrier height, $T_{\ell}(E)$ approaches one. Then as we increase the bombarding energy, reactions of higher ℓ values become more important because of the factor $(2\ell + 1)$. Since the spin, J, of the compound nucleus is the resultant of the <u>vector</u> sum of the spin of the target nucleus, I_x, the spin of the incoming particle, I_a, and ℓ; the larger the value of ℓ the wider the range of values J may take on.

Now consider the incoming particle bringing in a particular value of ℓ. The compound nucleus can have any spin possible by the

vector addition of I_x, I_a, and ℓ. However, the relative probability that the compound nucleus will be formed with the spin J will be proportional to the statistical weight of that state, $(2J + 1)$.

Thus, as we increase the energy of the incoming particle, reactions involving larger ℓ values become more and more important. The compound nucleus is formed over a wider and wider range of spin states with the higher spin states being somewhat favored because of their greater statistical weights.

Step 2, the break-up of the compound nucleus to give the excited residual nucleus, Y^*, can be treated in a similar manner. In leaving the compound nucleus, particle b must also penetrate a potential barrier. The greater the excitation energy of the compound nucleus, the greater the amount of energy that b can carry off, and also the greater the amount of angular momentum, ℓ', that it may leave with. The spin of the excited residual nucleus, I_{Y^*}, is the resultant of the vector sum of the spins of the compound nucleus and the particle b and the angular momentum carried off, ℓ'. Therefore, as the excitation energy of the compound nucleus is increased, the excited residual nucleus can be formed over an increasing range of spin states. Here again formation of the higher spins is favored because of their greater statistical weights.

Combining steps 1 and 2 the following result is obtained. As the energy of the bombarding particle is increased, the excited residual nucleus is formed over a wider and wider range of spin states. The formation of the states with the higher spins will be favored because of their greater statistical weights.

Now to consider step 3, the de-excitation of the excited residual nucleus by successive gamma ray emission. It is known that nuclei, even when excited to fairly high energies, in the process of de-excitation by gamma ray emission do so by the emission of a relatively small number of successive gamma rays. Furthermore, these gamma rays are of low multipole order, dipole or at most quadrupole. Therefore, one would not expect very great spin changes to occur in the de-excitation of Y^*. High spin states of Y^* should decay predominantly to the high spin isomer. Low spin states of Y^* should decay predominantly to the low spin isomer. Here, too, we have an additional factor to favor the formation of the high spin state. If an excited state has a choice of decaying to two lower states, all other factors contributing to the half-lives being the same, the decay to the state with the higher spin is slightly favored because of its greater statistical weight.

If we now combine the three steps, we obtain the following result. As we increase the energy of the bombarding particle, Y^* is formed over an increasing range of spin states, the formation of the higher spin states being favored over those with low spin. This in turn means that the amount of the high spin isomer finally formed will continue to increase relative to the amount of the low spin isomer. There would be <u>no</u> particular limiting value that the ratio σ_m/σ_g would approach at high energies.

Just how rapidly σ_m/σ_g will approach and cross the value $(2I_m + 1)/(2I_g + 1)$ as the bombarding energy is increased will, of course, depend largely on the spins of the target nucleus of the bombarding projectile, and of the isomeric states to be formed along

with the potential barrier to be penetrated. Another factor that may enter is the relative proportions of the different spin states lying between the upper state and the continuum in the final nucleus, Y. Still another factor would be the existence of a state lying close to the pair of isomeric states, and feeding predominantly into one of them. These effects would all serve to vary the rate at which σ_m/σ_g changes with energy.

Two bombardments were carried out by this author to produce 23-hr Hg^{197m_2} and 65-hr Hg^{197} by a (p, n) reaction on Au^{197}. Gold 197 has a spin of 3/2, while Hg^{197m_2} has a spin of 13/2 and Hg^{197} a spin of 1/2.[5] There is also a Hg^{197m_1} state lying between the other two states, possessing a spin of 5/2, and feeding almost instantaneously into the ground state Hg^{197}. The bombardments were carried out at proton energies of 10 and 20 Mev, respectively. The Hg^{197m_2} activity was followed by means of the 133 kev gamma ray which occurs in its decay to the ground state. The Hg^{197} activity was followed by means of the 77 kev gamma ray which occurs following the electron capture. The K x-rays of gold and mercury are in an energy range including the 77 kev peak, and it was estimated that about half of the electron capture decays of Hg^{197} produced K x-rays that counted in the 77 kev peak. After making corrections for x-rays and internal conversion of the gamma rays, it was found that at 10 Mev σ_{m_2}/σ_g was ~0.22, and at 20 Mev σ_{m_2}/σ_g was ~0.24. These data are in marked contrast to the results obtained for the Co^{58} isomers in this energy range, and demonstrate how the various factors mentioned above may affect the value of

σ_{m_2}/σ_g. In the case of the Hg^{197} isomers there are at least three factors present that would tend to retard the increase of the formation of the high spin isomer relative to the formation of the low spin isomer as one increases the energy, as compared to the case of the Co^{58} isomers. In the formation of the Hg^{197} isomers there is a greater overall spin change in going from the target nucleus to the high spin state while the overall spin change in going to the low spin state is still small. Also, the centrifugal potential for protons is greater than for alpha particles, making it more difficult for protons to bring in large angular momentum changes at these energies. Then, too, in Hg^{197} there is the presence of the 5/2 spin state which feeds directly into the low spin state. In addition to these factors, the difference in number and type of states in the region between the continuum and the low-lying isomeric states for the two nuclides, Co^{58} and Hg^{197}, may have an added effect. Hence, it is not surprising that in the Au^{197} (p, n) reaction one would have to go to higher energies than in the Mn^{55} (α, n) reaction to obtain the predominance of the high spin isomer in the product nucleus.

In the cases of spallation and high energy fission, the incoming particle brings in a great deal of energy, and many particles are boiled off before the final excited nucleus is formed. Each time a nucleon enters or is emitted, a spin change can take place, and with each spin change the proportion of high spin states formed should be increased relative to the number of low spin states formed. Thus, it would seem quite likely that in the formation of isomers by spallation and high energy fission the formation of the high spin

isomer would be highly favored over that of the low spin isomer, and that the ratio σ_m/σ_g would go well beyond the value of $(2I_m + 1)/(2I_g + 1)$.

III. ACKNOWLEDGMENTS

The author wishes to thank Dr. R. S. Livingston for the use of the Oak Ridge Calutron and Dr. D. C. Dunlavey for performing the alpha-electron coincidence experiment with the long-lived Bi^{210}. Thanks are due also to G. B. Rossi, Bart Jones, and the crew of the 60-inch cyclotron for their aid and cooperation in carrying out the bombardments for the cobalt experiments.

I would like to express my deepest gratitude to Professor I. Perlman, without whose inspiration and guidance this work could not have been accomplished.

IV. REFERENCES

1. H. M. Neumann, J. J. Howland, Jr., and I. Perlman, Phys. Rev. 77, 720 (1950)
2. H. Palevsky, D. J. Hughes, and C. Eggler, Phys. Rev. 83, 234 (A) (1951)
3. M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952)
4. W. J. Sturm and V. Johnson, Phys. Rev. 83, 542 (1951)
5. J. M. Hollander, I. Perlman, and G. T. Seaborg, Revs. Modern Phys. 25, 469 (1953)
6. M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951)
7. L. W. Nordheim, Revs. Modern Phys. 23, 322 (1951)
8. W. F. Biller, Ph. D. Thesis, University of California Radiation Laboratory Classified Report UCRL-2067 (Jan. 1953)
9. L. Seren, H. N. Friedlander, and S. H. Turkel, Phys. Rev. 72, 888 (1947)
10. E. Segrè and A. C. Helmholz, Revs. Modern Phys. 21, 271 (1949)
11. A. W. Fairhall, Massachusetts Institute of Technology Laboratory for Nuclear Science and Engineering Progress Report No. 131 (May 31, 1952)
12. L. Katz, L. Pease, and H. Moody, Can. J. Phys. 30, 476 (1952)
13. F. Boehm, P. Marmier, and P. Preiswerk, Helv. Phys. Acta 25, 599 (1952)
14. J. Goldenberg and L. Katz, Phys. Rev. 90, 308 (1953)
15. L. Katz, R. G. Baker, and R. Montabetti, Can. J. Phys. 31, 250 (1953)

16. C. J. Avery, K. C. Kaericher, and M. L. Pool, Phys. Rev. 87, 216 (A) (1952)

17. J. J. Livingood and G. T. Seaborg, Phys. Rev. 60, 913 (1941)

18. G. Friedlander and J. W. Kennedy, Introduction to Radiochemistry (J. Wiley and Sons, Inc., New York, 1949) p. 110

19. K. Strauch, Phys. Rev. 79, 487 (1950)