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# Long-Lived Isomer of RaE ($Bi^{210}$)†

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The mass assignment of a long-lived bismuth alpha activity previously reported as being an isomer of $Bi^{210}$ has been confirmed by means of electromagnetic mass separation. The half-life of the $Bi^{210}$ (long) is now reported to be $2.6\times10^6$ years. The alpha-decay energy of $Bi^{210}$ (long) has been redetermined and found to be $5.031\pm0.020$ Mev. Comparison with the alpha-decay energy of RaE places the $Bi^{210}$ (long) 25 kev below RaE with, however, an uncertainty of the same order of magnitude. $Bi^{210}$ (long) has measurable $\beta^-$ branching (1 part in 270) to give $Po^{210}$, and this gives a partial half-life of $7.0\times10^8$ years for this mode of decay. An unsuccessful search was made for $Bi^{210}$ (long) as a decay product of RaD ($Pb^{210}$), resulting in a lower limit of $2\times10^7$ years for the decay of RaD to this isomer. Also, an upper limit of $1.4\times10^4$ years was set on the half-life for the transition from RaE to $Bi^{210}$ (long). On the basis of decay data now available, the most probable designation for the $Bi^{210}$ (long) state is deduced to be 4− although a somewhat higher spin number could be possible.

## A. INTRODUCTION

WHEN bismuth ($Bi^{209}$) is irradiated with slow neutrons, a long-lived bismuth alpha emitter is noted after the 5.0-day RaE ($Bi^{210}$) has decayed and its alpha-emitting daughter $Po^{210}$ has been removed.[1] For a number of reasons it was thought to be $Bi^{210}$ and, therefore, an isomer of RaE: (1) the radioactivity was produced by slow neutrons on bismuth; (2) the alpha-particle energy (5.1 Mev) was about that expected for $Bi^{210}$ and not for $Bi^{208}$ or $Bi^{211}$; (3) the alpha emitter proved to be the parent of a $4.2\pm0.5$-min thallium beta emitter which seemed to be $Tl^{206}$ as well as could be discerned with the weak source available. No decay could be observed over an extended period of time and a lower limit of 25 years for the half-life could be set on the basis of this information. Actually a half-life of the order of $10^6$ years can be deduced from other considerations to be discussed.

It was deduced in the earlier report[1] on the basis of available decay data that the long-lived alpha emitter was the upper isomeric state of $Bi^{210}$. It will be seen that this conclusion must now be revised and that RaE is most likely the upper state although the uncertainties of the decay data upon which the decision is based are about the same as the energy difference. We shall designate the long-lived isomer simply as $Bi^{210}$ (long).

The present report also deals with a measurement of the $\beta^-$ branching of $Bi^{210}$ (long) and experiments which set lower limits of the decay periods for the isomeric transition and for the $\beta^-$ decay of RaD to $Bi^{210}$ (long). From these data it is possible to deduce that the probable spectroscopic state for $Bi^{210}$ (long) is 4−.

## B. CONFIRMATION OF THE MASS ASSIGNMENT OF $Bi^{210}$ (LONG)

A sample of neutron-irradiated bismuth was stored until the RaE had decayed and then was purified rigorously. The specific alpha activity was 4 disintegrations per minute per milligram of bismuth. This material was subjected to calutron isotope separation,[2] collecting the bismuth ions at positions corresponding to mass numbers 208, 209, and 210.

Samples were taken from each fraction and weighed after electrodeposition onto tared platinum disks. The disks then served as sources for alpha disintegration rate determinations. The results are shown in Table I where it is seen that the bismuth caught at mass number 210 is considerably enhanced in alpha activity while that at positions 208 and 209 is depleted. The finding of higher specific activity at mass number 208 than at 209 is not considered as indicating alpha activity as mass 208 and could result from the pattern of "spillover" of an isotope into successive mass number positions.

## C. ALPHA-DECAY ENERGY OF $Bi^{210}$ (LONG)

In the absence of the observation of an isomeric transition between RaE and $Bi^{210}$ (long) the way remains for deciding which is the higher energy state by comparing their alpha- or beta-decay energies. The alpha-decay energy of RaE can be calculated from other decay data and that for $Bi^{210}$ (long) can be measured. The beta-decay energies cannot be compared because the specific activity of $Bi^{210}$ (long) is far too low for beta-ray spectroscopy and the preponderance of the

TABLE I. Specific alpha activity of calutron separated neutron-irradiated bismuth.

| | Alpha disintegrations per minute per milligram of bismuth | |
|---|---|---|
| Mass fraction | Sample 1 | Sample 2 |
| Unseparated | 4 | |
| 208 | $1.2\pm0.6$ | $1.8\pm0.6$ |
| 209 | $0.6\pm0.6$ | $<0.2$ |
| 210 | $128\pm10$ | $142\pm10$ |

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[1] Neumann, Howland, and Perlman, Phys. Rev. 77, 720 (1950).

[2] We are indebted to Dr. R. S. Livingston and members of the Electromagnetic Research Division, Oak Ridge National Laboratory, and to Dr. C. P. Keim and the Stable Isotopes Research and Production Division for performing this separation.

alpha-decay daughter, $Tl^{206}$, would make measurement impossible in any case.

Using the calutron-enriched $Bi^{210}$ (long), thin samples were prepared by electroplating onto platinum disks. The alpha-particle energy distribution was then measured in an ionization chamber connected to a 48-channel differential pulse height analyzer. The results of five determinations gave values of 4.92, 4.94, 4.94, 4.93, and 4.945 Mev; and, although the last one cited is probably more reliable than the others, we shall take the average 4.935±0.020 Mev. For the first three runs, $Ra^{226}$ and daughters were used for energy calibration, and for the last two, $Po^{208}$ and $Po^{210}$ were run simultaneously with the $Bi^{210}$ (long) to provide an internal standard. The pulse-analysis curve for the last determination is shown in Fig. 1. There was no evidence for more than a single alpha group for $Bi^{210}$ (long); the greater width of the peak compared with those of the polonium isotopes can be explained by the sample thickness.

Despite the absence of complex alpha structure, the possibility would still exist that the observed group does not lead directly to the ground state of $Tl^{206}$ and therefore would not define the decay energy. If so, there should be conversion electrons or gamma-rays present in abundance comparable to the alpha group. Dr. D. C. Dunlavey of this laboratory looked for low-energy conversion electrons by impregnating an Ilford G5 photographic plate with some of the bismuth and counting the coincidences between alpha-particle and electron tracks. The number of electrons of $<150$ kev in coincidence with alpha particles was much too small to satisfy the above supposition, and the few observed probably could be accounted for by the low energy tail of the $Tl^{206}$ beta spectrum. The sample of $Bi^{210}$ (long) was also examined with a scintillation spectrometer, and no gamma rays were observed.

From these experiments it is fairly certain that the 4.935-Mev alpha group represents the ground-state transition, and, when correction for recoil energy is made, the decay energy becomes 5.031±0.020 Mev.

The alpha-decay energy for RaE is calculated from the measured $\beta^-$-decay energies of RaE and $Tl^{206}$ and the alpha energy of $Po^{210}$, as may be visualized from the decay cycle, Fig. 2. For this purpose the following decay energies are adopted:[3] RaE $\beta^-$ decay—1.165 Mev, $Po^{210}$ $\alpha$ decay—5.401 Mev, $Tl^{206}$ $\beta^-$ decay—1.51 Mev. The alpha-decay energy of RaE, therefore, becomes 5.056 Mev, which has an uncertainty of perhaps 20 kev. From these figures it would appear that RaE is heavier than $Bi^{210}$ (long) by 25 kev, but the uncertainties of the data are of the same order.

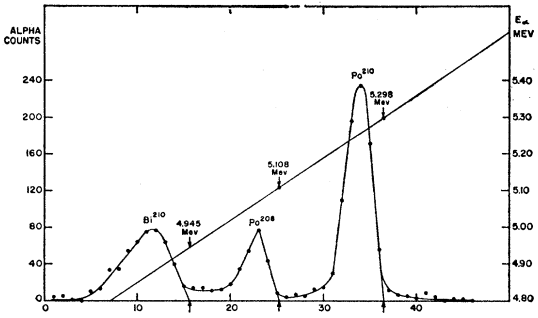

FIG. 1. Alpha pulse analysis of long-lived $Bi^{210}$ with $Po^{208}$ and $Po^{210}$ as standards.

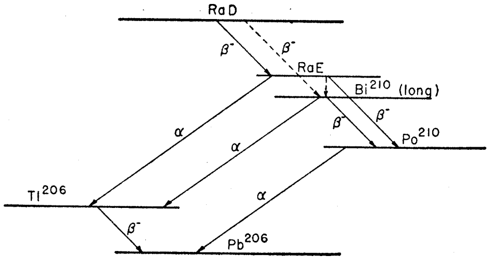

FIG. 2. Decay cycles involving RaE and $Bi^{210}$ (long). (Broken lines indicate modes of decay which are possible but which have not been observed directly or indirectly.)

If the beta-decay energy of RaD to RaE is taken to be 65 kev,[3] then RaD is unstable with respect to $Bi^{210}$ (long) by 90 kev. This difference is probably outside of the uncertainties of the data and the attempt to observe this decay will be mentioned below.

### D. ALPHA-DECAY HALF-LIFE OF $Bi^{210}$ (LONG)

Two groups[4–6] have reported a discrepancy between the neutron activation cross section of bismuth to give RaE and the total absorption cross section. Hughes and Palevsky[5] have adopted the value[7] 19 millibarns (mb) for the RaE activation cross section and have determined the total absorption cross section to be 33 mb. The difference, 14 mb, presumably goes into the formation of $Bi^{210}$ (long). Littler and Lockett[6] have found the RaE activation and absorption cross sections to be 20.5 and 30.8 mb, respectively, leaving 10 mb for the $Bi^{210}$ (long) activation.

Using the amount of $Po^{210}$ as an internal standard of neutron exposure and the specific activity of $Bi^{210}$ (long) found in this laboratory, Hughes and Palevsky have calculated the alpha-decay half-life for $Bi^{210}$ (long) to be $2.6\times10^6$ yr.

### E. BETA-DECAY HALF-LIFE FOR $Bi^{210}$ (LONG)

Since $Bi^{210}$ (long) has the same mass as RaE within about 25 kev, it must be beta-unstable. A previous attempt[1] to observe the beta branching of $Bi^{210}$ (long)

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[3] References to these data may be found in the "Table of Isotopes," Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[4] Palevsky, Hughes, and Eggler, Phys. Rev. **83**, 234 (1951).

[5] D. J. Hughes and H. Palevsky, Phys. Rev. **92**, 1206 (1953).

[6] D. J. Littler and E. E. Lockett, Proc. Phys. Soc. (London) **A66**, 700 (1953).

[7] *Neutron Cross Sections*, U. S. Atomic Energy Commission Report AECU-2040, Supplement 2 (Office of Technical Services, Department of Commerce, Washington, D. C., 1952).

by looking for $Po^{210}$ growth showed only that the branching is small.

In the present study a 50-g sample of bismuth oxychloride containing $2\times 10^5$ alpha disintegrations of $Bi^{210}$ (long) was dissolved in hydrochloric acid solution and purified rigorously from $Po^{210}$ by successive precipitations of added tellurium with stannous chloride as a reducing agent. After a growth period of 83 days some $Po^{208}$ was added as a tracer, and the polonium again removed with tellurium carrier. The tellurium was separated from polonium by dissolving the mixture and precipitating the tellurium alone with sulfur dioxide. The polonium was next extracted with 20 percent tributyl phosphate in dibutyl ether, back extracted into concentrated nitric acid and finally electroplated onto a platinum disk after the nitric acid had been removed by evaporation and the polonium taken up in dilute sulfuric acid.

The bismuth sample was allowed to remain for another period of 82 days, and a second polonium removal was carried out. The yields of $Po^{210}$ were determined by total alpha-particle counting in a standard alpha counter and $Po^{210}/Po^{208}$ ratio measurement with the pulse analyzer. After correcting for chemical yield (from $Po^{208}$ recovery) and growth of $Po^{210}$ to saturation, the equilibrium value of $Po^{210}$ in the two experiments was 685 and 661 disintegrations per minute. The bismuth contained $1.8\times 10^5$ alpha disintegrations per minute. From this the $\beta^-$ branching of $Bi^{210}$ (long) is seen to be $3.7\times 10^{-3}$ and the $\beta^-$-decay half-life is accordingly $7.0\times 10^8$ yr.

It is of course not completely out of the question that $Bi^{210}$ (long) is the upper isomer and that the growth of $Po^{210}$ resulted primarily through the isomeric transition to RaE. In this case, the value $7.0\times 10^8$ yr would apply to the isomeric transition, and the $\beta^-$ half-life would then be even longer. Since the evidence does point to $Bi^{210}$ (long) as the ground state, we shall assume that the growth of $Po^{210}$ arose directly from the beta decay.

### F. SEARCH FOR $Bi^{210}$ (LONG) IN NATURE

As already mentioned, RaD is undoubtedly unstable with respect to $Bi^{210}$ (long). Based on the energy relationship of the $Bi^{210}$ isomers and the 65-kev decay energy of RaD to RaE, the decay energy of RaD to $Bi^{210}$ (long) is 90 kev.

The most sensitive means of looking for this transition is by the examination of uranium ores in which the $Bi^{210}$ (long) would be at its equilibrium concentration. The bismuth fraction was removed from a sample of pitchblende and purified. Over a period of several weeks it showed only the decay period of RaE, and the amount at the time of separation was $5.5\times 10^6$ disintegrations per minute. After complete decay of the RaE, the $Po^{210}$ was removed and the sample examined for the alpha activity of $Bi^{210}$ (long). None was found, and the limits of detection corresponded to a maximum of 5 disintegrations per minute.

The fraction of RaD which decays to $Bi^{210}$ (long) is, therefore, $<10^{-6}$, and its partial half-life is $>2\times 10^7$ yr. $Bi^{210}$ (long) could also arise from the isomeric transition from RaE. By use of the same data, the half-life for this transition is $>10^4$ yr.

### G. DISCUSSION

In discussing the nuclear configuration for $Bi^{210}$ (long), we shall adopt the following properties for it and neighboring nuclei, most of which have been discussed in this paper:

(1) $Bi^{210}$ (long) is the ground state of $Bi^{210}$, and it is 25 kev lighter than RaE. The half-life for the isomeric transition is $>10^4$ yr.
(2) The alpha-decay half-life for $Bi^{210}$ (long) is $2.6\times 10^6$ yr and its decay energy is 5.03 Mev.
(3) The corresponding values for RaE are $3\times 10^4$ yr and 5.06 Mev.
(4) The $\beta^-$-decay half-life for $Bi^{210}$ (long) is $7.0\times 10^8$ yr, and its decay energy is 1.14 Mev.
(5) The corresponding values for RaE are 5.0 days and 1.17 Mev.
(6) The half-life for the decay process RaD→$Bi^{210}$ (long) is $>2\times 10^7$ yr and the decay energy is 90 kev.

According to the independent particle model, $Bi^{210}$ with one proton beyond the 82-proton shell and one neutron beyond the 126-neutron shell should have odd parity low-lying states. With this assumption, Petschek and Marshak[8] have assigned RaE to a 0− state on the basis of the shape of its beta spectrum. The other possibilities, 2− and 1−, gave a poorer fit. Detailed calculations on the possible low-lying states of $Bi^{210}$, according to the independent-particle model, have been made by Pryce[9] and his results are not at variance with this assignment for RaE.

Since the beta-decay transitions involving $Bi^{210}$ (long) differ so radically from those of RaE, it is fairly certain that $Bi^{210}$ (long) possesses a large spin number. This would also be borne out by the long lifetime for the isomeric transition between the two isomers of $Bi^{210}$. As pointed out by Pryce,[9] one should expect to find high angular momentum states among the low-lying levels of $Bi^{210}$, although his calculations would indicate that such states lie above the low-spin states. On this basis $Bi^{210}$ (long) would be a higher isomeric state than RaE, which is opposite to the indications of our experimental information. It is the purpose of this discussion to fix as precisely as possible the spectroscopic state of $Bi^{210}$ (long) on the basis of decay data.

We are assuming that $Bi^{210}$ (long) is the ground state of $Bi^{210}$ (or at least is a lower-lying level than RaE),

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[8] A. G. Petschek and R. E. Marshak, Phys. Rev. **85**, 698 (1952).

[9] M. H. L. Pryce, Proc. Phys. Soc. (London) **65**, 773 (1952).

and therefore the observed growth of $Po^{210}$, governed by a half-life of $7.0\times10^8$ yr, comes directly through beta emission. The log *ft* value for this transition is $\sim19$ which is similar to that for the $K^{40}$ beta decay. Since both $Bi^{210}$ (long) and $K^{40}$ probably decay directly to the ground states of even-even nuclei,[10–12] it is possible that $Bi^{210}$ (long), like $K^{40}$, has spin 4. Similarly, the failure to find $Bi^{210}$ (long) from the decay of RaD (partial half-life $>2\times10^7$ yr) means that the log *ft* value is $>13.6$ and that the spin for $Bi^{210}$ (long) is at least 3.

From the same measurement it could be concluded that the half-life for the 25-kev isomeric transition from RaE to $Bi^{210}$ (long) is $>1.4\times10^4$ yr. By use of the Weisskopf formula[13] the lifetimes for 25-kev transitions of multipole order may be calculated: $\tau_\gamma(E3)\sim9\times10^{-4}$ yr, $\tau_\gamma(M3)\sim0.2$ yr, $\tau_\gamma(E4)\sim8\times10^4$ yr, and $\tau_\gamma(M4)\sim2\times10^7$ yr. These lifetimes refer only to radiative transitions and internal conversion would make the transition half-lives shorter. It would seem, therefore, that spin changes of three or less for the isomeric transition are ruled out unless the energy difference is 5 kev or less rather than the indicated 25 kev. Therefore, if the spin for RaE is 0, that for $Bi^{210}$ (long) must be at least 4.

From the absence of $Bi^{210}$ (long) in the beta decay of RaD (decay energy 90 kev, half-life $>2\times10^7$ yr) the log *ft* value is $>13.6$. This large log *ft* value is consistent with the other information indicating that the spin for $Bi^{210}$ (long) is $>3$. The only known mode of decay for RaD is to a state 47 kev above that of RaE,[14] and since less than 1 event in $10^6$ results in $Bi^{210}$ (long), a limit can be set on the relative transition probabilities from the "47-kev state" to the RaE level and to the level of $Bi^{210}$ (long). Wu, Boehm, and Nagel[14] have shown the 47-kev transition is of the $M1$ type, so that if RaE is $0-$, the 47-kev level is $1-$. Let us suppose that $Bi^{210}$ (long) is $4-$ and see whether the failure to observe transitions from the 47-kev state to $Bi^{210}$ (long) is consistent. This would be an $M3$ transition of 72 kev $(47+25)$ for which the lifetime would be of the order of $10^2$ sec. The lifetime for the 47-kev $M1$ transition should be about $10^{-9}$ sec, so that the failure to observe $Bi^{210}$ (long) in the decay of RaD through this pathway is understandable. An assignment of $4-$ for $Bi^{210}$ (long) is seen to be consistent with the data presented here.

Mention should be made of the alpha-decay half-lives of the two isomers, although alpha-decay theory for such nuclei is not very helpful at present for assigning spectroscopic states. The two isomers have almost the same decay energy, but the partial alpha half-life of $Bi^{210}$ (long) is about 100-fold that of RaE. RaE itself is highly hindered as are other bismuth alpha emitters.[15] A large spin change for the decay of $Bi^{210}$ (long) could well be a factor, but also the rearrangement necessary in creating the alpha particle is probably involved when nucleons having such large spins are involved and when two closed shells are broken open as is the case here.

### H. ACKNOWLEDGMENT

We wish to acknowledge the part played by Dr. Henry Neumann (now at Northwestern University) in some of the early phases of this work.

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[10] The first excited state of $Po^{210}$ is probably at 1.19 Mev in accordance with a gamma ray observed by Hoff (see reference 11) in the electron capture decay of $At^{210}$. Neither RaE nor $Bi^{210}$ (long) could decay to this state. A level at this energy is also expected from the systematics of the first excited states of even-even nuclei (see reference 12).

[11] R. W. Hoff, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL-2325, September, 1953 (unpublished).

[12] F. Asaro and I. Perlman, Phys. Rev. **87**, 393 (1952).

[13] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[14] Wu, Boehm, and Nagel, Phys. Rev. **91**, 319 (1953).

[15] Perlman, Ghiorso, and Seaborg, Phys. Rev. **77**, 26 (1950).