The effect is small, however, and at worst could cause the observed cross sections to be in error by about 10 percent.

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Neutron Capture γ-Rays from Lead and Bismuth

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Only two strong γ-rays have been detected with a pair spectrometer during the bombardment of lead thermal neutrons. The weaker of the two is due to capture in Pb^{206} and has an energy of 6.734 ± 0.008 Mev, and the stronger is due to capture in Pb^{207} and has an energy of 7.380 ± 0.008 Mev. Apart from a very weak γ-ray with an energy of 6.90 ± 0.05 Mev, no other radiation has been detected. The 6.9-Mev γ-ray may be due to the excitation of a hitherto unknown state in Pb^{208}, or it may be due to an impurity.

Bismuth produces a γ-ray with an energy of 4.170 ± 0.015 Mev. The breadth of the coincidence peak is about 70 kev greater than would be expected for a homogeneous radiation. The γ-rays revealed by the coincidence spectra of lead and bismuth have intensities of about one quantum per capture.

The binding energies of four neutrons in Pb^{210} (RaD) have been calculated using the above experimental data and compared with that computed from the decay of Pb^{210} and its products. The latter exceeds the former by 0.37 ± 0.07 Mev. The origin of this discrepancy is discussed.

The binding energy of Pb^{210} is shown to be less than the energy of disintegration of RaC'', and experiments are described which prove that if neutrons are produced in the decay of the latter, the yield is less than one neutron per two hundred and fifty disintegrations.

No γ-ray with the binding energy of Pb^{205} (6.4 ± 0.2 Mev) was detected. Pb^{205} is shown to be unstable against electron capture to Tl^{205} by about 300 kev. Both Bi^{208} and Bi^{209} are shown to be unstable for α-decay by 3.25 Mev.

I. INTRODUCTION

A PRELIMINARY report on the energies of the γ-rays produced by capture of slow neutrons in lead and bismuth has already been published.[1] The present paper contains details of this work. The method of energy and intensity measurement will be discussed elsewhere.

A sample of the material to be examined is placed in a high neutron flux in a hole which traverses the concrete radiation shield of the pile. Gamma-rays from the pile are prevented from passing down the hole by a block of bismuth 5 in. long. The pair spectrometer is placed outside the concrete shield, and a series of lead collimators limits the γ-radiation reaching the spectrometer to that emitted from the central region of the sample. In this way, unwanted radiations emitted by the lining of the hole are eliminated.

II. RADIATIONS FROM LEAD

The first measurements with lead were made with two samples, one of chemically pure metallic lead, and the other of litharge (PbO) in a Dural container. The spectra obtained are shown in Fig. 1. It will be seen that the two characteristic peaks at 6.7 and 7.4 Mev appear in the same proportion in the spectra of both samples; and, in addition, the 7.7-Mev aluminum capture γ-ray appears in the litharge sample. Now, it is improbable that an impurity which might be responsible for the weaker of the two peaks should be present in both samples to the same extent; for this reason both radiations have been ascribed to lead. Since the energy released by neutron capture in the even mass number isotope will be less than that produced by the adjacent odd isotope, the γ-ray with the least energy is ascribed to capture in Pb^{206} and the γ-ray with the greatest energy to capture in Pb^{207}.

This assignment is confirmed by the more detailed

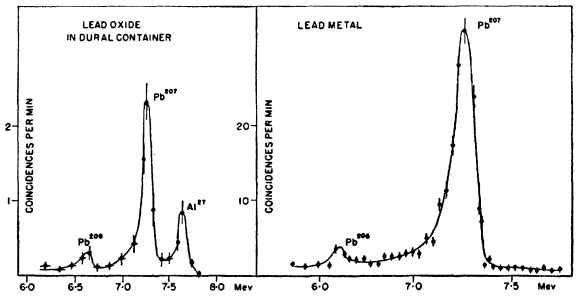

FIG. 1. Coincidence spectra for lead oxide, in a Dural container, and for lead metal, obtained with the pair spectrometer.

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[1] Kinsey, Bartholomew, and Walker, Phys. Rev. **78**, 77 (1950).

studies of the lead spectrum shown in Fig. 2, in which the upper curve represents the spectrum obtained with a target of natural lead, and the lower curve, the spectrum obtained with a sample of radiogenic lead,[2] enriched in Pb^{206} and depleted in Pb^{207}. The ordinates of Fig. 2 have been adjusted to correspond to the same pile power, and the peak coincidence counting rates should be in proportion to the isotopic content of the two samples.[3] It will be seen that the 6.7-Mev peak is strongly enhanced in the sample of radiogenic origin, while the 7.4-Mev peak, due to capture in Pb^{207}, is reduced in intensity. The magnitude of the effect is shown in the last row of Table I.

It will be seen that the observed ratio of the peak counting rates of the 7.38-Mev γ-ray (due to Pb^{207}) is in agreement with the value calculated from the ratio of the isotopic contents of the two samples. The ratio

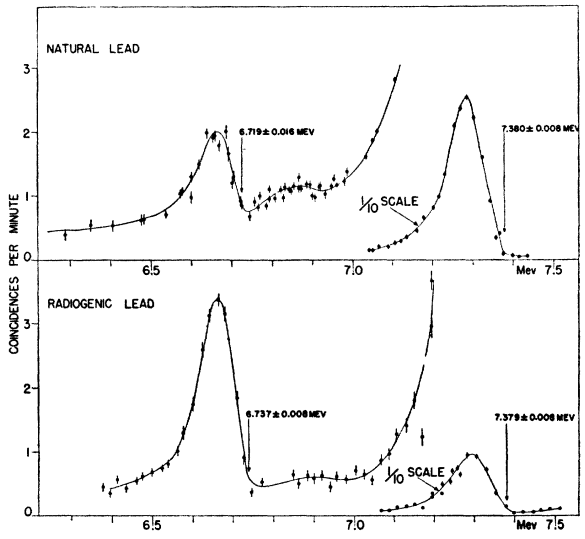

FIG. 2. Coincidence spectra for natural lead and for radiogenic lead, obtained with the pair spectrometer.

of the peak counting rates of the 6.7-Mev γ-ray, however, is much lower than the value expected. The discrepancy may arise from an additional weak γ-ray produced in the natural lead sample with the same energy as that due to capture of neutrons in Pb^{206}. It is possible that this γ-ray is part of the spectrum produced by capture in Pb^{207}; in that event, it must correspond to the creation in Pb^{208} of an excited state with an energy (0.65 Mev) equal to the difference between its energy and that of the binding energy of Pb^{208}, *viz.*, 7.38 Mev. No excited state is known to exist in Pb^{208} with this energy; and, as pointed out below, it is most unlikely that any excited states exist below 2.62 Mev. It is much more probable that the peak of 6.7 Mev in natural lead is enhanced by a γ-ray due to an impurity.

TABLE I. Peak coincidence counting rates for the lead γ-rays in relation to isotopic composition.

	Pb^{206}	Pb^{207}	Pb^{208}
Composition (percent):			
Natural lead (A)	23.6	22.6	52.3
Radiogenic lead (B)	88.2±1.8	8.76±0.17	3.04±0.06
Ratio of peak counting rates:			
Calculated (B/A)	3.75	0.39	
Observed (Fig. 2)	2.7	0.37	

Figure 2 shows the presence of a very weak γ-ray with an energy of 6.90±0.05 Mev. While this γ-ray is distinct in the upper curve of Fig. 2, its presence is indicated but not definitely established in the lower curve. Evidently, if this γ-ray is produced by lead, it cannot result from the capture of neutrons in Pb^{206} but must be associated with capture by another isotope. This could only be Pb^{207}, because the neutron binding energies in Pb^{205} and Pb^{209} are much lower than the energy of this γ-ray. If it is due to a transition leading to an excited state in Pb^{208}, the energy of this state must be 0.48±0.05 Mev. Now γ-rays with energies of 0.51 and 0.58 Mev are known to occur in the excitation of Pb^{208} produced by the decay of ThC″. These γ-rays are generally supposed to excite the 2.62-Mev level, which is assumed to be the first excited state in Pb^{208}. (See Fig. 6a.) The contrary assumption, that the emission of the 2.62-Mev γ-ray produces an excited state at 0.51 or 0.58 Mev, is inconsistent with the observed end point of the β-spectrum of ThC″ and leads to difficulties in the explanation of the other γ-rays which are emitted. It seems to be more probable that the 6.9-Mev γ-ray is also due to an impurity.

The energies of the two principal γ-rays are given in Fig. 2. The differences between the two sets of results are within the probable statistical errors. The coincidence counting rates produced by the radiogenic lead sample for the 6.7-Mev peak are higher than those of the natural lead sample; and also, as discussed above, the 6.7-Mev peak from natural lead may contain an extraneous component. For these reasons the energy measurement obtained with the radiogenic lead is the more accurate. The weighted mean energies are,

from capture in Pb^{206}: 6.734±0.008 Mev;
from capture in Pb^{207}: 7.380±0.008 Mev.

The residual error given is derived from various systematic errors which may be present in the measurement of the magnetic field of the spectrometer.

Apart from the radiations just described, no other γ-ray between 2.6 and 8 Mev has been detected in a survey of the lead spectrum. No γ-ray has been found with the binding energy of the neutron in Pb^{209} (about 4 Mev); such a γ-ray would not be expected, for the ground-state transition in that nucleus should be highly forbidden. The capture of neutrons in Pb^{207}, however,

[2] Prepared from uranium ore by Eldorado Mining and Refining, Ltd. (1944), Port Hope, Ontario.

[3] We are indebted to Dr. Thode of McMaster University for providing the facilities for the isotopic analysis of the radiogenic lead used in this experiment.

might produce the well-known excited state in Pb^{208} at 2.62 Mev. If a transition to this state occurs as often as does the transition to the ground state, it is possible that a γ-ray of this energy might escape detection, because the sensitivity of the pair spectrometer falls by a factor of 40 between 7.38 and 2.62 Mev. The creation of this state could take place as the result of the emission of a γ-ray of 4.76 Mev. A special search was made for a γ-ray of this energy, but none was found. A coincidence peak with one percent of the counting rate of the 7.38-Mev peak would have been detected; such a counting rate would be equivalent to a γ-ray intensity of about 5 percent of that of the ground-state transition.

We have found evidence for such radiations of low energy by comparison of the intensity measured with an ionization chamber with that obtained by counting the recoil electrons produced in an anthracene crystal. The intensity of the soft radiation cannot be very great, however, as it was not shown in a study of the absorption of the capture γ-radiation made with the ionization chamber.

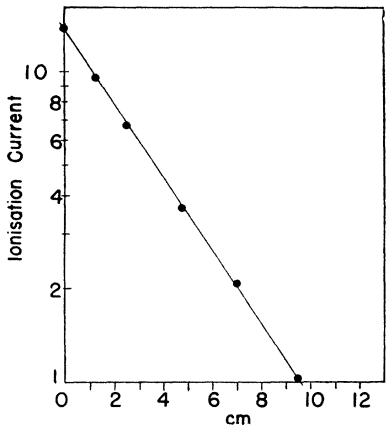

FIG. 3. Absorption of lead capture radiation in brass.

Figure 3 is an absorption curve in brass of the radiation from natural lead. The absorption was measured with the aid of a thick-walled ionization chamber, the small background obtained when the beam of γ-radiation was shut off being subtracted from the ionization current at each point. The logarithmic plot is linear, with an absorption coefficient of 0.271 ± 0.010 cm^{-1}. The measurements were made under exceptionally good geometrical conditions, and the measured absorption coefficient therefore represents the total absorption. The calculated absorption coefficient (0.257 cm^{-1}, which was computed on the basis of the two lead γ-rays with intensities in the ratio of 1:0.093) is in good agreement with this figure.

The linearity of the absorption curve suggests that there is no intense soft γ-radiation. However, the linearity does not prove unequivocally the absence of radiations other than the two γ-rays described above; because the response of a thick-walled ionization chamber is proportional to the energy of the radiation, and the absorption coefficient in most materials does not change much with energy for γ-rays of a million electron volts or more. If the 2.62-Mev γ-ray were present and if the flux of this radiation in photons per unit area per unit time were the same as that of the 7-Mev radiation, it would be only just detectable as a departure from linearity in Fig. 3.

Using an anthracene crystal with a photomultiplier and discriminator circuit, we have computed the total counting rate of recoiling electrons produced by the 7-Mev radiations by extrapolating to zero the curve in which the counting rate obtained is plotted against the bias of the discriminator.[4] The flux of the γ-radiation is readily computed from this counting rate, the number of electrons in the crystal, and the Compton cross section. This method was found to give correct results when applied to the γ-rays produced by a source of Co^{60} of known strength. If no softer γ-rays were present, the flux of the 7-Mev radiations measured with an ionization chamber should be the same as that measured by the counting rate in a crystal. A difference between the results obtained with these two methods should be a sensitive indication of the presence of softer radiations. We have measured the flux of the 7-Mev radiations with a thick-walled ionization chamber, using the figures given by Fowler, Lauritsen, and Lauritsen[5] for the number of ion pairs produced by a thick-walled chamber per incident photon. The flux obtained with the anthracene crystal was found to be twice as great as that obtained with the ionization chamber. The discrepancy indicates the presence of some softer radiation which was not detected with the pair spectrometer.

III. RADIATIONS FROM BISMUTH

Natural bismuth has only one isotope, Bi^{209}. With a bismuth sample a weak broad coincidence peak was found near 4 Mev. In this energy region strong γ-rays from aluminum occur. Aluminum is used in the construction of the pile, and its γ-rays will be superposed on the spectrum to be studied unless precautions are taken to insure that no aluminum lies in the field of view of the spectrometer. Aluminum radiation was successfully eliminated with the aid of extra lead collimators. The effectiveness of these measures was determined readily by setting the spectrometer to count the peak coincidences arising from the strong aluminum γ-ray at 7.72 Mev.

A survey of the γ-rays produced by bismuth showed a coincidence peak near 4 Mev and no evidence for γ-rays at any other energy. A very long and detailed investigation gave the results shown in Fig. 4. The end point of the bismuth spectrum corresponds to an energy of 4.170 ± 0.015 Mev.

[4] The authors are indebted to Dr. W. G. Cross for making these measurements.

[5] Fowler, Lauritsen, and Lauritsen, Revs. Modern Phys. **20**, 236 (1948).

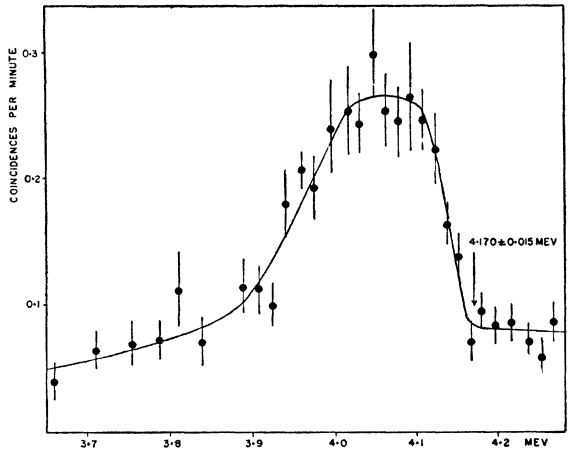

FIG. 4. Coincidence spectrum for bismuth obtained with the pair spectrometer.

Experience has shown that the width (at half-maximum) of the coincidence peak produced by a homogeneous γ-ray is, in energy units, nearly independent of the γ-ray energy. For the particular arrangement of slits which define the aperture of the Geiger counters in the present work, this width is about 130 kev. The width of the bismuth peak, however, is nearly 200 kev. It is certain, therefore, that more than one γ-ray contributes to the spectrum of Fig. 4. Two radiations with an energy difference of 70 kev and of about equal intensity could account for the width observed. This difference is greater than the energy of the highest of the known low-lying states in Bi^{210} (RaE) but much less than the energy of the first excited state produced in the (d,p) reaction.[6] It is possible that the lower energy capture γ-ray is responsible for the production of the isomeric state of RaE recently discussed by Neumann, Howland, and Perlman.[7]

IV. INTENSITIES OF THE LEAD AND BISMUTH γ-RADIATIONS

The simplicity of the γ-ray spectra of lead and bismuth indicates that roughly one photon is emitted per capture. This conclusion has been verified by a comparison of the intensities of these γ-rays with those produced by some light elements which are also responsible for apparently homogeneous radiations.

If is it assumed that the neutron flux is constant throughout the sample material, the peak coincidence counting rate (q_r) for a γ-ray of energy E_r is given by:

$$q_r = CN\sigma p_r T_r T_r' \epsilon_r,$$

where C is a constant, N is the number of atoms presented to the field of view of the spectrometer, σ is the capture cross section for thermal neutrons, p_r is the number of γ-rays produced per capture, T_r and T_r' are the fractions of this radiation transmitted through the sample itself and through the boron-paraffin block used to remove neutrons from the beam of radiation, and ϵ_r is the counting efficiency in the spectrometer (which is a known function of the γ-ray energy).

This expression can be used to compute p_r, for all other quantities contained in it can be measured or calculated. The constant C will depend on the neutron flux. If the neutron flux is assumed to be the same for different samples when the pile is operated at the same power level, C may be determined from a measurement on the 2.75-Mev γ-ray emitted by Na^{24} generated in a sodium sample. Because when radioactive equilibrium is established between the Na^{24} activity and the neutrons producing it, p is unity for this γ-ray, and the coincidence counting rate will measure the rate of neutron capture.

The number of γ-rays (p) per neutron capture found for lead and bismuth are given in Table II and compared with those obtained for two light elements, *viz.*, Be and C. The capture cross section of lead was taken to be 220 millibarns and that of bismuth 15 millibarns. The results shown in Table II are necessarily inaccurate; there is no significance in the fact that the number of quanta per capture is greater than unity in some cases. The method is subject to a large error (possibly 20 percent) involved in the assumption that the neutron flux in the samples is identical for the same pile power, because, in fact, the neutron flux varies rapidly along the axis of the hole in the pile and quite appreciably in a distance equal to the length of a sample container. In addition, other errors are introduced in the calculation of the counting efficiency of the spectrometer.

The capture cross section due to Pb^{206} has been ignored in the calculation of the yield of the γ-ray produced by Pb^{207}, because the intensity of the Pb^{206} γ-ray is only one-thirteenth of that of the former. It is possible that strong γ-rays of lower energy are produced by capture in Pb^{206} without being detected. Therefore, until the thermal neutron cross section of Pb^{206} has been separately measured, the absolute yield of the Pb^{206} γ-ray cannot be found.

V. CAPTURE OF NEUTRONS BY Pb^{204}

The energy of the ground state γ-ray due to neutron capture in Pb^{204} may be calculated from the disintegration energies of Tl^{204} and Tl^{206} and the data now available on the binding energies of lead and thallium isotopes. The sum of the binding energies of Pb^{205} and Pb^{206} less the sum of the binding energies of Tl^{205} and

TABLE II. Estimated number of quanta produced per capture in a direct transition to the ground state of the product nucleus.

Sample material	Number of quanta per capture
Pb^{207}	1.3
Bi	1.6
Be	1.7
C^{12}	1.0

[6] J. A. Harvey, Phys. Rev. **79**, 241(A) (1950).
[7] Neumann, Howland, and Perlman, Phys. Rev. **77**, 720 (1950).

Tl^{206} is equal to the difference in the disintegration energies of the two β-emitting bodies, Tl^{206} and Tl^{204}. For the binding energy of Pb^{206} we assume 8.15±0.05 Mev, an average of the results of Harvey and of Palevsky and Hanson (Table III); for that of Tl^{205}, 7.48±0.15 Mev, obtained by Hanson and his collaborators.[8] Our value for the binding energy[9] of Tl^{206} is 6.23±0.05 Mev. Neither Tl^{204} nor Tl^{206} emits γ-radiation[10,11] and the end points of their β-spectra[10–12] are at 0.80±0.03 and 1.63±0.1 Mev.[7] Thus, we obtain a binding energy of 6.4±0.2 Mev for Pb^{205}. No γ-ray has been found in the capture γ-ray spectrum at this energy (Fig. 2).

From these figures it follows that Pb^{205} is unstable against decay to Tl^{205} by 300±130 kev. Therefore, its decay of Tl^{205} is possible only by electron capture. The decay radiations of Pb^{205} have not yet been identified with certainty.

VI. THE NEUTRON BINDING ENERGIES

The experimental values of the neutron binding energies of lead and bismuth are listed in Table III. Harvey's results[13] were obtained from the Q-values of the highest energy groups of protons and tritons observed in (d,p) and (d,t) reactions using a 16-Mev deuteron beam. The results of Palevsky and Hanson[14] and of McElhinney and his collaborators[15] were obtained by the measurement of the thresholds of the (γ,n) reactions.

If $B_n(A, Z)$ is the neutron binding energy of a nucleus of mass A and charge Z and if B_d and B_t are the binding energies of the deuteron and the triton, respectively, the binding energy is related to the Q-values of the particle groups which lead to the ground states of the product nuclei through the equations:

$$B_n(A,Z)=B_d+Q[(A-1,Z)(d,p)(A,Z)]$$
$$B_n(A,Z)=B_t-Q[(A,Z)(d,t)(A-1,Z)].$$

B_d has the value 2.230±0.007 Mev, according to Bell and Elliott;[16] and B_t has the value 6.262±0.023 Mev, a quantity which can be deduced from the Q-value for the reaction $H^2(d,p)H^3$ obtained by Tollestrup, Fowler, and Lauritsen.[17] For both Pb^{207} and Pb^{208}, the equality of the binding energies obtained with these two reactions (Table III) shows that the highest energy proton and triton groups observed must, indeed, be those which lead to the ground states, because if this were not the case, the one expression would lead to a higher value and the other to a lower value of the binding energy.

For the binding energies of Pb^{207} and Pb^{208} our results are in close agreement with those obtained by Harvey. The two stronger γ-rays of Fig. 2 therefore represent the ground state transitions in these isotopes.

The binding energy of Pb^{208} determined by Palevsky and Hanson is in agreement with the other results. Their value for the binding energy of Pb^{207}, however, is about 220 kev too high. While the error in the absolute value of their measurements is rather high, the error in the difference of the binding energies should be much smaller. The difference in the binding energies of Pb^{208} and Pb^{207} is 0.49 Mev. If 7.38 Mev is assumed to be the more accurate value for the binding energy in Pb^{208}, this indicates that the binding energy of Pb^{207} should be 6.89±0.06 Mev. This value is considerably greater than the result of the present measurements; there seems to be no explanation of this discrepancy.

More recently, the thresholds of the (γ,n) reactions in Pb^{208} and Pb^{207} have been measured by Parsons, Lees, and Collie.[18] For Pb^{208} they find a binding energy of 8.1±0.3 Mev and for Pb^{207} a value 1 Mev less. Their results do not agree with any of those listed in Table II, although they were obtained with a method similar to that used by Palevsky and Hanson.

The binding energy of a neutron in Pb^{210} can be calculated from that of Bi^{210} using the disintegration energies of Pb^{209} (0.70 Mev)[10,19] and of Pb^{210} (RaD). There are difficulties in the interpretation of the γ-rays produced by RaD, but the total disintegration energy is not likely to differ much[20] from 0.06 Mev. With these figures, the binding energy of a neutron in Pb^{210} becomes 4.81±0.03 Mev.

TABLE III. Experimental values of neutron binding energies in Pb and Bi (in Mev).

Nucleus	Present work	Harvey[a] (d,p)	Harvey[a] (d,t)	Palevsky and Hanson[b]	McElhinney, *et al.*[c]
Pb^{206}			8.10±0.10	8.25±0.10	
Pb^{207}	6.734±0.008	6.71±0.03	6.70±0.05	6.95±0.10	
Pb^{208}	7.380±0.008	7.37±0.03	7.37±0.05	7.44±0.10	
Pb^{209}		3.87±0.05			
Bi^{209}			7.44±0.05		7.45±0.20
Bi^{210}	4.170±0.015	4.14±0.03			

[a] Reference 13. [b] Reference 14. [c] Reference 15.

[8] Hanson, Duffield, Knight, Diven, and Palevsky, Phys. Rev. **76**, 578 (1949).

[9] We are indebted to Dr. J. A. Harvey for the communication of his result for the binding energy of Tl^{206}, *viz.*, 6.16±0.15 Mev. We have found a strong γ-ray produced by capture of neutrons in thallium at 6.23±0.05 Mev which may be identified with the binding energy.

[10] K. Fajans and A. F. Voigt, Phys. Rev. **60**, 619 (1941).

[11] K. Fajans and A. F. Voigt, Phys. Rev. **58**, 177 (1940).

[12] R. S. Krishnan and E. A. Nahum, Proc. Camb. Phil. Soc. **36**, 490 (1940).

[13] Private communication. We are indebted to Dr. J. A. Harvey for the privilege of using these results before publication.

[14] H. Palevsky and A. O. Hanson, Phys. Rev. **79**, 242A (1950).

[15] McElhinney, Hanson, Becker, Duffield, and Diven, Phys. Rev. **75**, 542 (1949).

[16] R. E. Bell and L. G. Elliott, Phys. Rev. **79**, 282 (1950).

[17] Tollestrup, Fowler, and Lauritsen, Phys. Rev. **78**, 372 (1950).

[18] Parsons, Lees, and Collie, Proc. Phys. Soc. (London) **A63**, 915 (1950).

[19] W. Rall and R. G. Wilkinson, Phys. Rev. **71**, 321 (1947).

[20] H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc. (London) **A160**, 454 (1937).

VII. COMPARISON WITH DATA OF RADIOACTIVITY

The binding energy of four neutrons in Pb^{210} can be obtained in two ways: (1) by the addition of the experi-

mental binding energies listed in Table III, together with that calculated for Pb^{210} and referred to in the last paragraph, and (2) from the radioactivity data of RaD and its products.

(1) The results of the present γ-ray measurements are probably the most accurate values for the binding energies of Pb^{207} and Pb^{208}. For the binding energy of Pb^{209}, we use that obtained by Harvey and for the binding energy of Pb^{210}, the value 4.81 ± 0.03 Mev. The sum of these four quantities is 22.79 ± 0.05 Mev.

(2) The binding energy of four neutrons in Pb^{210} is equal to the energy equivalent of the difference in mass between four neutrons and the α-particle, less the total energy set free in the consecutive disintegrations between Pb^{210} and Pb^{206} (Fig. 5). Using the data recently compiled by Tollestrup, Fowler, and Lauritsen,[17] the mass difference $(4n-\alpha)$ is equivalent to 29.79 ± 0.04 Mev (1 mMU is equivalent to 0.93104 Mev).[21] For the disintegration of RaD, we assume[20] 0.06 Mev; for RaE, 1.17 Mev;[22–24] and for Po^{210}, 5.40 Mev.[25] The binding energy of four neutrons in Pb^{210} is therefore 23.16 ± 0.05 Mev. This quantity exceeds that obtained by method (1) by 0.37 Mev.

This discrepancy is difficult to explain. For the reasons given in a previous paragraph, there seems to be no doubt that the energies of the two principal γ-rays of Fig. 2 are indeed the binding energies of Pb^{207} and Pb^{208}. It is most improbable that the discrepancy can arise from errors contained in the disintegration energies of RaD, RaE, or Po and most unlikely that individual errors in these quantities could accumulate to this difference. Furthermore, the calculated binding energy of a neutron in Pb^{210} contains only the neutron binding energy of Bi^{210} and the β-disintegration energies of RaD and Pb^{209}; of the latter energies, the β-disintegration energy of RaD is unlikely to be in error by as much as 0.02 Mev, and the β-decay of Pb^{209}, which has been accurately measured, is not followed by the emission of γ-radiation. Therefore, either one or both of the neutron binding energies of Pb^{209} and Bi^{210} have been underestimated. We must conclude that the bismuth γ-ray or the higher energy proton group formed in the $Pb^{208}(d,p)Pb^{209}$ reaction, or both, do not represent the direct transitions to the ground states of the product nuclei.

A similar conclusion has been obtained independently by Huizenga, Magnusson, Simpson, and Winslow.[26] These authors suggest that the binding energy of Pb^{209} is given correctly by the energy balance in Harvey's experiments on the reaction $Pb^{208}(d,p)Pb^{209}$ and that the experimental value for the binding energy of Bi^{210} is deficient. Their argument is based in part on the normal position of Pb^{209} and the anomalous position of Bi^{210} in relation to the alternation of binding energies in a sequence of isotopes if these binding energies are correctly given in Table III. This is indeed a cogent argument if applied to a succession of isotopes of the same element for which no discontinuity in the binding energy is known to exist. However, some irregularity in the binding energy alternation might be expected at the commencement of the new shell which is created in the addition of the 127th neutron and the 83rd proton. For this shell is unique in that the addition of these nucleons halves the neutron and proton binding energy which obtained at the closure of the previous shell at the Pb^{208} nucleus. Huizenga and his associates also point out that if the true binding energy of Pb^{209} is greater than the experimental value by the whole of the deficiency, the binding energy of that isotope would be greater than that of the odd-odd nucleus Bi^{210}, which, on account of the interaction of the unpaired neutron and proton, would be expected to be more tightly bound. This argument, which is more convincing, suggests that the energy deficiency should be ascribed to Bi^{210}. It is supported by the fact that the last neutron or proton, respectively, in the even-even nuclei Pb^{210} and Po^{210} has higher binding energy than the odd neutron in Pb^{209}.

It is difficult to understand why the ground state of Pb^{209} or Bi^{210} should not be obtained as a product of the (d,p) reactions. The critical orbital angular momentum of the 16-Mev deuterons used in Harvey's experiments is about $10\hbar$. This is quite sufficient to produce the ground state of Pb^{209}, which according to Feather and Richardson[27] has a spin of 11/2. The spin of Bi^{210} is probably 2 units, and its ground state should be no more difficult to produce directly even if, as would seem very probable, it is compounded of a closed core together with a neutron and a proton in states of the same orbital angular momentum.

Too little is known about the mechanism which determines the intensity of γ-radiation to decide

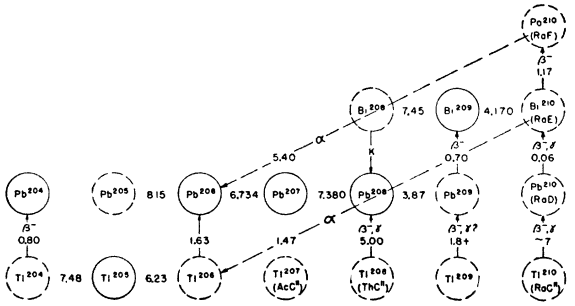

FIG. 5. Relations between certain isotopes of Tl, Pb, Bi, and Po. Experimental neutron binding energies are written between the symbols for the isotopes, and the decay energies between those for the isobars.

[21] J. W. M. DuMond and E. R. Cohen, Revs. Modern Phys. **21**, 651 (1949).
[22] L. M. Langer and M. D. Whitaker, Phys. Rev. **51**, 713 (1937).
[23] A. Flammersfeld, Z. Physik **112**, 727 (1939).
[24] G. J. Neary, Proc. Roy. Soc. (London) **A175**, 71 (1940).
[25] Perlman, Ghiorso, and Seaborg, Phys. Rev. **77**, 26 (1950).
[26] Huizenga, Magnusson, Simpson, and Winslow, Phys. Rev. **79**, 908 (1950).
[27] N. Feather and H. O. W. Richardson, Proc. Phys. Soc. (London) **61**, 452 (1948).

whether the radiative capture of a neutron by Bi^{209} should lead directly to the ground state of Bi^{210}. The spin of Bi^{209} (9/2) and its magnetic moment suggest that the odd proton in this nucleus has an orbital angular momentum of 5 units. If its parity is odd, the radiative transition to the ground state, which would result from the capture of a slow neutron, would produce a change of parity. This follows because the β-disintegration of Bi^{210} is second forbidden and produces no change in parity. The parity of the ground state of Bi^{210} is, therefore, that of the even-even nucleus Po^{210}, which is probably even, for it differs from the closed shell nucleus Pb^{208} only by the possession of the two extra protons. Since the spin of Bi^{210} is probably 2 units, the minimum spin radiated will be 2 units and the γ-ray will be magnetic quadrupole in type. Theoretically, such a third-order γ-ray should be forbidden as compared with first- and second-order radiations. Experimental evidence does not support this conclusion, however, for, as pointed out in another communication,[28] there seem to be instances (e.g., K^{40}) in which magnetic quadrupole radiation emitted in a transition directly to a ground state can compete successfully with γ-rays emitted in transitions to a variety of excited states.

VIII. THE DISINTEGRATION OF RaC″

The neutron binding energy of Pb^{210} (RaD) of 4.81 Mev is lower than the energy produced in the disintegration of its parent, Tl^{210} (RaC″). This fact suggests the possibility that neutrons may be emitted in the disintegration of RaC″. Inasmuch as the binding energy of Pb^{210} is calculated from that of Bi^{210}, this possibility still exists if the neutron binding energy of Bi^{210} is greater than the energy of the capture γ-ray from Bi^{209} by 0.37 Mev, the full amount of the energy discrepancy previously discussed.

Now the neutrons emitted in the disintegration of N^{17} are easily detected because the β-transition between the ground state of that nucleus and its product is forbidden; and, therefore, most of the transitions lead to neutron emission. However, the β-disintegration of RaC″ is of the allowed type and produces RaD in an excited state at about 4 Mev. The position of this state is not known exactly, but it certainly lies below the neutron binding energy. If a β-transition is possible to a higher level of RaD, neutron emission could occur; these neutrons, however, would be produced in measurable numbers only if the β-transition to that level were also of the allowed type and if the energy of the partial β-disintegration was not too small.

The product of neutron emission from RaC″ is Pb^{209} This body has a period of about 3 hr; and its β-activity would probably remain undetected when a source of RaC″ (prepared by recoil) had decayed to a few tenths of a percent of its original strength, for such sources invariably contain an appreciable contamination of RaC introduced by the process of aggregate recoil. No

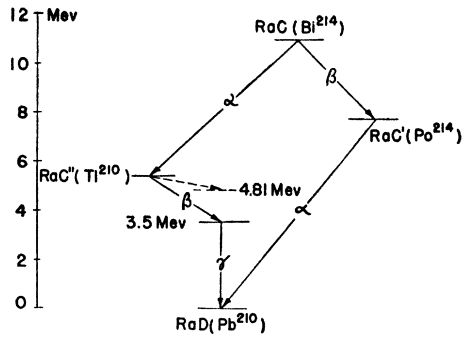

FIG. 6. The disintegration of RaC and its immediate products drawn on the assumption that the most energetic β-particles of RaC produce the ground state of RaC′.

residual activity ascribable to Pb^{209} can be discerned in the decay curves of RaC″ obtained by Fajans.[29] If neutrons are emitted, their presence, therefore, is likely to be established only by direct measurement.

RaC″ is produced by the α-decay of RaC only to the extent of one atom in 4000 disintegrations of that nucleus (Fig. 6). The remainder decay by β-emission to RaC′ and thence to RaD by α-disintegration. Owing to the small extent to which it is produced in the decay of RaC and owing to its short life of 1.32 min, the total disintegration energy of RaC″ has not been measured accurately by a direct method. The end point of the β-spectrum was found to be 1.7 Mev by Goldstein and Lecoin[30] and 1.95 Mev by Devons and Neary.[31] The latter experimenters found that the γ-ray energy emitted per disintegration was about 5 ± 1 Mev. The total disintegration energy is therefore about 7 ± 1 Mev.

In principle the total energy of disintegration of RaC″ can be found much more exactly by subtracting from the total energy set free in the branch RaC–C′–D, the energy of the most energetic group of α-particles due to the branch RaC–C″ (Fig. 6). At present this method is subject to some doubt because of an uncertainty in the energy of the disintegration of RaC. According to the scheme drawn up by Ellis,[32] the highest energy group of β-particles produces an excited state in RaC′ at 606 kev, and recently Feather and Richardson[27] have given theoretical reasons to show that the disintegration should follow this course. However, Bothe and Maier-Leibnitz[33] failed to find coincidences between the highest energy β-group and the 606-kev γ-ray. This indicates that the most energetic β-group leads directly to the ground state of RaC′. According to Constantinov and Latyshev,[34] the energy of this group is 3.17 Mev. Until further measurements are made on the disintegra-

[28] Kinsey, Bartholomew, and Walker, Phys. Rev. **78**, 481 (1950).

[29] K. Fajans, Physik. Z. **12**, 369 (1911).

[30] L. Goldstein and M. Lecoin, Compt. rend. **202**, 1169 (1936).

[31] S. Devons and G. J. Neary, Proc. Cambridge Phil. Soc. **33**, 154 (1937).

[32] C. D. Ellis, International Conference on Physics, London (1934).

[33] W. Bothe and H. Maier-Leibnitz, Z. Physik **104**, 604 (1937).

[34] A. A. Constantinov and G. D. Latyshev, J. Phys. (U.S.S.R.) **5**, 249 (1941).

tion of RaC, the total disintegration energy must be considered to be uncertain: its value may be 3.17 or 3.78 Mev. Using the α-particle energy measurements of Lewis and Bowden,[35] it follows that the total disintegration energy of RaC'' is 5.39 or 6.00 Mev. The latter figure, which corresponds to the direct production of the 606-kev level in RaC' by the most energetic β-particles of RaC, is in better agreement with the results of Devons and Neary than is the former figure. The total disintegration energy of RaC'', as mentioned at the beginning of this section, therefore is greater than the neutron binding energy of Pb^{210}.

In collaboration with Dr. C. H. Millar of this laboratory we have measured the total neutron emission from commercial sources of radium and radiothorium in equilibrium with their products, using specially sensitive boron trifluoride counters. While the neutron emission from radium was found to be about one neutron per million disintegrations of radium, that from the radiothorium source was considerably higher. Now no possibility exists for the emission of delayed neutrons in the thorium series, and so it is clear that the greater part of the neutrons observed from both sources are due to the disintegration of light elements in the container or in chemical combination with the active element. Since the fraction of atoms of RaC which disintegrate along the RaC'' branch is only 2.5×10^{-4}, the present measurement shows that the neutron yield is not more than 1 in 250 disintegrations of RaC''. If an excited state in RaD exists just above the neutron binding energy (which is assumed to be 4.81 Mev: the dotted line in Fig. 6), it would be excited in an allowed transition twenty times or twice per hundred disintegrations, according to whether the disintegration energy of RaC'' is 6.00 or 5.39 Mev. This calculated yield is much greater than that observed, and the low value of the latter indicates a low density of levels in RaD at 5 Mev with the spin and parity appropriate for an allowed β-transition from RaC''.

IX. INSTABILITY OF BISMUTH

The α-disintegration energy of Bi^{209} can be calculated from the neutron binding energies of Table III and certain disintegration energies. The α-disintegration energy is equal to the binding energy of the α-particle less the sum of the binding energies of the two neutrons and two protons which, when removed separately, create the product nucleus. The proton binding energy of Bi^{209} can be calculated from Harvey's value of the neutron binding energy of Pb^{209} and the β-disintegration energy of Pb^{209}. Assuming that the latter is 0.70 Mev, this proton binding energy is 3.79 Mev.[36] For the two neutron binding energies, the energies of the two lead γ-rays are used; finally, the proton binding energy of Pb^{206} is found to be 7.08 Mev. These figures lead to an α-disintegration energy of 3.25 Mev in Bi^{209}. In view of the discrepancy in the binding energies of four neutrons in Pb^{210}, discussed above, this result may be too high by 0.36 Mev.

The half-life of Bi^{209} can be calculated from this disintegration energy. The result obtained by using the Bethe formula,[37] corrected by a factor of a hundred to allow for the forbidden nature of α-emission from bismuth isotopes,[38] is about 10^{16} years. This value is of the same order as that obtained by Jenkner and Broda,[39] who, using a specially sensitive photographic method, have shown that the half-life must be greater than 3×10^{15} years. The calculated result, however, is a lower limit for the lifetime; if we use the lower value of the disintegration energy, *viz.*, 2.89 Mev, the half-life is of the order of 10^{18} years.

The range of the Bi^{209} α-particle should lie between 1.65 and 1.9 cm of air. This range is very close to the value equivalent to the radius of the unidentified K ring in pleochroic halos (1.74 cm of air).[40] It does not seem possible, however, to identify this halo with Bi^{209}, as has been suggested,[39] for the disintegration rate is probably insufficient to produce the effect.

The difference between the α-disintegration energy of Bi^{208} and that of Bi^{209} is equal to the difference in the neutron binding energies of Bi^{209} and Tl^{205}, and these are equal within the limits of experimental error. Therefore, the α-disintegration energy of Bi^{208} is equal to that of Bi^{209}. This energy is too low to identify Bi^{208} with the long lived α-emitter produced by neutron bombardment of bismuth which according to Neumann, Howland, and Perlman[7] has a disintegration energy of 5.12 Mev. The present results therefore confirm their conclusion that that body is an isomer of Bi^{210} and does not arise from the production of Bi^{208} by an $(n,2n)$ process.

X. SIMPLICITY OF THE LEAD AND BISMUTH γ-RAY SPECTRA

The simplicity of the γ-ray spectra produced by lead and bismuth is their most remarkable feature. It is quite unique among the heavy elements. In this respect, these nuclides resemble the lightest elements. The latter produce single homogeneous γ-rays because no excited states exist between the ground state and the neutron binding energy or because the probability of radiation to one of these states is greatly reduced by the small energy difference available. The latter explanation is probably sufficient to account for the simplicity of the γ-radiation produced by capture in bismuth. For the neutron binding energy is very low and, with the exception of the multiplicity of low-lying levels

[35] W. B. Lewis and B. V. Bowden, Proc. Roy. Soc. (London) **A145**, 235 (1934).

[36] It is significant that the energy required to remove the 83rd proton from Bi^{209} is so close to that required to remove the 127th neutron from Pb^{209}. This equality is also revealed by the close similarity of the disintegration energy of Pb^{209} with the energy equivalent of the mass difference between the neutron and the hydrogen atom.

[37] I. Perlman and T. J. Ypsilantis, Phys. Rev. **79**, 30 (1950).

[38] H. M. Neumann and I. Perlman, Phys. Rev. **78**, 191 (1950).

[39] K. Jenkner and E. Broda, Nature **164**, 412 (1949).

[40] G. H. Henderson and L. G. Turnbull, Proc. Roy. Soc. (London) **A145**, 582 (1934).

which have been revealed in the decay RaD, only three excited states are known to exist between the ground state of Bi^{210} and its neutron binding energy.[6] However, some different explanation is required for lead, because the energy levels available for excitation in Pb^{208} are more numerous. Clearly, the homogeneity of the lead radiations must be associated in some way with the closing of the nuclear shell at Pb^{208}. It will be shown below that the transitions which can arise between the neutron capturing state and the known excited states of Pb^{208} would produce, with one exception, γ-radiations of a high order of multipolarity; and this may be an explanation of the high probability of the direct transition to the ground state.

The system of levels in Pb^{208} which is excited in the decay of ThC″ is shown in Fig. 7(a). According to Martin and Richardson,[41] the spin of the 2.62-Mev level is 1, that of the higher levels 3 or 4, and all have the same parity as that of the ground state. Now a consideration of the magnetic moment of Pb^{207} suggests that its ground state has odd parity. If we take the parity of the closed shell nucleus, Pb^{208}, to be even, the direct transition to the ground state which follows the capture of a thermal neutron by Pb^{207} must produce a change of parity. It follows from the spin of Pb^{207} (1/2) that this γ-ray is of the electric dipole type. Likewise, direct transitions from the neutron capturing state of Pb^{208} to any of the levels of Fig. 7a must also produce a change of parity. Therefore, with the exception of the transition to the ground state or to the 2.62-Mev level, these transitions must produce γ-rays of the third multipole order or higher. In a previous paragraph we have shown that magnetic quadrupole radiation generally may not be a forbidden type. However, in the present instance, third-order radiations may well be forbidden by comparison with electric dipole radiation. It is very curious that there is no transition to the 2.62-Mev state, because there is no known selection rule which forbids this transition.

The assignment of spins to the excited states of Pb^{208} discussed by Martin and Richardson has been rendered doubtful by recent measurements of the angular correlation of the 2.62-Mev with the 0.58-Mev γ-rays.

[41] D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) **63**, 223 (1950).

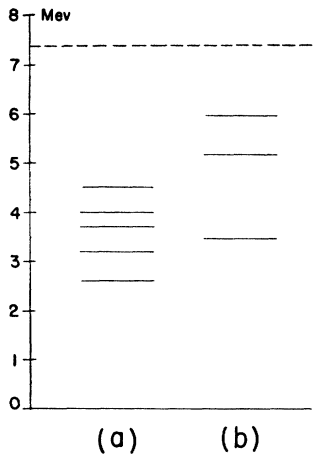

FIG. 7. The energy levels of Pb^{208}: (a) as found from the decay of ThC″ and (b) as revealed by the $Pb^{208}(d,p)Pb^{209}$ reaction.

These new results are compatible only with a spin of 2 units for the 2.62-Mev state and 4 units for the 3.2-Mev state.[42] If this is the correct assignment, there is still no explanation of the absence of the γ-ray leading to the 2.62-Mev state although the remaining γ-rays may be forbidden to the same degree.

A curious feature of the excitation of the Pb^{208} nucleus by the (d,p) reaction (Fig. 7b) is the absence of the excitation of the 2.62- and 3.20-Mev states. The first state to be excited in this reaction has an energy of about 3.5 Mev, but the accuracy of measurement is hardly sufficient to identify this level with any of those displayed in the decay of ThC″. Such a level, if it had a low spin, 0 or 1, would not be accessible to the β-decay of ThC″, for which the spin is at least 4 units. Now we have found a general correspondence between the excitation of nuclear levels by the (n,γ) and (d,p) reactions which would lead one to expect that a failure to excite the 2.62-Mev level in the (n,γ) reaction would have its counterpart in a similar failure to excite this state in the (d,p) reaction. On the other hand, the (d,p) reaction produces an excited state at 3.5 Mev in Pb^{208}; one would expect to find a γ-ray with this energy or with 3.9 Mev, the difference between it and the binding energy. These γ-rays have not been detected.

[42] We are indebted to Dr. M. Johns for the privilege of seeing his results before publication.