Alpha-Gamma Angular Correlation in Bi^{212}(ThC)†

JOHN W. HORTON*
Palmer Laboratory, Princeton, New Jersey
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The $\alpha-\gamma$ angular correlation function, $W(\theta)$, of the cascade

$$Bi^{212}(ThC) \xrightarrow{\alpha(6.04\text{ Mev})} Tl^{208*} \xrightarrow{\gamma(40\text{ kev})} Tl^{208}(ThC'')$$

was determined by experiment to be consistent with a predicted law of the form $W(\theta)=1+A\cos^2\theta$. The ratio $W(90°)/W(180°)$ was measured to be 1.2993, ± 0.0095 due to statistics, and ± 0.010 due to experimental corrections. Comparison of this result with predictions of $\alpha-\gamma$ angular correlation theory shows that Bi^{212} cannot have zero spin. Of fifty-four possibilities considered, it is shown that the following spin and parity assignments are most consistent with presently available experimental and theoretical evidence from several sources: the ground state of Bi^{212} is $1(-)$, the 40-kev state of Tl^{208} is $4(+)$, and the ground state of Tl^{208} is $5(+)$. These assignments determine the orbital momenta of α particles emitted in the decay of Bi^{212} to be a mixture of 3 and 5 units for the transition to the 40-kev state of Tl^{208} and to be 5 units for the transition to the ground state of Tl^{208}. Using these values of orbital momenta in the Gamow theory, and in the Weisskopf-Devaney theory of α fine-structure, the theoretical ratios of decay probabilities for the two groups, $\lambda(40\text{ kev})/\lambda(0,)$ are found to be in good agreement (10 to 50%) with the observed value. This agreement affords, in the case of Bi^{212}, an explanation for the prohibited decay to the ground state relative to the first excited state found for many non-even-even nuclei by Perlman, Ghiorso, and Seaborg.[1] Since the emission of the α particle with five units of orbital momentum hinders the decay $Bi^{212}\rightarrow Tl^{208}$ (ground state) by a factor of 16, this helps to explain the unusual size of the "departure factor" (1000) given to this decay by Perlman *et al.*

INTRODUCTION

PERLMAN, Ghiorso, and Seaborg[1] have shown that the partial half-lives for alpha decay of nuclei which are not of the even-even sort are generally longer than would be expected on the basis of a Gamow theory which holds well for even-even nuclei. They have shown further that the partial half-life of decay to the ground state of the daughter nucleus is usually much longer than the partial half-life for decay to the first excited state. To explain these two circumstances they suggested that the longer half-lives might be due, in part, to difficulties in forming the alpha particle when odd nucleons are present. Although such a mechanism may exist, it is important in specific cases to see whether the longer half-lives are not due to the shape and size of one or both of the nuclei involved in the alpha decay, or to the emission of the alpha particle with angular momentum.

The alpha decays of the nuclei $_{83}Bi^{212}$(ThC), $_{92}U^{235}$, and $_{95}Am^{241}$ exhibit to the most pronounced degree the two effects just mentioned. We have attempted to determine by the method of alpha-gamma angular correlation the angular momenta of the alpha particles emitted in the two most energetic alpha decays of bismuth-212 to determine to what extend the angular momenta can explain the long observed partial half-lives. Bismuth-212 is of particular interest because its decay scheme is well known (Fig. 1), and because the residual nucleus of the alpha decay, $_{81}Tl_{127}{}^{208}$, is expected to have a simple configuration in shell theory. A preliminary report of the results of this experiment was presented at the Cambridge meeting of the American Physical Society in 1953.[2] The first work on this problem was done by Kulchitskii.[3] His results differ significantly from those given here. Recently, Weale has published results on this problem[4] which are similar to the findings in the present experiment.

APPARATUS

In the present experiment, the detecting apparatus consisted of two end-window photomultiplier tubes (RCA 6199) situated inside an evacuated, steel drum. As is shown schematically in Fig. 2, the tubes were enclosed up to their photocathodes in a brass housing which permitted the tubes themselves and all electrical connections to them to be in air at atmospheric pressure. The large size of the drum and the lightweight construction of the tube housings made negligible the amount of gamma radiation scattered from these metal parts into the gamma-detecting crystal. The alpha counter was fixed in angle although its distance from the source could be varied so that apparatus tests could be made on decaying sources. The gamma counter was rotated in a horizontal plane by an arm which went through the center of the bottom of the drum. The source could be inserted into the vacuum through a $\frac{3}{4}$-in. ball-valve located at the center of the top of the drum. Both photomultiplier tubes employed as scintillators thallium-activated sodium iodide crystals. On the alpha counter, a $\frac{1}{2}$-in. square crystal was located $\frac{1}{8}$ in. away from the $1\frac{1}{4}$-in. diameter photocathode. A Lucite light-pipe of a shape shown in Fig. 2 transmitted light from the crystal to the face of the tube. Light-pipes longer than $\frac{1}{4}$ in. were found to scatter an excessive amount (4% for $\frac{1}{2}$ in.) of the 40-kev gamma rays into the gamma detector. Crystals of greater area $\frac{1}{8}$ in. distant

† Most of this work was done in partial fulfillment of the requirements for the Ph.D. degree at Princeton University.
* Now at the IBM Watson Laboratory at Columbia University, New York, New York.
[1] Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).
[2] J. Horton and R. Sherr, Phys. Rev. 90, 388(A) (1953).
[3] L. A. Kulchitskii, Doklady Akad. Nauk. S.S.S.R. 73, 1153 (1950).
[4] J. Weale, Proc. Phys. Soc. (London) 68, 35 (1955).

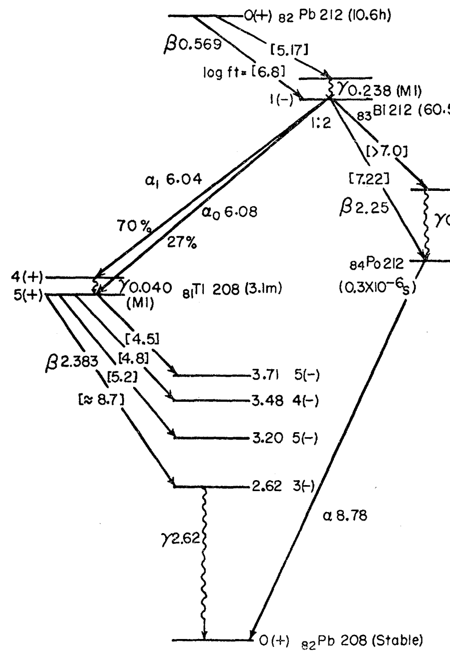

FIG. 1. Decay scheme of the thorium active deposit—$_{84}Po^{214}$(ThB), and the descendants shown above, in secular equilibrium, constituted the radioactive source of this experiment. Only levels pertinent to the experiment are shown. Energies are given in Mev. The spin-parity assignments in Pb^{208} are by Elliot, Graham, Walker, and Wolfson,[15] based upon the results of internal conversion and $\gamma-\gamma$ angular correlation measurements (these rays are not shown for simplicity); the remaining assignments are discussed in the text. The numbers in square brackets are log*ft*-values of beta transitions, computed from the results of work by Martin and Richardson,[9,10] and by Elliot *et al.*[15,19] Additional data were taken from *Nuclear Data*, National Bureau of Standards Circular No. 499 (U. S. Government Printing Office, Washington, D. C., 1950).

from the photocathode were found to give lower energy resolution because of the inhomogeneity of the photocathode. A crystal thickness of $\frac{1}{16}$ in. on the α counter was one-half the range of the most energetic beta particles (2.2 Mev), and permitted most of the gamma radiations with energies above 250 kev to pass through. On the gamma counter, a $\frac{11}{16}$ in.$\times\frac{11}{16}$ in. crystal with a

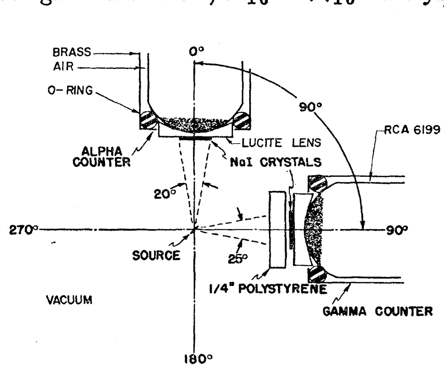

FIG. 2. Disposition of the counters—The tube mounts are situated near the center of an evacuated drum (not shown) which is 18 in. in diameter and 9 in. high. The source foil was attached to a light-weight Lucite frame and rod.

$\frac{1}{4}$ in. thick Lucite light-pipe gave an energy resolution expected for mounts of this sort as is indicated by the shape of the 78-kev γ ray in Fig. 4. This crystal was made $\frac{1}{32}$ in. thick so as to minimize counts due to gamma radiations more energetic than 40 kev; the 40-kev ray was completely absorbed. A $\frac{1}{4}$-in. piece of polystyrene was placed in front of the gamma crystal to stop alpha and beta particles. The thin crystal for the γ counter was prepared in a dry-box by rubbing the crystal on a glass flat covered with stretched silk. The flat was mounted just below the surface in a vessel containing butyl alcohol. The crystal was removed quickly from the alcohol with its surfaces clear and dry. The α-crystal, being of smaller area, could be cleaved to the desired size. It was found desirable for good resolution that all surfaces of this crystal be cleaved. The voltage pulses from the photomultipliers were shortened to 1 microsecond by means of delay lines, and then amplified to 30 volts. There were two output channels for each amplifier. The pulses in one channel were again amplified by a factor of three. All of these pulses with amplitudes greater than 3 v then triggered a blocking oscillator. The other output channel was fed into a differential pulse-height discriminator whose action was to pass through itself a pulse from the blocking oscillator whenever a pulse appeared at its input with an amplitude lying within specified limits. Pulses from the alpha and gamma differential discriminators were fed to scalers and also to a coincidence circuit. The pulses arriving at the coincidence circuit were triangularly shaped, 0.2 microsecond in duration and 20 volts in amplitude. Upon inserting delays between pulses initiated by 6-Mev alpha particles and 40-kev gamma rays (which follow within 10^{-10} sec), it was found that the coincidence rate fell to $\frac{1}{2}$ maximum in 0.04 microsecond.

SOURCE

Sources of ThC were prepared by electrostatically collecting its 11-hour parent ThB onto one side of strips of 0.0005-in. aluminum foils. The collection was carried out in a one-inch diameter hemispherical chamber of brass into which thoron was evolved through a grid forming the bottom of the chamber. The thoron came from a preparation consisting of radio-thorium in a hydrogel of ferric hydroxide.[5] The hydrogel was distributed in a thin layer upon the surface of a glass flat, and was situated 1 mm below the grid just mentioned. The emanating power of this preparation was about 100 percent. With a collecting voltage of 900 v, about one quarter of the ThB formed by the decay of the radiothorium could be collected in a 12-hour period onto the 2.4 mm$\times$7.2 mm source area generally employed. The purpose of the grid was to make the electrostatic field independent of the charging of the source material.

In the course of taking data it was discovered that

[5] O. Hahn, Ann. Chem. 440, 121 (1924).

ThC″ nuclei recoiling from the source were depositing on the front of the gamma counter when the source faced that counter. Since this effect disturbed the normalization of the data, the source was covered with a thin (0.17-mg/cm²) aluminum foil to prevent the escape of these recoils.

IDENTIFICATION OF THE RADIATIONS

The pulse-height spectrum for the alpha channel is shown in Fig. 3. The energies, relative intensity, and decay half-life of the peaks shown there identify them as being due to alpha particles of the decays ThC($_{83}Bi^{212}$) → ThC″($_{81}Tl^{208}$) and ThC′($_{84}Po^{212}$) → $_{82}Pb^{208}$. The 40-kev gamma peak could be seen only in the coincidence spectrum as shown in Fig. 4 because of the size of the 80-kev x-radiation arising in members of the thorium active deposit other than ThC″. The 80-kev peak was identified by comparison with the 72-kev x-radiation resulting from K capture in Tl^{204}.

PROCEDURE

For each run, the source was visually aligned in the chamber, and then oriented so that the normal to the source made either plus 45° or minus 45° with the direction of the alpha counter. The true coincident count divided by the total gamma count, C/N_γ, was determined at various angles. Division by the gamma count removed the effect of variations in the distance of the movable (gamma) counter from the source and the effect of the dependence of C upon time due to the radioactive decay of the source. After every few angles, a measurement was made at 180°, so that all measurements would be made relative to 180°, and this so within a period of about a half-hour. This was done to remove possible effects due to electronic instabilities, to counting losses in a decaying source, and to changes in the apparatus which would occur from source to source (crystal deterioration, position of the source). In all of the runs, the resolving time was around 1.7×10^{-7} sec and the ratio of true counts to accidentals increased from 2:1 to 5:1 as the source decayed. Although the initial counting rate at the output of the α-differential discriminator was 2000 per sec, no correction for losses was made because the dead time of the discriminator was 11×10^{-6} sec. A slight change in the pulse height of the 40-kev peak due to the rotation of the photomultiplier in the earth's magnetic field was compensated for by resetting the amount of pulse amplification.

RESULTS

Since the gamma transition is dipole, as will be shown below, the data should follow a law of the form $1+A\cos^2\theta$. To test the data against this law (see Fig. 5), we have drawn the line $W=1.30-0.30x$ ($x=\cos^2\theta$) through the normalizing value of 1 at $x=1$ and through the accurately determined point marked "A" at $x=0$. This latter point corresponds to the

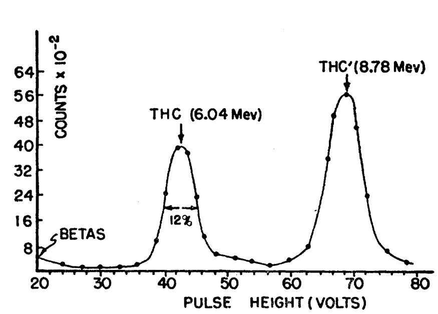

Fig. 3. Alpha-particle pulse-height spectrum of the thorium active deposit with sodium iodide scintillator. The shoulder on the high-energy side of the lower peak is due to the low-energy group of α particles.

average observed value of the anisotropy, that is, of $[(C/N_\gamma)_{90°}+(C/N_\gamma)_{270°}]/2(C/N_\gamma)_{180°}$ which was 1.298 ±0.0095. Of the data shown in Fig. 5, those referred to as "Series I" are early data taken when the instabilities were greater, and those referred to as "Series III" were taken under improved conditions. These data are meant to show that there is no significant departure from the straight lines described above.

Values of the anisotropy at 90° and 270°, i.e., of $(C/N_\gamma)_{90°}/(C/N_\gamma)_{180°}$ were found to agree with each other within ±1.5% statistics. The result of the test of symmetry about 90° is shown in Fig. 5 by the point at 50°.

This result disagrees with that of Kulchitskii[3] who found the correlation to have a maximum at 135°. This investigator used a proportional counter to detect the alpha particles and a Geiger counter to detect the gamma quanta. There was no electronic pulse-height discrimination. Weale's data,[4] taken with proportional counter and scintillator, are best fitted at several angles by the (corrected) correlation function $W(\theta)=1-(0.22$

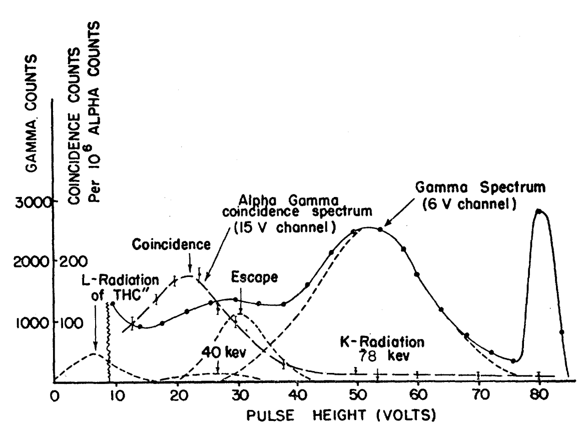

Fig. 4. Gamma-ray pulse-height spectrum of the thorium active deposit with a thin sodium iodide scintillator; and αγ-coincidence pulse-height spectrum. The 78-kev peak is K-radiation following internal conversion in elements other than Tl^{208}(ThC″). The dashed curves are based upon the tested resolution of the crystal scintillator at 50 kev and 80 kev. The curve of L-radiation is an estimated one. The peak at right is due to a pulse-height limiter. The crystal thickness was $\frac{1}{32}$ in. The 6.04-Mev alpha group (see Fig. 3) was used in obtaining the αγ-coincidence spectrum shown.

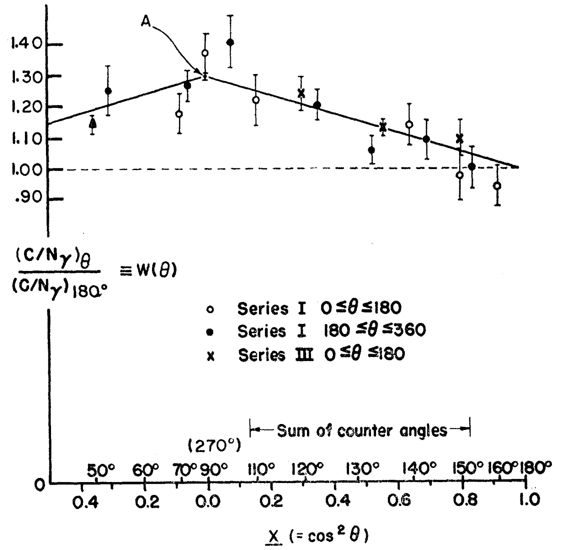

FIG. 5. Coincident counting rate *vs* θ, the angle between α and γ counters. Data from several series of runs, differing in minor respects of experimental conditions, are plotted against $x=\cos^2\theta$ to test a predicted correlation function of the form $W(\theta)=a+bx$. This function is shown as two straight lines passing through the normalizing points $W=1$ at $\theta=0°$ and $180°$, and through the accurately determined point at $\theta=90°$, marked A, $W(90°)=1.298 \pm 0.0095$. Corrections to the observed value of $W(90°)$ are described in the text.

$\pm 0.05) \cos^2\theta$, which is in agreement with the present result. Since both Weale and Kulchitskii used uncovered sources in gases at about the same pressure, the difference between Kulchitskii's results and those of Weale and the present experiment probably does not arise from the effect of recoil nuclei mentioned previously (see "Source" section).

CORRECTIONS

The mechanical alignment of the angular scale was checked by replacing the source with a small pellet of Na^{22}, and by studying the angular distribution of the annihilation radiation. Since it was found that all the angles had to be increased by 3.5°, a correction to the observed anisotropy of $+0.0026$ was required. A correction of $+0.0151$ was made for the effect of finite solid angles by integrating the function $1+A\cos^2\theta$ over square crystal areas.[6] A small, anisotropic, coincident background was observed after the 40-kev γ ray had been removed by absorbing foils. Although a number of tests were made, the origin of this effect is not entirely clear. A correction of $-0.0150(\pm 0.0048)$ was made for this. The effect of 40-kev gamma radiation scattered by the alpha counter housing into the gamma counter necessitated a negative correction ≤ 0.004. Since 10–12 kev L-radiation is emitted as an alternative in the de-excitation of the 40-kev state it was possible that some of this radiation contributed to the coincidence counting rate. This effect was investigated with 0.001-in. absorbing foils of Al and yielded a correction of $+0.0036 \pm 0.0071$. There was a small amount of absorption of 40-kev radiation in the source backing which necessitated a correction of -0.0019. Coincidence counts were sought due to α-particles striking the α crystal and producing a radiation which would be detected by the γ counter. Using Po^{210} as source, no coincidence counts were observed at any angle with the γ channel set as in the ThC experiment. At twice the amplifier gain setting coincidence counts were observed, however, and they were attributed to a radiation from the α crystal which could be stopped by 0.001 in. of Al foil. No correction had to be made for this effect.

The corrected value of the anisotropy, $W(90°)/W(180°)$, is 1.2993, ± 0.0095 due to statistics and ± 0.010 due to the above corrections. Weale's correlation function gives the anisotropy $W(90°)/W(180°) = 1.28 \pm 0.08$.

INTERPRETATION OF THE MEASUREMENT

(A) The gamma radiation is magnetic dipole. In Table I we give a number of internal conversion coefficients for the L-shell which have been calculated by Gellman *et al.*[7] These calculations have been verified experimentally by Mihelich and Church.[8] The entries on the next to the bottom line are the values measured by Surugue for the 40-kev ray of this experiment; the values have been normalized to the calculated value of the L_I subshell. We also give normalized values for the internal conversion of a 46.4-kev gamma ray following the beta decay of RaD as measured by Wu, Boehm, and Nagel. Wu believes that this latter gamma ray is pure magnetic dipole. It is seen that the 40-kev ray of this experiment is magnetic dipole, and further that an admixture of 1% electric quadrupole radiation would certainly be an upper limit. We conclude from the selection rules governing the emission of magnetic dipole radiation that the final two levels in Tl^{208} have the same parity and differ by 0, or 1 unit of spin.

(B) The spin of Bi^{212}(ThC) cannot be zero. The selection rule on alpha emission is that the alpha-particle orbital momentum quantum number, l, is even when there is no change in parity of the nuclear states connected by the alpha decay, and odd otherwise. The conservation of angular momentum in α decay requires that if the spin of the decaying nucleus is zero, that the alpha particle be emitted with $l=I$, where I is the spin of that level of the residual nucleus excited by the decay. Suppose that Bi^{212} has spin zero and that the 40 kev and ground states of Tl^{208} have spins I' and I'', respectively. Then the l values of the alpha groups to these two states would be I' and I'', respectively. If I' and I'' differ by one unit, the parity selection rule for alpha decay implies that the two final states differ in

[6] J. Horton, Princeton University thesis, 1953 (unpublished).

[7] Gellman, Griffith, and Stanley, Phys. Rev. 85, 944 (1952).
[8] J. Mihelich and E. Church, Phys. Rev. 85, 733 (1952).

parity. Since this is not so, in the present case, I' and I'' do not differ by one unit. The magnetic dipole character of the radiation requires that I' and I'' do not differ by two or more units; hence they cannot differ at all. Now the theoretical $\alpha-\gamma$ angular correlation functions for all cascade cases $0-I-I$ are lower at 90° than at 180°, so long as the admixture of electric quadrupole radiation is less than 10%, as is certainly the case here. Since the present experiment showed a higher correlation at 90° than at 180°, we conclude that the initial spin cannot be zero.

Martin and Richardson[9] conclude from a study of the shape of the beta spectrum of the transition between the ground states of $_{83}Bi^{212}$(ThC) and $_{84}Po^{212}$(ThC′), and from the assumption that the transition is first forbidden ($\log ft=7.2$) that the spin change is either 0 or ±1. Assuming that the ground state of $_{84}Po^{212}$(ThC′) has zero spin, the ground state of Bi^{212} has a spin of 0 or 1. Since the present experiment rules out the former, the spin of the ground state of Bi^{212} is 1.

But if the spin is 1, then the parity of this state is probably odd. Martin and Richardson's[10] measurement of the K-interval conversion coefficient of the 726 kev gamma ray in Po^{214}(ThC′), establishes it to be $E2$ and sets an upper limit of 7 β decays to the 726 kev level of Po^{212} per 100 disintegrations of Bi^{212}. This gives a $\log ft$ value >7.0 for this transition. Since the β decay of Bi^{208} to the ground state of Po^{212} has a $\log ft$ value of 7.2, first forbidden transitions and a change of parity are indicated according to Nordheim's classification.[11] Feather[12] has discussed this assignment and believes that only 0(−) or 1(−) are admissible values. Furthermore, the spin and parity of Bi^{210} which differs from $_{83}Bi^{212}$ only by a pair of neutrons is 1(−). Wu has discussed this assignment fully.[13] The odd parity of Bi^{210} is predicted by shell theory, and borne out by β decay evidence. The difficulty with the odd parity for Bi^{212} is the low ft value of the β decay to the 238-kev state of Bi^{212}, which has certainly the odd parity of the ground state because the 238-kev ray is very nearly pure $M1$.[14] It is assumed here that this decay is "favored" first forbidden (see the next paragraph). In this connection, it is worth noting that a "favored" decay occurs[13] to the first excited state of Bi^{210}.

(C) The spin and parity of $_{81}Tl^{208}$(ThC″)—Elliott, Graham, Walker, and Wolfson[15] have assigned spin and parity to the four excited states of Pb^{208} shown in Fig. 1. Using the intensities of the beta rays quoted by Richardson,[16] the $\log ft$ values in Fig. 1 were calculated.[17] These are all around 5 and hence indicate allowed transitions.[11] The evidence against this being so is strong since six β decays around $A=208$ are known[13,18] which are believed to be first forbidden and yet have $\log ft$ values in the range 5 to 6. A spin of Tl^{208} higher than 6 is certainly ruled out, however, in view of the intense β decay to the 4(−) state of Pb^{208}. A spin less than 4 would permit the β decay to the ground state of Pb^{208} to be observed,[12] whereas it is not found. Assignments of 6(+), 6(−), or 5(−) are also ruled out for the ground state of Tl^{208} because the β decay to the 2.62-Mev 3(−) state of Pb^{208} should then have, according to Nordheim's classification,[11] a $\log ft$ value in the range 13 to 18; whereas the $\log ft$ value is around 8.7 on the basis of the approximate intensity of this β ray (~0.1%) found by Elliott *et al.*[19] It seems unlikely on the same basis that Tl^{208} is 4(−) because this assignment would imply an allowed transition to the 3(−) state and a $\log ft$ value around 5. We conclude that possible values of spin and parity of the ground state of Tl^{208} are 4(+) or 5(+) on β-decay evidence, with 5(+) the more probable assignment.

TABLE I. A L-shell internal conversion coefficients (number of L-conversion electrons per quanta observed).

Type of γ-radiation	L_I	L_{II}	L_{III}	L shell
	Calculated values (40.0 kev)[a]			
$E1$	0.29	0.26	0.35	0.91
$E2$	5.3	195.	196.	397.
$M1$	27.8	2.4	0.030	30.3
	Observed values (normalized to 30 for L_I shell)			
ThC(40.0 kev)[b]	30(±3)	3(±0.6)	0.5(±0.25)	
RaE(46.4 kev)[c]	30	2.2	0.42	

[a] Gellman, Griffith, and Stanley, Phys. Rev. 85, 944 (1952).
[b] J. Surugue, Ann. Phys. 8, 485 (1937).
[c] Wu, Boehm, and Nagel, Phys. Rev. 90, 388(A) (1953).

COMPARISON WITH THEORY

It has been argued that the initial state, Bi^{212}(ThC) is 1(−), and that the final state in the $\alpha-\gamma$ cascade, Tl^{208}(ThC″), is 4(+) or 5(+). Theoretical values of the anisotropy are given in Table II for an initial spin I_0 of 1 or 2, and for final spin values, I_f, ranging from 3 to 6. The intermediate spin, I_i, can have the values I_f-1, I_f, or I_f+1. The α particle is assumed to be emitted with a single value of orbital momentum l_α; the γ ray is assumed to be pure dipole. Note that odd (even) values of l_α imply in the present cascade a (no) nuclear parity change between the initial state (Bi^{212}) and the two states of Tl^{208}. Under these assumptions, values of the anisotropy can be calculated mainly from

[9] D. G. E. Martin and H. O. W. Richardson, Proc. Roy. Soc. (London) 195, 287 (1949).
[10] D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) 63A, 223 (1950).
[11] L. Nordheim, Revs. Modern Phys. 23, 372 (1950).
[12] N. Feather, in *Beta and Gamma Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 746.
[13] C. Wu, reference 12, p. 343.
[14] D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) 63, 223 (1950).
[15] Elliott, Graham, Walker, and Wolfson, Phys. Rev. 93, 356 (1954); 94, 795(A) (1954).
[16] H. Richardson, Nature 161, 516 (1948).
[17] E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).
[18] E. Konopinski, Ann. Rev. Nuc. Sci. 2, 293 (1953).
[19] J. C. D. Milton (private communication).

TABLE II. Theoretical values of the $\alpha-\gamma$ angular correlation function anisotropy, $W(90°)/W(180°)$, for initial spin 0, 1, or 2 and dipole γ ray. By "case" is meant the $\alpha-\gamma$ cascade $I_0 \xrightarrow{l_\alpha} I_i \xrightarrow{l_\gamma=1} I_f$ in which a nucleus of spin I_0 emits an α particle with a single value of orbital momentum l_α, and the residual nucleus with spin I_i decays to a final state of spin I_f by emitting a dipole gamma ray.

Case $\frac{l_\alpha}{(I_0\rightarrow I_i\rightarrow I_f)}$	$\frac{W(90°)}{W(180°)}$	Case	$\frac{W(90°)}{W(180°)}$	Case	$\frac{W(90°)}{W(180°)}$
$1-{}^{1}2-3$	1.167	$1-{}^{3}2-3$	1.098	$1-{}^{2}2-3$	1.116
$1-{}^{3}4-3$	1.732				
$1-{}^{2}3-4$	1.250	$1-{}^{4}3-4$	1.262	$1-{}^{3}3-4$	1.215
$1-{}^{4}5-4$	1.680				
$1-{}^{3}4-5$	1.300	$1-{}^{5}4-5$	1.307	$1-{}^{4}4-5$	1.274
$1-{}^{5}6-5$	1.665				
$1-{}^{4}5-6$	1.335	$1-{}^{6}5-6$	1.344	$1-{}^{5}5-6$	1.313
$2-{}^{1}2-3$	<1				
$2-{}^{2}4-3$	1.588				
$2-{}^{1}3-4$	1.167	$2-{}^{3}3-4$	1.081	$2-{}^{2}3-4$	1.056
$2-{}^{3}5-4$	1.613				
$2-{}^{2}4-5$	1.250	$2-{}^{4}4-5$	1.172	$2-{}^{3}4-5$	1.165
$2-{}^{4}6-5$	1.610				
$2-{}^{3}5-6$	1.302				
$0-{}^{l}I-I$	<1 for $l=I$, $I>1$				
$1-{}^{l}I-I$	<1 for $l=I-1$, $I=3, 4, 5, 6$				
$2-{}^{l}I-I$	<1 for $l=I-2$, $I=3, 4, 5, 6$				

Lloyd's tables,[20] and from Thirion's formula[21] in the few cases where Lloyd's tables cannot be used. The Clebsch-Gordon coefficients in Thirion's formula were evaluated by a form due to Van der Waerden.[22] All three possible values of l_α are considered for $I_0=1$, *viz.*, $l_\alpha=I_i-1$, I_i, and I_i+1 to illustrate the effect of mixing (see the next section). Taking the corrected experimental value of the anisotropy as $1.299\pm0.042(0.042=3[(0.0095)^2+(0.010)^2]^{\frac{1}{2}})$, it is seen immediately from Table II that independent of the parity changes involved, $I_i=I_f-1$, and that $I_f\neq3$. If $I_0=2$, also $I_f\neq3$ or 4. Considering, however, the previous possibilities for initial and final spins, and parities, it is seen that $I_f\neq4$ if $I_0=1$. Hence, the only cascade not ruled out on the basis of either β-decay or angular correlation evidence is the cascade $(1-)-(4+)-(5+)$. This conclusion must now be qualified (but not altered) by a discussion of certain effects which are disregarded in the theoretical values of anisotropy given in Table II.

Alpha Mixing

If the spin of the initial state is 1, then the wave function of the emitted alpha particle may be a mixture of at most two states of orbital momenta. If the initial spin is 1 and the intermediate spin is, say, 3, the alpha particle may carry off 2, 3, or 4 units of angular momentum; however, only the 2 and 4 states mix because their wave functions have the same parity. With mixing, it is possible for the anisotropy to be larger than either of the values for the pure states. This comes about because of a term in the correlation function which depends upon the difference in the phase angles of the α-particle wave functions at large distances from the residual nucleus.[6,23,24] Calculations for the mixed $1-3-4$ case show that the anisotropy increases from 1.250 to a maximum value of 1.300, for instance.[23] This effect is not large enough here to be considered further.

Reorientation

Two effects which tend to destroy our knowledge of the orientation of the spin axis of the residual nucleus are precession in the magnetic field of the unpaired electron in Bi^{212} and in Tl^{208}, and the disturbances of the recoil process. The theory of Alder[25] can be applied to estimate the effect upon the anisotropy as the nuclear spin precesses in the field of the single unpaired electron of the thallium atom. Taking the nuclear spin in the intermediate state as 4, an electronic angular momentum of $\frac{1}{2}$, and a half-life of the intermediate state of 10^{-10} sec,[25] the theory of Alder predicts that the ratio $W(90°)/W(180°)$ would be reduced by less than 3.5%. During the recoil of the nucleus into the backing material, it may be supposed that the nuclear spin experiences torques of various directions and magnitudes which are associated with the collisions the recoiling atom makes with (aluminum) atoms of the backing. After the emission of the alpha particle, if the nucleus is left in a quantum state having magnetic quantum number m_0, the effect of the torques acting upon the nucleus is to induce transitions out of the m_0 state to states of different values of m. Assume for simplicity that only those transitions occur for which $\Delta m=\pm1$, and that the transition probability, p, is independent of m_0. Since torques occur randomly in time and magnitude during the recoil, suppose that coherence affects between the states m_0 and $m_0\pm1$ can be ignored. Calculations of the anisotropy according to this model with $p=0.5$ show that the anisotropy of the case $1-{}^{3}4-{}^{1}5$ $(l_\alpha=3, l_\gamma=1)$ is reduced to 1.273, as compared with 1.300 if $p=0$, and that in the case $1-{}^{5}6-{}^{1}5$ the anisotropy is 1.626, as compared with 1.665 if $p=0$. The value of $p=0.5$ is surely too large; for the observed $\alpha-\gamma$ correlation function in the decay[27] of Th^{228} can be fitted on the basis of this model by taking $p=0.2$, although for this case, transitions $\Delta m=\pm2$ must be introduced. Since the intermediate state for Th^{228} decay has a longer lifetime than that of the Bi^{212} decay by several orders of magnitude and since the latter state certainly has a smaller quadrupole moment, we conclude that even $p=0.2$ is too high a value to be

[20] S. Lloyd, Phys. Rev. **83**, 716 (1951).
[21] J. Thirion, Compt. rend. **230**, 2090 (1950).
[22] Van der Waerden, *Gruppentheoretische Methode in der Quantenmechanik* (Verlag Julius Springer, Berlin, 1932), p. 69.
[23] J. Beling, Massachusetts Institute of Technology, Ph.D. thesis, 1951 (unpublished).
[24] H. Frauenfelder, in *Beta and Gamma Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 531.
[25] K. Alder, Helv. Phys. Acta **25**, 235 (1951).
[26] R. Graham and R. Bell, Can. J. Phys. **31**, 377 (1953).
[27] Beling, Feld, and Halpern, Phys. Rev. **84**, 155 (1951).

used in the calculations concerning Bi^{212}. Although these two arguments are admittedly approximate they show that the reduction of anisotropy is small because of the large angular momentum of the intermediate state and because the anisotropy of cases such as $1-4-5$ is peculiarly insensitive to even moderately large changes in the population of the intermediate state.

It is concluded that the anisotropy calculated by ignoring reorientation processes would be lower by 3 or 4% at most if these processes were considered.

Shape of the Nucleus

It will be inferred in the next section that Bi^{212} and Tl^{208} have intrinsic quadrupole moments $|Q| \leq 1\times 10^{-24}$ cm^2. Obtaining the eccentricity, η, from Blatt and Weisskopf,[28] using this in formulas given by Hill and Wheeler[29] for the variation with direction of the Gamow barrier penetrability factor for α-particle emission, the following values of penetrability relative to unity for a sphere are found for emission in the direction of the two symmetry axes: pancake 1.09 (through edge) and 0.85; cigar 1.18 (tip) and 0.92. This effect, it is estimated, would change the anisotropies of Table II by less than $\frac{1}{2}$%.

Gamma Mixing

If there is a small amount of electric quadrupole radiation mixing with the dipole radiation, then the anisotropy can be appreciably different from the pure dipole case. The theory of this effect as given by Lloyd[20] involves the square of the mixing ratio δ^2, defined as (intensity electric quadrupole)/(intensity magnetic dipole), and the sign of δ which may be either positive or negative. The curves of anisotropy, $W(90°)/W(180°)$, *vs* δ^2 given in Fig. 6 were calculated from Lloyd's formulas for an initial spin of 1 and several different cases of intermediate and final spins. Only lowest values of l_α were used since the effect of α-mixing is of much less importance. Both positive and negative values of δ were considered. The Lloyd theory is approximate, since terms in δ^2 are neglected. An exact calculation of these terms for the $1-{}^23-{}^14$ case using formulas based upon those of Ling and Falkoff[30] shows that for $\delta^2 \leq 10^{-2}$, the values given in Fig. 6 are in error by not more than 2%. From a comparison of the theoretical and observed values of the $L_{\rm I}:L_{\rm II}:L_{\rm III}$ internal conversion ratios given in Table I, an upper limit of 1% can be set upon the amount of $E2$ mixing. This is sufficient to rule out all cases in which the first two spins are equal (this fact was used in the arguments which showed that the spin of ThC is not zero), since the mixing would have to be as high as 10% before these cases would be isotropic. Since it is only known that $\delta^2 \leq 10^{-2}$, it is

[28] J. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 27.

[29] D. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953). The author is indebted to Professor Wheeler for advice concerning the calculation of the penetrability ratios given here.

[30] D. Falkoff and D. Ling, Phys. Rev. **76**, 1639 (1949).

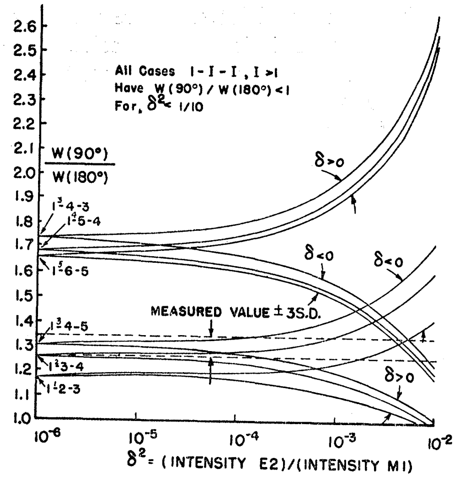

FIG. 6. Dependence of the anisotropy $W(90°)/W(180°)$ upon the extent of mixing of electric quadrupole and magnetic dipole radiations in the de-excitation of the intermediate state of an $\alpha-\gamma$ cascade. Various cascades are specified in Fig. 6 by the sequence of nuclear spins involved. An initial spin of 1 is assumed and the α particle is emitted with orbital momentum, l, equal to the difference in initial and intermediate spins. The number between the first two values of nuclear spins is the value of l for α emission to the intermediate state. The dashed lines indicate the corrected experimental value of the anisotropy ± three times the square root of the sum of the squares of the standard deviation of the counting statistics and the uncertainty of the applied corrections. These values are 1.299±0.042. From experiment $\delta^2 < 10^{-2}$. Taking the theoretical value $\delta^2 < 5\times 10^{-5}$, only cases $1-4-5$ and $1-3-4$ are consistent with the measured value.

seen from Fig. 6 that all the cases shown there are possible ones.

On theoretical grounds, however, it is to be expected that δ^2 is much smaller than 10^{-2}. On the basis of the unified nuclear shell model, Bohr and Mottelson[31] have calculated the intrinsic quadrupole moment Q_0 of ${}_{68}Er^{166}$ to be 10×10^{-24} cm^2. The observed transition probability $T_{E2}(\log T_{E2} = 7.91)$ of $E2$ radiation from the first excited 2+ state to the ground 0+ state was used, and collective excitations were assumed. The quadrupole moment and spin of ${}_{83}Bi^{209}$ are known to be -0.4×10^{-24} cm^2 and 9/2, respectively. Using these two values and the projection factor P_Q given by Bohr and Mottelson, $|Q_0| = |Q|/P_Q \approx 0.6\times 10^{-24} < 1\times 10^{-24}$ cm^2, independently of the strength of coupling of the $h_{9/2}$ proton to the 82–126 core (reference 31, Fig. 10). This value agrees with estimates of $Q_0 = 2\times 10^{-24}$ cm^2 for ${}_{84}Po^{212}$ and ${}_{84}Po^{214}$ (reference 31, p. 116). The intrinsic quadrupole moment of ${}_{81}Tl^{208}$ is not greater than this because the $g_{9/2}$ neutron of Tl^{208} would not be expected to distort the 82–126 core any more than the $h_{9/2}$ proton of Bi^{209}. Taking the value of the intrinsic quadrupole moment of Tl^{208} to be $< 1\times 10^{-24}$ cm^2, and extrapolating the erbium data from 80 kev to 40 kev [Eqs. VII (17) and (18)], we find that the transition probability of 40-kev quadrupole radiation is $T_{E2} < (1/2^5)(1/10)^2 \times (0.8)10^8$ $sec^{-1} = 2.4\times 10^4$ sec^{-1}. All other $E2$ transi-

[31] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

tions listed by Bohr and Mottelson are consistent with this value as an upper limit. Because collective excitations have been assumed in this estimate, this value is a safe limit by several orders of magnitude for the 40-kev state of Tl^{208}.

The mean life of the 40-kev state has been measured by Graham and Bell[26] to be $<7\times10^{-11}$ sec. Weale[4] has measured the L-shell internal conversion coefficient as 15.7 ± 1.6 (electrons per quanta observed), and a measurement of 21 ± 7 in the present experiment is consistent with this. The theoretical value is 30.3 (see Table I). To obtain the total internal conversion of the 40-kev ray, the value of 15.7 must be doubled to account for the conversion in the M, N, and O shells (see reference b of Table I). The transition probability T_{M1} for the magnetic dipole radiation from the 40-kev state is thus $>5\times10^{8}$ sec^{-1}. Hence the mixing ratio $\delta^2=T_{E2}/T_{M1}<5\times10^{-5}$. Reference to Fig. 6 shows that this value, taken with the measured value of the anisotropy, excludes all $\alpha-\gamma$ cascades except the cases $1-3-4$, and $1-4-5$. Using the β-decay evidence to decide the parities of initial and final states, and to eliminate $4(-)$ and $5(-)$ for the final state, these cases become $(1-)-(3+)-(4+)$, and $(1-)-(4+)-(5+)$. The first case $(1-)-^3(3+)-(4+)$ can now be ruled out for the following reason. No α-mixing is possible in this case; hence for $\delta^2=0$, $W(90°)/W(180°)=1.215$, uniquely. A curve drawn through 1.215 in Fig. 6 parallel to the curve $\delta<0$ for the $1-4-5$ case falls outside of the measured limits. Reorientation would increase this difference. The remaining case is $(1-)-(4+)-(5+)$.

DISCUSSION

From the evidence given above, the $\alpha-\gamma$ cascade

$$_{83}Bi^{212} \xrightarrow{\alpha(6.04)} {}_{81}Tl^{208*} \xrightarrow{\gamma(0.040)} {}_{81}Tl^{208}$$

has one of the following sets of spin-parity assignments: $(1-)-(4+)-(5+)$, or possibly $(1-)-(3+)-(4+)$. This result may be compared with shell theory and with the theory of α fine-structure. Pryce[32] has calculated the spins and parities of the first six states of $_{81}Tl_{127}{}^{208}$ on the basis of the j-j coupling model. Assuming a configuration $(s_{1/2})_P{}^{-1}(g_{9/2})_N{}^1$ for the ground state of Tl^{208}, the ground state is found to be $5(+)$ and the 40-kev state $4(+)$, in agreement with our $(1-)-(4+)-(5+)$ case. Pryce's calculations for $_{83}Bi_{127}{}^{210}$, which should pertain to $_{83}Bi_{129}{}^{212}$, indicate a low spin of 0, 1, or 2. If the spin is 0, or 1, an odd parity is indicated. This is in agreement with the assignment of $1(-)$ made here to Bi^{212}.

The Gamow theory of α fine-structure[33] predicts for spherical nuclei a decreasing emission probability with increasing orbital momentum, l_α, of the emitted α particle. Because $_{83}Bi^{212}$ and $_{81}Tl^{208}$ are very nearly spherical, for the reasons given above, and because of the large values of l (≥3) found in the decay of $_{83}Bi^{212}$, this theory may be accurately tested. For this purpose, we use the following formula for the decay probability, λ_l, given by Weisskopf[34] and Devaney[35]

$$\lambda_l=\frac{D}{2\pi}\cdot T_l(\alpha)=\frac{D}{2\pi}\cdot\frac{4(k/K)G_l{}^{-2}}{1+(k/K)^2(G_l'/G_l)^2}. \quad (1)$$

D is the level spacing of the daughter nucleus, G_l and G_l' are respectively the values at the nuclear radius, R, of the irregular Coulomb wave function and of its derivative with respect to kr, k is the observed α-particle wave number (corrected for recoil), and K is the wave number of the α particle in the nucleus [which is assumed[34] to have an energy of $\sim$5 Mev when Eq. (1) is used]. This formula is essentially the same as that given by Gamow except that the expressions given by Eq. (1) for formation factor and barrier penetrability are the result of a more detailed treatment of the α-decay process.

If Tyson's[36] exact formulas for G_l and G_l' are used in (1) with $D=40$ kev, $R=8.52\times10^{-13}$ cm, $l=3$, and $E_{40\text{ kev}}=6.163$ Mev[37] (after correction for recoil), we obtain the decay constant for emission to the 40-kev state: $\lambda(40\text{ kev})_{\text{theor}}=2.94\times10^{-5}$ sec^{-1}. The observed value is $\lambda(40\text{ kev})_{\text{exp}}=4.49\times10^{-5}$ sec^{-1}. This agreement is good enough to warrant a calculation of the ratio $\lambda(40\text{ kev})/\lambda(0\text{ kev})$ with $R=8.52\times10^{-13}$ cm. For $E_{0\text{ kev}}=6.203$ Mev (corrected for recoil) we obtain the values of λ_l/λ_0 given in Table III, again using exact formulas for the G_l in (1). Tyson's formulas for G_l and the numerical results of Table III were checked numerically by putting G_0 and G_0' into recursion formulas satisfied by Coulomb wave functions[38] and developing successive values of G_l and G_l' up to $l=5$. The agreement was 2% at $l=5$. For $l=5$, Gamow[34] gives $\lambda_5/\lambda_0=0.0814$. Winslow,[39] using wave functions for which an accuracy of a few percent is claimed and a Gamow-type theory gives $\lambda_5/\lambda_0=0.081$. Using Tyson's exact formula for λ_4, it was calculated that the value of λ_4 for decay to the 40-kev state is reduced because of the lower disintegration energy by a factor 1.54. The WKB approximation developed by Gamow gives 1.51. Applying the energy correction 1.54 to the values in Table III, one obtains in Table IV ratios of decay probabilities to the 40-kev and 0-kev states, $\lambda(40)/\lambda(0)$, for all spin and

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

[33] G. Gamow and C. Critchfield, *Atomic Nucleus and Nuclear Energy Sources* (Clarendon Press, Oxford, 1949), p. 174.

[34] J. Blatt and V. Weisskopf, see reference 28, Chap. XI.

[35] J. Devaney, Phys. Rev. **91**, 587 (1953).

[36] J. Tyson, Massachusetts Institute of Technology thesis, 1948 (unpublished); Tyson's penetrability formulas are given also in a table of Coulomb wave functions by Feshbach, Shapiro, and Weisskopf, Nuclear Development Associates, Report NDA-15B-5; U. S. Atomic Energy Commission Report NYO-3077 (unpublished).

[37] G. H. Briggs, Revs. Modern Phys. **26**, 1 (1954).

[38] L. Infeld, Phys. Rev. **72**, 1125 (1947).

[39] G. Winslow, Argonne National Laboratory Report ANL-5381, January, 1955 (unpublished), p. 33 ff.

TABLE III. Ratios of α-decay probabilities λ_l/λ_0 for α particles of disintegration energy $E_\alpha=6.203$ Mev, and residual nucleus having $Z=81$ and $R=8.52\times10^{-13}$ cm.

$l=$	0	1	2	3	4	5	6	7	8
λ_l/λ_0	1.000	0.833	0.579	0.336	0.163	0.0669	0.0251	0.0078	0.0021

parity combinations which have not yet been ruled out definitely, and for some combinations which probably have been eliminated. It is seen from Table IV that theoretical values are either too high by at most 50% or too low by a factor 4 to 5. The assumption about mixing is seen to be relatively unimportant. For the even-even nuclei, the Gamow theory predicts a ratio $\lambda_2(E)/\lambda_0(0)$ which agrees with experiment to within a factor of two in the range $A=208$ to 230.[40–42] We may perhaps infer from this that the theory in this case is not in error by as much as a factor of 4 to 5 and consequently rule out the cases without an asterisk in Table IV. It is seen from Table IV that initial spin 2 is not ruled out by α-decay theory; but it is ruled out by β-decay evidence above. Of the cases (1−)−(4+)−(5+), and possibly (1−)−(3+)−(4+) listed at the beginning of this section as not being ruled out by β-decay evidence, Table IV does not rule out the case (1−)−(4+)−(5+), and this case is in good agreement with the α-decay theory. Gamow's formula[33] for λ_l gives a ratio $\lambda_3(40)/\lambda_5(0)$ which is 1.14 higher than observed. Predictions of Preston's theory can be taken from a graph calculated by Winslow[42] since the parameters assumed there are approximately the same as the present ones. The graph gives a value of $\lambda_3(40)/\lambda_5(0)$ which is 0.7 below that observed. An energy correction of 1.54 was used in both cases.

On the basis of the single remaining case (1−)−(4+)−(5+), the l-values to the 40-kev and 0-kev states of Tl^{208} are, respectively, 3 and 5, if no mixing is assumed. For these values of l, the decay probabilities are, as shown in Table III, reduced below that for $l=0$ by the factors 3 and 16, respectively. These factors account, in the decay of Bi^{212} at least, for the prohibited decay of the ground state relative to the first excited state found by Perlman *et al.*[1] for many non-even-even nuclei, but only explain part of the large "departure factors" (350 for the 40-kev state and 1000 for the ground state) observed for Bi^{212}.

[40] P. Falk-Variant and J. Teillac, Compt. rend. **256**, 914 (1953).
[41] S. Rosenblum and M. Valadares, Compt. rend. **234**, 2359 (1952).
[42] G. Winslow, Argonne National Laboratory Report ANL-5277 (unpublished), p. 37.

TABLE IV. Ratios $[\lambda(40)/\lambda(0)]_{theor}/[\lambda(40)/\lambda(0)]_{exp}$ for certain values of spin and parity in the αγ cascade

$$Bi^{212} \xrightarrow{\alpha} Tl^{208*} \xrightarrow{\gamma(40\text{ kev})} Tl^{208}.$$

Possible orbital momenta l of α particles to the 40-kev and 0-kev state of Tl^{208} are given. Ratios are calculated for mixing of l-values and for lowest l-value (no-mixing). $\Delta\pi$ "yes" ("no") means that a (no) parity change is assumed between Bi^{212} and Tl^{208*} and between Bi^{212} and Tl^{208}. $[\lambda(40)/\lambda(0)]_{exp}=2.572$.[a]

Case ($I_0-I_i-I_f$)	$\Delta\pi$	l_{40}	l_0	Mixing	No-mixing[b] (lowest l)
1−3−4	yes	3	3,5	0.21	0.25
1−4−5	yes	3,5	5	1.52	1.27*
1−3−4	no	2,4	4	1.15	0.90*
1−4−5	no	4	4,6	0.22	0.25
2−3−4	yes	1,3,5	3,5	0.78	0.63*
2−4−5	yes	3,5	3,5,7	0.25	0.25
2−3−4	no	2,4	2,4,6	0.24	0.25
2−4−5	no	2,4,6	4,6	1.03	0.90*

[a] A. Rytz, Compt. rend. **233**, 790 (1951).
[b] Cases not marked with an asterisk are ruled out by α-decay theory, as discussed in the text.

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